

# Assessment of heavy metal contamination and spatial distribution in surface and subsurface sediment layers in the northern part of Lake Babrukas

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It is widely accepted that heavy metal contamination in sediment, soil, and groundwater is one of the largest threats to environmental and human health. Sediments are the principle sinks for heavy metals in aquatic environments and can result in a secondary contamination source affecting the ecosystem. Analysis of heavy metal amounts in sediments and comparison with reference levels is a reliable indicator of ecosystem health, however understanding the distribution of pollutants is among most essential information for environmental research and critical for environmental management and decision-making. 64 samples were collected from surface (0–0.6 m) and subsurface (0.6–1.2 m) layers of sediments in the northern part (bay) of Lake Babrukas, which had undergone pollution by municipal wastewater in 1964–2002. The level of pollution attributed to heavy metals was evaluated using X-Ray fluorescence analysis and comparison with several reference levels (maximum allowable concentrations in soil, background concentrations in Lithuanian soils and background concentrations in Lithuanian lake sediments) in order to determine anthropogenically derived sediment contamination of bottom sediments. Spatial distribution patterns of metals in sediments were demonstrated by employing ordinary kriging interpolation. Results of heavy metal pollution analysis reveal a significant anthropogenic impact on the northern part of Lake Babrukas with concentrations of several heavy metals in both surface and subsurface layers of sediments exceeding not only background concentrations in Lithuanian soils and lake sediments, but even maximum concentrations allowable by Lithuanian legislation. Detected levels of arsenic (As), cadmium (Cd), antimony (Sb), mercury (Hg) and tin (Sn) demonstrate a significant pollution anomaly and high potential threat to the water ecosystem and even human health by exceeding maximum allowable concentration up to 10.6 times, while detected levels of chromium (Cr), copper (Cu), nickel (Ni), and zinc (Zn) show the effects of anthropogenic activity on the lake by exceeding background concentrations in Lithuanian soils and lake sediments up to 7.25 times. A comparison of heavy metal amounts and spatial distribution patterns in the two layers of sediments reveals much higher pollution levels in the subsurface (0.6–1.2 m) than surface (0–0.6 m) level by most metals, namely As, Cd, Hg, Ni, Sb and Sn, while Zn was the only metal with higher concentrations in the surface level. Spatial distribution analysis demonstrates irregular distribution of most metals due to complexity of influencing physical and chemical processes, but the general trend of high concentration anomalies stretching through the south-eastern and north-eastern parts of the bay towards its northern point is explained by former discharges of wastewater into the littoral zone of the eastern shoreline and water currents towards excess water overflow into Lake Lovka (Olauka), interconnected with Lake Babrukas in the north.

**Key words:** lake sediments, heavy metals, spatial distribution, spatial interpolation, kriging, X-Ray fluorescence analysis

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## INTRODUCTION

Although metals and metal compounds are natural constituents of all ecosystems, moving between atmosphere, hydrosphere, lithosphere, and biosphere (Bargagli, 2000), it is widely accepted that heavy metal contamination in sediment, soil, and groundwater is one of the largest threats to environmental and human health (Salomons, Förstner, 1984). Toxic compounds including metals tend to accumulate in sediments of water bodies through complex physical and chemical adsorption mechanisms which depend on the nature of the sediment matrix and the properties of adsorbed compounds (Ankley et al., 1992; Leivouri, 1998; De Bartolomeo et al., 2004; Abul Kashem et al., 2007). Thus, sediments are the principle sinks for heavy metals in aquatic environments (Akçay et al., 2003). However, sediments can also release them back into the ecosystem when changes occur in environmental conditions such as pH, redox potential, the presence of organic chelators, resuspension, desorption or (bio) degradation of the sorptive substances, resulting in a secondary contamination source affecting the ecosystem (Linnik, Zubenko, 2000).

Heavy metals are considered serious pollutants not only because of their persistence and non-degradability in the environment but also because most of them have toxic effects on living organisms when they exceed a certain concentration (Nriagu, Pacyna, 1988; Enserink et al., 1991). Additionally, aquatic organisms can bioaccumulate, biomagnificate or biotransfer certain metals to concentrations high enough to bring about harmful effects, especially by the transfer of metals through the food chain (Naimo, 1995). Despite the differences in toxic effects of the metals, environmental conditions and the time of exposure, their concentrations are reliable indicators of ecosystem health (Singh et al., 2005).

Most heavy elements or heavy metals concern researchers because of environmental or human exposure, however 10 of them (As, Cd, Cr, Cu, Hg, Ni, Pb, Sb, Sn and Zn) are of major interest in bioavailability and toxicology studies because of the potential for increased environmental and health risk (McKinney, Rogers, 1992). Accordingly, these 10 elements were selected

as the specific metals of interest for the current study.

Both atmospheric and other, more direct, metal inputs are historically recorded worldwide in the sediments of lakes and rivers which have experienced significant human influence (Nriagu et al., 1979; Dominik et al., 1984; Carignan et al., 1994). Natural lakes and reservoirs have probably been most impacted by point source discharges since wastewater releases to flowing water are most common all over the world. Direct heavy metal discharges to surface waters from industrial or municipal sources have often exceeded atmospheric and other non-point source inputs (Nriagu et al., 1979; von Gunten et al., 1997). Although over the past 30 years such point source discharges have been largely diminished, in sediments accumulated metals have potential for serious environmental risks and should be examined extensively (Wildi et al., 2004).

One of the most common methods for assessing pollution levels and quantifying metal enrichment in bottom sediments is to compare identified concentrations with a background level or with a concentration posing serious environmental risks (e. g. maximum concentration defined in legal environmental regulations and guidelines). However, understanding distribution of pollutants is critical for the environmental management and decision-making (Liu et al., 2006), and spatial distribution of metals in sediments is among the most essential information for environmental research (Marvin et al., 2002).

Therefore, a method combining both data on concentrations of sedimentary metals and simulation using algorithms of spatial interpolation and a geographic information system was applied to assess distribution of heavy metals in the northern part of Lake Babrukas (Trakai district), which had undergone pollution by municipal wastewater in 1964–2002. Specific objectives of this study are twofold: (1) to analyze and assess contamination by heavy metals in surface (0–0.6 m) and subsurface (0.6–1.2 m) layers of bottom sediments, and (2) to demonstrate spatial distribution patterns of heavy metals in the two layers of sediments in the northern part of Lake Babrukas.

## MATERIALS AND METHODS

### Study area

Lake Babrukas is located in Trakai district, to the southwest of Trakai town (5357 inhabitants) and to the northeast of Babriškės settlement (132 inhabitants). The lake belongs to the Neris River basin. Its shape is elongated, stretching almost directly from the south to the north, where excess water overflows into a tiny (area of 1.21 ha) Lake Lovka (Olauka), interconnected with Lake Babrukas through a marshy watercourse with a length of 80 m. An island with an area of 0.21 ha is located almost in the centre of the lake, and the deepest part of the lake (with a maximum depth of 17.2 m) extends from the island to the north-northwest. Other main morphometric parameters of Lake Babrukas are provided in Table 1.

Table 1. Morphometric parameters of Lake Babrukas

Morphometric parameter	Value
Basin area	5.6 km <sup>2</sup>
Lake surface area	37.1 ha
Volume	2459.1 thousand m <sup>3</sup>
Maximum length	1.5 km
Maximum width	0.5 km
Shoreline length	3.5 km
Average depth	6.6 m
Maximum depth	17.2 m
Water circulation	4.85% per year

In 1964 a municipal wastewater treatment plant of Trakai town was constructed on the north-eastern shore of Lake Babrukas and biologically treated wastewater was directly discharged into the lake. In 1970–1980 discharges from the wastewater treatment plant into Lake Babrukas amounted up to 2000 m<sup>3</sup> of wastewater per day. This amount decreased in the nineties and reached 500–600 m<sup>3</sup> per day until 2002, when the wastewater treatment plant was closed (Taminskas, 2004).

### Sampling methodology

The location of sampling sites was influenced by two major assumptions: (1) to provide uniform spatial coverage of the northern bay of the lake, and (2) to allow assessment of sediment quality through the surface (0–0.6 m) and subsurface

(0.6–1.2 m) layers of bottom sediments. For this purpose, the area of the northern part of the lake (4 600 m<sup>2</sup>) was covered with a grid, consisting of equilateral blocks with a border length of 10 m, forming 47 sampling sites in the intersection points of the gridlines. The grid was then overlaid on the sediment distribution map and the points lacking sufficient sediment layer were screened out, resulting in 32 final sampling sites (Fig. 1). Samples from each of the two layers of bottom sediments were collected in the 32 sites constituting a total batch of 64 samples.

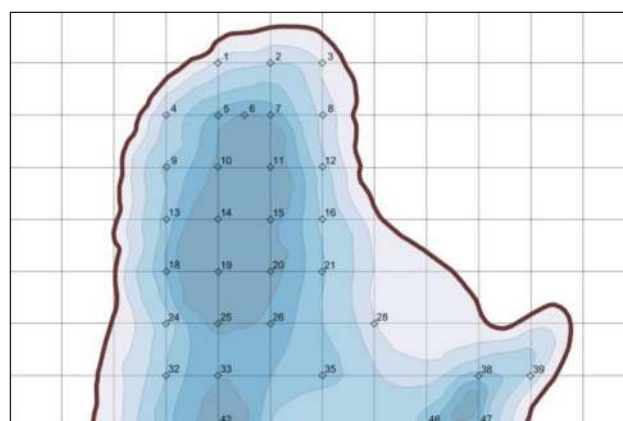


Fig. 1. 32 final sampling sites overlaid on the sediment distribution map

Sampling was performed from a boat in November 2011, using a stainless steel sediment corer with a protective valve to ensure isolation of the core from surrounding sediment and water. In each of 32 sampling sites, sediment core samples were taken from the surface (0–0.6 m) and subsurface (0.6–1.2 m) layers of lake bottom sediments. The length and diameter of a sampled sediment core amounted up to 60 cm and 8 cm, respectively. Each sediment core sample was placed in a sealed polyethylene bag and transported to the laboratory.

### Sample preparation and analysis

Each sediment core sample was homogenized to create a uniform batch, and a combined sample from each batch was taken and placed in a Petri dish to promote uniform air drying. Samples were dried for 11 hours at the temperature of 110 °C

and ground using a porcelain pestle and mortar. Grounded samples were sieved through a 2.0 mm and 250  $\mu\text{m}$  stainless steel sieves, as required for X-Ray Fluorescence analysis (National Exposure Research Laboratory of US EPA 2006). Sieved samples were analyzed using a Thermo Scientific Niton® XL2 X-Ray fluorescence spectrometer.

### Spatial interpolation methodology

The kriging interpolation technique was used to demonstrate spatial distribution patterns of heavy metals in the two layers of sediments in the northern part of Lake Babrukas. Kriging is a method of interpolation, which predicts unknown values from data observed at known locations. This method uses a variogram to express spatial variation, and minimizes the error of predicted values which are estimated by spatial distribution (Oliver, Webster, 1990). In a geostatistics context, kriging can be defined as a generalized linear regression technique used with a variogram model for spatial data interpolation. In this study, the spatial data are the measured concentrations of metals in sediment. The linear regression estimator  $Z^*(\underline{u})$  is the unknown concentration to be estimated by kriging. It is defined as:

$$Z^*(\underline{u}) - m(\underline{u}) = \sum_{\alpha=1}^{N(h)} \lambda_{\alpha} [Z(\underline{u}_{\alpha}) - m(\underline{u}_{\alpha})], \quad (1)$$

where  $\lambda_{\alpha}(\underline{u})$  is the weight assigned to the measured concentration  $z(\underline{u}_{\alpha})$ , which is interpreted as a realization of the random variable  $Z(\underline{u}_{\alpha})$ . The means  $m(\underline{u})$  and  $m(\underline{u}_{\alpha})$  are the expected values of the random variables,  $Z(\underline{u})$  and  $Z(\underline{u}_{\alpha})$ . The mean  $m(\underline{u})$  is used to represent the large scale variation (trend) in the data (Ramanitharan et al., 2005). Spatial interpolation in this study was performed by using ordinary kriging, where the mean  $m(\underline{u})$  is unknown and allowable to fluctuate globally, but assumed constant within the local neighbourhood. It is assumed that kriging provides statistically optimal and unbiased prediction (Tao, 1995; Wang et al., 2003). Kriging procedures were performed and spatial distribution contour maps were prepared by using Golden Software Surfer 9.0 software package. It should be noted that spatial distribution patterns are demonstrated only for those specific metals of interest, which were identified in more than 5 samples in one of the sediment layers and more than 2 samples in the other layer.

## RESULTS AND DISCUSSION

Results of the heavy metals X-Ray Fluorescence analysis are presented in Table 2, along with reference values, maximum allowable concentrations<sup>1</sup> (MAC) for each metal in soil and background concentrations (BC) in Lithuanian soil and in lake sediments. MAC in soil was used as a reference value, because Lithuanian legislation does not provide requirements (or permissible values) for HM concentrations in sediments of natural water bodies. As noted in the methodology section, spatial distribution patterns are demonstrated for those metals which were identified in more than 5 samples in one of the sediment layers and more than 2 samples in the other layer. The results of spatial interpolation for these metals (As, Cr, Cu and Zn) are presented in Figs. 2–9.

As presented in Table 2, **arsenic** (As) was identified in almost half of the 32 samples in both surface and subsurface layers of bottom sediments. In the surface layer, only the maximum identified value 1.24 times exceeded maximum allowable concentration, however both mean and even minimum identified values exceeded background concentration in Lithuanian soils 2.35 and 1.79 times, respectively. The maximum identified value of As in the subsurface layer of sediments exceeded maximum allowable concentration as much as 2.46 times, and even the mean identified value exceeded MAC 1.25 times. Both mean and minimum identified values exceeded background concentration of As in Lithuanian soils (respectively, 3.46 and 1.7 times). Although there is no data available regarding background concentrations of As in Lithuanian lakes, a comparison with background concentration in river sediments of Neris basin, which amounts to 2.4 mg/kg (Kadūnas et al. 1999), shows that concentrations of As in the lake sediments exceed natural background sediment levels. As shown in Fig. 2, arsenic is distributed over the whole surface layer with three maximum content areas found in the south-western, south-eastern and north-eastern parts of the investigated aquatory (bay) of Lake Babrukas. As illustrated in Fig. 3, arsenic concentrations were

<sup>1</sup> According to the Lithuanian legislation (Lithuanian Hygienic Norm HN 60:2004), maximum allowable concentration is the amount of a chemical substance expressed in mg/kg/dry weight of soil or sediments, which has no direct or indirect negative effect on human health.

less variable in the subsurface layer and concentrated mostly in the north-central and northern parts of the bay.

Although **cadmium** (Cd) was found only in 2 samples of the surface layer, both minimum and maximum identified values exceeded maximum allowable concentration 3.58 and as much as 8.52 times, respectively, with the mean value exceeding MAC 6.05 times. Naturally, minimum, mean and maximum identified values also exceeded background concentration of 0.2 mg/kg in Lithuanian soils 53.75, 90.8 and 127.8 times, respectively.

Cadmium was identified in 10 samples of the subsurface layer of bottom sediments and its amounts were even higher than in the surface level: minimum, mean and maximum identified values exceeded maximum allowable concentration 4.03, 5.86 and 10.60 times, and background concentration in Lithuanian soils 60.5, 87.85 and 158.95 times, respectively.

**Chromium** (Cr) was found in 6 and 5 samples of the surface and subsurface layers, respectively, though its identified values did not exceed maximum allowable concentration, and only the maximum identified value in the surface layer of sediments 1.75 times exceeded background concentration in Lithuanian soils. However, mean and maximum identified values of Cr in both layers exceeded background concentration in mineral sediments of Lithuanian lakes (1.34 and 1.45 times in the surface layer; 3.45 and 2.45 times in the subsurface layer, respectively).

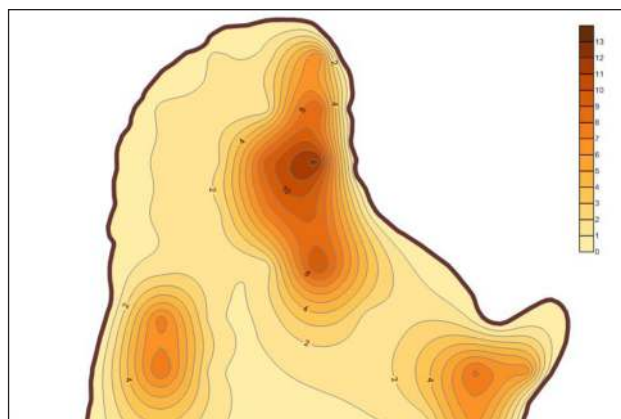


Fig. 2. Distribution of As in the surface (0–0.6 m) layer of sediments (mg/kg)

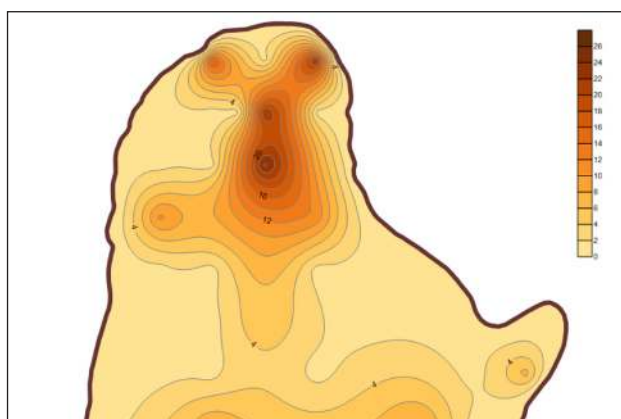


Fig. 3. Distribution of As in the subsurface (0.6–1.2 m) layer of sediments (mg/kg)

Table 2. Concentrations of specific metals of interest in the sediments of Lake Babrukas

Metal	As		Cd		Cr		Cu		Hg		Ni	
	0–0.6	0.6–1.2	0–0.6	0.6–1.2	0–0.6	0.6–1.2	0–0.6	0.6–1.2	0–0.6	0.6–1.2	0–0.6	0.6–1.2
Detected in (No. of samples)	12	14	2	10	6	5	13	7	0	1	0	1
Min., mg/kg	6.45	6.14	10.75	12.10	17.10	16.38	17.18	18.15	–	10.25	–	67.35
Max., mg/kg	12.35	24.59	25.56	31.79	76.97	56.30	32.10	32.41	–	10.25	–	67.35
Median, mg/kg	8.20	9.74	18.16	14.97	22.37	24.33	23.42	24.90	–	–	–	–
Mean, mg/kg	8.46	12.46	18.16	17.57	30.91	33.36	24.07	24.61	–	–	–	–
MAC <sup>1</sup> (in soil), mg/kg	10		3		100		100		1.5		75	
BC in soils <sup>2</sup> , mg/kg	3.6		0.2		44		11		0.1		18	
BC in lake sediments <sup>3</sup> , mg/kg	No data		No data		23.0		9.8		No data		11.3	

Table 2 (continued). Concentrations of specific metals of interest in the sediments of Lake Babrukas

Metal	Pb		Sb		Sn		Zn	
	0–0.6	0.6–1.2	0–0.6	0.6–1.2	0–0.6	0.6–1.2	0–0.6	0.6–1.2
Detected in (No. of samples)	1	0	1	5	1	10	15	12
Min., mg/kg	7.25	–	33.60	17.62	45.09	18.62	11.08	12.80
Max., mg/kg	7.25	–	33.60	59.34	45.09	70.73	261.11	38.47
Median, mg/kg	–	–	–	31.62	–	31.46	20.79	15.77
Mean, mg/kg	–	–	–	32.63	–	35.37	36.77	18.31
MAC <sup>1</sup> (in soil), mg/kg	100		10		10		300	
BC in soils <sup>2</sup> , mg/kg	15		1.5		2.3		36	
BC in lake sediments <sup>3</sup> , mg/kg	17.1		No data		2.3		50.2	

<sup>1</sup> Maximum allowable concentrations, mg/kg (Lithuanian Hygienic Norm HN 60: 2004);

<sup>2</sup> Background concentrations in loamy soils, mg/kg (Lithuanian Hygienic Norm HN 60: 2004);

<sup>3</sup> Background values (median) of trace elements in lake sediments of Lithuania, mg/kg (Kadūnas and Radzevičius 2003).

As shown in Fig. 4, the degree of chromium spatial variability in the surface layer of sediments was not very high however its distribution was similar to the above described arsenic distribution in the same layer, with medium content areas found in the south-western, south-eastern and north-eastern parts of the bay, but with a concentrated anomaly in the central-eastern part. A similar distribution pattern was observed in the subsurface layer (see Fig. 5), though maximum concentration areas shifted from the central-eastern part to the south-eastern and north-eastern parts of the bay.

**Copper (Cu)** was detected in 13 samples of the surface layer of bottom sediments and in 7 samples of the subsurface layer. None of the identified values approached maximum allowable concentration, however they exceeded background concentration in Lithuanian soils and in mineral sediments of Lithuanian lakes. Minimum, mean and maximum identified values of Cu in the surface layer of sediments 1.56, 2.18 and 2.92 times exceeded background concentration in Lithuanian soils, and 1.75, 2.46 and 3.28 times background concentration in mineral sediments of Lithuanian lakes. Similarly, minimum, mean and maximum identified values of Cu in the subsurface layer of sediments 1.65, 2.24 and 2.95 times exceeded background concentration in Lithuanian soils, and 1.85, 2.51 and 3.31 times background concentration in mineral sediments of Lithuanian lakes. As illustrated in Fig. 6 and Fig. 7, Cu was abundant both in the surface and subsurface layers of sedi-

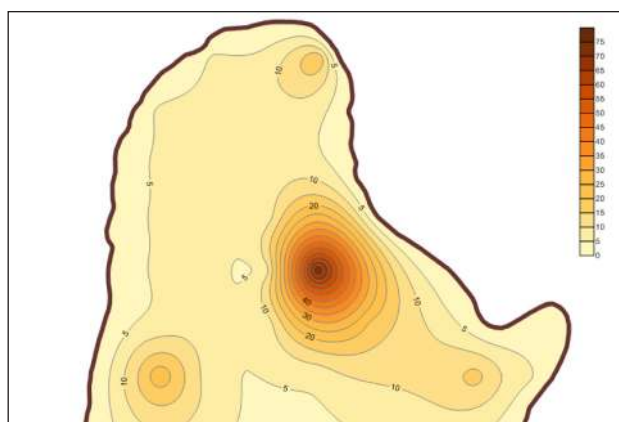


Fig. 4. Distribution of Cr in the surface (0–0.6 m) layer of sediments (mg/kg)

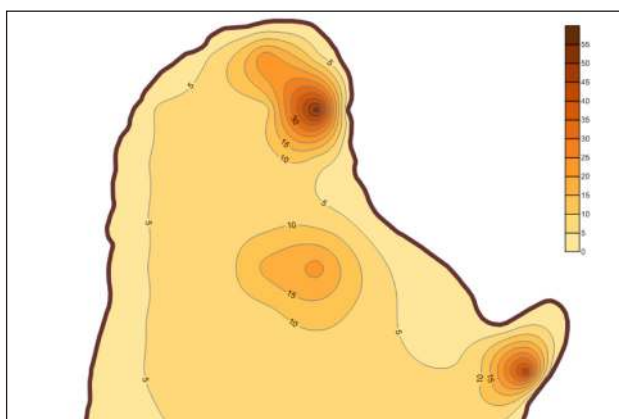


Fig. 5. Distribution of Cr in the subsurface (0.6–1.2 m) layer of sediments (mg/kg)

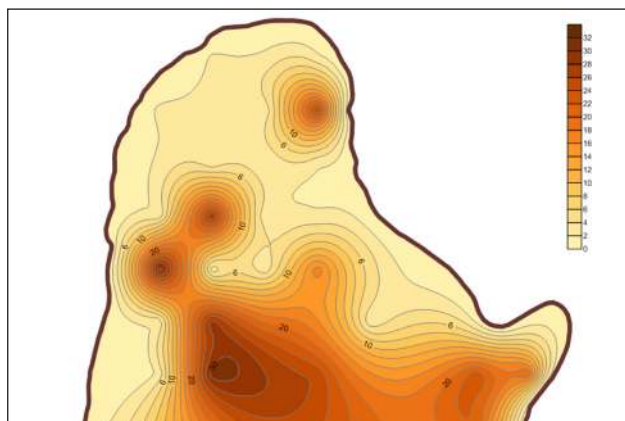


Fig. 6. Distribution of Cu in the surface (0–0.6 m) layer of sediments (mg/kg)

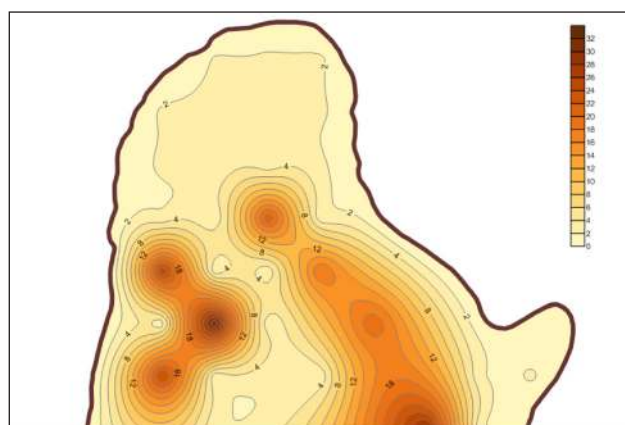


Fig. 7. Distribution of Cu in the subsurface (0.6–1.2 m) layer of sediments (mg/kg)

ments, covering the southern, western and south-eastern parts of the bay and showing increase in concentrations in the northern part of the surface layer and diminishing values in the central part of the subsurface layer.

**Mercury (Hg)** was not detected in any of the samples from the surface layer, and only in a single sample of the subsurface layer. The identified value of 10.25 mg/kg was however very high and 6.83 times exceeded maximum allowable concentration and even 102.5 times background concentration in Lithuanian soils. There is no available data regarding background concentrations of Hg either in Lithuanian lakes or in river sediments.

Similarly to Hg, **nickel (Ni)** was detected in a single sample of the subsurface layer. Contrary to

Hg, the identified value did not exceed maximum allowable concentration, but 3.74 times exceeded background concentration in Lithuanian soils, and 5.96 times background concentration in mineral sediments of Lithuanian lakes.

**Lead (Pb)** was also detected in a single sample of the surface layer, but the identified value of 4.25 mg/kg did not exceed either MAC or background concentrations in soils or sediments of Lithuanian lakes.

**Antimony (Sb)** was detected in one sample of the surface layer of bottom sediments and in five samples of the subsurface layer. The identified value of 33.60 mg/kg 3.36 times exceeded maximum allowable concentration and 22.4 times background concentration in Lithuanian soils. In the subsurface layer, minimum, mean and maximum identified values of antimony 1.76, 3.26 and 5.93 times exceeded maximum allowable concentration, and respectively 11.75, 21.75 and 39.56 times background concentration in Lithuanian soils.

**Tin (Sn)** was detected only in a single sample of the surface layer, but in 10 samples of the subsurface layer. The identified value of 45.09 mg/kg 4.51 times exceeded maximum allowable concentration and 19.61 times background concentration in Lithuanian soils. In the subsurface layer, minimum, mean and maximum identified values of Sn exceeded maximum allowable concentration 1.86, 3.54 and 7.01 times, while background concentration in soils and sediments of Lithuanian lakes was exceeded by minimum, mean and maximum identified values 8.01, 15.38 and 30.75 times, respectively.

**Zinc (Zn)** was found in 15 and 12 samples of the surface and subsurface layers, respectively, however identified values did not exceed maximum allowable concentration. In the surface layer, mean and maximum identified values of Zn 1.02 and 7.25 times exceeded background concentrations in soils, while only the maximum identified value 5.20 times exceeded background concentration in sediments of Lithuanian lakes. In the subsurface layer, only the maximum identified value of zinc 1.07 times exceeded background concentration in soil but did not exceed background concentration in sediments of Lithuanian lakes. Although detected in as many as 15 samples, distribution of zinc in the surface layer of sediments exhibited a clear consolidation of its amounts in the south-eastern

part (see Fig. 8) and a gradual distribution towards the central part of the bay. In the subsurface layer of sediments zinc demonstrated patchy distribution (Fig. 9) with patches of medium concentrations in the south-eastern, south-western and central parts of the bay and a high concentration anomaly in the north-eastern part.

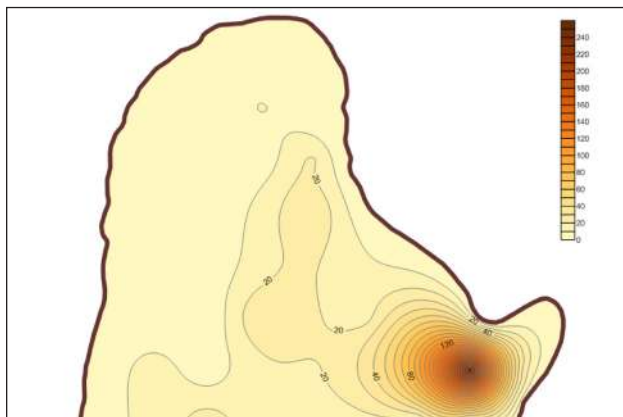


Fig. 8. Distribution of Zn in the surface (0–0.6 m) layer of sediments (mg/kg)

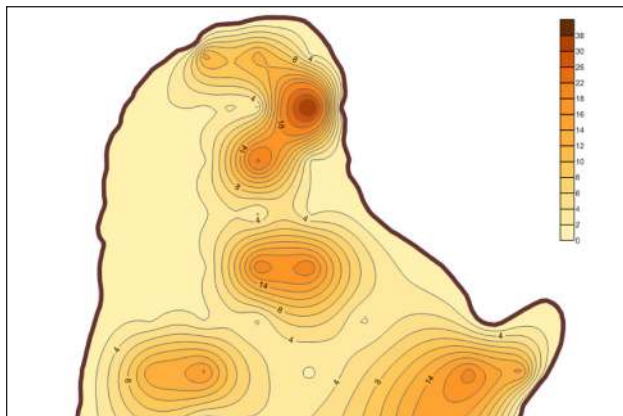


Fig. 9. Distribution of Zn in the subsurface (0.6–1.2 m) layer of sediments (mg/kg)

It is known that metals are released into aquatic systems both from natural and anthropogenic sources, however it is a proven fact that metal loads (especially heavy metals) from human activities are up to several times higher (Nriagu, Pacyna, 1988). Lake Babrukas has no direct inflowing streams and is mostly fed by groundwater

inflows and precipitation (Taminskas et al., 2004) hence it could not receive contaminants via riverine inputs, and discharged wastewater must have been a significant source of pollutants, including heavy metals. This assumption is supported by detected high levels of arsenic (As), cadmium (Cd), antimony (Sb), mercury (Hg), and tin (Sn), which demonstrate major pollution anomalies and a high potential threat to the water ecosystem and even human health by exceeding maximum allowed concentration up to 10.6 times. Furthermore, detected levels of chromium (Cr), copper (Cu), nickel (Ni), and zinc (Zn) show evidence of the effects of anthropogenic activity on the lake by exceeding background concentrations in Lithuanian soils and lake sediments up to 7.25 times. A comparison of these findings with the results acquired by other Lithuanian authors reveals further proof of the impacts from the former municipal wastewater treatment plant of Trakai town and maybe a potential vertical migration of metals. For example, metal concentrations in the bottom sediments of three south-east Lithuanian lakes (Malkėstas, Monis and Pakastas) were within the limits of backgrounds in Lithuanian soils and sediments (Kruopienė, 2004). Unfortunately, concentrations of only 4 heavy metals of interest (Zn, Cu, Pb and Ni) were investigated by the authors of the mentioned study and only in surficial sediments, thus comparison of pollution by heavy metals in deeper layers is not possible. A study of 5 heavy metals of interest (Cr, Ni, Cu, Pb and Zn) in bottom sediments of Lake Didžiulis, located in the same Trakai district, was performed in 2011 by Butkus and Šalčiūnienė. It should be noted that similarly to Lake Babrukas it was being polluted for several decades (until 2002) by Lentvaris town communal and industrial effluents. The study revealed similar contamination and vertical distribution patterns of heavy metals. The authors report high variability of identified heavy metal concentrations in the sediments of Lake Didžiulis: Cr ranged from 6.9 to 45.2 mg/kg, Ni from 20.4 to 55.7 mg/kg, Cu from 7.1 to 80.9 mg/kg, Pb from 24.4 to 87.8 mg/kg, Zn from 13.0 to 75.8 mg/kg. Concentrations of the metals did not exceed the maximum allowable concentrations (MAC) in Lithuanian soil, whereas the concentrations of copper and lead in some samples were close to MAC. Also, there was a significant difference between the amounts of me-



tals in the surface and deeper layers of bottom sediments, with concentrations increasing with the depth of the sediment layer, and levels of Cr and Zn in the near-bottom were even twice as high as in the surface layer. The authors explain this trend by the former pollution entering with effluents the lake water and later also the sediments as well as silt mixing and possible vertical migration of metals; this assumption is in line with the results of heavy metal analysis in Lake Babrukas.

Distribution and behaviour of metals in aquatic environments is very complex and determined by many physical and chemical processes, such as complexation with inorganic and organic ligands, formation of colloid complexes, participation in redox and biological reactions. These processes are further influenced by hydrodynamics, biogeochemical processes and environmental conditions (redox, pH, temperature) of the individual system (Cantwell et al., 2002; Canavan et al., 2007). Furthermore, different metals exhibit different affinities for the various solid-phase fractions of sediment (Calmano et al., 1993). This variability of factors may explain the fact that detected levels of heavy metals in Lake Babrukas vary within a wide range: As from 6.14 to 24.59 mg/kg, Cd from 10.75 to 31.79 mg/kg, Cr from 16.38 to 76.97 mg/kg, Cu from 17.18 to 32.41 mg/kg, Hg from 0 to 10.25 mg/kg, Ni from 0 to 67.35 mg/kg, Pb from 0 to 7.25 mg/kg, Sb from 17.62 to 59.34 mg/kg, Sn from 18.62 to 70.73 mg/kg, Zn from 11.08 to 261.11 mg/kg. Also, it may be assumed that a complex and multifactorial nature of the mentioned processes determines irregular and rather unpredictable spatial distribution of metals detected in both layers of bottom sediments in Lake Babrukas. However, the general trend of high concentration anomalies stretching through the south-eastern and north-eastern parts of the bay towards its northern point may be explained by former discharges of wastewater into the littoral zone of the eastern shoreline and water currents towards excess water overflow into Lake Lovka (Olauka), interconnected with Lake Babrukas in the north. The south-eastern patch of high heavy metal levels, which is recognizable in distribution patterns of most metals, is also in line with the above assumption and probably was caused by retention of suspended matter (and adsorbed metals) in the minor south-eastern embayment of the northern bay of Lake Babrukas.

## CONCLUSIONS

1. Detected levels of heavy metals in sediments of Lake Babrukas varied within a wide range: As from 6.14 to 24.59 mg/kg, Cd from 10.75 to 31.79 mg/kg, Cr from 16.38 to 76.97 mg/kg, Cu from 17.18 to 32.41 mg/kg, Hg from 0 to 10.25 mg/kg, Ni from 0 to 67.35 mg/kg, Pb from 0 to 7.25 mg/kg, Sb from 17.62 to 59.34 mg/kg, Sn from 18.62 to 70.73 mg/kg, Zn from 11.08 to 261.11 mg/kg.

2. Detected levels of most heavy metals: arsenic (As), cadmium (Cd), antimony (Sb), mercury (Hg) and tin (Sn), exceeded maximum allowable concentrations (which have no direct or indirect negative effect on human health) up to 10.6 times, thus high potential threat to the water ecosystem and even human health may be assumed.

3. Detected levels of chromium (Cr), copper (Cu), nickel (Ni) and zinc (Zn) exceeded background concentrations in Lithuanian soils and lake sediments up to 7.25 times and show evidence of the effects of anthropogenic activity (wastewater discharge) on the lake.

4. A comparison of heavy metal amounts and spatial distribution patterns in the two layers of sediments reveals higher pollution levels in the subsurface (0.6–1.2 m) than surface (0–0.6 m) level by most metals, namely As, Cd, Hg, Ni, Sb and Sn.

5. Although distribution of metals in aquatic environments is determined by many physical and chemical processes, the general trend of high concentration anomalies stretching through the south-eastern and north-eastern parts of the bay towards its northern point may be explained by former discharges of wastewater into the littoral zone of the eastern shoreline and water currents towards excess water overflow into Lake Lovka (Olauka), interconnected with Lake Babrukas in the north.

6. The south-eastern patch of high heavy metal levels, which is recognizable in distribution patterns of most metals, could be explained by retention of suspended matter (and adsorbed metals) in the minor south-eastern embayment of the northern bay of Lake Babrukas.

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#### SUNKIŲJŲ METALŲ TARŠOS IR ERDVINIO PASISKIRSTYMO VERTINIMAS BABRUKO EŽERO ŠIAURINĖS DALIES PAVIRŠINIAME IR POPAVIRŠINIAME DUGNO NUOSĖDŲ SLUOKSNIUOSE

##### *Santrauka*

Sunkiųjų metalų kaupimasis vandens telkinių dugno nuosėdose yra aktuali aplinkos užterštumo problema, ypač kai jų kiekiai viršija ne tik fonines koncentracijas, bet ir leistinas normas, nustatytas siekiant išvengti pavojaus gamtiniams aplinkos komponentams ir žmogaus sveikatai. Sunkiųjų metalų kiekių įvertinimas, lyginant juos su didžiausiomis leidžiamomis koncentracijomis ir foninėmis koncentracijomis dirvožemyje ir ežerų nuosėdose, yra patikimas vandens ekosistemos

užterštumo indikatorius, tačiau erdvinė teršalų sklaidos analizė atskleidžia papildomą informaciją apie jų pasiskirstymą nagrinėjamoje teritorijoje. Straipsnyje nagrinėjamas Babruko ežero, į kurį 1964–2002 m. buvo išleidžiamos komunalinės nuotekos, dugno nuosėdų užterštumas sunkiaisiais metalais. 2011 m. lapkričio mėn. ežero šiaurinės dalies paviršiniame (0–0,6 m) ir popaviršiniame (0,6–1,2 m) dugno nuosėdų sluoksniuose buvo paimti 64 mėginiai. Sunkiųjų metalų koncentracijos mėginiuose buvo nustatytos rentgeno fluorescencinės spektrometrijos metodu, o pasitelkus krigingo erdvinės interpoliacijos metodus buvo įvertintas erdvinis metalų pasiskirstymas abiejuose sluoksniuose. Siekiant įvertinti antropogeninės veiklos poveikį, išmatuotos metalų koncentracijos buvo palygintos su didžiausiomis leidžiamomis (DLK) ir foninėmis koncentracijomis Lietuvos dirvožemyje ir ežerų nuosėdose. Lyginamosios analizės rezultatai rodo, kad kai kurių sunkiųjų metalų (arseno (As), kadmio (Cd), stibio (Sb), gyvsidabrio (Hg) ir alavo (Sn)) koncentracijos viršija ne tik fonines vertes, bet ir Lietuvos higienos normos HN 60: 2004 nustatytas DLK, o chromo (Cr), vario (Cu), nikelio (Ni) ir cinko (Zn) koncentracijos viršija fonines koncentracijas Lietuvos dirvožemyje ir ežerų nuosėdose. Sunkiųjų metalų vertikalios pasiskirstymo vertinimas rodo didesnę popaviršinio dugno nuosėdų sluoksnio užterštumą daugeliu metalų (As, Cd, Hg, Ni, Sb ir Sn), sietiną su praeities tarša ir vertikalia teršalų migracija. Erdvinės sklaidos analizės rezultatai demonstruoja netolygų metalų pasiskirstymą dėl šiam procesui įtakos turinčių veiksnių įvairovės ir daugialypiškumo, tačiau anomalinių koncentracijų zonos, besidriekiančios pietrytinėje ir šiaurės rytų nagrinėjamos akvatorijos dalyse, gali būti paaiškinamos į litoralinę zoną 1964–2002 m. išleidžiamų komunalinių Trakų miesto nuotekų tėkme (kartu su ežero vandens pertekliumi) link šiaurėje esančios protakos į Olaukos ežerėlį.

**Raktažodžiai:** ežero dugno nuosėdos, sunkieji metalai, erdvinis pasiskirstymas, erdvinė interpoliacija, krigingas, rentgeno fluorescencinė spektrometrija

