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Ecosystem and human health assessment in relation to aquatic environment pollution by heavy metals: case study of the Murmansk region, northwest of the Kola Peninsula, Russia

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Abstract

Throughout the Euro-Arctic region of Russia (Murmansk region), there is a substantial increase of metal concentrations in water, which are related to local discharges from the metallurgical and mining industry, transboundary pollution, as well as indirect leaching of elements by acid precipitation. This study collates data to investigate the relationship between surface water contamination by metals, and fish and human health. Fish are used as a biological indicator to show the impact of water pollution by metals on the ecosystem's health. The etiology of fish and human diseases are related to the water pollution and accumulation of metals in organisms. High concentrations of Ni and Cd in water drives an accumulation of these elements in organs and tissues of fish, especially in kidneys. The relation between the accumulation of Ni in kidneys and the development of fish nephrocalcinosis and fibroelastosis was established. Statistical analysis demonstrated that human populations in cities close in proximity to smelters show the highest incidence of disease. The results of histological, clinical, and post-mortem examination of patients shows the highest content of toxic metals, especially Cd, in livers and kidneys. Our complex investigation of a set of disorders observed in fish and human populations indicates that there is a high probability that the negative impact on human health is caused by prolonged water contamination by heavy metals. As a novel finding, this paper shows that based on the similarity of pathological processes and bioaccumulation of metals in fish and humans, examining the content of heavy metals in fish can be used to confirm etiology and evaluate the potential risk to human health by pollution of surface waters.

1. Introduction

Assessment of the ecological consequences of changes in geochemical cycles due to the impact of mining and the metallurgical industry is of great importance for the health of the environment. There has been a great deal of research that has demonstrated the negative effect of environmental increases in concentrations of metals on animal and human health (Seiler *et al* 1994, Nishijo *et al* 2000, Watras and Huckabee 2002, Satarug *et al* 2003, Satarug *et al* 2010, Revich *et al* 2003, Hoffman *et al* 2005, Nordberg *et al* 2007).

It is not always possible to establish a direct correlation between water quality and prominent diseases within human populations. Therefore, it is important to find supporting evidence of adverse effects on aquatic bioindicators to determine the etiology of diseases, thereby confirming the connection with poor water quality. Numerous publications confirm that fish (*in situ*) are a good indicator of aquatic environmental and ecosystem health, especially in cases of toxic water pollution (Whitfield and Elliott 2002, Yeom and Adams 2007, Moiseenko and Kudryavtseva 2002, Moiseenko *et al* 2008). Pathological changes to the organs of fish give an indication of water toxicity and the potential danger of anthropogenic contaminants entering aquatic environments. Therefore, pathophysiological parameters of fish can be used as a criterion for assessing water quality and ecosystem health (Adams and Ryon 1994, Attrill and Depledge 1997). In our studies for several lakes and rivers in Russia, we use the ecotoxicological approach for the assessment of water quality and ecosystem health based on the registration of pathological changes in fish. These studies were performed for several arctic lakes (Moiseenko and Kudryavtseva 2002, Moiseenko *et al* 2006), as well as for the largest river in Europe, the Volga (Moiseenko *et al* 2008). If physiological disorders within fish are similar, and the etiology of their diseases are associated with toxic metals, then this could be one of the arguments in favor of a possible negative effect of water pollution on human health.

In 2017 the project 'Developing of methodologies for monitoring, assessment, forecasting and prevention of risks related to transfer of toxic pollutants through biological pathways capable of accumulating in trophic chains and spreading in Arctic ecosystems' was initiated in Russia. One of the main goals of this project is to provide scientific investigations to evaluate the demand of organizing systems for biological monitoring at the federal level.

Evidence of a solid relationship and interconnection between water quality, fish pathology, and human health in this article is realized for the first time using studies in the industrially developed Euro-Arctic region.

The objectives of this study were as follows:

i. identifying the levels of water contamination by metals, and the quality of drinking water;
ii. study of pathophysiological parameters of fish as a criterion for assessing water quality and environment health;
iii. analysis of population morbidity and metal accumulation within organs, and assessments of water quality within Arctic industrial regions.

2. Characteristics of the region

The Murmansk region is situated above the Arctic Circle in the Kola Peninsula, Russia. Among Arctic regions, this region is the most densely populated and has the highest industry development. Copper-nickel, ferrous, apatite-nepheline, and rare element ores are presently exploited. More than 70 years of mining and smelting operations by the companies Severonickel and Pechenganickel has led to the failure of purification treatments to remove toxic metals from the water. The annual emission of dust in the region up to the year 1990 was 64 000 t year^{-1}, including 2460 of Ni and 1600 of Cu t year^{-1}. In addition to the metal pollution, sulfur dioxide emission was about 500 000 t year^{-1} (Moiseenko 1999). Impact zones, pollution within a 30 km radius, emerged from where the concentrations of Cu and Ni were equal to toxic levels from 1980–1990 (Ni up to 30–40, and Cu10–15 μg l^{-1}) (Moiseenko 1999). For the assessment of metal toxicity in surface water for fisheries and aquatic life, the following concentrations (μg l^{-1}) are legislatively accepted in Russia and are used in this work (units: μg l^{-1}): Cd = 5, Ni = 10, Cu = 1, Pb = 10, Hg = 5, Cr = 1, Zn = 10, and As = 50 (Filatov 1989).

Here, large apatite-nepheline mining gave rise to the increase in pollution of Sr, Al, and other elements. Water acidification has resulted in mining waste rocks and tailings, and the increased mobility of Al and Cd from soil and rock geochemical anomalies (Moiseenko 1999). Moiseenko and Kudryavtseva (2002) described the zoning of the Murmansk region by type and concentration of metal pollution.

The transformation of economic conditions in Russia at the beginning of the 1990s brought a stop to many industrial activities, and, accordingly, slowed down lake pollution. Some economic revival in the last decade was spurred by technology modernization, and there are more restrictions on the pollution of lakes and the atmosphere. As a result, pollution entering surface water has decreased by more than ten times over the past 25 years (Moiseenko *et al* 2009).

As paradoxical as it may seem, the domestic water supply to major towns, encompassing approximately 300 000 residents, is often taken from surface water bodies receiving effluents from industrial plants or located in the zone of airborne pollution. Increased morbidity of nephrolithiasis and cholelithiasis in the local population was found in 1970 (Atlas of Murmansk region 1971). In certain cases (as with surface water contamination), it is difficult to show a direct relationship between water pollution and human health. Hazardous substances found in the human body may originate from food, water, and the atmosphere. Therefore, biological indicators may serve as evidence of the etiology of diseases caused by water contamination. Fish are good markers to use for the estimation of water quality (Whitfield and Elliott 2002, Yeom and Adams 2007, Moiseenko *et al* 2008).

3. Materials and methods

Multidisciplinary studies were carried out in industrially developed towns (Monchegorsk, Apatity, and Olenegorsk) and in more remote settlements (Alakurti and Lovozero) that used surface water sources for their drinking water supply. The schematic layout of major industrial facilities and population localities, water intakes for drinking water supply, and fish catching is shown in figure 1.

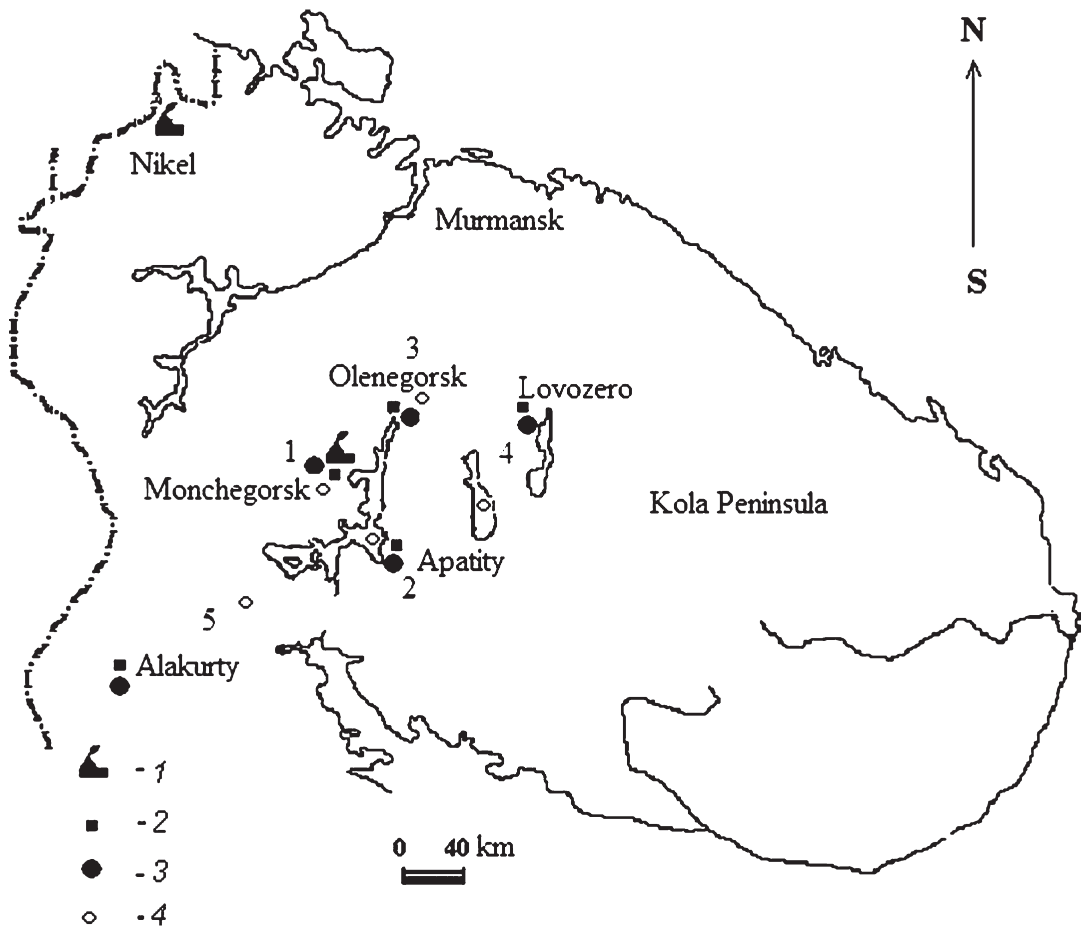

Figure 1. Layout of (1) copper-nickel smelters, (2) population localities, (3) drinking water intakes, and (4) fish catching.

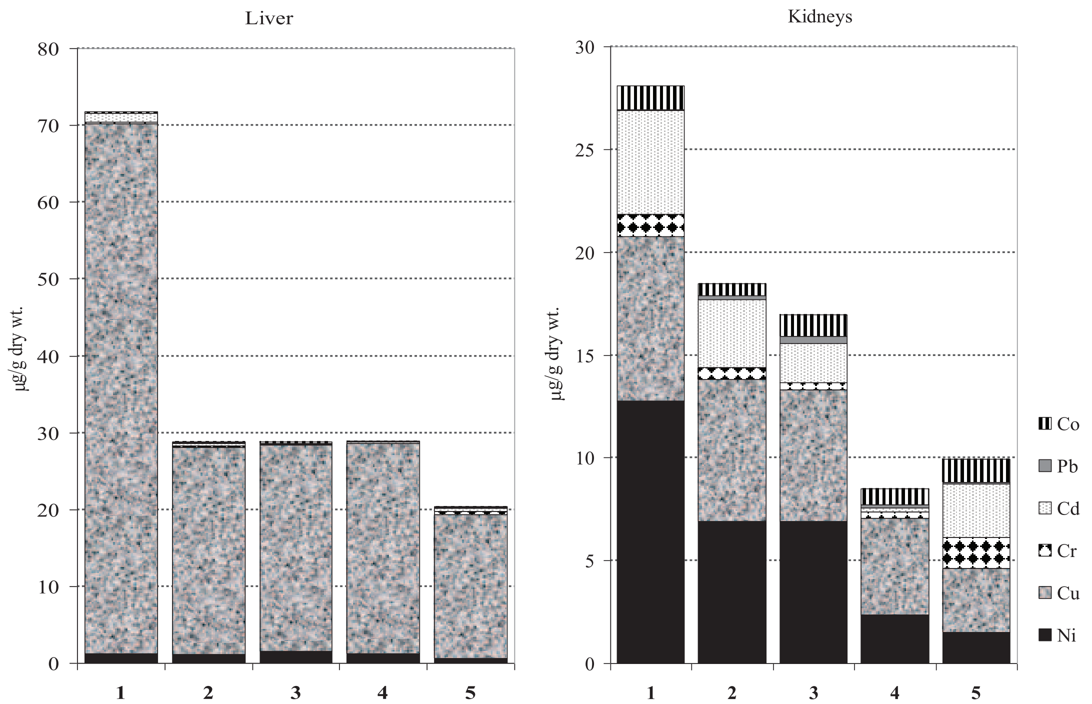

Figure 2. Metal content in liver and kidneys of the examined fish in lakes near the water intake stations: (1) Monche Lake, near the drinking water intake Monchegorsk; (2) Imandra Lake, near the water intake station Apatity; (3) Kolozero Lake, near the water intake Olenegorsk; (4) Umbozero Lake, near Lovozera; (5) Pirenga Lake, near Alakurti.

Domestic water supply to the population of Monchegorsk is taken from Monche Lake, which lies in the area (<10 km) of airborne pollution by emissions from the copper-nickel smelter Severonikel. The town of Olenegorsk is also situated within the propagation zone of smoke emission, but at a greater distance (>50 km). The town of Apatity is supplied with water originating from Imandra Lake. Water intake for Apatity is located in the zone of transit flow of wastewater from the copper-nickel smelter Severonikel (about 200 km away), and municipal wastewater is released into this lake. Water to the Lovozero and Alakurti settlements is supplied by surface water bodies; however, they are situated far enough from industrial centers (>200 km from the smoke emission) to be relatively weakly impacted by pollution.

In these cities during the period of 1998–2002, quasi-synchronous studies were conducted: (i) determining water chemistry in lakes (at water intake sites), (ii) assessing water quality in pipelines, (iii) catching fish for the investigation of metal content in organs and assessing fish morbidity, and (iv) collecting data of the population morbidity, and post-mortem examination of patients to investigate metal content in organs.

Studies of long-term dynamics of water quality and fish morbidity are based on the author's data collected over several years (from 1981–2013) at Imandra lake. This lake has been selected in accordance with our project goals and to meet the requirement to rate contamination over time (since the period of maximum pollution is in the 1980s). Data of this long-term investigation includes water quality, and health conditions of fish, including diseases and bioaccumulation of metals (Ni, Cu, Co, Zn, Fe, Mn). The content of metals, such as Hg, Cd, and Pb, was rated in water and fish from 1996.

3.1. Water sample collection

Water samples collected from water supply sources as well as from reticulation networks for water supply to the population were analyzed for water quality. Water samples were collected in Nalgen® polyethylene bottles. Water samples from lakes were always taken at the precise sites where fish were caught for examination. The samples were immediately placed in dark containers, and were cooled to about 4 °C while being transported to the laboratory. Water chemistry analysis for Ca^{2+}, Mg^{2+}, K^+, Na^+, alkalinity, $SO_4{}^{2-}$, Cl^-, color, $NO_3{}^-$, $NH_4{}^+$, total N, (TN), $PO_4{}^{3-}$, total P, and Si were carried out by standard techniques (APHA 1992). A Metrohm® pH meter was used to measure pH. Conductivity (20 °C) was measured with a Metrohm® conductivity meter. Alkalinity was measured using the Gran titration method. Organic matter content was measured using the Mn oxidation method. Al, Fe, Sr, Mn, Zn, Ni, Co, Cd, Pb, and As concentrations were determined by atomic absorption spectroscopy (PerkinElmer 5000 model, Corp., Norwalk, USA, equipped with a graphite furnace HGA-400). Standard solutions with appropriate concentrations of each element were made from 1000 ppm atomic absorption spectroscopy (AAS) stock standards (Merk, Darmstadt, Germany). Hg concentrations was determined using the PerkinElmer Hg analyzer FIMS-100. Standard solutions with appropriate concentrations of each element were made from 1000 ppm stock AAS standards (Merk, Darmstadt, Germany). Determination limits were 0.01, 0.07, 0.002, 0.05, and 0.6 $\mu g \cdot l^{-1}$ for Cu, Ni, Cd, Pb, and Hg, respectively. The quality of the analytical results was tested annually by intercomparisons (Intercomparison 1998–2002).

3.2. Fish study

Special permit for fish catching was obtained for research purposes annually from the Murmansk Fisheries Committee. Investigations of fish were conducted in lakes near the water intake stations: (1) Monche Lake, near the drinking water intake for Monchegorsk; (2) Imandra Lake, near the water intake station for Apatity; (3) Kolozero Lake, near the water intake for Olenegorsk; (4) Umbozero Lake, near Lovozero; and (5) Pirenga Lake, near Alakurti. We also analyzed the historical data on fish disease and metal content in fish in Lake Imandra during very high water pollution.

Whitefish (*Coregonus lavaretus* L.) were used as the bioindicator. The minimum number of fish observed was 200 of the same age (from 4–6 years old); all were free of internal parasites during the investigation period (September).

Macrodiagnostics to determine fish health were carried out under field conditions. The clinical and pathological anatomical signs of intoxication and any abnormalities were documented on the basis of visual examination of the fish during the first hour after fishing. The clinical symptoms of fish intoxication were as follows: de-pigmentation or change in body color, breach of fin edging, swelling of the skin, the presence of hematomas, dullness of eye cornea, anal inflammation, deformation of skull bones, low calcification, and scoliosis of the spine. By pathologo-anatomical dissection, we documented changes in organs: dark-red and oedema gills or pale ones with an anemia rim along the gill-arch; for the liver, color changes, and an increase of size, and friability, necrosis, and signs of atrophy; for kidneys, color changes, connective tissue growing inside the organ, state of ducts and the presence of nephritic calculi, and breaches of gonad form and structure.

For more precise microdiagnostics, the organs of fish with overt signs of pathology were sampled for histological analysis. Histological sections were prepared in the laboratory according to the standard method (Bucke 1994). For satisfactory histological preparations only freshly killed fish were considered. Diagnosis of disease was confirmed on the base of histopathological observations. The percentage of sick fish in the stock of each local polluted zone was documented. Based on our knowledge of histopathological alterations of organs, we selected the most informative intoxication symptoms: external signs of liver, gill, and kidney pathology. The kidney pathology was the most widespread for fish of Kola lakes (Moiseenko and Kudryavtseva 2002). The percentage of fish with intoxication symptoms in polluted lakes was used as the criterion (biomarker) of metal negative effects on the fish.

For determination of the metal content in the fish bodies, subsamples from at least ten individual fish were collected from the liver, kidneys, and muscle. Biological samples 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 for 1 g of tissue). Concentrations of elements (Cu, Ni, Cd, Pb) were determined using a graphite furnace atomic absorption spectrophotometer (Analyst-800) with a Zeeman background corrector. Hg concentration was determined using an FIMS-100 Hg analyzer (PerkinElmer). Determination limits were as follows: Cu = 0.05, Ni = 0.02, Cd = 0.002, Pb = 0.01, and Hg = 0.01 μg·g^{-1}.

3.3. Population morbidity

Before starting this research, consent was obtained from the Murmansk Committee of the Ministry of Health (Resolution number 1230–344451, P-645). This research was based on the classic, post-mortem examination of human pathology, and analytical chemistry (bioaccumulation) and histology. Examinations were carried out in pathology laboratories in local hospitals. Pathologo-anatomical autopsy, the description of pathology, and selection of biomaterial were performed 2 h after ascertaining the fact of death of patients according to the established order (Order of the Ministry of Health and Medical Industry of the USSR on 29 April 1994; No. 82 'On the procedure of post-mortem autopsy, section 4: The order of the autopsy'). The studies did not use biomaterials of living people, and no intravital biopsy or histology was performed. This research abided by the ethics presented in the Council of Europe Convention on the Protection of Human Rights and Dignity with regard to the Application of Biology and Medicine, the Convention on Human Rights and Biomedicine, adopted in Oviedo (Spain) in 1997; additional protocol followed the Council of Europe Convention on Biomedicine and Human Rights, concerning Biomedical research (Belousov 2005).

We selected dead patients who spent no less than 10 years in the cites and settlements of interest, who never worked at factories or other places with high health risks, and who had no chronic alcoholism or viral hepatitis. In total, 110 patients aged 35–60 were examined post-mortem. The main objective was to find out the forms of illnesses related to liver and kidney disturbances.

To assess toxic metal accumulation within the tissues of residents of these populated localities, post-mortem samples were taken (while carrying out routine post-mortem examinations) from the livers and kidneys. Post-mortem samples were additionally taken for histological examination, which was carried out by conventional methods. Determination of metal concentrations in liver and kidney samples was carried out by the same method as in fish (the method described above). A minimum of ten post-mortem kidney and liver samples were selected from each city for this analysis. Appropriate tissues from dead fetuses (22 stillborn and immature births) were considered to be the reference for trace element in organs.

Mathematical analyses of data were performed using Statistics Package 8. The following statistical methods were used: descriptive statistics, correlation matrix, and *t* test for independent samples. During the correlation analysis of the dependencies of elements accumulation in the organs of postmortem patients, as well as their diseases and pathologies of systems and organs, with concentration in drinking waters, the latter were used as mean values for each city or village, because it is impossible to relate (to correlate) a posthumous patient to a specific sample of tap water in each city or village.

4. Results

4.1. Water chemistry

The chemistry of lake water and the drinking water supply for towns and settlements is given in table 1. Water that is supplied to Monchegorsk is characterized by a relatively high content of Ni (11.6 ± 1.4 μg l^{-1}), Cu (12.1 ± 1.3 μg l^{-1}), Cd (0.30 ± 0.05 μg l^{-1}), and other metals. The concentrations of metals found in Lake Imandra (the water supply of Apatity) are lower (Ni = 8.1 ± 1.0 μg l^{-1}) than in the water supplied to Monchegorsk, but are higher than background values. Metal concentrations in the water supply for the Lovozero and Alakurti settlements are the lowest, and very close to regional background values.

The metal concentrations in drinking water after being processed in the water treatment system is only slightly lower than that in lake water. In urban areas, tap water is relatively more contaminated than water from natural sources. For instance, in Monchegorsk, the Ni content has increased on average to 15.6 ± 2.5μg l^{-1}. Fe in lake water was close to natural concentrations (30–50 μg l^{-1}), but in drinking water from the pipelines the Fe content was much higher, especially in the cities of Monchegorsk (162 ± 33 μg l^{-1}), Apatity (255 ± 49 μg l^{-1}), and Olenegorsk (106 ± 19μg l^{-1}).

Historical data of the metal concentration in waters of Imandra Lake over years of research are presented in table 2.

The highest metal concentration (Ni up to 50 μg l^{-1}) was observed in the 1980–1990s. In the last decades, the Ni contents decreased by up to 7–15 μg l^{-1}.

Table 1. Water chemistry of (1) water supply source and (2) drinking water for several cities and settlements of the Murmansk region. Present mean value and standard error; n—the number of water samples.

Parameter	Monchegorsk		Apatity		Olenegorsk		Lovozero		Alakurty	
	1	2	1	2	1	2	1	2	1	2
n	15	26	12	25	9	12	9	12	8	10
pH	6.90 ± 0.05	6.53 ± 0.07	7.28 ± 0.05	7.25 ± 0.03	6.99 ± 0.05	6.81 ± 0.10	6.70 ± 0.18	6.76 ± 0.20	7.53 ± 0.12	7.63 ± 0.06
Conductivity, μSm cm^{-1}	29 ± 1	30 ± 1	99 ± 3	105 ± 1	41 ± 2	37 ± 1	38 ± 7	49 ± 9	156 ± 11	144 ± 6
Ca, mg l^{-1}	2.45 ± 0.07	2.27 ± 0.03	4.19 ± 0.10	5.40 ± 0.09	2.50 ± 0.13	2.34 ± 0.10	2.63 ± 0.12	2.78 ± 0.27	14.7 ± 0.8	13.8 ± 0.6
Mg, mg l^{-1}	0.78 ± 0.02	0.74 ± 0.01	1.07 ± 0.03	1.22 ± 0.01	1.29 ± 0.08	1.15 ± 0.03	1.48 ± 0.06	1.60 ± 0.17	4.92 ± 0.26	4.50 ± 0.24
Na, mg l^{-1}	1.59 ± 0.07	1.88 ± 0.11	13.3 ± 0.3	13.9 ± 0.2	3.24 ± 0.27	2.81 ± 0.11	2.45 ± 0.19	2.44 ± 0.22	10.8 ± 0.51	9.80 ± 0.47
K, mg l^{-1}	0.45 ± 0.01	0.44 ± 0.01	2.50 ± 0.06	2.25 ± 0.01	0.72 ± 0.04	0.67 ± 0.03	0.59 ± 0.04	0.63 ± 0.04	2.00 ± 0.05	1.91 ± 0.04
HCO_3, μg l^{-1}	139 ± 6	100 ± 7	348 ± 4	379 ± 5	212 ± 8	167 ± 20	185 ± 22	148 ± 19	1395 ± 87	1295 ± 46
SO_4, mg l^{-1}	4.15 ± 0.14	4.00 ± 0.11	21.1 ± 0.5	21.0 ± 0.3	3.38 ± 0.18	2.82 ± 0.12	2.15 ± 0.27	1.98 ± 0.24	10.6 ± 0.44	9.92 ± 0.37
Cl, mg l^{-1}	1.41 ± 0.23	2.88 ± 0.27	4.93 ± 0.25	5.96 ± 0.11	3.02 ± 0.32	3.76 ± 0.43	3.76 ± 0.51	4.57 ± 0.55	3.73 ± 0.30	3.24 ± 0.23
ermanganate consumption, mgC l^{-1}	3.70 ± 0.33	3.06 ± 0.12	2.76 ± 0.15	1.96 ± 0.07	6.87 ± 0.40	6.93 ± 0.43	9.66 ± 0.68	8.28 ± 0.61	0.86 ± 0.18	0.81 ± 0.09
Fe, μg l^{-1}	49 ± 13	162 ± 33	31 ± 7	255 ± 49	61 ± 8	106 ± 19	815 ± 65	742 ± 59	1287 ± 129	739 ± 97
Al, μg l^{-1}	39 ± 6	38 ± 4	48 ± 10	13 ± 2	34 ± 9	31 ± 6	99 ± 22	78 ± 20	2 ± 1	3 ± 1
Ni, μg l^{-1}	11.6 ± 1.4	15.6 ± 2.5	8.1 ± 1.0	5.2 ± 0.2	1.5 ± 0.2	1.1 ± 0.2	0.5 ± 0.1	0.6 ± 0.2	0.5 ± 0.1	0.4 ± 0.1
Cu, μg l^{-1}	12.1 ± 1.3	15.4 ± 2.6	4.1 ± 0.5	3.2 ± 0.3	2.9 ± 0.4	1.9 ± 0.2	0.8 ± 0.2	1.2 ± 0.3	0.7 ± 0.2	0.8 ± 0.3
Cd, μg l^{-1}	0.30 ± 0.05	0.19 ± 0.02	0.16 ± 0.05	0.15 ± 0.03	0.10 ± 0.03	0.09 ± 0.02	0.13 ± 0.03	0.13 ± 0.02	0.11 ± 0.02	0.14 ± 0.02
Pb, μg l^{-1}	<0.5	<0.5	1.5 ± 0.4	0.6 ± 0.1	<0.5	<0.5	<0.5	<0.5	<0.5	<0.5
Sr, μg l^{-1}	20 ± 1	17 ± 1	72 ± 4	90 ± 2	37 ± 3	35 ± 2	51 ± 8	49 ± 4	160 ± 8	145 ± 6
Cr, μg l^{-1}	0.6 ± 0.1	0.3 ± 0.0	0.3 ± 0.0	0.2 ± 0.0	0.2 ± 0.0	0.3 ± 0.0	0.3 ± 0.0	0.3 ± 0.0	0.1 ± 0.0	0.1 ± 0.0
Co, μg l^{-1}	0.4 ± 0.1	0.5 ± 0.1	0.3 ± 0.0	0.3 ± 0.1	<0.2	<0.2	0.3 ± 0.0	<0.2	<0.2	<0.2

Note: The bold text in the table refers the Ni, Cu, and Cd concentrations that reliably differ (with significance level $p < 0.05$) in the sources of water supply and drinking water as compared to those in the village of Alakurty.

Table 2. Retrospective data reflected of Imandra Lake pollution over various years of research: characteristics of whitefish diseases (%), and heavy metals concentrations in water and accumulation in fish livers and kidneys (present mean values and standard errors).

	Year of investigation	1981	1986	1991	1996	2003	2013
Metal concentration in water, mean value and standard error, μg L−1							
	Ni	42.5 ± 3.6	46.0 ± 3.9	25.7 ± 3.1	15.6 ± 2.5	11.3 ± 1.1	7.7 ± 1.7
	Cu	3.8 ± 0.3	11.3 ± 0.5	15.6 ± 1.9	6.1 ± 0.6	5.5 ± 0.4	13 ± 0.5
	Pb	n/d	n/d	n/d	0.5 ± 0.1	0.3 ± 0.0	0.5 ± 0.0
	Cd	n/d	n/d	n/d	0.27 ± 0.03	0.15 ± 0.03	0.5 ± 0.01
	Hg	n/d	n/d	n/d	<0.01	<0.01	0.02
The main symptoms of fish diseases, % from number of the surveyed individuals							
	Nephrocalcinosis	52	47	45	14		
	Fibroelastosis	48	53	55	48	39	19
	Lipoid degeneration of liver, and cirrhosis	100	89	78	48	39	24
	Gonad structure anomalies	34	27	8			4
	Number of investigated fish	$n = 788$	$n = 721$	$n = 453$	$n = 462$	$n = 235$	$n = 206$
Metals accumulation in fish organs, mean value and standard error, μg g^{-1} dry weight							
Ni	Liver	3.8 ± 1.3	2.1 ± 0.2	2.6 ± 0.4	1.3 ± 0.1	1.2 ± 0.1	2.2 ± 0.3
	Kidney	18.9 ± 3.6	26.6 ± 3.8	13.1 ± 1.5	13.8 ± 2.1	7.4 ± 0.5	5.6 ± 0.6
Cu	Liver	20.4 ± 4.3	27.4 ± 2.4	34.8 ± 3.5	36.6 ± 4.0	33.2 ± 2.8	36.8 ± 3.9
	Kidney	6.7 ± 0.8	4.2 ± 0.3	6.6 ± 0.7	5.6 ± 0.5	6.9 ± 0.2	4.1 ± 0.4
	Muscles	2.3 ± 0.4	0.42 ± 0.04	1.2 ± 0.2	1.1 ± 0.1	0.81 ± 0.07	0.81 ± 0.04
Pb	Liver	n/d	n/d	n/d	0.21 ± 0.06	0.13 ± 0.03	0.07 ± 0.01
	Kidney	n/d	n/d	n/d	0.32 ± 0.05	0.33 ± 0.06	0.06 ± 0.01
Cd	Liver	n/d	n/d	n/d	0.83 ± 0.15	0.39 ± 0.06	0.42 ± 0.11
	Kidney	n/d	n/d	n/d	2.9 ± 0.6	3.0 ± 0.4	2.8 ± 0.6
Hg	Liver	n/d	n/d	n/d	0.14 ± 0.01	0.09 ± 0.01	0.38 ± 0.03
	Kidney	n/d	n/d	n/d	0.19 ± 0.05	0.04 ± 0.00	0.22 ± 0.02

n/d—no data.

At the same time, other toxic metals decreased in the water.

4.2. Metal accumulation and fish pathology

Figure 2 shows the metal content in the liver and kidneys of fish in lakes near the studied cities and towns in the last period of study.

A very high content of Ni, Cu, and Cd are found in fish from lakes near Monchegorsk. The following metals accumulated most in the fish livers: Cu (up to 60–70 μg g^{-1} dry weight), Ni (up to 12–15 μg g^{-1} dry weight), and Cd (up to 5–8 μg g^{-1} dry weight). In the kidneys, mostly Ni (up to 12–15 μg g^{-1} dry weight) and Cd (up to 10–15 μg g^{-1} dry weight) accumulated. The content of these elements in the fish organs is much lower (especially Ni) in the lakes located far from the city (near Lovozera and Alakurti). At the same time, despite the distance, the Cd content in the kidneys of fish is too high, reaching to 1.9–5.1 μg g^{-1} dry weight (except for the fish from the Lovozero Lake). According to the *t* test, the accumulations of Cu in the liver, and Ni and Cd in the kidneys of fish from the lake near Monchegorsk are significantly higher (with significance level $p < 0.05$) than in fish from lakes in other cities and towns.

Table 2 summarizes the historical data concerning fish pathology in Imandra Lake, and the metal concentrations in fish over the past 30 years. Kidneys of fish are capable of accumulating high levels of Ni and Cd. Ni content in the kidney during a period of strong pollution (1981) exceeded the level of 2013 by three to five times. The content of Cd in the kidney as compared with whitefish from other northern regions was an order of magnitude higher. A tendency towards lower Cd and Pb content in fish was observed between 1996 and 2013. However, there was a reverse tendency for Hg. The highest liver concentrations of Hg were observed during the last period of the study.

In areas of the Kola Peninsula polluted by Ni–Co smelters, the most serious abnormalities—nephrocalcinosis and fibroelastosis—were exhibited in kidneys. Historically the most frequently occurring fish pathology is kidney stone illnesses. As the level of water pollution decreases, the frequency of identifiable diseases is also decreased. In the case of fibroelastosis, visually determined granulation on the hystological cut-offs is verified by connective tissue growing inside the parenchyma, and fibrous kidney texture. Morphological changes in the liver manifest in the form of lipoid dystrophy, which is a symptom of progressive hepatopathy. In the case of lipoid dystrophy, fat occlusions, which almost completely fill the cells, appear in the hepatocytes. Diffuse disruptions of fish liver, accompanied by disturbances in the morphological structure of liver lobules and pronounced necrosis of liver tissue, were also diagnosed. Histological analysis of hepatic diseases shows that different liver abnormalities (from initial to debilitative) were frequently detected in fish living in water contaminated by metals. The fish morbidity during various periods of time is presented in table 2.

In the 1980s, when the level of pollution was insignificant, almost all species examined had signs of morbidity in varying degrees of severity; during the last time period, only 24% of fish examined had signs of intoxication.

4.3. Metal accumulation and human population morbidity

Figure 3 shows the metal content in post-mortem samples of the liver and kidneys of inhabitants of the studied cities and towns.

According to the *t* test, the accumulation of Cu in the liver of residents from all the cities and towns is significantly higher (with a significance level $p < 0.05$) than in the control group. Furthermore, the content of Ni is significantly higher in the inhabitants of Monchegorsk, Olenegorsk, and Alakurtti, Co is significantly higher in the inhabitants of Monchegorsk, Apatit, and Olenegorsk, Cd is significantly higher in the inhabitants of Apatit and Monchegorsk, and Pb is significantly higher among the inhabitants of Monchegorsk and Alakurtti. At the same time, the accumulation of Cu, Cd, and Pb in the kidneys in the inhabitants of Monchegorsk, Apatit, and Olenegorsk is significantly higher than in the control group, Ni is significantly higher in the inhabitants of Monchegorsk, Apatit, and Olenegorsk, and Co is significantly higher in the inhabitants of Monchegorsk and Olenegorsk.

The concentration of heavy metals in livers of post-mortem samples of Monchegorsk's inhabitants was >2 times higher than the normal concentrations expected for many metals, especially for Ni, Cu, Co, Cd, and Pb. Cd concentrations, found in kidney tissues, were higher than in reference groups by >5 times. Compared with the reference group, accumulation of Cu and Cd within livers and kidneys is typical for Apatity inhabitants, accumulation of Ni, Cu, Co, and Cd is typical for Olenegorsk inhabitants, and accumulation of Cu is typical for Lovozero and Alakurti inhabitants.

The primary objective of this human population morbidity investigation was to determine pathological processes within the liver and kidneys that developed latently and were not diagnosed in the patient's lifetime. In 24 of the 110 cases observed, pathological processes were not diagnosed in the patient's lifetime. The histological pattern of liver pathogenesis was mostly represented by fatty degeneration of hepatocytes, its sporadic necrosis, and autolytic degradation in the peripheral area with the formation of fat–protein detritus. In the 110 patient examinations, one case of hemosiderosis, five cases of toxic destruction, four different types of dystrophy, and ten cases of fatty degeneration were reported.

The histological picture of kidney pathogenesis was represented by nephrosclerosis at the initial development stage by glomerulonephritis and amyloidosis events. These diseases are noninfectious and polyetiologic, which does not exclude its toxic pathogenesis impacted by metals. In addition to inflammatory changes in kidneys, specific focal dystrophic and necrobiotic changes in blood vessels (capillaries, precapillaries, capillary veins) were detected. Moreover, 36 cases of urolithiasis among the patients were identified during post-mortem examination.

5. Discussion

5.1. Water quality assessment

Surface water in Arctic regions is characterized by low Ca content and low salt content; moreover, this water is vulnerable to airborne contamination or contamination from industrial effluent (Arctic Pollution Issues 1997, Henriksen *et al* 1998). In the past, surface waters in the Murmansk region were not contaminated, and were characterized by low salt and nutrient content (e.g. total phosphorus $< 2\ \mu g\ l^{-1}$), suspended material ($0.7–1.0\ mg\ l^{-1}$), and microelements ($<1\ \mu g\ l^{-1}$), compared to the background values of reference lakes: Ni and Cu $< 1\ \mu g\ l^{-1}$ and Cd $< 0.01\ \mu g\ l^{-1}$ (Moiseenko *et al* 2009).

The main elements of water pollutants of the Kola region are Ni and Cu because of activities of large copper-nickel smelters. Hg content in water is low. High levels of pollution are defined within a radius of 30 km around the smelters, with background levels of the metals detected up to 100 km due to the spread of smoke emissions, including dust particles and sulfur dioxide. Acid deposition can favor mining waste rocks and tailings, as well as the release of trace metals from soil and rock geochemical anomalies (Moiseenko and Kudryavtseva 2002).

The drinking water supply in this region originates from lakes. The water purification system used by water treatment stations is ineffective with respect to removing toxic metals. A comparison of metal content in water (the source of drinking water) and the metal content in the final domestic water supply demonstrates that toxic metals are not removed from the treated water. In fact, metal concentrations increase within the water pipeline, especially in respect to the concentrations of Fe and Mn. This is especially seen in the towns located on Lake Imandra: Apatity and Monchegorsk. Increasing the concentration of Fe in the water was observed during its migration through the conduit due to leaching in the steel pipe. In comparison to lake water, the concentration of Fe in water (in pipelines in Monchegorsk) increases by more than three times, and in Apatity by more than five times. The concentration of many elements in the water was not reduced from the pipeline, compared to the concentrations in lake water. This can be an indicator of a poor water purification system. Higher Ni concentrations were detected in water from Monchegorsk ($15.6 \pm 2.5\ \mu g\ l^{-1}$) compared to the lake water ($11.6 \pm 1.4\ \mu g\ l^{-1}$).

Our results did not find excess metal concentrations in the water, according Russian guidelines, for drinking water in any population locality (Bespamyatnov and Krotov 1985). The results of our previous

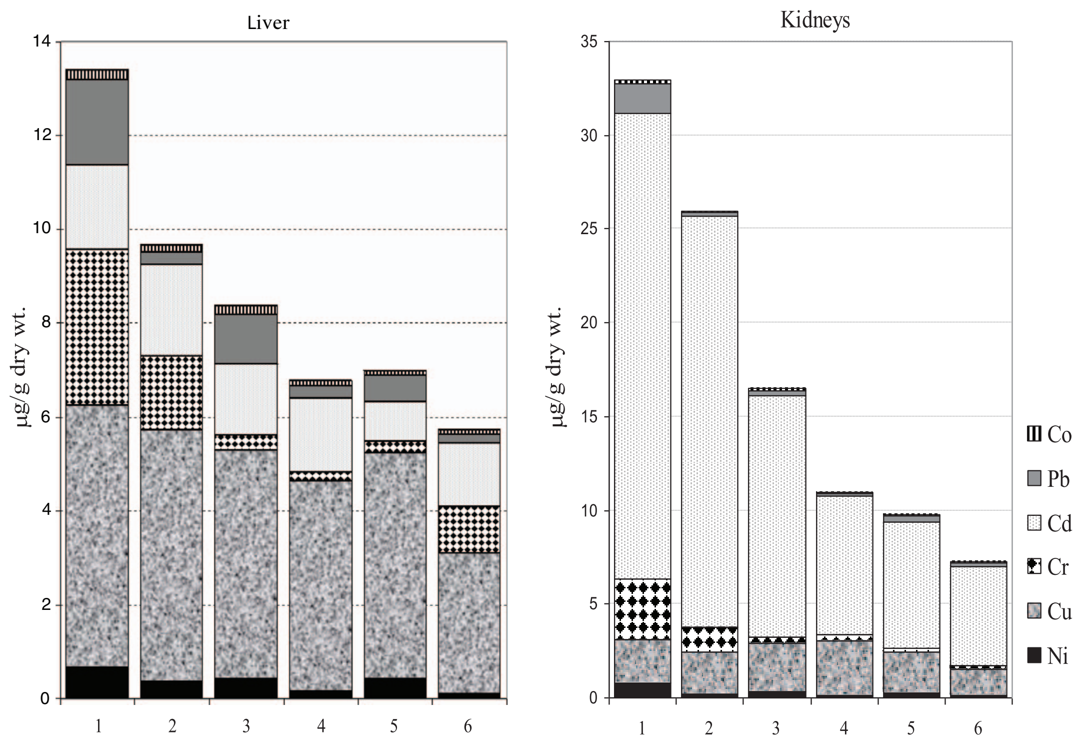

Figure 3. Metal content in liver and kidneys of the examined patients from the cities and settlements of the Murmansk region: (1) Monchegorsk, (2) Apatity, (3) Olenegorsk, (4) Lovozero, and (5) Alakurti. (6) Dead fetuses (still-born and immature births).

study indicate that the metals mainly exist in ionic form (for instance, Ni, Cd, Zn, and Pb) (Moiseenko 1999). It should be taken into account, however, that water is polluted by a complex of metals. These metals are active in soft water due to a very low Ca concentration. The low Ca concentration is known to increase the assimilation potential and negative impact of metals on living organisms, including humans (Seiler *et al* 1994, Nordberg *et al* 2007, Munk and Faure 2004).

5.2. Fish as bioindicators of water quality and ecosystem health

Pathological changes in the organs of the studied fish allowed the determination of water toxicity and the potential dangers of water contamination. In our comprehensive studies performed in different sites of the Volga basin and Arctic lakes, fish morbidity was used as the indicator of ecosystem health (Moiseenko and Kudryavtseva 2002, 2006, 2008). The results show that water quality in polluted sites is unfavorable, and the calculated dose–effect relationships confirm that the main cause of pathophysiological disturbances in fish is water pollution with toxic elements, which have a prolonged effect.

The stenobiotic character of whitefish in Arctic waters explains their high demand for water quality and their fast response to a change in water environment. During the period of intensive pollution, fish health clearly indicated strong toxic stress in the water environment. Under lake pollution by metals, nephrocalcinosis was first detected in Imandra Lake (Moiseenko and Kudryavtseva 2002). A similar disease encountered by freshwater fish within fish ponds showing high salinity is discussed in the book *Fish Pathology* (Roberts 1994).

The large amount of statistical data concerning fish diseases can confirm the connection between water quality and diseases. High concentrations of Ni in water leads to the accumulation of this element in fish organs and tissues, with the maximum accumulation occurring in the kidney. The relationship between the frequency of nephrological pathology ($\mathrm{Path}_{\mathrm{nephr}}$, %) and the level of Ni accumulation in fish (in zones with differing dissolved Ni concentrations) is approximated by the following equation (Moiseenko and Kudryavtseva 2002):

$$\mathbf{Path}_{\mathrm{nephr}} = 2.32(\mathbf{C}_{\mathrm{Ni\,kidney}}) - 4.72,\ r = 81,\ p = 0.0001$$

where C_{Nikidney} is the content of Ni in the kidney ($\mu g\, g^{-1}$ dry weight).

The equation shows that if dissolved Ni concentration reaches 3–5 $\mu g\, l^{-1}$, there is a risk of kidney pathology occurring within fish. Moreover, the contamination from metallurgical processing units is quite integrated, involving multiple metal elements. Data

Table 3. Average statistical data on occurrence of major diseases in adult population in cities and the Murmansk region as a whole. Occurrence measured as the number of diagnosed per 1000 population (OIP 2008, CPH 2009).

Region/city	Diseases of organs of digestion	Diseases of genitourinary system	Urolithiasis	Neoplasms
Monchegorsk	0.7	43.4	1.5	13
Apatity	0.3	53.9	1.8	18.1
Olenegorsk	0.3	33.7	3.0	10.4
Lovozero	0.1	38.9	1.4	6.8
Murmansk region	0.4	48.0	1.4	12.3
Russia	0.3	38.8	1.3	9.6

for the last decade demonstrate that Ni contamination is accompanied by Cd, the most toxic element of the studied contaminants. The latter occurs in much smaller concentrations than Ni in sewage; however, an additional source may be leaching from rock by acid rain. Accumulation of Cd in the kidney of whitefish is more important in acidified lakes. Sub-toxic doses of Cd can cause disease in fish (Hollis *et al* 1999).

Hg concentrations in fish are comparable with those of other fish in Arctic regions. For example, contents of Hg in the liver and muscles of whitefish of Imandra are comparable to those in lake Taimyr, where their contents in whitefish liver was 0.38–1.15 $\mu g\,g^{-1}$ dry weight, and in muscle 0.1–0.38 $\mu g\,g^{-1}$ dry weight (Allen-Gil *et al* 2003). Hg content in fish is not caused by local contamination, but is the result of the enrichment of the northern chemosphere with elements (Watras and Huckabee 2002). The average life expectancy of fish at the moment of sampling was five years. However, metal contamination of water was much higher thirty years ago, and high morbidity in fish was observed. Using this bioindicator, it is possible to conclude that the negative impact on people 30–35 years ago was a stronger. Based on the concept of ecosystem health (Yeom and Adams 2007, Adams and Ryon 1994) and the rate of fish intoxication symptoms in recent periods, we can conclude that the water quality and ecosystem conditions are better, but are still far from full recovery.

5.3. Assessment of human morbidity to freshwater pollution by metals

Hazardous substances infiltrate in humans through food, water, and air; therefore, it is often difficult to establish a reliable correlation between the quality of drinking water and population health. However, it should be taken into account that people in transpolar regions frequently consume products delivered from southern regions; this is why input of pollutants through consuming food made from local agricultural products cause insignificant impact on population morbidity. We recognize the possible influence of other negative factors. However, analysis shows that metals are the leading contaminants in this case.

In the northern regions covered by the scope of this study, with widely developed metallurgical and ore mining and processing industries, the leading environmental factor adversely affecting population health is the pollution of water bodies used as drinking water sources. Pollutants can penetrate humans through drinking water, and accumulate, thus provoking diseases.

The toxic impact of chemical elements on humans is caused by their chemical nature, amount, and composition, as well as by individual features of the organism. The threshold concentrations of individual elements vary depending on other elements present in the environment and organism. Surface continental waters in the extreme north regions have generally low Ca concentrations. The toxicity of metals increases in low-Ca water, especially in ionic forms, which ensures their maximal penetrating capacity and toxicity for living organisms.

According to statistical data (OIP 2008, CPH 2009) the most significant disease types in the Murmansk region include the following: diseases of the blood circulatory system; neoplasms; and diseases of respiratory organs, the urogenital system, and digestion organs, including hepatic cirrhosis (table 3). Comparative analysis of the main abnormalities in fish and humans shows their similarity.

It should be emphasized that the collected historical data characterize the level of pollution and fish pathology more than 30–35 years ago, and the human population has already suffered the effects of higher doses of pollution.

The highest morbidity rates are typical for the populations of towns consuming water from lakes Imandra and Monche, where the highest metal concentrations were recorded in drinking water. The most unfavorable is the situation of the growth of malignant neoplasm. According to statistics, the number of reported cases of tumors averaged 13.0 per 1000 people in Monchegorsk, 18.1 per 1000 people in Apatity, and 10.4 per 1000 people in Olenegorsk (OIP 2008, CPH 2009). Analysis of the current concentrations of chemical elements in water is needed to determine possible exposure to metals over the whole period of drinking water contamination. However, statistics do not always reflect the objective state of the health of the population, especially in northern regions where migration is high. A more accurate analysis reflects an accurate picture of the results recorded during post-mortem dissection.

The fact of accumulation of metals in human livers and kidneys has been established. It is possible that the patients examined post-mortem received their main dose of contamination over a more prolonged period of time. The high levels of metal concentrations

Table 4. Correlation coefficients between metals accumulated in the liver and kidneys of people, and their concentrations in drinking water in different towns and settlements in the Murmansk region (significant correlation coefficients ($p \le 0.05$) are shown in bold type).

Organ	Ni	Cu	Cd	Pb	Sr	Cr	Co
Liver	0.78	0.78	0.30	−0.45	−0.30	0.26	0.81
Kidney	0.87	−0.19	0.62	−0.29	−0.30	0.31	0.83

in the livers and kidneys are typical for Monchegorsk residents, where domestic water intake is located in the propagation zone of smoke emissions from copper-nickel smelting works. Notwithstanding the fact that the highest concentrations of Ni occur in drinking water, the most abundant element found in human kidney tissue is Cd (five times greater than that of the reference group). This element is highly toxic and induces various pathologies in organisms (Satarug *et al* 2003, Hoffman *et al* 2005, Li *et al* 2011).

Despite the many factors involved, we have established a correlation between the concentrations of metals in water and their concentrations in human liver (table 4), and diseases and pathologies of systems and organs in the population of Kola Peninsula towns (table 5).

These facts confirm the etiology of the aqueous metal accumulation and disease in humans. This confirms the penetration of these metals in water. Co is an element associated with copper-nickel ores, which is melted at the Monchegorsk plant. Cd in the kidney also has a credible relationship with its concentrations in the water.

Ni is known to have carcinogenic and gonadotoxic potential (Sidorenko and Itskova 1980). The accumulation of high concentrations of Cd in kidneys of examined patients should be pointed out. This element is nonessential: therefore, the human body does not normally contain Cd (Nordberg *et al* 2007). Despite the fact that Ni and Cu are the major drinking water contaminants in the region, Cd accumulation in human kidneys with high concentration was observed. The towns are ranked by toxic metal accumulation in the liver and kidneys of inhabitants: Monchegorsk > Apatity > Olenegorsk > Lovozero > Alakurti.

Research by Sakamoto *et al* (2013) (study of trace element concentration in chorionic tissue and umbilical cord tissue of the placenta) shows that among toxic elements the placental barrier worked most strongly against Cd, followed by Hg, and then Se, Zn, and Cu. In spite of the results of Sakamoto *et al* (2013), the results of this study show that small quantities of Cd were observed in newborns. Therefore, it must be considered that Cd may penetrate the placental barrier (see figure 3). Cd may cause malignant organ and tissue changes, and affects the progression of diabetes, hypertension, osteoporosis, leukemia, and neoplasm development (Nishijo *et al* 2000, Satarug *et al* 2003, Revich *et al* 2003, Hoffman *et al* 2005, Li *et al* 2011). The authors of studies on Cd contamination argue that Cd in low doses accumulates within the lungs, kidneys, and adrenal glands, leading to various disorders. For instance, Li *et al* (2011) showed that the increase in cardiovascular disorders, including hypertension, in the studied group of people was associated with a renal disorder caused by Cd.

Many other confounding factors can affect the content of elements in the human body, including lifestyle (food, air quality, smoking, etc.). Bernhard *et al* (2005) presented a critical review of the effects of smoking on the content of many metals (Cd, Pb, Zn, Ni, Al, etc.). Satarug *et al* (2010) reported that smokers have a 1.7-fold increase of serum Cd content in comparison to non-smokers ($0.92 \pm 0.83\ \mu g\,l^{-1}$ and $0.55 \pm 0.48\ \mu g\,l^{-1}$, average age 36 years). However, in our studies there was no data about smoking status of the examined patients. At the same time, in the historical data there is a confirmation of the influence of environmental factors on the morbidity of the population. In the 1970s, the first medical-geographical examination of population in the Murmansk region was held, which showed that the population of Monchegorsk and Apatity is characterized by diseases such as kidney stones and gallstones (Atlas of Murmansk Region 1971).

6. Conclusions

The towns and settlements in the Murmansk region use drinking water from polluted lakes. The major polluting element in this region is Ni and Cu. Cd contamination in surface waters is associated with leaching by acid rain. The existing water treatment systems in this region fail to remove metals from drinking water supplies.

During the period of severe pollution of Imandra, there were mass incidents of fish diseases (nephrocalcinosis, lipid liver degeneration, cirrhosis, anemia, scoliosis, and others). The highest content of metals in water (especially Ni and Cu) was observed in fish from the water bodies that were exposed to smoke emissions of Cu and Ni smelters. Nephrocalcinosis of fish was mediated by the accumulation of toxic metals and their toxic impact. Based on fish intoxication symptoms, we can conclude that water quality has improved in recent periods, but is still far from recovery. The histological examination of fish shows that the main disorders are within the kidneys. Hepatic disorder is the result of the general toxic action of the metal contaminant mixture.

The highest accumulation of metals within the kidney and liver were recorded in inhabitants of Monchegorsk (the concentrations of many metals are high, especially, Ni, Cu, Cd, and Pb). The highest concentrations in the kidney tissue were Cd. This element is present in the kidneys of newborn children, indicating high penetration capability into the human organism. The results of this study show that while concentrations of metals in drinking water are quite

Table 5. Correlation coefficients between diseases and pathologies of systems and organs in the population of Kola Peninsula towns, and microelement concentrations in drinking water ($\sum C_i/MPC_i$—sum of exceedances of microelements in water in relation to the maximum permitting concentrations for aquatic life) (significant correlation coefficients ($p \leq 0.05$) are shown in bold type).

Concentration in water	Diseases and pathologies of systems and organs in the population				
	Organ		System		
Kidney	Liver	Gastrointestinal	Hematopoietic	Cardiovascular	
Ni	0.40	**0.63**	**0.89**	**0.64**	**0.73**
Cu	0.43	**0.64**	**0.91**	**0.67**	**0.77**
Co	**0.93**	**0.75**	**0.85**	**0.90**	**0.91**
Cr	**0.60**	0.53	0.51	**0.76**	**0.70**
Sr	0.56	0.18	0.03	0.08	0.01
Cd	**0.87**	**0.76**	**0.87**	**0.77**	**0.83**
Pb	**0.97**	0.58	**0.58**	**0.69**	**0.70**
Zn	0.49	0.59	**0.81**	**0.61**	**0.68**
$\sum$Ci/MPCi for aquatic life	**0.76**	**0.77**	**0.95**	**0.79**	**0.86**

low according to the accepted standards in Russia, they still cause high rates of diseases in the human population. Kidney pathologies are abundant within populations using a contaminated water supply for drinking water.

Comparative analysis of abnormalities in fish and humans shows their similarity, and thus, the fish can serve as bioindicators of pollution, and help us determine future directions for medical and environmental studies, which are labor intensive and costly. Therefore, one may arrive at the conclusion that an unsatisfactory quality of drinking water within the industrial area of the Murmansk region may be the cause of population morbidity. These conclusions suggest the need to carry out more thorough studies in order to correct the existing standards for drinking water, taking into account specific regional conditions, and to develop more elaborate water purification treatment systems to reduce the risk of diseases within the population.

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