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Heavy Metals in Sediments of the Vasyugan River Basin (Russian Federation), Chemical Composition and Environmental Risk

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Abstract: The study aims to analyze the state of bottom sediments of the Vasyugan river. The river's pollution because of oil production is considered. Samples have been taken during 2018. To determine the concentration of microelements, semi-quantitative spectral analysis, atomic absorption, and inversion voltammetric analysis methods have been used. The concentration of petroleum products has been determined by infrared spectrophotometry. Sediments of metals have been found in Vasyugan, mainly iron, aluminum, silver, magnesium, calcium and barium. On average, it is 0.5-2.0 times higher concentration compared to tributaries ($p \leq 0.05$). River basin Vasyugan is characterized by a moderate level of pollution resulting from oil production. The horizontal (surface water) and vertical migration of heavy metals and oil products (bottom sediments in rivers, peat in marshy areas) have been revealed. In total, 23 chemical elements are found, with a predominance in the Vasyugan riverbed and its tributaries.

Key words: Vasyugan, oil production, heavy metals, pollution, surface water, bottom sediments.

Introduction

The modern oil industry is characterized by a constant increase in the rate of hydrocarbon production (Bai et al., 2011, 2012). The negative consequences are increased pollution of ecosystems, including rivers by oil products (Dou et al., 2013; Pomortsev, 2019). The relationship of surface runoff and bottom sediments of rivers and lakes is a proven fact (Yu et al., 2011; Ergenekon and Ulutaş, 2014). Given the frequency of environmental pollution by oil and oil products, as well as various emergency situations, it becomes an extremely urgent issue to study the problems of river pollution (Ferreira-Baptista and De Miguel, 2005; Fu et al., 2012).

Rivers are a convenient model object, in the basins of which there are oil enterprises. As it is known, two processes can occur during river pollution – self-cleaning of rivers, as well as their secondary pollution (Gilham et al., 2008; Gan et al., 2010). In order to predict the use and protection of water bodies, it is necessary to identify mechanisms of anthropogenic transformation of river waters and bottom sediments (Yuan et al., 2008; Guan et al., 2017).

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The authors have selected the Vasyugan River as a model object. This river has a length of 1082 km. Vasyugan is one of the largest tributaries of the Ob River, which is the central waterway of Western Siberia. Vasyugan has a significant drainage basin – about 62,000 km^2. The river originates in Vasyugan swamps, whose area is the largest among the swamps in the whole world. The entire basin of the Vasyugan River is included in the taiga zone. Within the river basin, there are about 50,000 lakes. The entire system of reservoirs has an extremely branched and complex structure, combined into a single river. Therefore, any massive impact will inevitably be displayed not only on the river, but also on its entire system as a whole. Considering the fact that Vasyugan flows into the Ob at a distance of more than two thousand km from its mouth, the effect of pollution will spread to the Ob, and then to the ocean. Thus, by studying the effect of pollution on the tributary (Vasyugan), it is possible to predict the long-term effect on larger objects (Ob, the Arctic Ocean). Intensive oil production is carried out in the study area, which determines a high level of pollution, including in emergency situations (breakthrough of the oil pipeline).

Considering the enormous area of bogs (about 5.3 million ha) in the Vasyugan basin, a significant increase in the thickness of the peat layer is observed, from 1 to 1.5 mm per year. In the bulk, it is 0.05 km^3 of peat per year. With an increase in the volume of peat deposits, the water accumulated in the swamp increases to 0.045 km^3 per year. Such indicators, undoubtedly, impact the climate of the region. Pollutants accumulate in the swamps, from which it is practically impossible to extract them. The latter determines, ultimately, the long-term effects of pollution (Han, 2009; Guan et al., 2017).

Bottom sediments are an integral component of aquatic ecosystems (Beijing et al., 2011; Yuan et al., 2014). In the bottom sediments, there is a cycle of chemical compounds. Sediments are the environment in which various organisms live (algae, higher vascular plants, protozoa, mollusks, crustaceans, insects, vertebrates) (Hu et al., 2013, 2016).

The formation of bottom sediments is associated with a group of processes that occur in the drainage basin, mainly erosion (Ermakov et al., 2017; Hu et al., 2017). In addition, processes occurring directly in water bodies influence the bottom sediments, namely, the mechanical processing of coasts by humans, as well as the activities of organisms (Yuswir et al., 2013; Hu et al., 2017). The nature of bottom sediments is also determined by the regime of the reservoir – in a moving body of water (river) it will be more dynamic compared to reservoirs that hold water (lakes, swamps) (Hua et al., 2012, Zaccone et al., 2010). At the same time, in reservoirs that hold water, the accumulative effect for bottom sediments is more pronounced (Kong, 2014). Thus, if one is aware of the state of bottom sediments, it is possible to obtain information and a forecast on changes in aquatic ecosystems and the environment (Zhao et al., 2010; Lai et al., 2013).

The topic of the impact of pollutants (heavy metals and related compounds) on the ecological state of rivers (e.g. the Vasyugan River) is of relevance nowadays. However, there are issues that are not fully clarified. Namely, vertical and horizontal movement of heavy metals and other pollutants in various environments – water, bottom sediments and in peat layers.

The purposes of this article are to study the state of bottom sediments of the Vasyugan river, the composition of heavy metals and related compounds that are contained in these sediments, as well as to predict environmental risk.

Materials and Methods

Materials

Samples have been taken during 2018. The Vasyugan river basin and its tributaries, the Nyurolka and Parabel rivers (Figure 1) have been taken as the research region. Samples have been taken in these water bodies, 200 samples in each Vasyugan's tributary and 500 in Vasyugan itself. A total of 1100 samples have been taken, of which 100 in the upper reaches of Vasyugan, in the middle and lower reaches of this river – 200 each.

Samples have been taken in areas typical for these rivers. The structure of the pollution areas, as well as the geological and chemical features (that have led to the formation of these areas) have been taken into account. Samples have been of two types – from surface waters and from bottom sediments. A total of 1100 samples have been taken from surface waters and from bottom sediments.

Surface water samples have been taken from a depth of 0.5 m from the surface (with the help of sample tubes that are designed exclusively for sampling in order to avoid the ingress of impurities) and then poured into a sealed plastic bag. A label has been also placed with the following information:

- the number of the sample;
- coordinates;
- date of sampling;
- depth of sampling from the surface of the river;
- geological composition of bottom sediments.

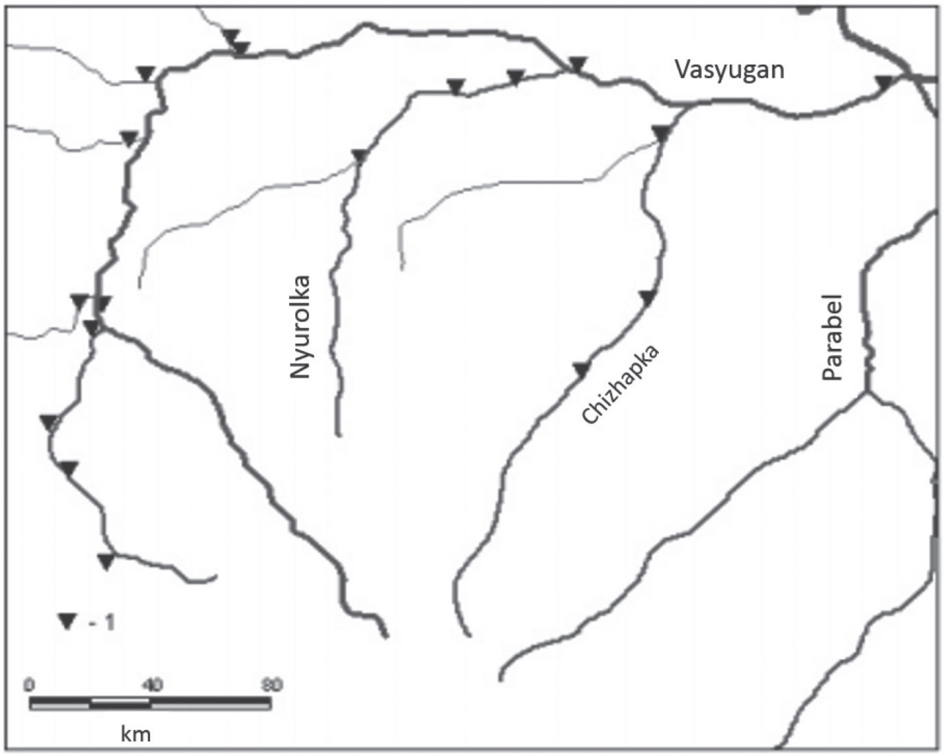

Figure 1: The research region – Vasyugan River and its tributaries.

A passport for each sample has been filled in place.

Bottom samples, including for the purpose of assessing the level of pollution with oil products, as well as with chlorine anions (in a water extract) have been taken immediately after surface water samples. For this, special bottom grabs have been used. To compare the results, the selection of surface water and bottom sediments has been carried out in the same places. The concentration of chlorine and oil products has been determined in both bottom and surface samples. The concentration of microelements has been determined separately from each other, or with the assessment of not matching indicators. After sampling, bottom samples have been dried in laboratory conditions. In addition, in the vicinity of oil-producing enterprises, peat samples have been taken (core samples) at depths of up to 15 cm, from 15 to 20 cm, from 21 to 25 cm, and from 26 to 50 cm. A total of 100 peat samples have been taken.

Methods

The sampling is determined by the indicators that need to be obtained. Thus, water samples from the surface layers are taken in the central parts of river beds. The oil content is determined by sampling from the upper layers of bottom sediments. When determining heavy metals, samples are taken from different layers of bottom sediments, but are ultimately combined into one sample. Sampling is carried out according to the protocol GOST (state standard) 17.1.5.01-80.

The analysis of microelements has been carried out in the laboratories of the Polytechnic Institute, as well as in the Institute of Geochemistry of the Russian Academy of Sciences. The following standard methods have been used to determine the concentration of microelements: semi-quantitative spectral analysis, atomic adsorption method, and also the inversion voltammetric analysis method. The presence and concentration of petroleum products have been determined using infrared spectrophotometry.

Peat samples have been dried using ovens; subsequently, after burning in a muffle furnace, the chemical composition has been determined by spectral analysis.

Statistical Analysis

Data analysis has been carried out using the program Origin v. 8.0. The differences are significant at $p \leq 0.05$. Differences between the parameters have been revealed using the Fisher two-sample *t*-test for independent samples. The table shows the average values. To determine the relationships between element concentrations, Pearson correlations have been calculated.

Results

In addition to petroleum products, the presence of 23 chemical elements has been discovered in the bottom sediments of the Vasyugan River and its tributaries (Table 1). Moreover, in small rivers, whose dense network forms the basis of water circulation in the Vasyugan basin, only five elements have been found, namely copper, lead, zinc, nickel and chlorine anions, and also petroleum products. This is almost five times less than in larger tributaries and in Vasyugan itself ($p \leq 0.05$). Between Vasyugan and its tributaries, differences in the number of chemical elements are unreliable. In all 23 elements have been found in Vasyugan, then in the tributaries – 22-23, with the exception of chlorine anions in two cases (Table 1).

The maximum amount of iron, aluminum, silver, magnesium, calcium and barium has been found in Vasyugan. On average, it is 0.5-2.0 times higher concentration compared to tributaries ($p \leq 0.05$).

At the same time, some elements predominate in the tributaries, for example, the concentration of zinc, vanadium, lead, phosphorus, nickel, manganese, copper and cobalt (Chertal tributary, Table 1).

In the Ellenkulunyakh tributary, titanium and chromium prevail, in the Makhnya tributary – antimony and molybdenum. The concentration of chemical elements in the tributaries may exceed those for Vasyugan river 2.0 or more times ($p \leq 0.05$). The level of oil products has also been recorded in the Makhnya tributary, 3.0 times higher than that in the Vasyugan ($p \leq 0.05$) and 5.0 times higher in the Chizhapka

Table 1: The concentration of chemical compounds in the bottom sediments of the Vasyugan river (in µg/kg)

Chemical element	*1*	*2*	*3*	*4*	*5*	*6*	*7*
Iron	31000	29000	21000	16000	11000	20000	-
Manganese	550	650	70	350	100	400	-
Titanium	5233	4850	4065	6120	4035	3010	-
Chromium	115	109	202	403	210	145	-
Copper	29	32	6	21	20	11	5
Lead	21	22	5	11	8	20	3
Zinc	68	75	15	41	32	40	15
Nickel	35	42	31	16	11	20	36
Cobalt	17	20	10	5	7	7	-
Aluminum	61000	60000	42000	40000	41000	60000	-
Magnesium	7000	6000	3000	2000	900	2000	-
Silver	0.67	0.32	0.02	0.05	0.05	0.05	-
Molybdenum	1.5	1.0	1.5	1.0	1.5	2.5	-
Vanadium	87	99	62	45	45	45	-
Antimony	285	222	303	307	310	405	-
Lithium	25	35	35	35	35	20	-
Silicon	300000	300000	350000	450000	450000	300000	-
Phosphorus	1093	1666	662	884	808	810	-
Potassium	11000	16000	15000	16000	15000	10000	-
Calcium	16000	11000	11000	3000	3000	7000	-
Sodium	11000	10000	11000	10000	10000	7000	
Barium	701	550	610	600	505	525	-
Chlorine anions 2-	26.5	-	31.7	-	-	20.9	19.1
Oil products	69.4	-	47.5	-	-	235.9	123.1

Note: Rivers: 1 – Vasyugan, 2 – Chertala, 3 – Chizhapka, 4 – Ellenkulunyakh, 5 – Lontyn-Yah, 6 – Makhnya, 7 – small rivers.

tributary. In small rivers, there is a fairly high level of concentration of oil products, only 2.0 times lower than the concentration in Makhnya ($p \leq 0.05$). In other rivers of the Vasyugan basin, oil products, as well as chlorine anions, have not been found.

Pearson correlations are found between elements in the waters of the Vasyugan basin:

- between the concentrations of iron and manganese (0.82);
- iron and cobalt (0.91);
- iron and aluminum (0.9);
- iron and magnesium (0.92);
- vanadium (0.95);
- calcium (0.92).

For chromium, a negative correlation is found with nickel (–0.66), and a positive correlation with sodium (0.91). One-Way ANOVA analysis has shown that between groups the difference is more significant than within groups (6.3 vs. 1.4). This indicates that the variation in the concentration of elements is stronger between different elements than one element, but between different points.

In comparison with data on other rivers of the world, in the Vasyugan basin, the concentration of chemical elements falls within the minimum-maximum range (Table 2).

The exception is silicon, the concentration of which is 1.5 times higher than the maximum values for the rivers of the world ($p \leq 0.05$). According to the threshold limit value (TLV) indicators, in the Vasyugan basin, there is an excess of zinc concentration of 2.0 times ($p \leq 0.05$). The remaining three substances do not exceed the TLV limits.

Table 2: Comparison of the concentration of chemical elements in the Vasyugan river basin and other rivers of the world (according to Yanin, 2002a, 2002b)

Chemical element	*Average concentration for the Vasyugan basin*	*Data on the rivers of the world, the minimum-maximum range*	*The threshold limit value (TLV)*
Silicon	365000	from 3 to 210000	-
Aluminum	51000	from 2 to 89000	-
Iron	21000	from 800 to 70000	-
Potassium	13000	from 2500 to 35000	-
Sodium	9500	from 1500 to 10000	-
Calcium	8000	from 400 to 80000	-
Titanium	4500	from 2000 to 5500	-
Magnesium	3500	from 10000 to 15000	-
Phosphorus	1000	from 50 to 10000	-
Barium	500	from 100 to 2500	-
Manganese	450	from 100 to 8000	1500
Antimony	250	from 50 to 2600	-
Chromium	150	from 5 to 1700	-
Vanadium	75	from 30 to 200	-
Zinc	50	from 4 to 25000	23
Nickel	30	from 5 to 1500	-
Lithium	30	from 20 to 35	-
Chlorine	22	-	-
Copper	20	from 1 to 1000	3
Lead	15	from 15 to 30	32
Cobalt	15	from 1 to 45	-
Molybdenum	1	from 0.1 to 125	-
Silver	0.30	from 0.01 to 135	-
Oil products	85	-	-

As it is known, peat has a wide adsorption capacity, as well as the ability to accumulate chemical compounds that come with water. The data obtained by authors show that there is an accumulation of chemicals in peat, however, with some differences depending on peat depth (Table 3).

In peat, the authors have discovered arsenic, which is also present in bottom sediments and in the surface waters of rivers. This indicates the process of its accumulation in peat deposits. Some elements, including petroleum products, have a tendency to accumulate in the outer layer of peat (chromium and chlorine, a concentration is 0.5-3.0 times higher than in deeper layers, $p \leq 0.05$). The bulk of heavy metals accumulate in the middle layer of peat, at a depth of up to 20 cm, here their concentration is 2-10 times higher than that in other layers ($p \leq 0.05$). Some elements, such as calcium, have a maximum concentration at a depth of 25 cm. A second significant layer of oil products is found at a depth of 0.5 m.

Discussion

When the hydrological regime changes (spring floods, changes in riverbeds, etc.), particles of peat can be substantially transported with water (Li et al., 2014a, 2014b). Together with them, all heavy metals, oil products and other pollutants deposited in peat fall into the watercourse (Lu Guang and Min, 2017). In the authors' opinion, rivers with a concentration of oil products in excess of 700 mg/kg can be considered the most polluted; in the case of Vasyugan river basin, one can talk about a moderate pollution level. At the same time, during various emergency situations (accidents), a sharp increase in the level of pollution is possible (Ma et al., 2016).

The movement of heavy metals along river beds can be of different nature. For example, iron moves with the bottom masses as a result of the accumulation or decrease in the concentration of organics of natural origin (Mezamontenegro et al., 2012). The presence of zinc may be due to leaching of this metal from oil pipelines that cross rivers (Xie et al., 2014). The copper presence is of the same reason as that of iron, which is reflected in the high positive correlation between these heavy metals (Miao et al., 2019). Manganese is also of natural origin (Miao et al., 2014, 2019).

The presence of rare-earth or heavy metals such as nickel, molybdenum, lead is directly related to industrial oil production (Park and Choi, 2013).

The appearance in food chains of various heavy metals can have dire consequences for local ecosystems. A certain concentration of lead, arsenic and antimony can result in severe poisoning (Talbi et al., 2018). The presence of chemical elements in surface waters and their deposition in bottom sediments, and, ultimately, in the ocean, lead to their ingress and accumulation in zoobenthos (Wang et al., 2007, 2010a, 2010b). Eating such seafood can also cause severe poisoning in people (Wang et al., 2010b).

The role of bottom sediments, like swamps, is accumulating. On the one hand, the accumulation of heavy metals and other oilfield products leads to self-purification of river and swamp waters (Wang et al., 2013). On the other hand, all of the above water bodies may be subject to secondary pollution (Wu et al., 2015). This is similar to the action of a time bomb. The territory may already be depleted in terms of oil production, but water pollution will occur anyway, and in increasing concentration. In the authors' opinion, as a result of studies, it can be argued that bottom sediments, as well as peat deposits, can be a

Table 3: Chemical elements in the composition of peat at different depths in the vicinity of oil companies

Chemical element	*From 1 to 15 cm*	*15-20 cm*	*21-25 cm*	*26-50 cm*
Calcium	4500	14300	11000	1500
Silicon	Up to 1	150	Up to 1	15
Manganese	95	110	70	15
Aluminum	1	10	4	5.5
Barium	12	40	15	6
Antimony	16	155	55	10
Arsenic	0.9	2	Up to 0.1	Up to 0.1
Chromium	1.5	Up to 0.4	Up to 0.4	Up to 0.4
Chlorine	290	190	130	130
Oil products	850	1600	930	1200

promising object for studying the level of pollution of water bodies. This primarily relates to bodies of water with low water flow. Bottom sediments and the peat layer can thus act as an indicator of the level of pollution of the whole territory. Bottom sediments are the result of geochemical processes associated with the characteristics of the drainage basin (Yao et al., 2016). Bottom sediments can be an indicator of anthropogenic flows and anthropogenic pressure on a watercourse. As a result of chemical reactions, a chemical element can transfer to the aqueous phase and subsequently precipitate, as a result of which secondary pollution of the reservoir occurs (Yongquan, 2007). At the same time, the volume of migratory elements associated with pollution can be related to topography. The latter in turn can determine the hydrodynamic features of water flows and the geochemistry of bottom sediments in river beds.

Conclusions

Vasyugan river basin is characterized by a moderate level of pollution resulting from oil production. The horizontal (surface water) and vertical migration of heavy metals and oil products (bottom sediments in rivers, peat in marshy areas) are revealed. In total, 23 chemical elements have been found, with a predominance in the Vasyugan riverbed and its tributaries. In small rivers, the number of chemical elements acting as pollutants is five, in marshes – 9. Thus, part of the elements (arsenic) accumulates in the swamps, and the main part enters the river channels through direct industrial emissions.

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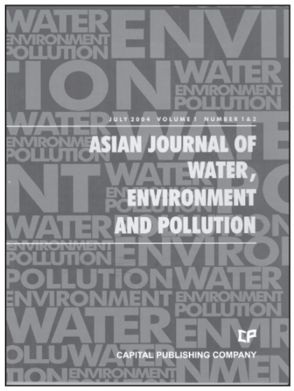

Aims and Scope

Asia, as a whole region, faces severe stress on water availability, primarily due to high population density. Many regions of the continent face severe problems of water pollution on local as well as regional scale and these have to be tackled with a pan-Asian approach. However, the available literature on the subject is generally based on research done in Europe and North America. Therefore, there is an urgent and strong need for an Asian journal with its focus on the region and wherein the region specific problems are addressed in an intelligent manner. In Asia, besides water, there are several other issues related to environment, such as; global warming and its impact; intense land/use and shifting pattern of agriculture; issues related to fertilizer applications and pesticide residues in soil and water; and solid and liquid waste management particularly in industrial and urban areas.

Asia is also a region with intense mining activities whereby serious environmental problems related to land/use, loss of top soil, water pollution and acid mine drainage are faced by various communities. Essentially, Asians are confronted with environmental problems on many fronts. Many pressing issues in the region interlink various aspects of environmental problems faced by population in this densely habited region in the world. Pollution is one such serious issue for many countries since there are many transnational water bodies that spread the pollutants across the entire region. Water, environment and pollution together constitute a three axial problem that all concerned people in the region would like to focus on.

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