Distribution and bioaccumulation of heavy metals (Hg, Cd and Pb) in fish: influence of the aquatic environment and climate

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Abstract
Mercury (Hg), cadmium (Cd) and lead (Pb) are toxic metals that continue to attract much attention because they are prone to be accumulated in fish tissues and can harm human health if taken up with food. Data acquired by studying the bioaccumulation of these metals in the various fish species from water bodies along a latitudinal gradient in Russia (from northern Arctic lakes to the southern mouth segments of the Volga River) are utilized to identify general tendencies and specifics in the accumulation of toxic metals depending on the aquatic environment and temperature. Results demonstrate that small quantities of the metals are accumulated in various functionally important organs: Hg is enriched in the liver and muscles, Cd in the kidneys and gills, and Pb in the kidneys and liver. The metals are proved to be simultaneously accumulated in all organs and tissues of the organism, and this reflects the uptake of the metals by the organism and their subsequent distribution in it. The aquatic environment and fish habitats affect the elements’ bioavailability. The metals are more significantly accumulated in predatory fish. At low Hg concentrations in the water, statistically significant dependences were identified between Hg accumulated in predatory fish organisms and concentrations of organic matter in the water. Cd is more bioavailable in waters with low pH. Pb displays the strongest dependence of its bioaccumulation in low-salinity water. Extensive data on fish in water bodies occurring in large territories in Russia, from the Arctic to warm southern latitudes, indicate that climate affects the intensity of Hg accumulation, whereas the accumulation of the other metals also depends on the Ca concentrations, with the uptake of these metals being more significant at low Ca concentrations. Concentrations of toxic metals in the muscles of the fish were below the values critical to food to be consumed by humans.

1. Introduction
The assessment of ecological consequences of changes in the geochemical cycles of elements under the industrial impact is of paramount importance for the environment. Aquatic environments are the final collectors of all kinds of pollution. In contrast to terrestrial environments, life in water bodies is characterized by stronger relations between aquatic organisms and environmental factors because of the important role of ecological metabolism in aquatic ecosystems and the high mobility of polluting substances in water. In comparison to invertebrates, fish are more sensitive to many toxicants and are thus a convenient target of studies of aquatic geochemistry and pollution with metals (Reynders et al 2008, Rajeshkumar and Li 2018, Kuczyńska et al 2018, Gashkina et al 2020).

Metal accumulation in fish is a serious problem as it hazardously impacts the health of human populations feeding on fish. Strong accumulation of metals in organisms leads to long-term consequences such as the organisms becoming mutagenic, embryotoxic, gonadotoxic, carcinogenic, etc (Satarug et al 2010, Sakamoto et al 2013, Bjerregaard and Andersen 2014, Handbook… 2014, Moiseenko et al 2018a, Ali et al 2019).

In the context of contamination with metals, much attention of researchers worldwide is now focused on hazardous metals such as Cd, Pb and Hg (AMAP 2002, Wood et al 2012, AMAP/UNEP 2013, Renieri et al 2014, Atobatele and Olutona 2015).
Extensive literature data demonstrate the hazardous features of these metals. Although Hg is dangerous due to its direct toxic characteristics, the most serious problems stem from its ability to be significantly accumulated in living organisms, with this accumulation becoming stronger up the food webs, and from remote gonado- and neuro-toxic features and carcinogenic properties (Wratras et al. 1998, Gochfeld 2003, William and Fitzgerald 2007, Soto et al. 2011, Jardine et al. 2013, World Health Organization 2013, Rice et al. 2014).

Cd is, in a sense, a metabolic antagonist of Zn. It was hypothesized (Wood 2001) that the primary mechanism of toxic action of Cd may be related to the inhibition of Ca transfer with proteins. Hypocalcemia is a hypothesis most commonly invoked to explain the toxic action of Cd (Li et al. 2011, Annabi et al. 2013, Sigel et al. 2013). Similar to Cd, Pb is analogous to Ca in processes of uptake by and accumulation in the fish. Ca\(^{2+}\) is adsorbed through the apical membrane Ca\(^{2+}\)-channel in the gills, and is transported across the basolateral membrane with the Ca\(^{2+}\)-ATPase (Griffith 2017). Because of its strong similarity to Ca, Pb penetrates the organism and gets involved in metabolic processes. Some data indicate that Pb shifts the nitrogen exchange in carp towards more active catabolism and also triggers neurotoxic and gonadotoxic effects (Garcia-Leston et al. 2010, Wood et al. 2012, Li et al. 2020).

The bioaccumulation and toxic properties of metals largely depend both on their own characteristics and on environmental conditions, which control the bioavailability of these metals (Gandhi et al. 2011, Magalhaes et al. 2015, Viânnêna et al. 2018, Moiseenko et al. 2020). Experimental studies in the natural environments enable understanding the effects of local contamination on the accumulation of metals in the fish. Thereby, the most interesting issue is how a small increase in the concentration levels of metals in water (due to global fluxes of elements, diffuse contamination of major streams, and/or leaching with acidification atmospheric precipitation) can affect the bioaccumulation of these metals by fish. At low concentrations of metals in water, their toxic properties are dependent on such ecological factors as pH and concentrations of Ca and organic ligands (Gandhi et al. 2011, Magalhaes et al. 2015, Amde et al. 2017, Liu et al. 2017, Viânnêna et al. 2018). Literature data indicate that bioaccumulation and toxic properties manifest themselves more clearly in low-alkaline water (Wijdevelda et al. 2018, Moiseenko et al. 2020).

Along with other factors, temperature influences the biogeochemical cycles of elements, and models for evaluating the temperature effects on the penetration and bioaccumulation of elements in fish are broadly discussed in the literature. A review of this literature indicates that a temperature increase can affect the bioaccumulation of metals through an increase in the vegetation period and metabolism of fish, with this effect being most clearly discernible in northern territories (Reist et al. 2006, Stern et al. 2012, Gouin et al. 2013, Moe et al. 2013, Hudelson et al. 2019, Gashkina and Moiseenko 2020).

Possessing extensive databases, we formulated our goal as studying the bioaccumulation by various fish species in water bodies along a latitudinal gradient (from Arctic northern lakes to the southern mouth parts of the Volga River) in order to identify the distribution and specifics in the bioaccumulation of toxic metals depending on aquatic environments and temperature. To do this, the following tasks have been carried out:

- studying the specifics of the water chemistry, including the concentrations of toxic metals (Hg, Cd and Pb) in the water of small lakes and large rivers (Northern Dvina, Pechora and Volga) in the European part of Russia;
- assessing the bioaccumulation of toxic metals in various fish species and to understand how these metals are distributed between fish organs and tissues;
- evaluating the effects of the aquatic environments and climate on the bioaccumulation of metals;
- determining the potential danger of consumption of fish at specified concentrations for humans.

2. Materials and methods

The research work in the European part of Russia was carried out at various water bodies: large rivers and small lakes located in a huge territory from the northern tundra and taiga to semiarid regions, where the mouth of the Volga River is situated. In the course of this research, we did our best to avoid local contamination sources and made it possible to evaluate the effect of diffuse contamination. A location map of the observation and sampling sites is presented in figure 1.

The Pechora is a large river in Russia, which flows into Pechora Bay of the Pechora Sea. The river is 1809 km long and its catchment area is 322 000 km\(^2\). Oil producing facilities on the shores contaminate the river. Other contamination sources are logging and wood-working enterprises. Our studies were conducted in the vicinity of the inflow of its tributary Usa (upstream of the Usa delta), and these data thus demonstrate the overall contamination of the river.

The Northern Dvina is one of the largest rivers in northwestern Russia and flows into the White Sea. The length of the river is 1302 km and its catchment area is 357 000 km\(^2\). The dominant contamination sources of the mouth segment of the Northern Dvina, where our studies were carried out, are cellulose- and paper-making and wood-working enterprises, which are located upstream of our study area.

The Volga is the longest river of Europe and flows through the western part of Russia. The length of
the Volga is 3690 km. Because dams for hydroelectric power plants were erected across the river, the Volga is navigable for most of its length. The basin of the Volga River, which is inhabited by 40% of the population of Russia, hosts 45% of the country’s industry and 50% of its agriculture. The biggest environmental problems stem from major industrial complexes, big dams, large cities, and from the need to maintain the navigability of this river (National Atlas of Russia 2007).

Along with large rivers, we have examined small lakes (in the tundra, taiga and mountainous territories) in the Barents region, which spans the Murmansk and Arkhangelsk region and Karelia. The small lakes are not directly environmentally impacted and receive only atmospheric fallouts with metals and acid-forming compounds, which are brought from local production facilities and provided by trans-boundary transfer (Moiseenko et al. 2018b, 2019).

Water chemistry sampling was aimed at assessing the disseminated contamination of major streams (the effects of spot contamination sources were eliminated) and the airborne effects on catchments and leaching of elements by acid precipitation in lakes. Water samples were always taken exactly at the sites where fish were caught for examination. Water samples were collected into Nalgen® Polyethylene
bottles (1 l and 60 ml). After their collecting, all samples were kept cool (ca. +4 °C) in dark containers.

Analyses of the water samples were carried out, according to recommendations in (Eaton et al 1992), as follows. The pH was measured using a Metrohm® pH meter; conductivity (20 °C) by Metrohm® conductivity; alkalinity was analyzed using the Gran titration method; and organic matter content was determined by the Mn oxidation method. Cd and Pb were analyzed by atomic absorption in a graphite furnace (GFAAS, PerkinElmer 5000, HGA-400, AAnalyst-800, Corp., Norwalk, USA). Hg was determined using fluorescence (Fl, model Merlin®).

The fish study was aimed at identifying the effects of metal bioaccumulation. The fish were studied during the period of active feeding in September. The fish were caught with standard gillnet series, consisting of eight fleets 10–45 mm bar mesh size. To minimize internal factors, such as sample maturation and age, we sampled impuberal fish (age of 4+ to 5+) in September. The number of fish individuals examined at each site was varied and was usually five or more. Whitefish (Coregonus lavaretus L.) is the most widespread species in the northern part of Russia. Bream (Abramis brama L.) is the most widespread fish species in the Volga River basin. The typical predators (which we sampled) are pike (Esox lucius L.) and perch (Perca fluviatilis L.), and in the northern areas it is salmon trout (Salmo trutta L.). These fish species do not migrate for long distances, a fact that enabled us to sample the material with regard to a particular zone of pollution. The methods applied in our work with fish is described in more detail in (Moiseenko et al 2018a)

Samples of fish organs and tissues to be analyzed for the metals were dried to their constant weight at 105 °C. Dry samples were prepared for analysis by wet digestion in ultrapure nitric acid (10 ml acid per 1 g of tissue). The fish solutions were also analyzed for Cd and Pb, also by atomic absorption, in a graphite furnace, on (GFAAS, PerkinElmer 5000, HGA-400, AAnalyst-800, Corp., Norwalk, USA). Hg was determined using atomic fluorescence (Fl, model Merlin®). Standard solutions with appropriate concentrations for each element were manufactured from 1000 ppm AAS stock standards (Merk, Darmstadt, Germany). The quality of the analysis was controlled by means of replicate analyses.

3. Results

3.1. Water chemistry

Data on the chemical composition of the waters and on Hg, Cd and Pb concentrations in waters sampled at the study areas are summarized in table 1. The water bodies significantly vary in the chemical composition of their water. Water in the lower stream of the Northern Dvina River is characterized by a fairly high conductivity and high Ca concentrations as a consequence of the composition of sedimentary rocks that make up the catchment area. Waters in the study area possessed fairly high values of the color index and total organic carbon (TOC) concentrations because of widespread woodlands and swamps in the catchment areas. Water in the lower stream of the Pechora River contains fewer salts and less organic matter.

The water of the largest of the rivers, the Volga, had the highest conductivity, which almost doubled from the upper to lower (near the mouth) stream of this river. In the latter, the Ca concentrations were at a maximum. In its upper stream, the river flows through woodlands, and its water there possesses the highest color indexes and TOC.

The chemistry of the waters of the small lakes broadly varies. The water of lakes in the tundra zone on the shores of the Barents Sea has a low salinity. The water of mountainous Chuna Lake in the tundra was very poor in Ca and organic matter, and compositional characteristics of the water were close to those of atmospheric precipitation. Towards the northern taiga, concentrations of humic acids in the waters increase because of more and more widespread woodlands. The salt composition of the waters broadly varies. Ca concentrations in the lacustrine waters vary by factors of 2–4 from one lake to another, with the lowest Ca concentration and pH found in lakes in the tundra zone (Chuna, B. Khariusnoe and Enozero), which are habitats of salmon trout.

Hg concentrations in the waters are very low in all of the water bodies sampled for fish: usually below the detection limit (<0.01 µg l⁻¹), i.e. the most thoroughly examined and sampled areas have not shown anomalously high levels of Hg contamination. The Cd concentrations were within the range of <0.05–0.52 µg l⁻¹. It should be mentioned that Ca concentrations in the waters of the examined lakes and rivers were lower than 60 mg l⁻¹, which is generally typical of water bodies in most of Russia’s territory. In the middle and lower stream of the Volga, Cd concentrations are higher, as they also are in the Northern Dvina River. Inasmuch as this metal is relatively labile, its migration into water bodies may be related to acid leaching.

Pb concentrations in the waters of the Volga and Northern Dvina broadly vary from 0.02–3.2 µg l⁻¹. There are many reasons for an increase in concentrations of this metal in aquatic systems: transport, combustion of fuels, industrial contamination, transboundary transfer, urbanization, etc.

3.2. Hg in the fish

Hg concentrations in organs and tissues of fish in various water bodies in European Russia vary within
### Table 1. Main indicators of the aquatic environment of lakes and rivers in the areas of fish catching.

| Water body         | Metal concentration in the water, µg l⁻¹ | pH  | Conductivity, µCm cm⁻¹ | Ca, mg l⁻¹ | Color index, °Pt-Co | TOC, mg C Ll⁻¹ | ΣT > 10 °C* |
|--------------------|------------------------------------------|-----|------------------------|------------|---------------------|----------------|-------------|
| **Large rivers:**  |                                          |     |                        |            |                     |                |             |
| Pechora            |                                          |     |                        |            |                     |                |             |
| lower stream       | <0.01                                    |     | 0.08 ± 0.07            | 0.5 ± 0.2  | 7.4 ± 0.1           | 80 ± 17        | 9.6 ± 2.6   | 350        |
| Severnaya Dvina    |                                          |     | 0.07 ± 0.05            | 0.8 ± 0.1  | 7.4 ± 0.1           | 176 ± 19       | 16.4 ± 1.8  | 1250       |
| lower stream       |                                          |     | 0.13 ± 0.04            | 0.5 ± 0.02 | 7.7 ± 0.1           | 195 ± 35       | 29.2 ± 4.6  | 1800       |
| Volga              |                                          |     | 0.12 ± 0.11            | <0.1       | 6.8 ± 0.3           | 255 ± 21       | 35.5 ± 2.1  | 2100       |
| upper stream       |                                          |     | 1.72 ± 0.9             | 7.4 ± 0.3  | 364 ± 8             | 34.6 ± 1.5     | 12.9 ± 2.2  | –          |
| middle stream      |                                          |     |                        |            |                     |                |             |
| lower stream       |                                          |     | 0.13 ± 0.07            | 1.72 ± 0.9 | 7.4 ± 0.3           | 364 ± 8        | 34.6 ± 1.5  | 3200       |
| **Lakes:**         |                                          |     |                        |            |                     |                |             |
| Kola Mountain      |                                          |     |                        |            |                     |                |             |
| Chuna              | <0.01                                    |     | <0.05                  | 0.1        | 6.3 ± 0.1           | 9 ± 0.5        | 0.8 ± 0.05  | 1.6 ± 1.3  | 1.9 ± 1.7 | 350        |
| Kola Tundra        |                                          |     |                        |            |                     |                |             |
| Enozero            | <0.01                                    |     | <0.05                  | 0.1        | 6.3 ± 0.3           | 27 ± 4         | 0.5 ± 0.2   | 23 ± 11   | 3.9 ± 2.3 | 350        |
| Kola Taiga         |                                          |     |                        |            |                     |                |             |
| B. Hariysnoe       | <0.01                                    |     | <0.05                  | 0.1        | 6.3 ± 0.2           | 9 ± 0.7        | 0.8 ± 0.1   | 3.9 ± 0.2 | 2.0 ± 1.7 | 350        |
| Pirenga, Okhtozo   |                                          |     | 0.08 ± 0.05            | 0.1        | 7.0 ± 0.2           | 33 ± 7         | 2.8 ± 0.5   | 35 ± 6   | 4.5 ± 1.7 | 800        |
| Arhangelsk Taiga   |                                          |     |                        |            |                     |                |             |
| Chernoe, Vokh'e    | <0.01                                    |     | 0.06 ± 0.05            | <0.1       | 7.3 ± 0.2           | 56 ± 23        | 6.4 ± 2.6   | 89 ± 8   | 13.7 ± 2.5 | 1150       |
| Karelia Taiga      |                                          |     |                        |            |                     |                |             |
| Padmozero          | <0.01                                    |     | 0.10 ± 0.01            | 1.97 ± 1.29| 7.9 ± 0.1           | 157 ± 1        | 17.9 ± 0.1  | 41 ± 1   | 9.5 ± 2.6 | 1400       |

Note: * — the sum of air temperatures above 10 °C for the region.
ally developed territories, and the catchment area is the river flows through urbanized and well industri-

cal of the middle stream of the Volga. This part of Hg bioaccumulation in the organism of bream is typ-

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tions are relatively high in the liver of pike from the concentrations in the liver of both the pike and perch were found in salmon trout not only in its liver, i.e. Hg accumulation is fish-species specific. Hg concentrations in predatory fish, such as salmon trout, perch and pike, are higher than in benthos-feeder fish species, such as white fish and bream, in all of the studied water bod-
ies. According to Hg concentrations in them, the fish species can be arranged as the following generalized sequence: salmon trout > pike > perch > whitefish > bream

Analysis of the data indicates that Hg is most sig-

ficantly accumulated in the liver of predatory fish. A vivid example of this is offered by salmon trout in B. Khariusnoe Lake (0.196 µg g⁻¹ dry weight), as well as pike and perch in forest lakes in the Kola Peninsula and Karelia. It should be mentioned that B. Kharius-

noe Lake is distant from any contamination sources; neither could Hg be brought to this lake by atmo-
spheric transboundary transfer. High Hg concentra-
tions were found in salmon trout not only in its liver, but in the kidneys, i.e. organs in which exchange pro-
cesses are active. Salmon trout is known to be an active predator, and its metabolism is at a high level. Hg concentrations in the liver of both the pike and perch are comparable to those in their muscles.

Among fish species in the rivers, Hg concentra-
tions are relatively high in the liver of pike from the Pechora and Northern Dvina rivers and from the lower stream of the Volga. Hg concentration in the organism of bream inhabiting the Volga River varied from <0.001–0.127 µg g⁻¹ dry weight. A high Hg bioaccumulation in the organism of bream is typical of the middle stream of the Volga. This part of the river flows through urbanized and well industri-
ally developed territories, and the catchment area is impacted by composite anthropogenic loads, which may result in Hg migration to these parts of the river.

Analysis of our data shows that various fish spe-
cies differently accumulate Hg, i.e. Hg accumulation is fish-species specific. Hg concentrations in predato-
ry fish, such as salmon trout, perch and pike, are higher than in benthos-feeder fish species, such as white fish and bream, in all of the studied water bod-
ies. According to Hg concentrations in them, the fish species can be arranged as the following generalized sequence:

| Water body | Species          | Gills       | Muscles     | Liver µg g⁻¹ dry weight | Kidney          |
|------------|------------------|-------------|-------------|-------------------------|-----------------|
| Pechora    |                   |             |             |                         |                 |
| lower stream | Whitefish       | 0.023 ± 0.013 | 0.018 ± 0.008 | 0.118 ± 0.041 | 0.012 ± 0.007  |
|            | Pike             | 0.088 ± 0.013 | 0.190 ± 0.046 | 0.144 ± 0.030 | 0.102 ± 0.017  |
| Severnaya Dvina | Whitefish      | 0.016 ± 0.002 | 0.076 ± 0.042 | 0.091 ± 0.012 | 0.013 ± 0.004  |
| lower stream | Bream           | 0.012 ± 0.003 | 0.072 ± 0.018 | 0.037 ± 0.028 | 0.007 ± 0.001  |
|            | Pike             | 0.146 ± 0.051 | 0.337 ± 0.085 | 0.248 ± 0.103 | –               |
| Volga      | upper stream     | Bream       | 0.011 ± 0.001 | 0.019 ± 0.003 | 0.053 ± 0.005  | 0.022 ± 0.003  |
|            | middle stream    | Bream       | 0.011 ± 0.002 | 0.049 ± 0.005 | 0.048 ± 0.009  | 0.030 ± 0.003  |
| lower stream | Bream           | 0.005 ± 0.001 | 0.031 ± 0.004 | 0.054 ± 0.010 | 0.012 ± 0.002  |
|            | Perch            | 0.035 ± 0.010 | 0.261 ± 0.055 | 0.341 ± 0.069 | 0.024           |
|            | Pike             | 0.046 ± 0.015 | 0.305 ± 0.022 | 0.243 ± 0.045 | 0.238 ± 0.060  |
| Lakes:     |                  |             |             |                         |                 |
| Kola Taiga | B.Hariysnoe      | Salmon trout | 0.086 ± 0.015 | 0.048 ± 0.010 | 0.196 ± 0.034  | 0.197 ± 0.028  |
|            |                  | Whitefish    | –            | 0.079 ± 0.023 | 0.120 ± 0.028  | 0.128 ± 0.071  |
|            |                  | Perch        | 0.039 ± 0.007 | 0.195 ± 0.046 | 0.109 ± 0.061  | 0.091           |
|            |                  | Pike         | 0.039 ± 0.023 | 0.088 ± 0.027 | 0.122 ± 0.011  | 0.126 ± 0.022  |
| Arhangelsk Taiga | Chernoe, Volch'e | Perch       | 0.010 ± 0.001 | 0.086 ± 0.020 | 0.026 ± 0.008  | –               |
|            |                  | Pike         | 0.009 ± 0.001 | 0.092 ± 0.017 | 0.037 ± 0.006  | –               |
| Karelia Taiga | Padmozero       | Perch        | 0.030 ± 0.007 | 0.126 ± 0.030 | 0.041 ± 0.013  | –               |
|            |                  | Pike         | 0.021 ± 0.014 | 0.260 ± 0.052 | 0.275 ± 0.065  | 0.209 ± 0.062  |
of most of the species strongly correlate with one another: $r = 0.90$, $n = 15$ for whitefish, $r = 0.82$, $n = 30$ for pike, $r = 0.68$, $n = 5$ for salmon trout and $r = 0.80$, $n > 50$ for all of the fish species considered collectively. Our analysis has also demonstrated that Hg accumulation in the kidneys proceeds in a manner analogous to the accumulation of the element in the liver and muscles of fish. A strong correlation was found between Hg concentrations in the liver and muscles of fish: $r = 0.85$ for salmon trout, $r = 0.80$ for perch and $r = 0.83$ for pike. According to their ability to accumulate Hg, fish organs and muscles can be arranged as:

liver $>$ muscles $>$ kidneys $>$ gills

These results demonstrate that even low contaminating Hg concentrations in a water body result in the accumulation of this element in all systems of living organisms, most strongly in the liver and muscles of predator fish species.

### 3.3. Cd in the fish

Table 3 shows Cd concentrations in systems of various fish species living in the territory of European Russia. In our study, the highest Cd concentrations were detected in the organs and tissues of salmon trout (22.6 μgg\(^{-1}\)dry weight) in the northern lakes of Chuna and B. Harriusnoe, which may be explained by the fact that this fish species lives in northern low-salinity waters and by the effect of the acid leaching of this metal or its transboundary transfer.

Cd concentrations in the organs and tissues of whitefish in such large northern rivers as Northern Dvina and Pechora are lower than in whitefish living in lakes in the northern Kola Peninsula. Cd accumulation in salmon trout in water bodies in the northern Kola Peninsula is also higher, particularly in the fish in mountainous Chuna Lake. Cd accumulation in perch and pike is also more intense in lakes in the northern Kola Peninsula. Cd accumulation in perch is also more intense in lakes in the northern Kola Peninsula.

Fish species define the following sequence of their decreasing ability of Cd accumulation:

salmon trout $>$ whitefish $>$ bream $>$ perch $>$ pike

This sequence does not always clearly characterize fish species-specific Cd accumulation, because Cd concentrations may vary from one fish organ to another:

**Table 3.** Average values and standard errors of Cd concentrations in the organs and tissues of fish species from different water bodies. A dash means no data.

| Water body               | Species       | Gills           | Muscles          | Liver            | Kidney           |
|--------------------------|---------------|-----------------|------------------|------------------|------------------|
| **Large rivers:**        |               |                 |                  |                  |                  |
| Pechora                  | Whitefish     | 0.10 ± 0.02     | <0.01            | 0.19 ± 0.05      | 0.75 ± 0.23      |
|                          | Pike          | 0.02 ± 0.01     | <0.01            | 0.06 ± 0.02      | 0.48 ± 0.13      |
| Severnaya Dvina          | Whitefish     | 0.14 ± 0.04     | 0.03 ± 0.02      | 0.07 ± 0.02      | 0.72 ± 0.14      |
|                          | Bream         | 0.26 ± 0.11     | 0.11 ± 0.06      | 0.82 ± 0.54      | 0.61 ± 0.33      |
|                          | Pike          | 0.01 ± 0.00     | 0.01 ± 0.00      | 0.02 ± 0.01      | 0.12 ± 0.12      |
| Volga                    | Bream         | 0.05 ± 0.01     | 0.03 ± 0.01      | 0.25 ± 0.04      | 1.09 ± 0.19      |
|                          | Pike          | 0.19 ± 0.02     | <0.01            | 0.05 ± 0.01      | 0.07 ± 0.04      |
|                          | Bream         | 0.01 ± 0.00     | <0.01            | 0.26 ± 0.04      | 1.94 ± 0.28      |
|                          | Perch         | 0.03 ± 0.01     | 0.01 ± 0.00      | 0.48 ± 0.05      | 0.88 ± 0.11      |
|                          | Pike          | 0.05 ± 0.01     | 0.03 ± 0.01      | 0.25 ± 0.04      | 1.09 ± 0.19      |
| Lakes:                   |               |                 |                  |                  |                  |
| Kola Mountain Chuna      | Salmon trout  | 2.94 ± 0.27     | 0.05 ± 0.02      | 10.35 ± 0.79     | 22.56 ± 1.85     |
| Kola Tundra Enznozero    | Salmon trout  | 0.86 ± 0.04     | 0.16 ± 0.01      | 1.23 ± 0.16      | 6.26 ± 0.90      |
| Kola Taiga B.Harriusnoe  | Salmon trout  | 0.98 ± 0.13     | 0.03 ± 0.01      | 5.48 ± 0.77      | 5.51 ± 0.28      |
| Pirenga, Okhtozero       | Whitefish     | 0.37 ± 0.07     | 0.22 ± 0.02      | 1.34 ± 0.29      | 5.31 ± 0.71      |
|                          | Perch         | 0.32 ± 0.12     | 0.21 ± 0.04      | 1.22 ± 0.21      | 1.08 ± 0.41      |
|                          | Pike          | 0.37 ± 0.07     | 0.26 ± 0.04      | 0.11 ± 0.02      | 0.81 ± 0.23      |
| Arhangelsk Taiga Chernoe| Perch         | 0.08 ± 0.01     | 0.01 ± 0.01      | 0.58 ± 0.14      | 0.60 ± 0.25      |
| Volch’e                   | Pike          | 0.04 ± 0.01     | 0.01 ± 0.00      | 0.07 ± 0.02      | -                |
| Karelia Taiga Padmozero  | Perch         | 0.19 ± 0.04     | 0.01 ± 0.00      | 1.12 ± 0.25      | 0.84 ± 0.28      |
|                          | Pike          | 0.34 ± 0.02     | <0.01            | 0.16 ± 0.01      | 0.67 ± 0.08      |
another. Therewith, Cd concentrations in the kidneys of all of the fish species are practically always more than double that in their liver. Cd concentrations in muscles are two to three orders of magnitude lower than in the kidneys and are rarely higher than 0.20 µg g\(^{-1}\) dry weight.

Coupled Cd accumulation was detected in the kidneys and liver of most of the species. The correlation coefficients between Cd concentrations in the kidneys and liver are \( r = 0.94, n = 40 \) for salmon trout and \( r = 0.83, n = 15 \) for whitefish. Accumulation in the kidneys and liver of salmon trout is strongly related to accumulation in its gills; the correlation coefficient of metal concentrations in the gills and liver is 0.96, and the coefficient for the gills and kidneys is 0.99, \( n = 40 \). Analogous correlations were detected in the whitefish and perch.

Cd concentration in the kidneys of bream living in the Volga basin is strongly correlated with the concentration of this element in other systems of the organism; the liver \( (r = 0.78, n = 52) \) and gills \( (r = 0.56, n = 52) \). Variations in Cd concentrations in the muscles, kidney, and gills of the perch are also coupled with one another, and this indicates that the metal is simultaneously accumulated in various systems of the fish organism if concentrations of this metal in the aquatic environment are elevated. Our data do not reveal any differences between Cd concentrations in the organisms of predatory and nonpredatory fish (the only exception is salmon trout). The salmon trout is likely able to accumulate very high Cd concentrations, particularly in the kidneys.

Fish organs and tissues can be arranged in the following sequence according to their Cd accumulating ability, regardless of fish species:

- Kidneys > liver > gills > skeleton > muscles

These results show that Cd can be accumulated in fairly high concentrations in fish inhabiting low-salinity waters in northern areas. For example, although Cd concentrations in the water of the Northern Dvina are half that in the studied parts of the Volga, Cd concentrations in the organs and tissues of bream living in the Northern Dvina are almost double that in this fish in the Volga. Cd is more significantly accumulated in whitefish and salmon species inhabiting small lakes in the Kola Peninsula.

### 3.4. Pb in the fish

Table 4 presents data on Pb concentrations in fish from various regions. The highest Pb concentrations in the various systems of the bream organism were found in the Northern Dvina and in the upper and middle stream of the Volga. Pb is maximally accumulated in the kidneys and liver. Although Pb concentration is higher in water in the lower stream of the Volga, this metal is maximally accumulated in fish organisms in the upper and middle stream of this river. Pb concentrations broadly vary in the organs and tissues of various fish species. The accumulation of Pb is not obviously specific to the species of fish. However, high Pb concentrations in the liver and kidneys are more frequently found in perch than in other fish species, particularly in forest lakes.

### Table 4. Average values and standard errors of Pb concentrations in the organs and tissues of fish species from different water bodies. A dash means no data.

| Water body | Species          | Gills µg g\(^{-1}\) dry weight | Muscles µg g\(^{-1}\) dry weight | Liver µg g\(^{-1}\) dry weight | Kidney µg g\(^{-1}\) dry weight |
|------------|------------------|-------------------------------|---------------------------------|-------------------------------|-------------------------------|
| **Large rivers:** |                  |                               |                                 |                               |                               |
| Severnaya Dvina |                 |                               |                                 |                               |                               |
| lower stream  | Whitefish        | 0.31 ± 0.21                   | 0.12 ± 0.01                     | 0.20 ± 0.12                   | 0.31 ± 0.24                   |
|              | Bream            | 0.19 ± 0.15                   | 0.19 ± 0.01                     | 0.75 ± 0.31                   | 0.23 ± 0.02                   |
| Volga        | upper stream     |                               |                                 |                               |                               |
|              | Bream            | 0.33 ± 0.07                   | 0.07 ± 0.02                     | 0.25 ± 0.07                   | 0.37 ± 0.12                   |
|              | Pike             | 0.19 ± 0.15                   | <0.01                           | 0.15 ± 0.08                   | 0.07 ± 0.04                   |
| middle stream| Bream            | 0.04 ± 0.01                   | 0.06 ± 0.01                     | 0.19 ± 0.04                   | 0.48 ± 0.10                   |
|              | Pike             | 0.05 ± 0.04                   | 0.08 ± 0.01                     | 0.06 ± 0.01                   | 1.49 ± 0.63                   |
| lower stream | Bream            | 0.07 ± 0.01                   | 0.02 ± 0.00                     | 0.06 ± 0.01                   | 0.13 ± 0.02                   |
|              | Perch            | 0.12 ± 0.06                   | 0.01 ± 0.00                     | 0.05 ± 0.02                   | 0.18                         |
|              | Pike             | 0.08 ± 0.05                   | <0.01                           | 0.01 ± 0.00                   | 0.18 ± 0.06                   |
| **Lakes:**   |                  |                               |                                 |                               |                               |
| Kola Taiga   |                  |                               |                                 |                               |                               |
| B.Hariysnoe  | Salmon trout     | 0.06 ± 0.02                   | 0.02 ± 0.01                     | 0.21 ± 0.08                   | 0.89 ± 0.12                   |
| Pirenga, Okhtozero |           |                               |                                 |                               |                               |
| Whitefish    | Salmon trout     | 0.08 ± 0.04                   | 0.05 ± 0.02                     | 0.25 ± 0.15                   | 0.18 ± 0.03                   |
|              | Perch            | –                             | 0.01 ± 0.00                     | 0.05 ± 0.01                   | 0.80 ± 0.18                   |
| Arhangelsk Taiga |             |                               |                                 |                               |                               |
| Chernoe, Volch’e |            | 0.26 ± 0.07                   | 0.10 ± 0.03                     | 0.19 ± 0.04                   | –                            |
|              | Pike             | 0.16 ± 0.08                   | 0.19 ± 0.15                     | 0.18 ± 0.06                   | –                            |
| Karelia Taiga |                  |                               |                                 |                               |                               |
| Padmozero    | Perch            | 0.19 ± 0.05                   | 0.08 ± 0.02                     | 0.38 ± 0.11                   | –                            |
Fish in all of the examined water bodies accumulate the highest Pb concentrations in functionally important organs: in the kidneys and liver, whereas Pb concentrations in the muscles and gills are low. According to their Pb concentrations, the organs define the following sequence:

\[ \text{kidneys} \geq \text{liver} \geq \text{gills} \geq \text{muscles} \]

Higher (above all other values) Pb concentrations in the kidneys of perch were found in the middle stream of the Volga. In this segment of the river, high Pb concentrations were also detected in the kidneys of bream. Among our study areas, the catchment area of the middle reaches of the Volga is characterized by better-developed industry infrastructure and is highly urbanized. Although we did our best to avoid polluted areas when conducting this study, the situation with the middle stream of the Volga shows the effect of the overall diffuse contamination caused by industrial activities and transport in the catchment area.

4. Discussion

4.1. Effects of geochemical factors on the bioaccumulation of metals

Literature data definitely indicate that fish are informative indicators of water contamination with metals and can accumulate these metals even when their concentration in the water is relatively low (Monroy et al 2014, Wei et al 2014, Chetelat et al 2015, Kuczyńska et al 2018, Moiseenko et al 2018a, Hudelson et al 2019). Our study was fairly comprehensive and was carried out using various fish species and involved studying a wide range of their habitats, from the tundra to semiarid zone and from large rivers to small lakes. We stress here once again that the study was focused primarily on the bioaccumulation of metals by fish inhabiting lakes and rivers that are impacted merely by diffuse and airborne contamination (outside local contamination zones). Because of this, we did not encounter critical contamination levels (with only a few exceptions). At the same time, the habitats broadly varied in living conditions and such parameters as concentrations of salts, Ca and humic organic compounds (table 1).

Analysis of our data shows that various fish species differently accumulate toxic metals, which may be explained by the effects of both endogenic and exogenic factors on this process.

Lifespan and age affect the metal content in fish, and with age the concentration is typically increased as a consequence of the prolonged exposure of fish in water with high metal content (Heath 2018). It has been proved that the Hg content increases with age in large fish, which is especially typical for perch and pike (Haines et al 1995, Backstrom et al 2020). Because samples of fish organs and tissues were taken from impuberual individuals of relatively similar age, during the summer–autumn feeding period, the effect of the latter on the bioaccumulation of nonessential elements was minimized.

It is known that elements find their way into the fish organism with water and food. Metals enter waters and migrate through the aquatic food webs (Wood et al 2012, Heath 2018). When arriving at a water body, metals are variably accumulated in the bottom sediments and food webs, and their bioaccumulation in the benthophage and predator species is controlled, first of all, by concentrations in the water and habitat conditions. In view of this, below we analyze how various parameters of the aquatic environment (water hardness, low pH, and organic matter) can affect the bioaccumulation of metals in the fish organism.

Mercury occurs in aquatic environments as numerous physical and chemical species, whose properties vary vastly, and this predetermines the complicated mechanisms of Hg distribution and accumulation in living organisms and its toxic properties. The most important Hg species in aquatic systems are elementary mercury (Hg\(^0\)), inorganic mercury (Hg\(^{2+}\)), monomethyl mercury (CH\(_3\)Hg\(^+\)) and dimethyl mercury Hg(CH\(_3\))\(_2\). Hg adsorbents and bioaccumulators are, particularly in contaminated areas, particulate matter and bottom sediments in water bodies. The bacterial conversion of Hg\(^{2+}\) in CH\(_3\)Hg\(^+\) is an important feature of the Hg cycle in any aquatic ecosystem and the very first link of the long transfer of this element along food webs and in bioaccumulation processes (Gochfeld 2003, William and Fitzgerald 2007, Lavoie et al 2013). Methylation processes are the most intense in the uppermost layers of bottom sediments in water bodies. These sediments are rich in organic matter, in particular matter, and in the mucus covering fish (Heath 2018). In the course of its migration through the food structure of ecosystems, concentrations of Hg increase in living organisms and reach a maximum in predatory fish species (Watras et al 1998, Rask et al 2007). Bioaccumulation in predatory fish can be as significant as by factors of 1000–10,000 relative to concentration in the water (William and Fitzgerald 2007). Data were published that Hg concentrations in fish organisms increase with their age (Farkas et al 2003, Li et al 2011, Chetelat et al 2015, Backstrom et al 2020). Bioaccumulation is most clearly discernible in large individuals of pike inhabiting water bodies in Scandinavia, and this is hazardous for the local human population that feeds on fish (Sharma et al 2008). In the high Arctic, Hg was determined to be accumulated in aquatic systems and fish (Norton et al 1990, AMAP 2002).

Hg leads to damage of the tertiary and quaternary protein structure and cellular function due to the connection with selenohydryl and sulfhydryl groups, as a result of reaction with methylmercury and destruction of the cellular structure. Moreover, the processes of transcription and translation are changed:
ribosemes disappear and endoplasmic reticulum and the activity of natural killer cells are eradicated, and there is free radical formation. Even though the Hg sulfhydryl bond is stable and divided to surrounding sulfhydryl consisting ligands, it also contributes free sulfhydryl groups to promote metal mobility within the ligands (Bernholt 2012; Wood et al. 2012, Jaishankar et al. 2014).

Animals that are exposed to toxic Hg have shown adverse neurological and behavioral changes. Our data indicate that Hg is accumulated in the liver, muscles and kidneys of fish. It has been demonstrated (William and Fitzgerald 2007, Rice et al. 2014, Kuczynska et al. 2018, Heath 2018) that Hg is enriched to the highest concentrations in fish liver and muscles, and hence, these organs are recommended in most studies to be used in biomonitoring for Hg. Our data indicate that Hg is also contained in high concentrations in the kidneys, which reflects the bioaccumulation of this element in the fish organism. A significant role is played by Hg received with food (Health 2018). This may explain why our data show relatively low Hg concentrations in the gills and its preferable accumulation in the liver of the fish. Hg can also be received by wild fish species with food organisms, because this element is prone to be accumulated in food webs in contaminated water bodies (Jardine et al. 2013).

At very low Hg concentrations in waters, Hg contamination can be identified by studying concentrations of this element in the fish (Cyr et al. 2019). The accumulation of this metal in fish strongly depends on characteristics of their habitat. The two key factors that activate the process of Hg methylation and, hence, its accumulation in fish are (1) the acid–base balance and low pH of the waters and (2) the occurrence of organic compounds in these waters (Braaten et al. 2014).

Predatory fish (pike and perch) caught in the lower stream of the Volga River contained four to five times more Hg than the benthophages did. This is explained by the specifics of their habitats; these habitats are constrained to thickets of aquatic plants and to silty bottom sediments, in which Hg methylation is active. It was pointed out in (Moiseenko et al. 2020) that this element shows strong affinity to humic organic compounds. In waters with a high color index and rich in humic acids, Hg is dominated by its methylated species, which are more actively accumulated in the fish organism (Chételat et al. 2015, Rahmanikolah et al. 2020). This has been reliably confirmed by our data on dependences of Hg concentrations in the muscles and liver of predators (pike and perch) on the TOC concentrations in the waters (figure 2).

Another factor that intensifies Hg methylation is low pH of the waters. Mercury methylation and its bioavailability in acidified water bodies have been convincingly proved in (Jardine et al. 2013). Data in (Moiseenko and Gashkina 2016) indicate that Hg concentrations in the liver and kidneys of loach inhabiting an acidified tundra lake in the northern Kola Peninsula are many times higher than in this fish species living in a neutral lake in the same area. A high level of Hg bioaccumulation was also detected in perch from acid lakes in Karelia and in lakes with high concentrations of humic acids (Haines et al. 1995). In Finland, correlations were detected between Hg accumulation in the perch and low pH of the waters (Manio 2001).

Although Hg concentrations in the studied fish organisms varied broadly, concentrations of this element in their muscles were no higher than the values established as a limiting standard for fish consumption by humans (World Health 2013).

Being a labile metal, cadmium is readily mobilized from minerals under the effect of acid atmospheric precipitation and can be accumulated to high concentrations in water bodies. Data on streams in Sweden demonstrate that Cd concentrations in natural waters increase with a decrease in their pH during the high-water season (Johansson et al. 1995). Cd occurs in natural waters mostly in an activated state and also as low-molecular inorganic complexes (Moiseenko et al. 2020). Eutrophic waters can also contain Cd complexes with organic matter or Cd adsorbed on particulate matter. Oligotrophic waters with pH ≤ 7.3 contain Cd mostly in the form of ions, with [Cd\(^{2+}\)]/[Cd\(_{tot}\)] = 0.8 and more (Cullen and Maldonado 2013). Similar data reported in (Moiseenko et al 2020) indicate that Cd ions make up more than 90% of the total Cd concentrations in low-salinity oligotrophic waters, and the bioavailability of this metal is high. Data on Arctic ice indicate that Cd can be transported with aerosols and fine particles for long distances and can then fall out far away from the contamination sources and thus cause elevated Cd concentrations in areas distant from any industrial centers (Norton et al. 1990, AMAP 2002).

Cd concentrations in the water bodies we studied in the course of this research are very low, and these concentrations are more broadly variable in the northern waters. This is likely explained by the acid leaching of this element in zones impacted by smoke emissions from copper–nickel smelters in the Kola Peninsula. Small lakes in the Kola Peninsula are affected by regionally spread airborne contamination with acid-forming compounds, primarily sulfates (Moiseenko et al. 2019).

Cd in the liver leads to hepatotoxicity and circulates to the kidneys and gets accumulated in the renal tissue causing nephrotoxicity. Cd can bind with cystein, glutamate, histidine and aspartate ligands and lead to the deficiency of iron. Cd and zinc have the same oxidation states and hence Cd can replace zinc in metallothionein and inhibiting sorption of free radical within the cell (Satarug et al. 2010, Sigel et al. 2013, Anaabi et al. 2013).
Cd is able to enter the fish organism through the gill epithelia, similar to Ca transfer in chloride cells (Bennet-Chamber et al 1999, Reynders et al 2008), and is accumulated in the liver and kidneys because of its strong interaction with cysteine and binding with metallothioneins (Satarug et al 2010). Because Cd metabolism is closely related to essential metals, first of all, Zn, Cd is able to substitute Zn in many vitally important enzymatic reactions and can break and inhibit them (Bzroska and Moniszko-Jakoniuk 2001, Satarug et al 2010, Annabi et al 2013, et al 2013, Li et al 2020).

Our studies demonstrate that this element is most significantly accumulated in the kidneys of fish (table 3). Most fish species do not show any reliably identified correlations between Cd concentrations in the waters and its accumulation in fish organisms. This is explained by strong effects of other exogenic factors (pH and Ca concentrations in the waters) on Cd accumulation. Note that the samples of whitefish organs and tissues were taken from organisms living in water bodies with pH from 6.5–8, and the samples of salmon trout organs and tissues were taken from fish inhabiting water bodies with pH 6.2–7.
More active Cd accumulation was found in fish organisms at lower pH, and this is confirmed by a correlation between the accumulation of Cd in the kidneys of the northern species and the pH of the waters (figure 3). Along with pH, Ca concentration in the water also affects the bioavailability of the metal and its accumulation in the fish. We have derived two-parameter equations that demonstrate relationships between more active Cd accumulation in the kidneys of benthophages at low pH and Ca concentrations. This equation for benthophages is,

\[
\text{Cd}_{\text{kidney}} = 16.7 - 1.81 \times \text{pH} - 0.050 \times \text{Ca}_{\text{water}},
\]

\[ r = 0.761, \quad p < 0.005. \]

Predatory fish species exhibit a more significant correlation only with pH, whereas the Ca concentration does not appreciably influence Cd uptake by the fish organism.

\[
\text{Cd}_{\text{kidney}} = 39.8 - 5.31 \times \text{pH} + 0.007 \times \text{Ca}_{\text{water}},
\]

\[ r = 0.638, \quad p < 0.01. \]

Hence, Cd accumulation in the kidneys of benthophages more strongly depends on Ca concentration in the water, whereas this accumulation in predatory fish is more dependent on the pH of the water.

Our data confirm that Cd penetration in the fish organism influences the supply of essential elements, for example, Zn, to the organism. A negative correlation is acquired for Cd and Zn concentrations in the kidneys of salmon trout (figure 4). ‘Utilizing’ penetration and metabolism mechanisms similar to those of Zn, Cd can substitute Zn in a number of biochemical functions and induce functional disturbances (Satarug et al 2010, Perez and Hoang 2017). The aforementioned dependence obviously shows that Zn availability for an organism diminishes as Cd is accumulated in the kidneys.

Muscles do not accumulate much Cd, and concentrations of this metal in them are lower than the standards established for food materials consumed by humans (World Health, 2013). However, the strong ability of this element to be accumulated in the kidneys may result in human pathologies, as was demonstrated in (Moiseenko et al 2018a).

It is a well-established fact that lead concentrations increase (similar to those of Hg and Cd) in continental waters, particularly in the northern regions (Norton et al 1990, AMAP 2002, Garcia-Leston et al 2010, Wood et al 2012). This metal is contained in low-salinity waters mostly in the form of ions, and an increase in the salt concentrations decreases its bioavailability (Moiseenko et al 2020). Because of this, Pb accumulation depends both on its total concentrations in the waters and on such parameters as pH and Ca concentration. With Ca shortage, Pb is more actively involved in metabolic processes, and this inevitably leads to their disturbances and eventually to the development of pathologies in the organism (Wood et al 2012). Pb concentrations in the studied waters varied broadly and were higher in waters at urbanized areas.

Pb toxicity combined with the ability to replace other bivalent cations such as Ca\(^{2+}\), Mg\(^{2+}\), Fe\(^{3+}\) and monovalent cations such as Na\(^{+}\), disturbs the biological metabolism of the cell. The ionic mechanism of Pb toxicity leads to significant changes in various biological processes such as cell adhesion, intracellular and inter-cellular signaling, protein folding, maturation, apoptosis, ionic transportation, enzyme regulation and the release of neurotransmitters (Garcia-Leston et al 2010, Wood et al 2012, Jaishankar et al 2014).
Pb concentrations in the gills, muscles and kidneys of pike and perch, as well as in the kidneys of salmon trout, were negatively correlated with pH and Ca concentrations in the waters. This indicates that Pb is more actively accumulated in the fish organism at low Ca concentrations in the waters and at their low pH, i.e. Pb accumulation more strongly depends on its concentration in the water and is enhanced in waters with lower pH (figure 5). Our data prove that Pb accumulation is more active in the kidneys, similar to Cd, and is controlled primarily by Pb concentration in the water.

The following dependence was derived for benthophages:

\[
Pb_{\text{kidney}} = 4.19 + 0.676 \times Pb_{\text{water}} + 0.013 \times \text{pH}, \quad r = 0.645, \quad p < 0.01.
\]

In this particular instance, a positive correlation between Pb accumulation in bream and pH of the water is explained by the fact that Pb concentrations are higher in waters in the southern areas, which are more mineralized and have higher pH, i.e. Pb concentrations in the kidneys of benthophages more strongly depend on Pb concentration in the water.

For predatory fish living within a broad range of environments, Pb accumulation is positively correlated with Pb concentration in the waters and negatively with pH:

\[
Pb_{\text{kidney}} = 0.358 + 0.225 \times Pb_{\text{water}} - 0.568 \times \text{pH}, \quad r = 0.725, \quad p < 0.02.
\]

In contrast to Pb concentrations in European lakes, those in the northern lakes are lower than 0.5 µg l\(^{-1}\), whereas these concentrations in some mountainous lakes impacted by anthropogenic contamination increase to 3 µg l\(^{-1}\). Correspondingly, Pb concentrations are lower in fish living in lakes in the northern Kola Peninsula and Karelia. For example, Pb concentrations in the liver are no higher than 0.5 ppm, whereas these concentrations in fish inhabiting lakes in Central Europe are higher than 1 ppm (Acidification... 1995). Hence, water contamination with Pb results in the accumulation of this metal in the organisms of fish. Pb is most actively accumulated in the fish organism in low-salt waters.

### 4.2. Human health risk assessment

Non-essential heavy metals (Cd, Pb and Hg) may be toxic even at quite low concentrations. Significant scientific reviews of the literature are devoted to the toxic effects of non-essential elements on the fish physiology (Wood et al 2012, Bjerregaard and Andersen 2014, Heath 2018. Handbook... 2014). However, in natural conditions, wild fish are affected by multielement pollution, so it is difficult to isolate the effect of each specific metal. The relationship between the accumulation of elements in fish and pathological disturbances in the organs and tissues of bream in the Volga River basin was established. Histological analysis of fish organs and tissues revealed serious disturbances in the morphology and function of the liver and kidneys, as well as in the hematopoietic system. (Moiseenko et al 2008).

The investigation of fish in the Kola lakes showed mass incidents of fish diseases (nephrocalcinosis, lipid liver degeneration, cirrhosis, anemia, etc). The highest content of metals was observed in fish from the lakes near areas exposed to smoke emissions of copper and nickel smelters. Nephrocalcinosis of fish by the accumulation of toxic metals and their toxic
impact (especially Ni and Cd) are found. The main disorders are within the kidneys, as shown by histological analysis. Hepatic disorder is the result of the general toxicity of the mixture of metals. Comparative analysis of abnormalities in fish and humans shows similarity, so the fish can serve as bioindicators of pollution used to predict the direction of medical studies in the search for toxic effects (Moiseenko et al. 2018a).

Accumulation of Hg, Cd and Pb in fish is a potential health threat to humans. Long-term consumption of foodstuff contaminated with metals may lead to the accumulation of toxic metals in several vital organs. This accumulation can result in perturbation of biochemical processes (Renier et al. 2014). Human health risk appears via accumulation of toxic heavy metals in fish causing a potential health threat to their consumers.

The human health risk assessment of heavy metals from fish consumption can be estimated using hazard quotient (US EPA 1999), when an estimated daily intake (EDI) is compared to the oral reference dose (RfD). The RfD of a daily exposure to the human population was reported as 0.1, 1 and 1.5 µg kg day$^{-1}$, for methylmercury, Cd and Pb, respectively (IRIS 2020). According to the Federal State Statistics Service of Russia, fish consumption in Russia averaged 21.5 kg year$^{-1}$ in 2015–2018 (ROSTAT 2019), which corresponds to 58.9 g day$^{-1}$, while the average human weight in Europe is 71 kg. Based on these data, we have determined the critical metal concentration in fish muscles in terms of dry weight at which the EDI does not exceed the RfD, which for Russia was 0.482, 4.82 and 7.23 µg g$^{-1}$ dry weight for methylmercury, Cd and Pb, respectively.

Critical concentrations of elements were not exceeded for all fish organs in our research (tables 2, 3, 4). However, the highest Hg concentrations are found in pike and perch in the lower reaches of the Volga and Northern Dvina rivers. Although the concentration of elements in fish does not exceed the maximum permissible daily intake of this fish, the coefficient of daily intake reaches 0.8 of the permissible amount; in other waters—from 0.2–0.5. The content of Cd and Pb in fish is more than ten times lower than the critical limits.

### 4.3. Temperature effects

Much effort is focused worldwide on studying how climate can affect the bioaccumulation and ecological toxicity of metals (Stern et al. 2012, Bulbus et al. 2013, Landis et al. 2013, Gouin et al. 2013, Ahonen et al. 2018, Hudelson et al. 2019, Lan et al. 2020). These studies indicate that the dominant adverse effect of climate variations on ecosystems and the bioaccumulation of elements are related to the risks of the emergence of new stress conditions under which organisms are more susceptible and vulnerable to chronic intoxication. In these situations, the accumulation of toxic compounds and elements in living organisms will result in more evident adverse consequences. Model calculations in (Moe et al. 2013) indicate that climate warming accelerates the cycling of toxic elements in aquatic ecosystems and enhances toxic properties of these elements.

In the context of Hg biogeochemistry in Arctic seas and lakes, climate warming will activate methylation processes and, hence, the consumption of the element and simultaneously enhance the elimination of gaseous Hg. An increase in the size of living organisms will influence the adsorption of this element (Stern et al. 2012). Data on a char population in northern Canadian lakes indicate that Hg is more actively accumulated by large individuals, which live in more

![Figure 5. Dependence of the Pb concentration in the kidneys on water pH for benthos feeders (1, dashed line) and predators (2, solid line).](image-url)
Figure 6. Dependences of Hg content in the muscles and liver of predatory fish (pike and perch) on the sum of air temperatures above 10 °C for the region.

It is known that the intensity of the metabolism of poikilothermal animals depends on the ambient temperature, and a temperature increase intensifies the ventilation of the gills and, hence, water flow, together with dissolved toxic trace elements (Maulvaul et al 2016). It is pertinent to mention that among the examined fish species, pike and perch are spread most widely (from the Arctic to the lower reaches of the Volga River). Analysis of concentrations of elements in the fish organism provides a clue to understanding the zonal specifics of the bioaccumulation of metals and, hence, the effect of temperature. These data allowed us to derive dependences of Hg concentrations in the muscles and liver on the southern lakes (Hudelson et al 2019). Climate warming extends the vegetation period and thus widens the food resources and, hence, accelerates Hg bioaccumulation (Chételat et al 2015). Simulations of the effect of climate warming on the bioaccumulation of toxic elements (Gouin et al 2013) indicate that the immediate consequences of this warming is higher levels of metabolism and food consumption by fish.
Figure 7. Dependences of Cd content in the kidneys of predatory fish or the sum of air temperatures above 10 °C for the region.

Climatic changes significantly indirectly affect aquatic environments through changing parameters of their water chemistry such as pH, concentrations of salts and biogenic elements, and oxygen concentration (Freitas et al 2019). Prediction data indicate that an increase in the air temperature by 2 °C will induce an increase in the phosphorus concentration by 25%–30% in the northern regions and 40% and more in the southern ones. A temperature increase of 2 °C was predicted to result in a more than 30% salinity increase in the northern regions (Moiseenko et al 2013). The eutrophication of water bodies will also lead to intense Hg methylation, and this will make this element more bioavailable.

An increase in the Ca concentration suppresses the accumulation of some elements in living organisms, and organic matter deactivates most metals (Moiseenko et al 2020). A temperature increase will lead to an increase in the salt concentrations, and the penetration abilities of metals such as Cd and Pb will decrease in more strongly mineralized waters. Model calculations in (Gouing et al 2013) indicate that the penetration ability of metals into the organism of codfish (Gadus morhua L.) will decrease with climate warming because of an increase in concentrations of inorganic salts and nutrients in the water.

Some associated factors may also result in even more clearly discernible effects than a temperature increase. For example, the bioaccumulation of Cd is more active at low pH and low Ca concentrations. Our data indicate that an increase in the Ca concentration from the northern to southern regions is associated with an increase in the sum of active temperatures of the vegetation period. Consequently, Cd accumulation in the kidneys was determined to be negatively correlated with the temperature sum (figure 7).

The bioaccumulation of Pb displays no temperature dependences. For example, the thermal effect of the Kola Nuclear Power Plant did not in any way modify the bioaccumulation of Pb in the tissues and organs of whitefish (Coregonus lavaretus L.) in Imandra Lake (Gashkina and Moiseenko 2020). Neither does thermal pollution in any way affect Pb bioaccumulation by concentrators of oysters in a littoral zone impacted by the discharge of warm water from the Houshi Power Plant in China (Lan et al 2020). At a significant temperature gradient in the examined water bodies, we acquired a dependence of Pb accumulation in the gills of benthophage and predator fish on the temperature sum in the region above 10 °C and on Ca concentration in the water. These dependences indicate that Pb accumulation in fish more strongly depends on the salt composition of the water. Inasmuch as an increase in the effective temperature in this region is associated with an increase in Ca concentrations in the waters, it seems to be the high Ca concentrations in the waters that suppress Pb penetration in the fish organism.

For predators, the dependence showing how temperature and Ca concentration in the water affect Pb bioaccumulation is as follows:

\[
\text{Pb}_{\text{gills}} = 0.210 - 0.011 \times \text{Ca}_{\text{water}} + 0.074 \times 10^{-3} \times \Sigma T > 10 \degree \text{C}, \ r = 0.769, \ p < 0.05
\]
more intense at higher temperatures. The effects of climate warming on Cd and Pb accumulation may perhaps manifest themselves in waters of similar salt composition and with low Ca concentrations.

5. Conclusion

It has been obviously proved that various fish species differently accumulate metals (Hg, Cd and Pb) at their low concentrations in the waters. Predator fish (salmon trout, pike and perch) usually accumulate more metals than benthophages do (whitefish and bream). If the water contamination is relatively insignificant, the elements are simultaneously accumulated in fish organs and tissues, and the concentrations of the metals reach higher levels in the kidneys and liver. The exception is Hg, which is almost equally intensely accumulated in the liver and muscles.

Our data acquired on the bioaccumulation of metals in fish in various water bodies in Russia along a latitudinal gradient (from northern Arctic lakes to southern mouth parts of the Volga River) show how the accumulation of elements depends on the aquatic environments and temperature.

Our results demonstrate that the aquatic environment and habitats of fish significantly affect the bioavailability of elements. At low Hg concentrations in the waters, its accumulation in pike and perch exhibits a definite and reliable dependence on organic matter content in the water. In the habitats of the predators (pike and perch), which are constrained to thickets of aquatic plants, Hg is more intensely methylated, and this results in its more intense accumulation in the fish. Low pH of the waters can also significantly increase Hg concentrations in the organisms of northern predatory fish.

Higher Cd concentrations are accumulated in the kidneys of fish at low pH and low Ca concentrations in the waters. The highest Cd concentrations are reached in the kidneys of fish in northern regions, in which Cd accumulation is likely facilitated by the leaching of the element by acid atmospheric precipitation. The accumulation of this metal in the kidneys of fish is reliably proved to be negatively correlated with the Zn concentration, i.e. the availability of Zn (an essential element) for the organism may decrease if the waters are contaminated with Cd.

Pb is accumulated in fish in water bodies in industrially developed and urban regions, such as the middle and lower stream of the Volga. Another factor on which Pb accumulation depends is Ca concentrations in the waters, with high concentrations of the latter element in the waters suppressing Pb accumulation.

Multipollution of the investigated waters leads to pathological disorders in functionally important fish organs: liver, kidneys, gills. Comparison of the calculated data on the critical values of the admissible daily fish muscle intake showed that the content of Hg, Cd and Pb in the muscles of fish that are eaten is below the critical levels.

Our data on fish from numerous water bodies scattered over a large territory of Russia indicate that climate affects the accumulation intensity of toxic elements. We acquired reliable dependences of Hg accumulation in fish on water temperature. These dependences indicate that an increase in the water temperature results in more intense Hg accumulation in the fish. The accumulation of the other metals strongly depends on Ca concentration in the waters, and the elements are more actively accumulated in the fish at low Ca concentrations. Climate warming leads to an increase in salt concentrations in the waters and, hence, suppresses the penetration of such metals as Cd and Pb into the fish organism.

Studies of the bioaccumulation of metals and their toxic impact must be carried out with regard to the effects of aquatic environments and temperature, which can significantly modify the bioavailability of the metals. This will also be taken into accounts in developing standards for water quality and hazards that emerge for the human population with climate warming.

Hg, Cd and Pb concentrations found in the muscles of the fish used for food were much lower than the standards recommended for the maximal concentrations of the elements that are not harmful to human health. However, it is necessary to take into account the possible consequences of the long-term use of fish for food and the hazards of bioaccumulation for human health when fish is consumed.

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Data availability statement

All data that support the findings of this study are included within the article (and any supplementary information files).

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