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EFFECTS OF HYDROMECHANICAL LAKE REMEDIATION ON DISTRIBUTION OF METALS AND METALLOIDS IN BOTTOM SEDIMENTS

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VILNIAUS UNIVERSITETAS

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HIDROMECHANINIO EŽERŲ VALYMO ĮTAKA METALŲ IR METALOIDŲ PASISKIRSTYMUI DUGNO NUOSöDOSE

Daktaro disertacija Biomedicinos mokslai, ekologija ir aplinkotyra (03B)

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INTRODUCTION

Relevance of the study. While metals and metalloids are natural constituents of all ecosystems, moving between atmosphere, hydrosphere, lithosphere, and biosphere (Bargagli 2000), it is widely accepted that contamination by some of these elements in sediment, soil, and groundwater is one of the largest threats to environmental and human health (Salomons and Förstner 1984; Nriagu and Pacyna, 1988). This issue is especially significant for lymnosystems, since these elements tend to accumulate in sediments of still water bodies through complex physical and chemical adsorption mechanisms which depend on the nature of the sediment matrix and the properties of adsorbed compounds (Salomons and Brils, 2004; Kilkus, 2005). 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 and Zubenko, 2000; Salomons and Brils, 2004).

Although accumulation of sediments is a natural part of lymnosystem ontogenesis, human induced eutrophication can result in trophic explosion and rapid lake degradation. According to the data, collected by researchers of Lithuanian University of Agriculture, almost 80% of Lithuanian lakes, with an area smaller than 50 ha are unnaturally oversilted (Balevičius et al., 2007). Although processes of excessive nutrient and sediment accumulation can be slowed down by nutrient input and pollution control in the lake basin, in some cases more radical methods of lake restoration are needed. One of such methods – hydromechanical bottom sediment removal (suction dredging) is becoming more and more popular in Lithuania, especially since financial assistance of the European Union became available for such projects.

However, there is scientific evidence that hydromechanical sediment removal results in disruption of natural layers of sediments, and thus potentially can cause resuspension, dispersion and redistribution of trace elements, most of which have toxic effects on living organisms when they exceed a certain concentration. Although the environmental risks related to dredging of contaminated sediments are being discussed in scientific literature, there is a lack of studies documenting the effectiveness of this strategy in removing potentially toxic chemical elements as well as resulting ecosystem effects (Zwolsman and Van Eck, 1993; Zhuang et al., 1994; Singh et al. 2005).

 Hence **the objective of the study** is to investigate and assess changes in distribution of metals and metalloids caused by hydromechanical bottom sediment removal (suction dredging) in two separate parts of a lake, which had undergone different levels of anthropogenic impact. To achieve this objective, the following **tasks of the study** were determined:

- 1. By applying methods of geochemical field work and laboratory analysis to investigate contents of metals and metalloids of interest in bottom sediments of two separate parts of Lake Babrukas: the northern part, which had undergone long-term pollution by municipal wastewater and the southern part, which is located farthest from the former municipal wastewater discharge point.
- 2. To assess sediment quality (contamination level with metals and metalloids of interest) in the surface (0-0.6 m) and subsurface (0.6-1.2 m) layers in the two different parts of the lake before hydromechanical lake remediation (suction dredging).
- 3. To assess sediment quality (contamination level with metals and metalloids of interest) of the newly formed surface sediment layer in the two parts of the lake after hydromechanical lake remediation.
- 4. By employing methods of mathematical statistics, to assess significance of the changes in contents of metals and metalloids caused by hydromechanical bottom sediment removal (suction dredging) in the two parts of the lake.
- 5. By employing methods of spatial statistics, to perform surface interpolation of the laboratory data and to assess spatial redistribution of metals and metalloids caused by hydromechanical bottom sediment removal (suction dredging) in the two parts of the lake.

Scientific novelty of the study. Although hydromechanical lake remediation projects have been carried out over several decades, there still is a lack of evidence about the changes in environmental status after such projects, while scientific literature regarding redistribution of metals and metalloids caused by hydromechanical bottom sediment removal is especially scarce both in Lithuania and in other countries. Assessment of the quality of lake bottom sediments during hydromechanical lake remediation projects and most related studies is usually restricted to measurements of concentrations of pollutants in a few discrete points. Therefore, one of the majors aspects of novelty of this work is compilation and application of a combined assessment system, consisting not only of extensive geochemical field work and laboratory analysis, but also methods of mathematical statistics and spatial interpolation. Additionally, to the best knowledge of the author, there were no studies performed in Lithuania regarding the effects of local lake remediation projects on the quality of bottom sediments hence results provided in this dissertation are novel and especially significant in preparation and design of future sediment removal projects.

Scientific and practical significance. Results of this work supplement scientific knowledge about the impacts of anthropogenic activities on lake sediments and provide novel information about the effects of hydromechanical lake remediation projects on the changes in environmental status of remediated lakes and changes in distribution of metals and metalloids in bottom sediments caused by bottom sediment removal (suction dredging). This data is also practically significant in preparation and design of future sediment removal projects and in determining their feasibility.

Defended statements:

- 1. Hydromechanical lake remediation results in changes of the contents of metals and metalloids of interest and their spatial redistribution in lake bottom sediments.
- 2. Hydromechanical lake remediation causes changes in vertical and horizontal redistribution which are specific to each metal and metalloid, thus a cumulative indicator should be used to assess overall changes in sediment quality of remediated lakes.
- 3. Calculation and statistical analysis of one such indicator total sediment contamination index (Zd) and surface interpolation of its values allow to evaluate statistical significance of changes in contamination degree of the newly formed surface sediment layer and to assess cumulative spatial

redistribution of metals and metalloids caused by hydromechanical lake remediation.

4. Changes in sediment contamination degree caused by hydromechanical lake remediation are statistically more significant in the northern part of the lake, which had undergone long-term anthropogenic impact of municipal wastewater discharge and these changes are evident in vertical migration of metals and metalloids of interest into the newly formed surface layer of sediments and a general increase of their amounts throughout most of the investigated aquatory.

Dissertation structure. The dissertation is written in Lithuanian language and consists of 162 pages, 42 tables and 45 figures, which are presented in the following chapters: Introduction, Literature Review, Information about the Object of Investigation - Lake Babrukas and its Remediation, Materials and Methods, Results and their Discussion, Conclusions, Recommendations, References, and List of Publications of the Author. Reference list contains 277 reference sources.

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LITERATURE REVIEW

This chapter presents a review of information about hydromechanical lake remediation projects, their effects on behavior of metals and metalloids in water and sediments, resuspension of these chemical elements from secondary contamination sources and their ecotoxicologic effects. Also, information about the object of this study - Lake Babrukas and its remediation is provided in a subsequent chapter.

MATERIALS AND METHODS

Study area

Lake Babrukas, which had undergone pollution by municipal wastewater in 1964- 2002 and partial mechanical restoration in 2011 was selected as an object for investigation of the effects of hydromechanical dredging on distribution of selected chemical elements in lake bottom sediments.

Lake Babrukas (Fig. 1) is located in Trakai district, to the southwest of Trakai town (5357) inhabitants) and to the northeast of the Babriškės settlement (132) inhabitants). The lake belongs to Neris River basin.

Figure 1. Location of Lake Babrukas

The shape of Lake Babrukas 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 (Fig. 2). The lake is attributed to the group of lakes with a medium depth. Other main morphometric parameters of Lake Babrukas are provided in Table 1.

Drainage basin area of Lake Babrukas spreads to the west of the lake and according to Lithuanian CORINE land cover database, consists of agricultural land with patches of natural vegetation (38%), forests (33%), urban areas (22%) and water bodies (7%). The lake has no tributaries, thus the main source of its water balance is precipitation $\left(\sim 233.8\right)$ thousand $m³$ per year). Surface inflow is negligible because of modest basin area and dominant light soils, causing fast infiltration of surface runoff, thus underground inflows are more significant and reach up to 14.6 thousand $m³$ of water per year (Zuzevičius, 1996).

Since the lake has no permanent surface outflow, the main water outgo is evaporation (\sim 241,5 thousand m³ per year) and underground outflow (\sim 13,8 thousand m³ per year) (Taminskas et al., 2004).

In 1964 municipal wastewater treatment plant of Trakai town was constructed on the north-eastern shore of Lake Babrukas and biologically treated wastewater was discharged directly into the lake. In 1970-1980 discharges from the wastewater treatment plant into the lake Babrukas amounted up to 2000 m^3 of wastewater per day and constituted a significant input in water balance of the lake. This amount decreased in the nineties and reached 500-600 $m³$ per day until 2002, when the wastewater treatment plant was closed (Taminskas, 2004).

Table 1. Main morphometric parameters of Lake Babrukas

Morphometric	Value		
parameter			
Basin area	5.6 km^2		
Lake surface area	37.1 ha		
Volume	2459100 m^3		
Maximum length	1.5 km		
Maximum width	$0.5 \mathrm{km}$		
Shoreline length	3.5 km		
Average depth	6.6 _m		
Maximum depth	17.2 m		
Water circulation	4.85% per		
	vear		

Figure 2. Bathymetric outline of Lake Babrukas (Taminskas et al., 2004)

Bottom sediment layers in Lake Babrukas were investigated in 2004 by researchers of Institute of Geography and Geology. They measured the thickness of the layers in 53 points of 8 profiles and several single spots. Based on these measurements, a map of bottom sediment distribution was prepared (provided in Fig. 3). According to the measurements, the largest amounts of bottom sediments are located in the northern and southern parts of the lake; the thickness of sediment layers in these parts are accordingly 4- 5 and 5,5-6 meters. Large deposits of bottom sediments were also detected in the deepest locations of the lake: to the north and to the south of the island, located in the center. Total amount of bottom sediments in Lake Babrukas were estimated at about 972,3 thousand $m³$. Hydromechanical removal of bottom lake sediments from the lake was started in July 2011 and finished in November 2011.

Multipurpose dredger "Watermaster Classic III" was used to remove sediments from the northern, southern and partially eastern parts of the lake; total remediation area constituted 6.95 ha, total length of remediated shoreline - 2,78 km, total amount of removed sediment (65-75% humidity) - 53 thousand m^3 .

Suction dredging attachment with a cutter crown and submersible pump was used to disrupt the layers of bottom sediments, mix them with water and discharge to temporary sedimentation sites constructed on the shore. After sedimentation, cleared water was discharged back to the lake, dried sediments (sludge) was removed from the sites an transported for the final utilization in Trakai forestry nursery gardens, rehabilitation of a local quarry and a municipal landfill.

Sampling site selection

Locations of sampling sites were influenced by the following major assumptions: to provide uniform spatial coverage of the investigated areas, and to allow assessment of sediment quality in two separate parts of the lake, which had undergone different levels of anthropogenic impact. For this purpose, two sampling areas were defined: (1) the northern part, which had undergone direct long-term impact by municipal wastewater discharge, and (2) the southern part, which is located farthest from the wastewater discharge point.

Figure 3. Distribution of bottom sediments in Lake Babrukas (Taminskas et al., 2004)

Sampling area of the northern part -4600 m^2 was covered with a grid, consisting of equilateral blocks with a border length of 9 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 as well as those where the depth exceeds 4.5 m (i.e. maximum dredger depth capacity) were screened out, resulting in 32 final sampling sites (see Fig. 4). Sampling area of the southern part of the lake (5100 m^2) was covered with a grid, consisting of equilateral blocks with a border length of 8 m. However, because of bathymetric features of the site (immediate and large increase in water depth towards the center of the area) most intersection points of the gridlines had to be screened out and the border length of the blocks within the bottom sediment deposit had to be reduced to 4 m in order to increase the number of sampling points. Although reducing the blocks resulted in 86 intersection points of the gridlines, the screening procedures produced 31 final sampling site (see Fig. 5).

Figure 4. 32 final sampling sites of the northern part of the lake overlaid on the sediment distribution map

Figure 5. 31 final sampling site of the southern part of the lake overlaid on the sediment distribution map

Sampling methodology

Sampling was performed by the author of this dissertation, both before and after the lake remediation works - respectively in early November, 2011 (from a boat) and in late February, 2012 (through ice), using a stainless steel sediment corer with a protective valve to ensure isolation of the core from surrounding sediment and water. Sediment cores (with a length and diameter of 60 cm and 8 cm respectively) were taken in each of the above described sampling sites (32 in the northern, and 31 in the southern part of the lake). A combined sediment sample was formed from three subsamples of each core, each combined sample was placed in a sealed polyethylene bag.

Before lake remediation, in each of the 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. After lake remediation, sediment core samples were taken in the same sampling sites from the surface (0-0.6 m) layer of lake bottom sediments, i.e. from the newly formed layer, which was directly impacted by hydromechanical (suction) dredging. Total number of combined samples in each investigated part of the lake is provided in Table 2.

Table 2. Total number of combined samples in each of the investigated parts of the lake

Sample preparation and analysis

Within 24 hours, combined sediment samples in sealed polyethylene bags were transported to the laboratory of Ecology and Environmental Center (Vilnius University) and prepared according to the procedures recommended for X-Ray Fluorescence analysis in the Report of US National Exposure Research Laboratory (US EPA, 2006). Preparation and laboratory analysis were performed by the author.

Each combined sample was placed in a Petri dish to promote uniform air drying. Samples were dried for 11 hours at the temperature of 110° C, until constant mass was reached. Dry samples were ground using porcelain pestle and mortar and sieved through 2.0 mm and 250 µm stainless steel sieves. Sieved samples were packed in capsules with ", Mylar" RFS thin $(6 \mu m)$ film and analyzed using a Thermo Scientific Niton® XL2 X-Ray fluorescence spectrometer.

Methods of statistical analysis and evaluation of sediment contamination degree

Statistical analysis of the data was performed by using StatSoft STATISTICA v. 9.1 software package. Data was checked for the condition of normality by using Shapiro–Wilk test. Since the condition was not met, a non-parametric test, specifically Wilcoxon signed ranks test was used for comparison of dependent samples. StatSoft STATISTICA package was also used for descriptive statistics and for drawing the Boxand-Whisker Plots.

Total sediment contamination index (Zd) was used in order to assess cumulative distribution of metals and metalloids before and after the lake remediation works (bottom sediment removal). This index is considered as one of the most precise indicators of the degree of sediment contamination with metals or other microelements (Dumčius et al., 2011), it is the most popular index among Lithuanian researchers thus allowing direct comparison with the results of other studies (Taraškevičius and Gregorauskas, 1993; Budavičius, 2003). Also, Zd was used for compilation of Lithuanian Geochemical Atlas (Kadūnas et al., 1999). Total sediment contamination index (Zd) is defined as:

$$
Z_d = \sum_{k=1}^{n} K_k - (n-1)
$$

where n - number of chemical elements of interest, K_K - concentration coefficient of each element, which is calculated as following:

$$
K_K = \frac{C_M}{C_F}
$$

where C_M - concentration of chemical element in the sample, C_F - background concentration of the element.

Average background concentration values of metals and metalloids in Lithuanian lake sediments (Kadūnas, 2003) were used for calculation of K_K and Zd; average background concentration values in Lithuanian soils were used for those elements, which have no established concentrations in sediments (Lithuanian Hygiene Norm HN 60:2004).

Spatial interpolation methodology

Kriging interpolation technique was used to demonstrate spatial distribution patterns of metals and metalloids in bottom sediments of the northern and southern parts of Lake Babrukas before and after lake remediation. 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 and Webster, 1990; Ramanitharan et al., 2005, Borisenko, 2009). 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 and metalloids in sediment. The linear regression estimator $Z(\mathbf{s}_0)$ is the unknown concentration to be estimated by kriging, which is defined as:

$$
\hat{Z}(\mathbf{S}_0) = \sum_{i=1}^n \lambda_i Z(\mathbf{S}_i)
$$

where $\hat{Z}(\mathbf{s}_0)$ - kriging estimation, $Z(\mathbf{s}_i)$ - the measured concentration in the sampling point, λi - the weight assigned to the measured concentration in the sampling point S_i , *n* - number of measured values.

Spatial interpolation was performed by ordinary kriging, where the mean is unknown and allowable to fluctuate globally, but assumed constant within the local neighbourhood.

Golden Software Surfer 9.0 software package was used to perform Kriging procedures and prepare spatial distribution contour maps. It should be noted that spatial distribution patterns were considered feasible for those metals and metalloids of interest, which were identified in 4 or more samples of the investigated layers of lake bottom sediments.

RESULTS

Distribution of each specific metal and metalloid before and after lake remediation (partial removal of bottom sediments)

As indicated in the methodological chapter, spatial distribution patterns were analyzed only for those metals and metalloids of interest, which were detected in 4 or more samples of the investigated layers of lake bottom sediments (i.e. arsenic, chromium, copper and zinc). Results of these analyses are provided below. It should be noted, that full text of the dissertation also includes analysis and comparison of detected concentrations of all 12 metals and metalloids with reference values (background concentrations and maximum allowed concentrations, MAC) before and after lake remediation.

As (arsenic)

Before lake remediation, in the **northern** part of the lake arsenic was detected in 12 of 32 total samples of the surface (0-0.6 m) layer and in 14 samples of the subsurface layer (0.6-1.2 m). As shown in Table 3, although concentrations of As in the surface layer range from 6.45 to 12.35 mg/kg, and in the subsurface - from 6.14 to 24.59 mg/kg, median concentrations are similar in both layers (respectively 8.20 and 9.74 mg/kg). However, high standard deviation value (6.52) indicates more irregular distribution of arsenic in the subsurface layer, compared to the surface layer (SD of 1.71).

Table 3. Concentrations of arsenic in the two layers of sediments in the northern part of Lake Babrukas before and after lake remediation

Max. conc. exceeds MAC, times 1.24 2.46 0.84

Comparison of detected concentrations (before remediation) with reference values (background concentration and maximum allowed concentration, MAC) clearly indicates impact of anthropogenic activity - discharge of municipal wastewater into the lake: maximum detected concentrations of arsenic in both surface and subsurface layers of sediments exceed MAC respectively 1.24 and 2.46 times, while background concentrations in Lithuanian soils are exceeded respectively 2.69 and 3.19 times.

Although Fig. 6 illustrates higher median concentration and larger interquartile range in the subsurface layer, this difference is not significant according to the results of Wilcoxon signed ranks test (see Table 4) - P value equals to 0.895860 (while $P \le 0.0500$ is considered significant).

After lake remediation, in the northern part of the lake arsenic was detected in 17 of 32 total samples of the surface (0-0.6 m) layer. As shown in Fig. 6, detected concentrations of arsenic after remediation are lower according to all descriptive statistical parameters, and this difference is considered significant (P=0.000069) according to the results of Wilcoxon signed ranks test (Table 4).

Table 4. Results of Wilcoxon signed ranks test, comparing arsenic concentrations in the northern part of Lake Babrukas

Figure 6. Comparison of arsenic concentrations in sediments of the northern part of Lake Babrukas before and after lake remediation

Spatial interpolation of the results of sample analysis demonstrates distribution of arsenic in the two layers of sediments and allows to evaluate changes caused by sediment removal during lake restoration. As shown in the spatial distribution contour map (Fig. 7), three maximum arsenic content areas were detected in the surface layer of sediments before lake remediation.

The largest area with highest concentrations of arsenic (up to 12.35 mg/kg) is located in the north-eastern part of the investigated aquatory and stretching towards minor Lake Olauka, interconnected with Lake Babrukas in the north. The other two, smaller patches are located respectively in the south-eastern (close to the small silted embayment) and south-western parts. Concentrations of arsenic in these two patches are only slightly higher than median - 8.20 mg/kg.

Largest amounts of arsenic in the subsurface layer were detected in the northern part of the aquatory (Fig. 8) - this irregular patch also stretches towards the already mentioned interconnection with Lake Olauka in the north. Maximum arsenic concentrations (up to 24.59 mg/kg) were found in the central part of the patch, while concentrations up to 14.00 and 20.00 mg/kg - respectively in the north-western and the north-eastern parts.

Figure 9 illustrates significant changes in arsenic distribution in the northern part of the lake after partial sediment removal (remediation). Although maximum concentrations are lower (up to 8.36 mg/kg), they were detected in a multitude of patches, distributed all over the investigated aquatory. These results of spatial analysis support and complement the results of statistical and comparative analysis provided above.

Figure 7. Distribution of arsenic in the surface (0-0.6 m) sediment layer of the northern part of Lake Babrukas (before lake remediation)

Figure 8. Distribution of arsenic in the subsurface (0.6-1.2 m) sediment layer of the northern part of Lake Babrukas (before lake remediation)

Figure 9. Distribution of arsenic in the surface (0-0.6 m) sediment layer of the northern part of Lake Babrukas (after lake remediation)

Before lake remediation, in the **southern** part of the lake arsenic was detected in 6 of 31 total sample of the surface (0-0.6 m) layer and in 12 samples of 28 in the subsurface layer (0.6-1.2 m). As shown in Table 5 and Fig. 10, concentrations of As are similar in both layers and range from 6.72 to 12.37 mg/kg in the surface layer and from 5.45 to 14.26 mg/kg in the subsurface; median concentrations are respectively 8.34 and 8.77 mg/kg. Variability of concentrations is rather low in both layers as implied by standard deviation of 1.92 in the surface, and 2.42 in the subsurface layer.

Although concentrations of arsenic are lower in the southern part, which had no direct long-term impact from discharge of wastewater are lower than in the northern part of the lake, maximum detected concentrations of arsenic in the surface and subsurface layers of sediments still exceed MAC respectively 1.24 and 1.43 times, while background concentrations in Lithuanian soils are exceeded 2.73 and 2.87 times respectively. Though Figure 10 illustrates higher median concentration and larger interquartile range in the subsurface layer, this difference is not significant according to the results of Wilcoxon signed ranks test (see Table 6) - P value equals to 0.492790 (while P<0.0500 is considered significant).

After lake remediation, in the southern part of the lake arsenic was detected in 16 of 31 total sample of the surface (0-0.6 m) layer. As shown in Fig. 10, detected concentrations of arsenic after remediation are lower according to almost all descriptive statistical parameters (except maximum value), and this difference is considered significant (P=0.004222) according to the results of Wilcoxon signed ranks test (Table 6).

Table 5. Concentrations of arsenic in the two layers of sediments in the southern part of Lake Babrukas before and after lake remediation

Table 6. Results of Wilcoxon signed ranks test, comparing arsenic concentrations in the southern part of Lake Babrukas

Figure 10. Comparison of arsenic concentrations in sediments of the southern part of Lake Babrukas before and after lake remediation

Changes caused by sediment removal during lake restoration on spatial distribution of arsenic in the southern part of the lake are illustrated in Figures 11-13.

Figure 11. Distribution of arsenic in the surface (0-0.6 m) sediment layer of the southern part of Lake Babrukas (before lake remediation)

As shown in the distribution contour map (Fig. 11), there were six separate patches of elevated arsenic concentrations detected in the surface layer of sediments before lake remediation, while highest concentrations (up to 14.21 mg/kg) are observed in the eastern part of the investigated aquatory. This eastern patch prevails in the subsurface layer (Fig. 12), while two additional areas of elevated concentrations are revealed in the south-eastern and central-western parts.

Figure 12. Distribution of arsenic in the subsurface (0.6-1.2 m) sediment layer of the southern part of Lake Babrukas (before lake remediation)

Figure 13. Distribution of arsenic in the surface (0-0.6 m) sediment layer of the southern part of Lake Babrukas (after lake remediation)

Differently from the northern part of the lake, spatial distribution of arsenic in the southern part of the lake after partial sediment removal (remediation) remains similar to the distribution observed in the subsurface layer before remediation (Fig. 13): pattern of the elevated concentrations remains almost identical, though average and median concentrations tend to be lower and maximum amounts are concentrated in the centralwestern instead of eastern and south-eastern parts.

Cr (chromium)

Before lake remediation, in the **northern** part of the lake chromium was detected in 6 of 32 total samples of the surface (0-0.6 m) layer and in 5 samples of 32 - in the subsurface layer (0.6-1.2 m). As shown in Table 7 and Fig. 14, median concentrations of Cr are similar in both surface and subsurface layers (22.37 and 24.33 mg/kg respectively), however very high variability of concentrations is observed in the surface layer as implied by standard deviation of 22.84. Also, maximum detected concentration (76.97 mg/kg) in the surface layer is significantly higher than in the subsurface (56.30 m) mg/kg). Variability of concentrations is clearly illustrated in Fig. 14 by a very low interquartile range of concentrations in the surface layer, and significance of the difference in concentrations of chromium between the two layers of sediments before lake remediation is proved by the results of Wilcoxon signed ranks test (P=0.000998, see Table 8). However, comparative analysis of detected concentrations (before remediation) with reference values shows no anomalous increase neither in the surface nor subsurface layers of sediments: median concentrations in both layers exceed average background concentrations in Lithuanian lake sediments 1.4 and 1.53 times respectively, while maximum allowed concentrations of chromium were not exceeded in any of the investigated samples.

Table 7. Concentrations of chromium in the two layers of sediments in the northern part of Lake Babrukas before and after lake remediation

After lake remediation, in the northern part of the lake chromium was detected in 4 of 32 total samples of the surface (0-0.6 m) layer. Concentrations of chromium fluctuate from 10.79 to 71.54 mg/kg (standard deviation - 25.76) and opposite to the arsenic, median concentration of chromium in the surface layer are almost two times higher after partial sediment removal (see Table 7 and Fig. 14) and this difference is considered significant (P=0.001140) according to the results of Wilcoxon signed ranks test (Table 8).

Figure 14. Comparison of chromium concentrations in sediments of the northern part of Lake Babrukas before and after lake remediation

Analysis of spatial distribution of chromium in the surface layer of sediments of the northern part of the lake reveals resemblance with distribution of arsenic in the corresponding layer (see Fig. 15): there are three main patches of elevated chromium concentrations, the largest and with highest concentrations (up to maximum value of 76.97 mg/kg) is located in the north-eastern part of the investigated aquatory and connects with a smaller patch of lower concentrations (up to 30.50 mg/kg) located in the south-eastern part (close to the small silted embayment).

Table 8. Results of Wilcoxon signed ranks test, comparing chromium concentrations in the northern part of Lake Babrukas

The third patch with concentrations reaching median value - 22.37 mg/kg is located in the south-western part. Also, a minor patch in the surface layer of northern part should be noted, since this is where maximum amounts of chromium (up to 56.30 mg/kg) prevail in the subsurface layer (see Fig. 16). The above mentioned patch of chromium in the north-eastern part of the investigated aquatory can be also seen in the subsurface layer, though its area is smaller and concentrations are lower. On the other hand, the patch in the south-eastern part (close to the small silted embayment) is more

pronounced in the subsurface layer, and concentrations of chromium in this patch amount up to 45.50 mg/kg and exceed median value almost two times.

Figure 15. Distribution of chromium in the surface (0-0.6 m) sediment layer of the northern part of Lake Babrukas (before lake remediation)

Figure 16. Distribution of chromium in the subsurface (0.6-1.2 m) sediment layer of the northern part of Lake Babrukas (before lake remediation)

Figure 17. Distribution of arsenic in the surface (0-0.6 m) sediment layer of the northern part of Lake Babrukas (after lake remediation)

Differently than in case of arsenic, no diffusion of chromium was observed after lake remediation in the northern part. As illustrated in Figure 17, two discrete patches still can be seen in the north-eastern and south-eastern parts of the investigated aquatory. The north-eastern patch, which was seen in the surface layer has diminished, while the area of the south-eastern patch (close to the small silted embayment) has increased in size.

Before lake remediation, in the **southern** part of the lake chromium was detected in 1 of 31 total sample of the surface (0-0.6 m) layer and in 1 sample of 28 in the subsurface layer (0.6-1.2 m). As shown in Table 9, detected concentrations (respectively 81.12 and 23.75 mg/kg) do not exceed maximum allowed concentration, however they are higher than background concentration in Lithuanian lake sediments - 15.95 mg/kg.

Table 9. Concentrations of chromium in the two layers of sediments in the southern part of Lake Babrukas before and after lake remediation

After lake remediation, in the southern part of the lake chromium was detected in 3 of 31 total sample of the surface (0-0.6 m) layer. Detected concentrations range from 11.17 to 62.65 mg/kg and neither maximum nor mean value exceeds maximum allowed concentration or background concentration of chromium in Lithuanian lake sediments.

Cu (copper)

Before lake remediation, in the **northern** part of the lake copper was detected in 13 of 32 total samples of the surface (0-0.6 m) layer and in 7 samples of 32 - in the subsurface layer (0.6-1.2 m). As shown in Table 10 and Fig. 18, variability of copper concentrations is very similar in both layers (standard deviation values are 5.00 and 5.02 respectively in the surface and subsurface layer), and there is only a minor difference in median concentrations - 23.42 and 24.90 mg/kg. Naturally, there is no significant difference between copper amounts in the two layers according to the results of Wilcoxon signed ranks test (P=0.793487, see Table 11). Comparative analysis of detected concentrations (before remediation) with reference values shows no anomalous increase neither in the surface nor subsurface layers of sediments: maximum allowed concentrations of copper were not exceeded in any of the investigated samples (actually, they are respectively 3.11 and 3.08 times lower), and median concentrations in both layers only exceed average background concentrations in Lithuanian lake sediments respectively 2.96 and 3.15 times. After lake remediation, in the northern part of the lake copper was detected in 10 of 32 total samples of the surface (0-0.6 m) layer. Concentrations of copper after remediation fluctuate within a higher range than before remediation - from 13.45 to 24.07 mg/kg (standard deviation - 7.43), however detected concentrations of copper after remediation are lower according to all descriptive statistical parameters (see Fig. 18), and this difference is considered significant (P=0.000010) according to the results of Wilcoxon signed ranks test (Table 11).

Figure 18. Comparison of copper concentrations in sediments of the northern part of Lake Babrukas before and after lake remediation

Table 10. Concentrations of copper in the two layers of sediments in the northern part of Lake Babrukas before and after lake remediation

Table 11. Results of Wilcoxon signed ranks test, comparing copper concentrations in the northern part of Lake Babrukas

Results of spatial distribution of copper in the surface layer of sediments of the northern part of the lake reveals different patterns of distribution compared with above described arsenic and chromium: major amounts of copper were found in large patches covering most of the south-central and western parts of the investigated aquatory (see Fig. 19).

Also, similarly to other elements of interest, there can be seen a smaller patch, located in the south-eastern part (close to the small silted embayment) and a patch of lower concentrations in the north-eastern part. However, practically no copper was detected in the northern part of the aquatory in the subsurface layer (Fig. 20) - instead two streaks of elevated copper concentrations ranging along the eastern and western littoral zones were distinguished.

Distinctive patterns of copper were also observed after lake remediation. There were negligible amounts of copper detected in the entire eastern side of the aquatory, significantly lower than background concentrations. Also, the patch located in the southeastern part (close to the small silted embayment) was clearly removed. However, the streak ranging along the western littoral zone has prevailed, while major amounts of copper were moved to the north-eastern part of the aquatory (see Fig. 21).

Figure 19. Distribution of copper in the surface (0-0.6 m) sediment layer of the northern part of Lake Babrukas (before lake remediation)

Figure 20. Distribution of copper in the subsurface (0.6-1.2 m) sediment layer of the northern part of Lake Babrukas (before lake remediation)

Figure 21. Distribution of copper in the surface (0-0.6 m) sediment layer of the northern part of Lake Babrukas (after lake remediation)

Before lake remediation, in the **southern** part of the lake copper was detected in 7 of 31 total sample of the surface (0-0.6 m) layer and in only 1 sample of 28 in the subsurface layer (0.6-1.2 m). As shown in Table 12, detected concentrations in the surface layer range from 15.28 to 25.85 mg/kg, with a rather low variability of concentrations (SD=3.24). Median concentration 2.72 times exceeds background concentration in Lithuanian lake sediments, however maximum detected concentration is 3.87 times lower than maximum allowed concentration. Concentration of 21.39 mg/kg, detected in one of the samples of subsurface layer is very close to median concentration of the surface layer (21.49 mg/kg).

Table 12. Concentrations of copper in the two layers of sediments in the southern part of Lake Babrukas before and after lake remediation

After lake remediation, in the southern part of the lake copper was detected in 12 of 31 total sample of the surface (0-0.6 m) layer. As shown in Table 12, detected concentrations of copper after remediation are slightly lower and range from 12.18 to 22.76 mg/kg. Median concentration - 17.52 mg/kg is higher (2.22 times) than background concentration in Lithuanian lake sediments, however maximum detected concentration is significantly (4.39 times) lower than maximum allowed concentration.

Zn (zinc)

Before lake remediation, in the **northern** part of the lake zinc was detected in 15 of 32 total samples of the surface (0-0.6 m) layer and in 12 samples of 32 - in the subsurface layer (0.6-1.2 m). As shown in Table 13 and Fig. 22, median concentration is slightly higher in the surface layer (20.79 vs. 15.77 mg/kg), however this difference is mainly caused by a high maximum value, detected in a single sample of the surface layer (261.11 mg/kg). This value also causes very high standard deviation of 62.40 (SD of concentration in the subsurface layer is much lower - 7.18). Detected zinc concentrations are almost two times lower than background concentration in Lithuanian lake sediments, and maximum detected concentration is significantly (4.39 times) lower than maximum allowed concentration. Even the above mentioned single maximum peak in the surface layer does not reach maximum allowed concentration of 300 mg/kg, while maximum detected concentration in the subsurface is 7.79 times lower than MAC. Results of Wilcoxon signed ranks test (see Table 14) show no significant difference between zinc concentrations in the two layers. After lake remediation, in the northern part of the lake zinc was detected in 21 of 32 total samples of the surface (0-0.6 m) layer. Concentrations of zinc after remediation range from 7.77 to 55.85 mg/kg (standard deviation of 14.54 is lower than its value in the surface layer, but higher than in the subsurface layer.

Figure 22. Comparison of zinc concentrations in sediments of the northern part of Lake Babrukas before and after lake remediation

Table 13. Concentrations of zinc in the two layers of sediments in the northern part of Lake Babrukas before and after lake remediation

Though median value (22.63) after remediation is slightly higher, results of Wilcoxon signed ranks test (see Table 14) show no significant difference between zinc concentrations before and after sediment removal.

Table 14. Results of Wilcoxon signed ranks test, comparing zinc concentrations in the northern part of Lake Babrukas

Analysis of spatial distribution of zinc in the surface layer of sediments of the northern part of the lake reveals a large single agglomeration of zinc located in the southeastern part (adjacent to the small silted embayment, see Fig. 23), where concentrations reach their maximum of 261.11 mg/kg.

In the subsurface layer (Fig 24), a large irregular patch of elevated zinc concentrations is located in the northern part of the investigated aquatory, and stretches (similarly to the one of arsenic) along its central part. Another agglomeration of zinc is located in the south-eastern part, adjacent to the small silted embayment, however concentrations here are lower and only slightly exceed median value. Even lower concentrations were detected in the third patch, located in the south-western part.

Although (as mentioned above) results of Wilcoxon signed ranks test show no significant difference between zinc concentrations before and after sediment removal, evident spatial redistribution of zinc can be seen in the contour map provided in Figure 25. Although the agglomeration of zinc located in the south-eastern part, adjacent to the small silted embayment still can be seen, its area and concentrations are much lower after remediation. However, a large new patch of elevated zinc concentrations is now located in the western littoral, stretching along the entire investigated aquatory.

Figure 23. Distribution of zinc in the surface (0-0.6 m) sediment layer of the northern part of Lake Babrukas (before lake remediation)

Figure 24. Distribution of zinc in the subsurface (0.6-1.2 m) sediment layer of the northern part of Lake Babrukas (before lake remediation)

Figure 25. Distribution of zinc in the surface (0-0.6 m) sediment layer of the northern part of Lake Babrukas (after lake remediation)

Before lake remediation, in the **southern** part of the lake zinc was detected in 30 of 31 total sample of the surface (0-0.6 m) layer and in 25 samples of 28 in the subsurface layer (0.6-1.2 m). Similarly to the northern part of the lake, median concentration of Zn in the surface layer is higher than the subsurface (30.49 vs. 23.01 mg/kg, see Table 15 and Fig. 26), and this difference is also mainly caused by a high maximum value, detected in a single sample of the surface layer (118.22 mg/kg). This also results in a high standard deviation value of 23.17 (SD of concentration in the subsurface layer is much lower - 5.69).

Table 15. Concentrations of zinc in the two layers of sediments in the southern part of Lake Babrukas before and after lake remediation

However, zinc concentrations, detected in both layers of sediments in the southern part of the lake are relatively low - 1.58 and 2.09 lower than background concentration in Lithuanian lake sediments, and even the above mentioned single maximum peak in the surface layer is 2.53 times lower than maximum allowed concentration of 300 mg/kg. Difference of zinc concentrations between the two layers before lake remediation is considered significant (P=0.000248) according to the results of Wilcoxon signed ranks test (Table 16).

Figure 26. Comparison of zinc concentrations in sediments of the southern part of Lake Babrukas before and after lake remediation

After lake remediation, in the southern part of the lake zinc was detected in 30 of 32 total samples of the surface (0-0.6 m) layer. Minimum and maximum concentrations (respectively 18.95 and 73.07 mg/kg), median value (29.27) and standard deviation (15.00) are similar to respective values in the surface layer before remediation, as illustrated in Fig. 26. This statement is supported by the results of Wilcoxon signed ranks test, which show no significant difference between zinc concentrations before and after sediment removal (P=0.798912). Also, as shown in Figures 27-29, spatial distribution of zinc in the surface layer of sediments after remediation has partly maintained its patterns

- though concentrations in the major agglomeration in the south-eastern part decreased from maximum of 118.22 to 65.25 mg/kg, and an increase of zinc amounts in the central and south-western part can be seen.

Figure 27. Distribution of zinc in the surface (0-0.6 m) sediment layer of the southern part of Lake Babrukas (before lake remediation)

Figure 28. Distribution of zinc in the subsurface (0.6-1.2 m) sediment layer of the southern part of Lake Babrukas (before lake remediation)

Figure 29. Distribution of zinc in the surface (0-0.6 m) sediment layer of the southern part of Lake Babrukas (after lake remediation)

Distribution of metals and metalloids before and after lake remediation (partial removal of bottom sediments) according to total sediment contamination index (Zd)

Firstly, it should be noted that concentrations of all 12 metal and metalloids of interest (As, Hg, Cd, Sb, Sn, Pb, Zn, Cu, Ni, Co, Cr and V) were used in calculation of Zd, regardless of the number of samples in which they were detected. Results of Zd calculations and spatial distribution of above mentioned elements before and after lake remediation are provided below.

Before lake remediation, in the **northern** part of the lake total sediment contamination index (Zd) values calculated for the surface layer sediments fluctuate within a very wide range - from 0.23 to 210.46 (see Table 17). Although maximum calculated Zd value of 210.46 is anomalously high (sediment with Zd value >128 is classified as highly hazardous), it is associated with a single sample, while median Zd value is 4.02, and interquartile margins range from 1 to 21.85 (Fig. 30). However it should be noted that total contamination degrees of 11 from total 32 samples of the surface layer are higher than permitted (i.e. exceed Zd=16).

As shown in Table 17 and Fig. 30, total contamination index values are significantly higher in the subsurface layer of sediments: median Zd value equals to 27.63, i. e. is 6.9 higher than in the surface layer, while maximum Zd value reaches 288.89. Also, in 5 samples (from total 32) Zd value exceeds 128, thus sediments are considered as being of highly hazardous contamination degree. Interquartile range of Zd values in the subsurface layer is also much wider - from 4.6 to 91.8 (Fig. 30). Significance of the difference between Zd values in the surface and subsurface layers of sediments is confirmed by the results of Wilcoxon signed ranks test (P=0.001965, see Table 18).

Although in the northern part of the lake maximum total contamination index Zd value after partial sediment removal during the lake restoration has decreased to 114.96, its median value - 15.60 has increased up to three times and approaches upper limit of permitted contamination degree (Zd=16).

Table 17. Total sediment contamination index (Zd) values in the two layers of sediments in the northern part of Lake Babrukas before and after lake remediation

Table 18. Results of Wilcoxon signed ranks test, comparing Zd values in the northern part of Lake Babrukas

Figure 30. Comparison of Zd values in sediments of the northern part of Lake Babrukas before and after lake remediation

Also, there is less variability in Zd values (standard deviation decreased from 38.18 to 22.38) and the limits of interquartile range are higher (from 13.11 to 18.6). Additionally, changes can be seen in the lower range of total contamination index values: before lake restoration, Zd values of 20 (from 32 total) samples ranged from 0.23 to 6, however, after the restoration there were only 4 samples in which Zd values were lower than 6. Significance of the above described increase in Zd values in the surface layer of sediments before and after lake restoration is confirmed by the results of Wilcoxon signed ranks test (P=0.039698, see Table 18).

Spatial interpolation of calculated total contamination index values demonstrates cumulative distribution of the 12 metals and metalloids of interest in the two layers of sediments and allows to evaluate changes caused by sediment removal during lake restoration. As shown in the spatial distribution contour map (Fig. 31), largest amounts (reaching maximum Zd value of 210.46) of investigated elements in the surface layer of sediments before lake remediation were concentrated in an oval patch with a diameter of \sim 15 m in the north-eastern part of the investigated aquatory, adjacent to interconnection of Lake Babrukas with minor Lake Olauka.

A patch of similar spatial extent but with lower values of Zd (up to 60.25) was detected in the center-eastern part; this patch falls into the U shaped larger area of elevated Zd values (ranging from 10 to 20) that stretches through the central part of the aquatory. However, as described above, sediment with Zd values below 10 was dominant in the surface layer of sediments before the lake restoration.

As can be seen in Figure 32, total contamination index values in the subsurface layer of sediments are not only significantly higher but also form more anomalous contamination areas.

Figure 31. Distribution of Zd values in the surface (0-0.6 m) sediment layer of the northern part of Lake Babrukas (before lake remediation)

Figure 32. Distribution of Zd values in the subsurface (0.6-1.2 m) sediment layer of the northern part of Lake Babrukas (before lake remediation)

Figure 33. Distribution of Zd values in the surface (0-0.6 m) sediment layer of the northern part of Lake Babrukas (after lake remediation)

Four such major areas (or patches) should be distinguished, since Zd values in these patches exceed 128, indicating that sediments there should be considered as being of highly hazardous contamination degree.

As mentioned above, Wilcoxon signed ranks test has shown that after lake remediation total contamination index values in the surface layer of sediments have changed significantly. Spatially, significant changes in cumulative distribution of the 12 metals and metalloids of interest after lake remediation can be seen in Figure 33. Although the patch of anomalously high total contamination index values detected before remediation in the surface layer of sediments of the north-eastern part of the investigated aquatory has been removed, there are two newly formed anomalies in the central and western parts of the aquatory with areas similar to the removed patch and with Zd values reaching respectively 114.96 and 85.56. Additionally, spatial interpolation clearly shows general increase in total contamination index values throughout the whole central part of the aquatory, especially within a patch of elevated Zd values stretching from the south to the north and exceeding the upper limit of permitted contamination degree (Zd=16).

Taking into account the above provided results it must be concluded that hydromechanical sediment removal has caused not only significant horizontal redistribution of the 12 metals and metalloids of interest but also vertical migration into the surface layer of sediments and a general increase of their amounts throughout most of the investigated aquatory.

Before lake remediation, in the **southern** part of the lake total sediment contamination index (Zd) values calculated for the surface layer sediments fluctuate within a lower (compared to the northern part) range from 0.33 to 105.09 (see Table 19). Maximum calculated Zd value of 105.09 is within the range of hazardous contamination degree (Zd 32-128), however it is associated with a single sample. It should be noted that median value of Zd - 15.11 is 3.76 times higher than in respective layer of sediment in the northern part of the lake and margins with the upper limit of permitted contamination degree (Zd=16), while the limits of interquartile range are 2.5 and 24.4 (Fig. 34). Also, total contamination values of 14 samples (from total 31) of the surface layer are higher than the permitted level (i.e. exceed Zd=16).

Table 19. Total sediment contamination index (Zd) values in the two layers of sediments in the southern part of Lake Babrukas before and after lake remediation

Figure 34. Comparison of Zd values in sediments of the southern part of Lake Babrukas before and after lake remediation

Opposite than in the northern part of the lake, total sediment contamination index (Zd) values calculated for the subsurface layer sediments are lower than the surface: median Zd value is 9.18, i.e. 40% lower, while maximum Zd value of 46.58 is 2.26 time lower than in the surface (see Table 19 and Fig. 34). However, similarly to the surface layer, total contamination values of 12 from total 28 samples of the subsurface layer are higher than the permitted level (i.e. exceed $Zd=16$) and the limits of interquartile range are only slightly lower - 0.68 and 20.1 (Fig. 34). Lack of significant difference between the two layers is also indicated by the results of Wilcoxon signed ranks test (see Table 20) - P value equals to 0.350496 (while P<0.0500 is considered significant).

Although in the southern part of the lake maximum total contamination index value after partial sediment removal during the lake restoration has decreased from 105.09 to 92.31, its median value has increased from 15.11 to 18.30 and exceeds the upper limit of permitted contamination degree (Zd=16). No changes were observed in variability of total contamination index values before and after lake restoration (standard deviation equals respectively 25.94 and 26.84), while lower limit of the interquartile range increased from 2.5 to 11.06 (upper limit decreased from 24.4 to 21.64). However, opposite than in the northern part of the lake, the difference in total sediment contamination index (Zd) values before and after lake remediation is not significant according to the results of Wilcoxon signed ranks test (P=0.468408, Table 20).

Spatial interpolation of calculated total contamination index values demonstrates irregular cumulative distribution of the 12 metals and metalloids of interest in the surface layer of sediments in the southern part of the lake (before lake remediation). Largest amounts (reaching maximum Zd value of 105.09) of investigated elements in the surface layer of sediments before lake remediation were concentrated in two oval patches with a diameter of \sim 15 m in the western and south-central parts of the investigated southern aquatory (see Fig. 35).

Table 20. Results of Wilcoxon signed ranks test, comparing Zd values in the southern part of Lake Babrukas

$P < 0.0500$ considered significant				
Pairs of variables	N	Z criterion	P value	
Zd values in surface $(0-0.6 \text{ m})$ layer and subsurface $(0.6-1.2 \text{ m})$ layer, before lake remediation	28	0.933628	0.350496	
Zd values in surface $(0-0.6 \text{ m})$ layer, before and after lake remediation	31	0.725072	0.468408	

As shown in Figure 36, total contamination index values in the subsurface layer of sediments in the southern part of the lake are distributed in a different pattern: there are three major areas of elevated Zd values forming an ring shaped anomaly in the center of the investigated aquatory. Maximum Zd values (up to 46.58) were identified in the western and eastern parts, while in the southern part Zd value equals 16, i.e. the upper limit of the permitted contamination degree.

Figure 35. Distribution of Zd values in the surface (0-0.6 m) sediment layer of the southern part of Lake Babrukas (before lake remediation)

Although, as mentioned above, the difference in total sediment contamination index (Zd) values before and after lake remediation is not significant according to the results of Wilcoxon signed ranks test, Figure 37 illustrates spatial redistribution of the 12 metals and metalloids of interest, expressed in Zd values. An evident shift of the areas with elevated Zd values towards the north-western part of the investigated southern aquatory can be seen in the contour map, while the number of these areas has increased up to four, and maximum Zd values of 92.31 were detected in three of them.

Figure 36. Distribution of Zd values in the subsurface (0.6-1.2 m) sediment layer of the southern part of Lake Babrukas (before lake remediation)

Figure 37. Distribution of Zd values in the surface (0-0.6 m) sediment layer of the southern part of Lake Babrukas (after lake remediation)

Additionally, results of interpolation clearly show an increase in median Zd values and an overall increase of the amounts of investigated chemical elements up to the upper limit of moderately hazardous contamination degree (Zd=32) throughout most of the investigated aquatory, especially in the north-western and northern parts of the southern aquatory of Lake Babrukas.

CONCLUSIONS

1. Review of scientific literature reveals that accumulation of excessive amounts of bottom sediments in Lithuanian lakes may be considered a serious environmental problem and the number of hydromechanical (suction dredging) lake remediation projects is constantly increasing, especially since availability of the European Union financial assistance.

2. Analysis of scientific literature also proves that behavior of metals and metalloids of interest in bottom sediments strongly depends on their chemical form and phase, which affect their solubility, mobility, toxicity, processes of adsorption, desorption and formation of chemical compounds, while hydromechanical sediment removal results in disruption of natural layers of sediments, and potentially in resuspension, dispersion and redistribution of these elements.

3. Although most of the metals and metalloids of interest have known physiological activities and are considered trace elements required by living organisms, literature review provides evidence of ecotoxicological risks related to concentrations beyond those necessary for their biological functions.

4. Analysis of concentrations of metals and metalloids of interest in two separate parts of lake Babrukas, which had undergone different levels of anthropogenic impact shows significant effects of long-term discharge of municipal wastewater on the quality of sediments in the northern part of the lake: detected levels of the investigated elements exceed background concentrations up to 103.74 times and maximum allowable concentrations (which have no direct or indirect negative effect on human health) up to 10.6 times.

5. Analysis of samples taken from the surface (0-0.6 m) and subsurface (0.6-1.2 m) layers of lake bottom sediments in the two different parts of the lake before and after hydromechanical (suction dredging) lake remediation and spatial interpolation of the results indicates vertical and horizontal redistribution specific to each investigated metal and metalloid, thus it is concluded that a cumulative indicator should be used for the assessment.

6. Calculation of total sediment contamination index (Zd) and spatial interpolation of its values allow to evaluate cumulative distribution of metals and metalloids in the investigated layers of bottom sediments and to assess sediment contamination degree before and after the lake remediation (partial bottom sediment removal).

7. Statistical analysis and spatial interpolation of total sediment contamination index (Zd) values in the *northern* part of Lake Babrukas, which had undergone longterm pollution by municipal wastewater allow to conclude that (1) before lake remediation, amounts of metals and metalloids of interest were significantly higher in the subsurface (0.6-1.2 m) layer of lake bottom sediments; (2) hydromechanical sediment removal had caused not only significant horizontal redistribution of the 12 metals and metalloids of interest but also vertical migration of these elements into the surface layer of sediments and a general increase of their amounts throughout most of the northern aquatory.

8. Statistical analysis and spatial interpolation of total sediment contamination index (Zd) values in the *southern* part of Lake Babrukas, which is located farthest from the former municipal wastewater discharge point allow to conclude that (1) before lake remediation, amounts of metals and metalloids of interest were lower in the subsurface

(0.6-1.2 m) layer of lake bottom sediments, although this difference is not statistically significant; (2) although the results of employed statistical test show no significant difference in total sediment contamination index (Zd) values before and after lake remediation, interpolation results illustrate clear spatial redistribution of the 12 metals and metalloids of interest, an increase of the number of areas with elevated Zd values and an overall increase of the amounts of investigated chemical elements throughout most of the southern aquatory.

SANTRAUKA

Darbo aktualumas. Nors metalai ir metaloidai (t.y. elementai, pasižymintys ir metalų, ir nemetalų savybėmis), migruojantys tarp atmosferos, hidrosferos, litosferos ir biosferos yra sudėtinė visų ekosistemų dalis (Bargagli, 2000), gamtinės aplinkos užterštumas žalingais jų kiekiais daugiau kaip du dešimtmečius pripažintas pasaulinio masto problema (Salomons and Förstner 1984; Nriagu and Pacyna,1988), kuri ypač aktuali limnosistemoms, nes ežerų dugno nuosėdų sluoksnyje akumuliuojasi maitinamojo baseino dirvožemio, oro bei vandens šaltinių užterštumo sąlygoti teršalų srautai (Salomons and Brils, 2004; Kilkus, 2005). Šiuose elementų akumuliacijos procesuose išskirtini metalai ir metaloidai, kurie aplinkoje natūraliai nesuyra, o gali būti tik praskiedžiami, arba įjungiami į laikinus santykinai nepavojingus kompleksus. Be to, šie elementai linkę biokoncentruotis mitybos grandin÷se ir pasižymi daugiau ar mažiau išreikštu lėtiniu ar ūminiu toksiniu, kancerogeniniu, mutageniniu, genotoksiniu ir kitokio pobūdžio neigiamu poveikiu biotiniams ekosistemos komponentams (Nriagu and Pacyna, 1988; Enserink et al., 1991; Naimo, 1995; Eggleton and Thomas, 2004). Nors ežerų dugno nuosėdose didžioji dalis metalų ir metaloidų yra imobilizuoti įvairiuose junginiuose ir kompleksuose, jie sudaro potencialius antrinės taršos židinius, iš kurių susikaupę teršalai dėl fizikinių–cheminių ir gamtinių sąlygų pokyčių (pvz., antropogenin÷s resuspensijos, pH, oksidacijos-redukcijos (redokso) potencialo, mikrobiologinio aktyvumo) gali patekti į vandens storymę ar dugno nuosėdų vandens frakciją ir tapti pavojingais vandens ekosistemai ir ypač hidrobiontams (Linnik and Zubenko, 2000; Salomons and Brils, 2004).

Kadangi vykstant limnosistemos ontogenezei, dugne kaupiasi vis daugiau nuosėdų, kurios susidaro tiek dėl medžiagos srautų iš maitinamojo baseino ir atmosferos, tiek dėl didėjančio sistemos trofiškumo, kiekvienas ežeras natūraliai evoliucionuoja sausumos geokomplekso kryptimi (Kilkus, 2005). Tačiau žmogaus veiklos sąlygota perteklin÷ maisto medžiagų prietaka (ypač kartu su klimato pokyčiais, sukeliančiais gruntinio vandens lygio pažemėjimą) sutrikdo natūralią, lėtą limnosistemos ontogenezę ir sukelia trofiškumo sprogimą, dėl kurio net ir oligotrofinė sistema per keletą dešimtmečių ar net metų (priklausomai nuo tūrio ir maistmedžiagių apkrovos) gali tapti eutrofine. Lietuvos žemės ūkio universiteto vandentvarkos katedros duomenimis, daugelį Lietuvos ežerų reikėtų priskirti senatvės stadijai, nors dažniausiai nėra galimybių nustatyti ar ežero eutrofikacijos procesai sąlygoti antropogeninės veiklos, ar yra natūralios ontogenzės dalis. Apibendrinus daugiamečius ežerų tyrimo rezultatus, matyti, kad beveik 80% mažesnių kaip 50 ha ploto šalies ežerų yra uždumblėje (Balevičius et al., 2007). Nors dumblo kaupimosi procesus vidutiniškai ar mažai uždumblėjusiuose ežeruose galima sulėtinti ar net stabilizuoti nutraukus teršalų prietaką iš baseino,

optimizuojant baseino žemėnaudą, periodiškai šienaujant antvandeninę augaliją ir pašalinant ją iš vandens telkinio, bei taikant kitus mažiau invazinius būdus, seklesniems vandens telkiniams dažnai prireikia žymiai radikalesnių priemonių – dalinio (arba visiško) nuosėdų pašalinimo (hidro–)mechaninėmis priemonėmis (žemsiurbių pagalba). Šis uždumblėjusių ežerų valymo metodas Lietuvoje taikomas jau nuo 1960-ųjų metų, jis ypač išpopuliarėjo atsiradus galimybei tam panaudoti Europos Sąjungos finansinę paramą.

Kadangi hidromechaninis ežerų dugno nuosėdų šalinimas yra pagrįstas nuosėdų sluoksnių struktūros suardymu ir sumaišymu su vandeniu, vyksta sąlyginai stabilių metalų ir metaloidų junginių ir kompleksų suardymas, įvairių medžiagų vertikalios ir horizontalios migracijos bei resuspensijos procesai. Nors sąsajos tarp vandens telkinių dugno nuosėdų šalinimo darbų ir vandens taršos visuotinai pripažistamos, informacijos apie teršalų elgseną ir jų potencialią grėsmę aplinkai darbų metu bei juos užbaigus yra stebėtinai mažai. Šis informacijos trūkumas dažniausiai siejamas su staigiais fizikiniais bei cheminiais vandens ir dugno terpių pokyčiais ir jų kintamumu laike ir erdvėje (Goossens and Zwolsman, 1996). Nors pats dugno suardymas ir jo sąlygojamas nuosėdų dalelių perskirstymas vienu metu apima 50–100 m pločio poveikio zoną ir tetrunka nuo kelių iki keliolikos valandų (Pennekamp and Quaak, 1990), tačiau dideli ir netolygūs nuosėdų dalelių koncentracijų gradientai, sedimentacijos ir antrinės taršos persiskirstymo procesai yra labai sudėtingi ir nepakankamai ištirti (Zwolsman and Van Eck, 1993; Zhuang et al., 1994; Singh et al. 2005).

Autoriaus žiniomis, Lietuvoje ežerų valymo darbų įtakotų dugno nuosėdų sluoksnių užterštumo pokyčiai iki šiol apskritai nebuvo tiriami, todėl šiame darbe pateikiama informacija yra ypač aktuali vertinant Lietuvos ežerų būklę, planuojant ežerų dugno nuosėdų šalinimo darbus ir nustatant jų tikslingumą.

Darbo tikslas ir uždaviniai. Šio darbo tikslas yra ištirti ir įvertinti hidromechaninio valymo darbų sąlygotus metalų ir metaloidų pasiskirstymo pokyčius antropogeninio poveikio atžvilgiu skirtingose ežero akvatorijose. Tikslui pasiekti buvo iškelti šie uždaviniai:

- 1. Taikant geocheminių lauko tyrimų ir laboratorinės analizės metodus ištirti dviejų priešingų Babruko ežero dalių (šiaurinės, patyrusios tiesioginį ilgalaikį antropogeninį poveikį – Trakų m. komunalinių nuotekų išleidimą ir pietinės – labiausiai nutolusi nuo buvusio tiesioginio antropogeninio poveikio vietos) dugno nuosėdų užterštumą nagrinėjamais metalais ir metaloidais.
- 2. Iki hidromechaninio valymo darbų pradžios, įvertinti abiejų ežero dalių dugno nuosėdų užterštumą paviršiniame $(0-0.6 \text{ m})$ ir popaviršiniame $(0.6-1.2 \text{ m})$ nuosėdų sluoksniuose.
- 3. Pasibaigus hidromechaninio valymo darbams, įvertinti abiejų ežero dalių dugno nuosėdų užterštumą naujai susiformavusiame paviršiniame nuosėdų sluoksnyje.
- 4. Taikant matematinės statistikos metodus, įvertinti valymo darbų įtakotų dugno nuosėdų sluoksnių užterštumo nagrinėjamais metalais ir metaloidais pokyčių reikšmingumą.
- 5. Taikant erdvinės statistikos metodus, atlikti laboratorinių tyrimų ir duomenų prognozių tarpiniuose taškuose interpoliaciją į ištisinį paviršių ir įvertinti

valymo darbų įtaką nagrinėjamų metalų ir metaloidų erdviniam pasiskirstymui.

Darbo mokslinis naujumas. Nors hidromechaninio ežerų valymo darbai vykdomi jau daugelį metų, iki šiol nėra aišku, kaip pasikeičia ežero aplinkosauginė būklė po jo išvalymo, o Lietuvos ir kitų šalių mokslinėje literatūroje informacijos apie valymo sąlygotus metalų ir metaloidų pasiskirstymo dugno nuosėdose pokyčius yra stebėtinai mažai. Valymo projektų ir ežerų dugno nuosėdų būklės vertinimo metu dažniausiai apsiribojama geocheminiais–laboratoriniais tyrimais, t.y. vertinamų cheminių elementų koncentracijų nustatymu pavieniuose taškuose. Todėl esminis šio darbo naujumas yra kompleksinės nagrinėjamų pokyčių vertinimo sistemos sudarymas ir jos pritaikymas, pasitelkiant ne tik išsamius geocheminius lauko ir laboratorinius tyrimus, bet ir matematinę statistinę analizę bei erdvinę duomenų prognozę ir interpoliaciją. Bet to, autoriaus žiniomis, Lietuvoje ežerų valymo darbų įtakotų dugno nuosėdų sluoksnių užterštumo pokyčiai iki šiol apskritai nebuvo tiriami, todėl šiame darbe pateikiama informacija yra nauja ir ypač aktuali vertinant Lietuvos ežerų būklę, planuojant ežerų dugno nuosėdų šalinimo darbus ir nustatant jų tikslingumą.

Mokslinė ir praktinė darbo reikšmė. Darbe pateikiami rezultatai ne tik papildo mokslinę informaciją ir žinias apie antropogeninės taršos šaltinių poveikį ežerų dugno nuos÷dų užterštumui, metalų ir metaloidų pasiskirstymą antropogeninio poveikio atžvilgiu skirtingų ežero akvatorijų dugno nuosėdose, bet ir suteikia naujos informacijos apie hidromechaninio ežerų valymo darbų įtaką dugno nuosėdų užterštumui. Ši informacija yra ypač aktuali vertinant Lietuvos ežerų būklę, planuojant ežerų dugno nuos÷dų šalinimo darbus ir nustatant jų tikslingumą.

Ginamieji darbo teiginiai:

- 1. Hidromechaninis ežero valymas sąlygoja nagrinėjamų metalų ir metaloidų kiekių pokyčius ir jų persiskirstymą dugno nuosėdose.
- 2. Hidromechaninio ežero valymo įtakotas nagrinėjamų metalų ir metaloidų persiskirstymas yra chaotiškas ir skirtingas atskiriems cheminiams elementams, todėl siekiant nustatyti valymo darbų itaką būtina naudoti indikatorius, leidžiančius įvertinti bendro, kumuliacinio nuosėdų užterštumo lygio pokyčius visų nagrinėjamų elementų atžvilgiu.
- 3. Vieno iš tokių indikatorių suminio užterštumo rodiklio Zd verčių statistinė analizė ir erdvinė interpoliacija leidžia ne tik nustatyti statistiškai patikimą hidromechaninio ežero valymo įtakotą metalų ir metaloidų pokyčių reikšmingumą naujai susiformavusiame paviršiniame dugno nuosėdų sluoksnyje, bet ir pademonstruoti erdvinį šių elementų perskirstymą dugno paviršiaus plote.
- 4. Hidromechaninio ežero valymo sąlygoti dugno nuosėdų užterštumo pokyčiai yra statistiškai reikšmingesni antropogeninį poveikį patyrusioje ežero dalyje, kurioje stebima tiek tiriamų metalų ir metaloidų vertikali migracija į naujai susiformavusį paviršinį nuosėdų sluoksnį, tiek ir bendras jų persiskirstymas visame dugno paviršiuje.

Disertacijos struktūra ir apimtis. Disertacija parašyta lietuvių kalba, ją sudaro įvadas, 4 skyriai (literatūros apžvalga, informacija apie tyrimų objektą – Babruko ežerą

ir jo hidromechaninį valymą, tyrimų medžiaga ir darbo metodai, darbo rezultatai ir jų aptarimas), išvados, rekomendacijos, literatūros sąrašas ir autoriaus mokslinių publikacijų sąrašas. Literatūros sąraše cituoti 277 literatūros šaltiniai. Disertacijos apimtis – 162 puslapiai, 42 lentelės ir 45 paveikslai.

Išvados:

- 1. Atlikus literatūros apžvalgą nustatyta, kad Lietuvos ežerų uždumblėjimas yra opi aplinkosauginė problema, o hidromechaninio ežerų dugno nuosėdų šalinimo projektu Lietuvoje vis daugėja, ypač atsiradus galimybei tam panaudoti Europos Sąjungos finansinę paramą.
- 2. Literatūros šaltinių analizė rodo, kad dugno nuosėdų aplinkoje nagrinėjamų metalų ir metaloidų sklaida priklauso nuo jų cheminės formos ir būsenos, kuri įtakoja tirpumą, judrumą, toksiškumą, adsorbcijos, desorbcijos ir cheminių junginių susidarymo procesus, o hidromechaninis dugno nuosėdų šalinimas, pagristas nuosėdų sluoksnių struktūros suardymu ir sumaišymu su vandeniu sąlygoja sąlyginai stabilių metalų ir metaloidų junginių ir kompleksų suardymą, įvairių medžiagų vertikalios ir horizontalios migracijos bei resuspensijos procesus.
- 3. Literatūros apžvalgos rezultatai leidžia teigti, kad nors dauguma nagrinėjamų metalų ir metaloidų yra mikroelementai, būtini daugumai gyvų organizmų, pernelyg didelės jų koncentracijos yra toksiškos hidrobiontams.
- 4. Atlikus nagrinėjamų metalų ir metaloidų tyrimus antropogeninio poveikio atžvilgiu skirtingose Babruko ežero akvatorijose, nustatytas reikšmingas Trakų m. komunalinių nuotekų išleidimo poveikis ežero šiaurinės dalies dugno nuosėdoms, kuriose aptiktos daugumos tiriamų elementų kiekių anomalijos: kai kurių elementų koncentracijos viršija fonines iki 103,74 karto, o didžiausias leidžiamas koncentracijas – iki 10,6 karto.
- 5. Dugno nuos÷dų paviršiniame (0–0,6 m) ir popaviršiniame (0,6–1,2 m) sluoksniuose tyrimų rezultatai ir jų interpoliacija į ištisinį paviršių prieš ir po hidromechaninio ežero valymo darbų rodo dugno nuosėdų šalinimo įtakotą tiriamų metalų ir metaloidų vertikalų ir horizontalų persiskirstymą, kuris yra chaotiškas ir skirtingas atskiriems elementams, todėl valymo darbų įtakos vertinimui būtina naudoti indikatorių, leidžiantį nustatyti bendrą, kumuliacinį tiriamų mėginių užterštumo lygį visų nagrinėjamų elementų atžvilgiu.
- 6. Apskaičiuotas suminis užterštumo rodiklis Zd ir atlikta jo verčių interpoliacija į ištisinį paviršių leidžia įvertinti kumuliacinį nagrinėjamų elementų vertikalų ir horizontalų pasiskirstymą atskiruose nuosėdų sluoksniuose prieš ir po hidromechaninio ežero valymo.
- 7. Tiesioginį ilgalaikį antropogeninį poveikį patyrusios šiaurinės ežero dalies dugno nuosėdų suminio užterštumo rodiklio Zd verčių statistinė analizė ir interpoliacija parodė, kad (1) prieš atliekant hidromechaninio valymo darbus, bendras nagrinėjamų metalų ir metaloidų kiekis buvo reikšmingai didesnis popaviršiniame dugno nuosėdų sluoksnyje; (2) hidromechaninis valymas šiaurinėje akvatorijoje sąlygojo ne tik reikšmingą tiriamų metalų ir metaloidų vertikalią migraciją į paviršinį dugno nuosėdų sluoksnį, bet ir erdvinį jų perskirstymą bei bendrą kiekių padidėjimą beveik visame paviršiaus plote.

8. Pietinės ežero dalies, kuri yra labiausiai nutolusi nuo buvusio tiesioginio antropogeninio poveikio vietos – nuotekų valymo įrenginių išleistuvo dalies dugno nuosėdų suminio užterštumo rodiklio Zd verčių statistinė analizė ir interpoliacija parodė, kad (1) prieš atliekant hidromechaninio valymo darbus, bendras nagrinėjamų metalų ir metaloidų kiekis popaviršiniame dugno nuosėdų sluoksnyje buvo mažesnis nei paviršiniame, tačiau šis skirtumas nėra statistiškai reikšmingas; (2) nors naudotas statistinis testas neparodė reikšmingo skirtumo tarp suminio užterštumo rodiklio verčių paviršiniame sluoksnyje prieš ir po hidromechaninio valymo, interpoliacijos rezultatai rodo aiškų valymo darbų įtakotą tiriamų elementų suminių kiekių erdvinį persiskirstymą, anomaliai aukštų Zd verčių židinių skaičiaus padidėjimą ir bendra suminio užterštumo rodiklio verčiu padidėjimą.

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CURRICULUM VITAE

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