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ORIGINAL PAPER

LONG-RANGE TRANSPORT OF HEAVY METALS AS A FACTOR OF THE FORMATION OF THE GEOCHEMISTRY OF SEDIMENTS IN THE SOUTHWEST OF THE REPUBLIC OF KARELIA, RUSSIA*

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Abstract

Bottom sediments are an integral part of any water body and a key to the current state of a hydrosystem, particularly the upper layers of lake sediments. The chemical composition of bottom sediments from small Lake Ukonlampi (Lahdenpohja District, Republic of Karelia, Russia) is discussed. Evidence for considerable historical heavy metal pollution of the lake and its surroundings was obtained, based on analysis of 30 cm long bottom sediment core. Elements concentrations in the bottom sediment samples were estimated using the mass-spectral method on a XSeries-2 ICP-MS. The influence of the long-range transport of pollutants on the formation of the geochemical features of current bottom sediments from the study area was assessed. It should be noted that the main sources of heavy metals are industrial emissions from St. Petersburg, some cities of the Leningrad Region and Finland. The heavy metal behaviour of bottom sediments from other small and large lakes of the Republic of Karelia and the Murmansk Region was shown to display a distinctive pattern. All the lakes contain similar heavy metals, which are accumulated most actively in the surface layers of current lake sediments. Sediment lake pollution in the present study was assessed using a geoaccumulation index (Igeo) of noted elements. It was shown that Ukonlampi lake sediments have uncontaminated and moderately contaminated P, Cu, Zn, W, and Tl levels. Other elements (Cd, Sn, Sb, Pb, and Bi) displayed Igeo from the uncontaminated and moderately contaminated level to the moderately contaminated level.

Keywords: bottom sediments, small lake, heavy metals, the Republic of Karelia.

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INTRODUCTION

Bottom sediments are an integral part of any water body and a key to the current state of a hydrosystem, particularly the upper layers of lake sediments (from 0 to 50 cm, depending on a study area). Various events that have occurred over the past two to three centuries are reflected in these lake layers (Ukonmaanaho et al. 1998, Dauvalter et al. 2011, Michinobu et al. 2013). Numerous paleolimnological studies conducted around the world show that heavy metal concentrations in bottom sedimentary sequences begin to increase in the lower layers that formed in the mid- to late 19^{th} century. The major geochemical agents of historical environmental pollution are Pb, Cd, Sb, Sn and Hg (McConnell, Edwards 2008, Dauvalter et al. 2011, Vinogradova et al. 2017). In urban or industrial areas, they occur together with other elements potentially dangerous for living organisms. Major heavy metal pollutants in the Republic of Karelia and the Murmansk Region, in addition to those listed above, include Ni, Cu, Zn, Co, and V (Dauvalter et al. 2011, Slukovskii et al. 2018).

Emissions from mining and metallurgical plants, heat and power plants, as well as road and rail transport, contribute mostly to the impact of heavy metals on the environment, including water ecosystems of some regions of Russia. The Republic of Karelia is adversely affected by pollution from St. Petersburg, Russia's second largest city (Vinogradova et al. 2017). Annual monitoring of air quality in the world's biggest cities shows that St. Petersburg is the most heavily polluted city in North Europe, far ahead of Helsinki, the capital of Finland. Petrozavodsk and Murmansk are the largest industrial urban areas in the Republic of Karelia and the Murmansk Region. The Petrozavodsk area has been studied thoroughly (Slukovskii et al. 2018, 2019). The goal of the present study was to estimate the contribution of the long-range transport of heavy metal aerosols to the formation of the geochemical characteristics of current sedimentation in small lakes in southwestern Karelia (Ladoga Lake catchment). The research was conducted on small Ukonlampi Lake, used as an example.

MATERIALS AND METHODS

Ukonlampi Lake is a small lake located in the southwestern Republic of Karelia (Lahdenpohja District). This lake lies about 10 km from the town Lahdenpohja, the capital of the Lahdenpohja District (Figure 1). The lake has an elongated oval shape. Ukonlampi Lake is 660 m long, about 175 m wide and covers an area of 0.11 km^2. There is a small island in the northern part of the lake.

A 30-cm-long core of bottom sediments from Lake Ukonlampi was col-

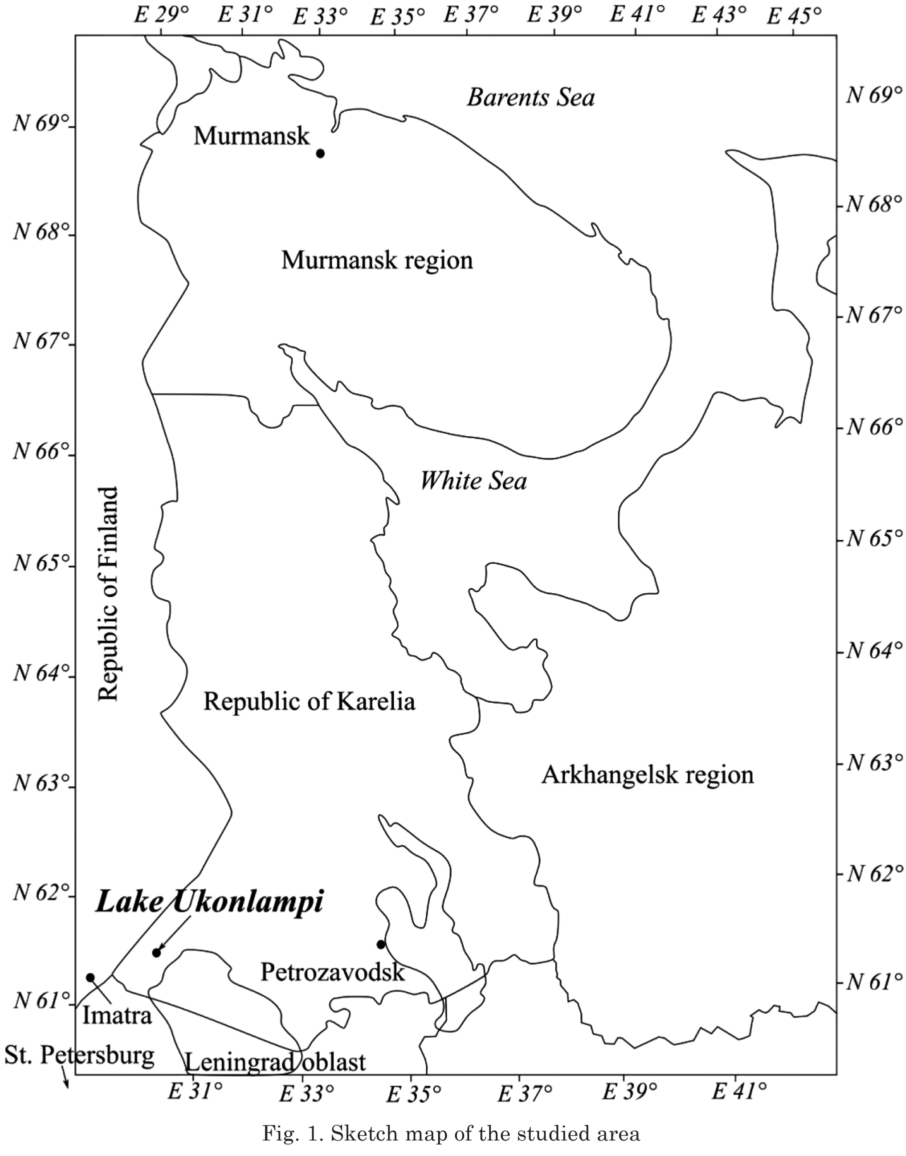

Fig. 1. Sketch map of the studied area

lected on 20 July 2017 with a Limnos sampler (made in Finland). The depth of the lake at the sampling site is about 3 m. It is the accumulation zone, where it is recommended to carry out sampling of lake sediments (DAUVALTER et al. 2011, MICHINOBU et al. 2013). The coordinates of the sampling site are: 61.583526°N and 30.035948°E. After the sampling, the core was divided into fourteen layers (0-2, 2-4, 4-6, 6-8, 8-10, 10-12, 12-14, 14-16, 16-18, 18-20, 20-22, 22-24, 24-26 and 26-30 cm). The divided samples were placed in plastic boxes and transported in a cooler bag to the laboratory. All the samples were kept in a laboratory refrigerator at a temperature is about from 3 to 6 degrees above zero before analysis. Values of pH and redox potential (E_h) were measured in the field using a portable pH-420 pH millivoltmeter (made in Russia) immediately after sampling bottom sediments.

All sediment samples were dried at a temperature of about 110 degrees above zero. Then they were mineralised by acid autopsy with an acid mixture in an open system. The method for preparing the samples for chemical analysis was described in SLUKOVSKII, POLYAKOVA (2016). Concentrations of elements in the bottom sediment samples were estimated using the mass-spectral method on a XSeries-2 ICP-MS instrument at the Analytical Centre of the Institute of Geology, KarRC, RAS, Petrozavodsk. The sediment age estimation was determined using the method of ^{210}Pb dating. The determination of the content of gamma-active radionuclides was carried out using a gamma spectrometer, ORTEC GEM-C5060P4-B, at the Department of Radiochemistry, Moscow State University.

The results were processed statistically by calculating the median, standard deviation of the median, minimum and maximum values and variation coefficients of sample values. MS Excel 2016 and Inkscape 0.48.4 software were used to illustrate the results in diagrams.

The geo-accumulation index I_{geo} of heavy metals in bottom sediments was calculated using the formula:

$$I_{geo} = \log_2\left(\frac{C}{1.5 * B}\right),$$

where: C is the metal concentration in a given layer of, B is the background metal concentration (Table 1) measured in the deepest layer of the core (MÜLLER 1979).

RESULTS AND DISCUSSION

The bottom sediments from Ukonlampi Lake are dark brown in colour. They are identified as sapropel, the most common type of freshwater sediments in Karelia. The Ukonlampi sapropel is commonly about 2 m thick. These lake sediments can be used as beneficial organo-mineral raw materials. They contain about 80% organic matter and less than 2% total Fe. The pH values of sediments vary from 5.22 to 5.88, increasing from the upper layers to the lower layers in the Ukonlampi sedimentary core, due to the increasing content of organic matter in the upper layers of sediments. The redox potential (Eh) values of the lower layers were about 0. On the other hand, the Eh values of the upper layers were up to -294 mV.

^{210}Pb radioactivity in the lake sediment core showed that all layers were accumulated during the 19th, 20th and 21st centuries (industrial period).

Table 1

Descriptive statistics of chemical element concentrations of a 30-cm-long core of bottom sediments from Ukonlampi Lake, the Republic of Karelia (mg k-1 d. m.)

Elements	x	Me	x_{max}	x_{min}	S_{med}	V (%)	B
P	1659	1403	2335	1169	427	25.8	1389
Ti	1593	1616	2347	987	443	27.8	1561
Zr	493.3	463.6	862.3	312.5	149.4	30.3	350.1
Mn	406.1	415.6	443.7	356.4	28.3	7.0	414.0
Ba	358.1	380.9	452.3	247.5	75.3	21.0	374.8
Zn	174.3	149.1	263.8	111.4	58.3	33.5	111.4
Sr	124.8	131.6	173.8	78.8	33.0	26.4	131.0
Pb	47.16	34.21	91.14	18.97	29.42	62.4	18.97
Cr	37.99	36.77	51.17	27.95	6.56	17.3	37.57
Rb	36.50	35.25	52.91	24.75	9.35	25.6	32.07
Cu	33.40	31.80	51.18	24.69	7.19	21.5	27.85
V	30.73	30.01	40.73	22.94	5.59	18.2	22.94
Ni	28.98	28.82	32.45	26.32	1.68	5.8	28.19
Sc	12.38	12.20	18.08	8.99	2.26	18.3	12.20
Li	9.825	9.722	13.870	6.927	2.399	24.4	8.394
Co	5.958	6.110	7.386	4.592	0.878	14.7	5.471
Sn	2.781	2.669	5.341	1.260	1.406	50.6	1.260
Mo	1.932	1.931	2.242	1.701	0.140	7.2	1.952
Cd	1.567	1.410	2.687	0.640	0.771	49.2	0.772
U	1.538	1.509	2.102	1.044	0.349	22.7	1.506
Cs	1.429	1.367	1.886	1.115	0.257	18.0	1.115
Sb	0.982	0.993	1.838	0.344	0.600	61.1	0.344
W	0.620	0.544	1.108	0.361	0.227	36.7	0.392
Tl	0.376	0.401	0.582	0.227	0.123	32.6	0.227
Bi	0.295	0.239	0.497	0.148	0.145	49.3	0.159

Explanations: x – average, Me – median, S_{med} – the standard deviation of the average, x_{max} and x_{min} – maximum and minimum values, V – variation coefficient, B – background metal concentration for geo-accumulation index I_{geo}

The medium rate of accumulation in Ukonlampi Lake during this time has been about 1.3 mm per year. These data are consistent with the results obtained in the study of modern deposits of lakes in Finland and the Murmansk region of Russia (Verta, Tolonen 1989, Dauvalter, Kashulin 2010).

All the above chemical elements, including heavy metals, were found in bottom sediment samples from Ukonlampi Lake. The average and median values and variation coefficients are shown in Table 1. Maximum median values of Ti, P, Zr, Mn and Ba were noted. Among heavy metals, Zn, Pb, Cu, V and Ni concentrations displayed maximum median values. Most of the elements analysed exhibited variation coefficient values of 5 to 30%. Eleven elements, including Pb, Sb, Sn and Cd, displayed variation coefficient values of 32 to 62%.

The vertical distribution of the elements revealed in the Ukonlampi core is shown in Figures 2 and 3. All the elements were divided into two large

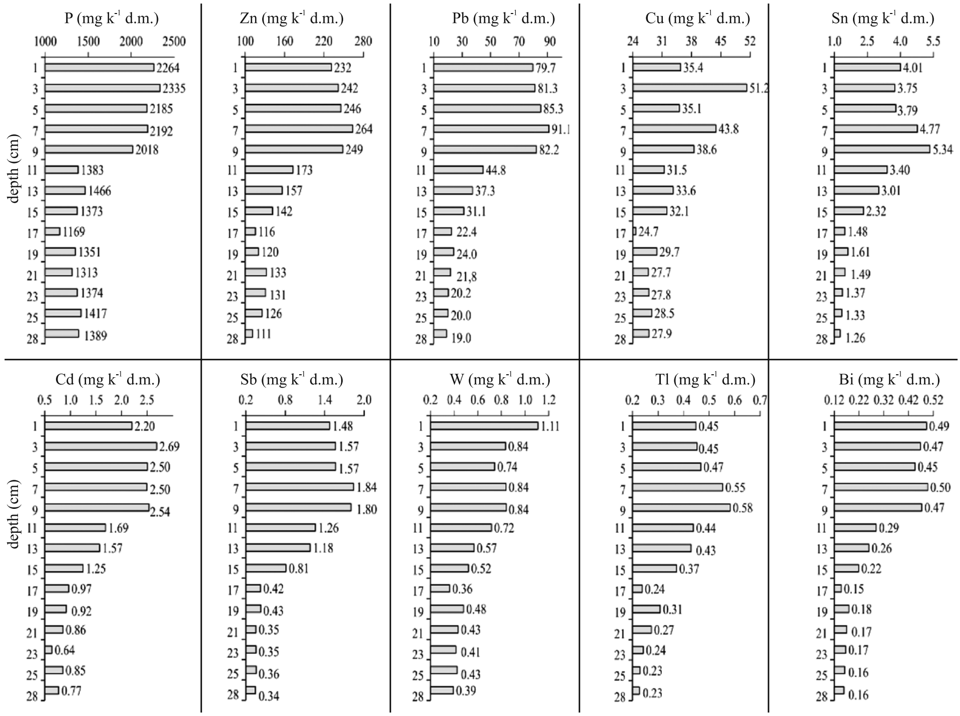

Fig. 2. Vertical concentrations of elements of Group I in the sediment core from Ukonlampi Lake, Karelia

groups. Group I included P, Zn, Pb, Cu, Ni, Cd, Mo, Sb, W and Tl, which had high concentrations in the upper layers of sediments (0 to 14 cm, depending on an element). Pb, Cd, Sb, Zn, P, W and Tl concentrations increased sharply in the upper layers, as compared to those in the lower layers. All other elements were included in group II. These elements display small concentration peaks in the middle or lower parts of the Ukonlampi sediment.

The southwestern Republic of Karelia, located between the Russian-Finnish border and the Ladoga Lake catchment, is considered to be an area most heavily affected by human activities. There are two small Karelian towns, Sortavala and Lahdenpohja, and some stone-quarrying companies there. The region is affected by St. Petersburg and its infrastructure, the dense transport network of the Leningrad Region and large industrial zones near the cities Svetogorsk, Kamennogorsk, Vyborg and Primorsk. Human activi-

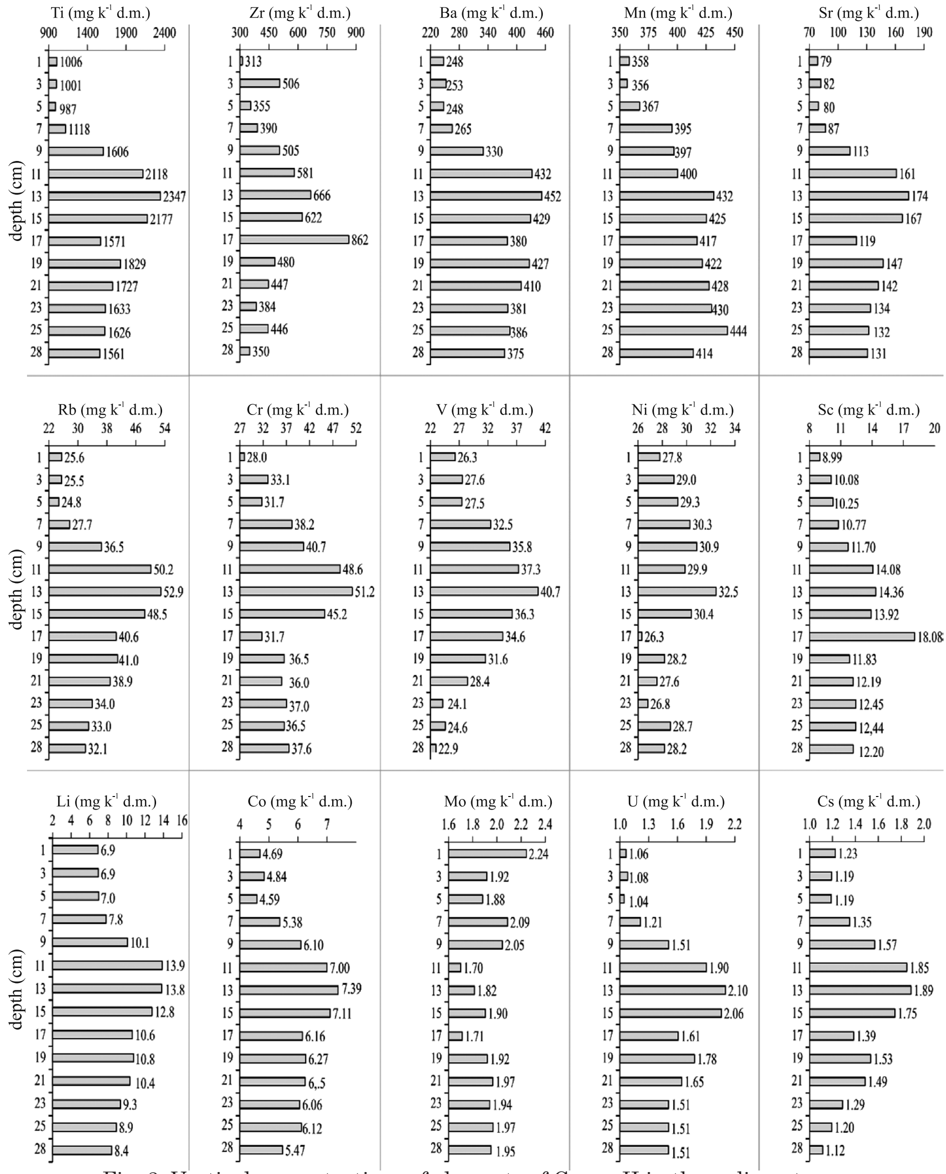

Fig. 3. Vertical concentrations of elements of Group II in the sediment core from Ukonlampi Lake, Karelia

ties in St. Petersburg are responsible for the geochemical anomalies of Cu, Zn, Ni, Cr, Pb, Cd, Co, Mo, Mn, Fe, Ba and Hg in bottom sediments from the small water bodies of St. Petersburg and the Gulf of Finland (Vallius, Leivuori 2003). According to that publication, principally these heavy metals in water bodies of St. Petersburg are a result of emissions from the chemical, mining, leather-textile, food, pulp-paper industries.

The long-range atmospheric transport of pollutants from Finland, primarily from metal production in Imatra and Kotka, may also have a harmful effect on the environment in the southwestern Republic of Karelia (Ukonmaanaho et al. 1998, Virkutyte et al. 2008). The main pollutants emitted by these factories are Pb, Cd, Cu, Zn, Ni, Cr and Sb. Such heavy metals as Pb, Cu, Ni and Cd are carried by air from the St. Petersburg area and the Leningrad Region to Karelia. The annual entry of these elements is about 1.5 (Vinogradova et al. 2017). They are accumulated in bottom sediments from small and large lakes in the northern and south-western Republic of Karelia and adjacent areas (Verta, Tolonen 1989, Davydova et al. 1999).

Thus, the elements in Ukonlampi Lake bottom sediments (Figure 2), particularly those in the upper layers of the lake sediments, have an anthropogenic origin. The corresponding behaviour of Pb, Zn, Cd and Cu was shown for sediments from small lakes in Finland: Hirvilampi, Valkjärvi, Vuorilampi, Laukunlampi, etc. (Ukonmaanaho et al. 1998). It has been noted that the heavy metal level in sediment cores began to rise in the 1900s due to the impact of the long-range atmospheric transport of pollutants from the industrial areas of Finland, Russia and other former USSR countries (Verta, Tolonen 1989, Dauvalter et al. 2011, Stankevica et al. 2012). The concentrations of the above elements in the lower layers of the sediment cores were recognised as background concentrations for southern Finland. A similar heavy metal behaviour in the upper layers (0-20 cm) of Ladoga Lake sediments has been noted earlier (Davydova et al. 1999). Pb, Cu and Zn concentrations in the bottom sediments, affected by human activities, tend to rise. This pattern is observed not only for the open water areas of Ladoga Lake but also for its near-shore zones, where elevated Pb, Ni, Cu, Cr, Zn, W and Co concentrations are accumulated in sediments that have just been formed (Ivanter et al. 2016).

Air and water pollution is known to occur as a result of the burning of coal and oil products, emissions from metallurgical, machine-building and instrument-making industries and vehicle emissions. In addition to common elements, such as Pb, Zn, Cu, Ni, etc., Tl, Sb, Sn and Bi anomalies in lake bottom sediments have been revealed by many experts. These experts assume that the noted elements enter the environment from the above sources too (Michinobu et al. 2013). Almost all of these elements display elevated levels in the Ukonlampi core, suggesting man-induced changes in sedimentation in the lake in the past two to three centuries. The main sources can be the emissions from different types of factories and from vehicles, same as in the case of other territories of the north.

The influence of the long-range atmospheric transport of heavy metals was already recognised as a pollution factor in small and large lakes in the Onega Lake catchment basin, southern Karelia (Slukovskii, Medvedev 2015, Slukovskii et al. 2018). The behaviour of chalcophile elements, like Cd and Pb, in current lake sediments from the various northern territories of the world, including Russia, the USA and Finland, is analysed to understand the long-range air transport of pollutants (Norton et al. 1990, Keinonen et al. 1992, Thomas, 1995, Dauvalter, Kashulin 2010).

It has been noted that the Pb accumulation level in the surface (0-10 cm) layers of the Ukonlampi sediments is almost close to that in the surface layers of lake sediments from the urbanised areas of Karelia with local sources of environmental pollution (Medvedev et al. 2019). Earlier studies have shown that Pb, Sb, Sn and Cd are major pollutants of the aquatic environment in the urbanised zones of the studied area. These heavy metals and other elements shown in Figure 2 could also be considered priority pollutants of the aquatic environment in the non-urbanised areas of Karelia, particularly southwestern Karelia. This has been confirmed by a high correlation level between elements from the technogeneous group of elements shown in Figure 2. For example, Pb and Sb concentrations in the sediments analysed are closely correlated (Figure 4). Both metals from lake sediments

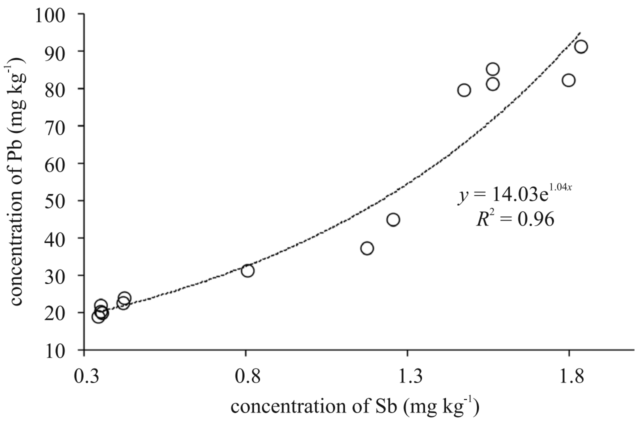

Fig. 4. Ratio of concentrations of Pb and Sb in the sediment core from Ukonlampi Lake, Karelia

in an urbanised environment (Slukovskii et al. 2018) behave in a similar manner. The influence of the long-distance air transport of heavy metals is observed in both cases.

Analysis of the geo-accumulation index (I_{geo}) of elements from Figure 2 in the Ukonlampi sediments has revealed uncontaminated and moderately contaminated P, Cu, Zn, W and Tl levels (Figure 5). Other elements (Cd, Sn, Sb, Pb and Bi) displayed I_{geo} from the uncontaminated and moderately contaminated level to the moderately contaminated level. Maximum I_{geo} values (>1.5) were calculated for Pb in 0-10 cm layer, for Sb in 0-10 cm layer and for Sn in 8-10 cm layer, because these elements have significant concentrations in the upper layers compared to the background. In the lower 16-26 cm layer of the sediments analysed, an uncontaminated pollution level of Ukonlampi Lake was revealed. A similar behaviour of I_{geo} values was

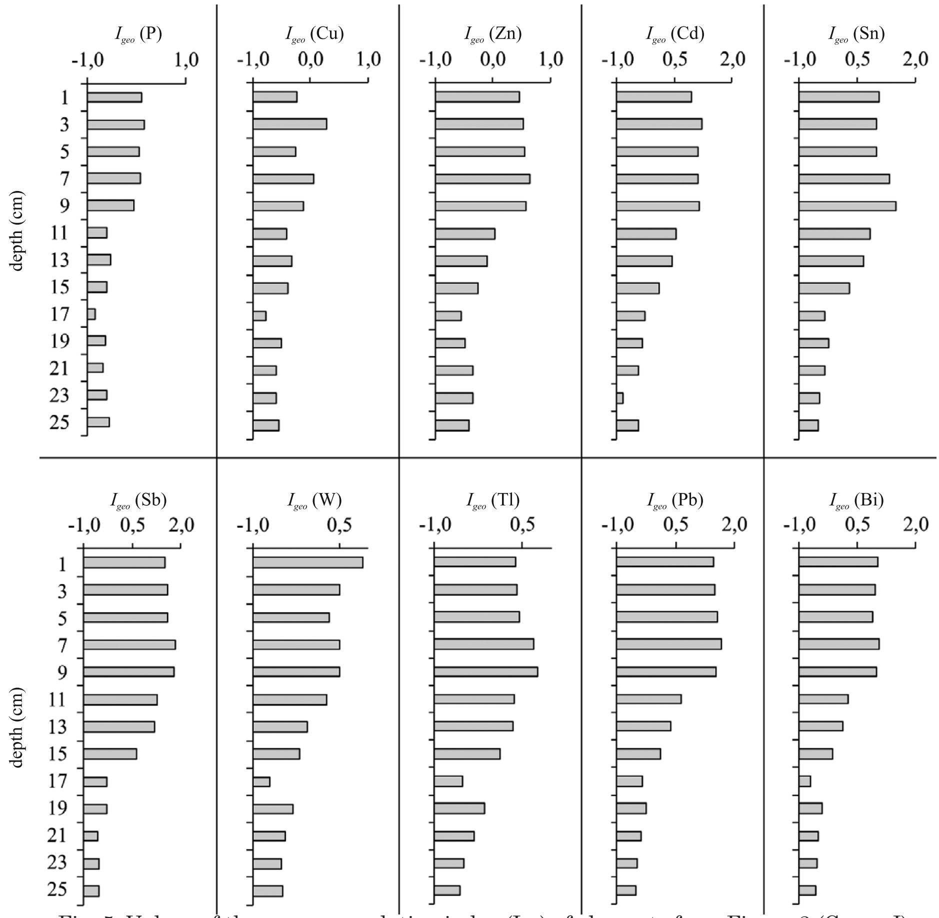

Fig. 5. Values of the geo-accumulation index (I_{geo}) of elements from Figure 2 (Group I) in the Ukonlampi sediments

reported for lake sediments from the urbanized areas of Karelia. However, the I_{geo} contaminated level of Pb, V, Cu, Ni and W in urbanized lakes was slightly higher than that in Lake Ukonlampi, because there are many direct industrial sources of pollution in the cities (Slukovskii, Medvedev 2015).

The vertical distribution of the elements shown in Figure 3 was attributed to natural processes, including catastrophic events (Andronikov et al. 2014). Most of the above elements are non-hazardous and low-hazardous for living organisms. However, considering that the layers from 0 to 16 cm were accumulated later than 1900, the peaks of Ti, Zr, Sr, Rb, Cr, V, Ni, Sc, Li, U, Cs and rare-earth elements in layers from 10 to 16 cm could also be associated with human influence on the environment around Ukonlampi Lake. This could be confirmed indirectly by the study of small rivers in Karelia, where extremely high alkaline metal concentrations were observed in modern bottom sediments. Dust particles of emissions from factories may contain these metals (Slukovskii et al. 2018). Besides, the soils in the nearby area display a vanadium anomaly associated presumably with steel production in Imatra, Finland (Rühling, Steinnes 1998). This plant of Ovako Group AB started to operate in the 1930s. Thus, the peaks of V and other elements shown in Figure 3 could be the result of the opening of a steel-producing plant in Imatra. However, this hypothesis should be verified during further studies of current sedimentation processes in southwestern Karelia.

Returning to the interpretation of the data in Figure 2, the phosphorus concentration in the upper layers was observed to be twice as high as in the lower layers of the Ukonlampi sediments. Phosphorus fertilisers used in agriculture are a major anthropogenic source of this element. The silting and eutrophication of the lake could be the consequence of this process. Firstly, as a result, the upper layers have become more acidic than the lower ones. Secondly, it has contributed to heavy metal accumulation, because organic matter is a significant sorbent of pollutants in water ecosystems (Förstner et al. 2004). Therefore, heavy metals are strongly bound by organic molecules in lake sediments, and they are unable to migrate into water again. But there is still an environmental risk of heavy metals migrating from bottom sediments to lake water. It may depend on the redox potential (Eh) values of the upper bottom sediment layers of Ukonlampi Lake because a system with more negative Eh can give electrons into a system with more positive Eh, as is the case of Ukonlampi Lake.

CONCLUSIONS

To examine the historical level of pollution of the southwestern area of the Republic of Karelia, the multiple element analysis of the current bottom-sediment core was performed. The study focused on heavy metal behaviour in sediments of small Ukonlampi Lake in the Lahdenpohja District, Republic of Karelia, Russia. It was shown that the long-range transport of heavy metal aerosols is the principal factor of pollution researched in the lake and catchment area. It was proven that the main source of heavy metals is industrial emissions from St. Petersburg, some cities of the Leningrad Region, and Finland. According to studies of small lakes in Finland and the Murmansk region, it was demonstrated that the heavy metal level in lake sediment cores sampled in the territory of northern Europe began to rise around the 1900s. In the core of sediment from Ukonlampi Lake there were high concentrations of P, Zn, Pb, Cu, Ni, Cd, Mo, Sb, W and Tl in the upper layers of sediment (0 to 14 cm, depending on an element). Lake sediment pollution in the present study was assessed using a geoaccumulation index (I_{geo}) of the noted elements. It was shown that Ukonlampi Lake sediments have uncontaminated and moderately contaminated P, Cu, Zn, W, and Tl levels. Other elements (Cd, Sn, Sb, Pb, and Bi) displayed I_{geo} from the uncontaminated and moderately contaminated level to the moderately contaminated level. These metals from lake sediment in the urbanised environment of Karelia behave in a similar manner.

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