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Metals in bottom sediments of Šventoji Port area (Lithuania)

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The article deals with the distribution of metals (Me) Cr, Pb, Cd, Ni, Cu, Zn, Hg, V, Al and the causality of their spatial dynamics in the bottom sediments of Šventoji Port waters. Areas of highest geochemical anomalies where Me / Al values exceed the calculated background values 1.1 times were distinguished in the zones of marine sand, alluvium and thindispersed material of sedimentation by the method of aluminium normalization. The highest load of all metals, except Cd, falls on the area of the branch to the Eastern basin in the zone of alluvial sedimentation and the sedimentation zone of thindispersed material in the Eastern basin.

Key words: bottom sediments, metals, normalization, Šventoji Port, zones of sedimentation

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INTRODUCTION

The water area of Šventoji Port, built in the inter-war years, has obtained its present contours gradually. Greatest transformations occurred in the part of the port that was closest to the sea. In the course of time, the sand from the Baltic Sea near-shore filled up the external basin of the port and covered the jetties. Alluvial sediments have covered the bottom of the internal port basins. As the Šventoji Port has actually been standing for a few decades, scientific research of its water area has been almost forgotten. The first survey of the composition of bottom sediments in the Šventoji River mouth and port basins was carried out by the authors of the present article in June 1996 (Galkus et al., 1997). It was then determined that the investigated bottom sediments had a variegated composition and that the concentration of metals mainly depended on the grain size of mineral sediments and on the content of total organic carbon (TOC). In 1996, the highest technogenic pollution was determined in the eastern basin of the port. In June 2004, repeated investigations were carried out in the Šventoji Port water area: geodetic, bathymetric, measuring of water indices (transparency, colour, concentration of dregs, etc.), sampling of surface bottom sediments, and measuring of water and particulate drift yields in the Šventoji River (Galkus et al., 2006). Based on the analysis and generalization of obtained data, the following aspects were evaluated: the main geographical factors responsible for the character of sedimentation processes, specific sedimentogenetic zones, formation patterns of the lithological composition of bottom sediments, the influence of bottom relief dynamics on sedimentation processes and the ability of water indices to reflect changes in sedimentation conditions (Galkus et al., 2006). Samples of bottom sediments, taken in a dense network of stations in 2004, allowed correcting the conclusions about the technogenic pollution of Šventoji Port made in 1996 and a detailed examination of specific features of certain groups of metals in the soils. Heavy metals represent one of the mentioned groups. Heavy metals are a group of metals and metalloids having specific gravity greater than 4 g/cm^3 (Connell, Miller, 1984).

Heavy metals get into water basins from a variety of natural and anthropogenic sources. In environments, metal pollution can result from direct atmospheric deposition, geological weathering or through the discharge of agricultural, municipal, residential or industrial waste products (Dawson, Macklin, 1998). Combustion processes are the most important sources of heavy metals, particularly, power generation, smelting, incineration and the internal combustion engine (Hutton, Symon, 1986; Nriagu, 1989; Nriagu, Pacyna, 1988; Wagner, Boman, 2003).

The main sources of heavy metals in the Šventoji Port are the Baltic Sea, the Šventoji River waters and pollution sources in the port itself. The patterns of their accumulation in the Šventoji Port bottom sediments have not been investigated. The authors are the first who undertook to determine the accumulation patterns of heavy metals (Cu, Zn, Pb, Hg, Cd, Cr, Ni, and V) in bottom sediments in the interface of different sedimentation environments applying modern research methods.

The present article analyses the distribution patterns of metals and the causality of their spatial dynamics in the surface bottom sediments of the Šventoji Port water area. Attention is focused on analysis of heavy metals, determining their technogenic constituents and distinguishing the areas of most highly polluted bottom sediments in the Šventoji Port water area.

MATERIALS AND METHODS

Sediment samples were collected in an area of 7.75 ha of the Šventoji Port in June 2004 (Fig. 1). A Van Veen grab sampler was used for collecting sediment samples. Surface (0–5 cm) sediment samples were transferred to plastic bags and frozen. The percentage of fractions from <0.01 mm to >10 mm was determined (Galkus et al., 2006).

The samples (total 71) were dried at room temperature and homogenized before analysis. Sediments were analysed for concentrations of copper (Cu), zinc (Zn), lead (Pb), mercury (Hg), cadmium (Cd), chromium (Cr), nickel (Ni), aluminium (Al), vanadium (V) and total organic carbon (TOC). Mercury was analysed by aqua regia digestion followed by ICP-MS analysis. For the rest elements, a 0.25 g split was heated in HNO_3-$HClO_4$-HF to fuming and taken to dryness. The residue was dissolved in HCl. The solutions were analysed by ICP-MS in ACME Analytical Laboratories LTD (Canada) (Loring, Rantala, 1992; Inductively…, 1998). Blank and standard reference materials (standard DST6/DS) were included in each analytical batch and processed identically to the samples. Analytical results for the standard reference material samples were about ±15% of the value for each metal established.

The content of total organic carbon (TOC) was determined by destruction of carbonates contained in sediments, using mineral acid, and oxidation of organic material at 900 °C using a carbon-analyser (Tiessen, Moir, 1993).

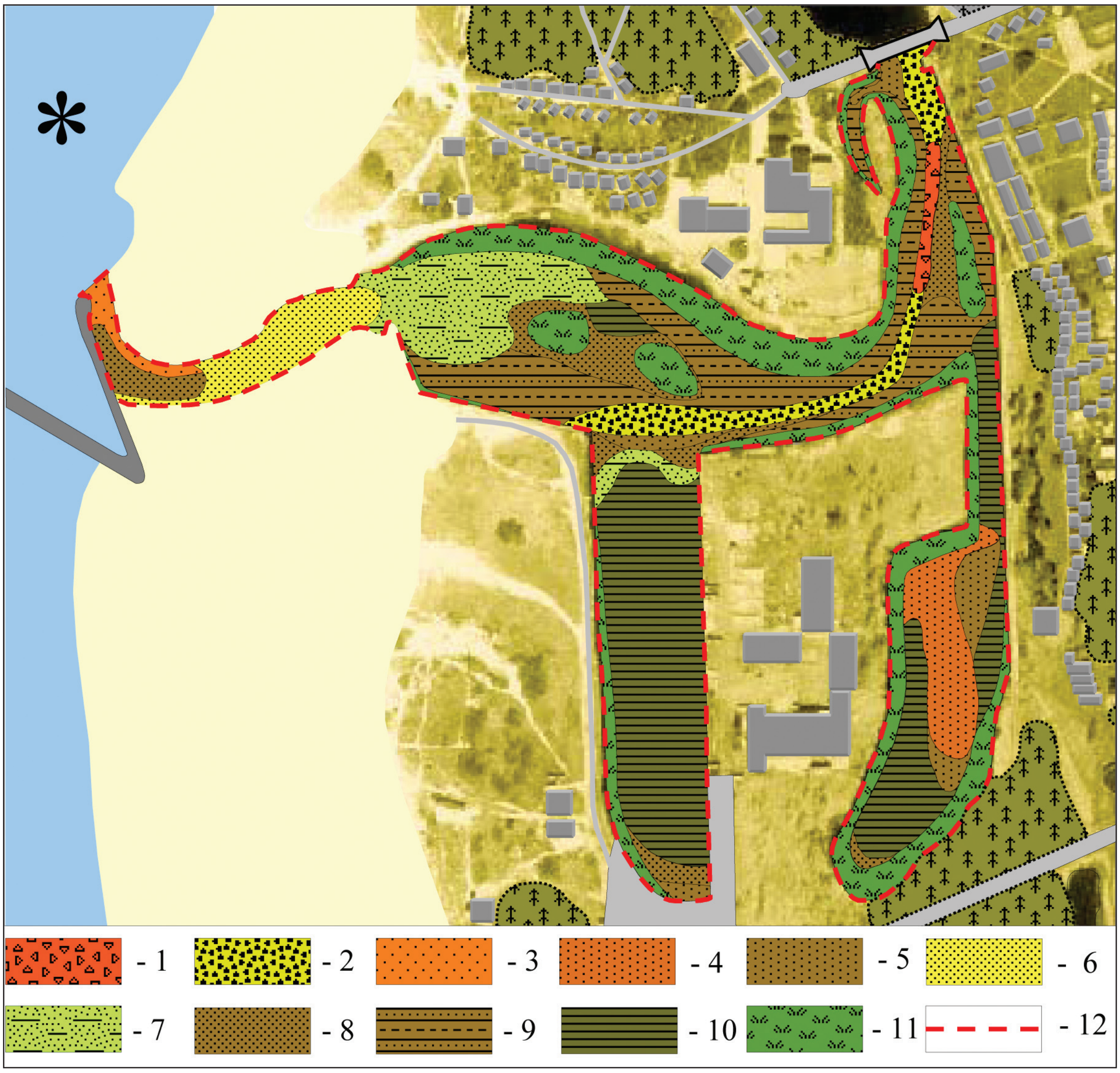

Fig. 1. Surface bottom sediments (0–5 cm) of Šventoji Port (Galkus et al., 2006):
1 – gravel; *2* – sand with gravel; *3* – coarse-grained sand; *4* – medium-grained sand; *5* – muddy medium-grained sand; *6* – fine-grained sand; *7* – silty sand; *8* – muddy fine-grained sand; *9* – muddy sandy silt; *10* – silty-clayey mud; *11* – overgrowths of bulrush and reed; *12* – boundaries of the study area

1 pav. Šventosios uosto akvatorijos paviršinės (0–5 cm) dugno nuosėdos (Galkus et al., 2006):
1 – žvirgždas; *2* – žvirgždinis smėlis; *3* – stambiagrūdis smėlis; *4* – vidutingrūdis smėlis; *5* – dumblingas vidutingrūdis smėlis; *6* – smulkiagrūdis smėlis; *7* – aleuritingas smėlis; *8* – dumblingas smulkiagrūdis smėlis; *9* – dumblingas smėlinis aleuritas; *10* – aleuritins pelitinis dumblas; *11* – plotai, apaugę meldų ir nendrių sąžalynais; *12* – tyrimų rajono riba

The obtained data were analysed and generalized by GIS methods and statistical analysis.

The interrelations of lithological composition of bottom sediments in the Šventoji Port and the Šventoji River, total organics and concentration of metals were determined by calculating (using Microsoft Excel 2000 program) the correlation coefficients (r). When the variables are in direct correlation, the correlation coefficient ranges from 0 to +1, and when they are in inverse correlation it ranges from 0 to –1. When $r = 0$, there is no interrelation among the variables. The determination coefficient (d_{xy}) showed the dependence of Al and Cu, Pb, Zn, Ni, Cd, Cr, Hg, V concentrations. The determination coefficient was calculated using the formula (d_{xy}) = r^2. The mentioned coefficient shows which variables are in a linear correlation (Чертко, 1987).

The method of normalization of metal concentrations with aluminium concentration for revealing geochemical anomalies was chosen because inert aluminium is the main constituent of clay minerals and can serve as a reference element of clay fraction. The concentration of metals in sediments is in direct dependence on the content of clay fraction. Pollution can be identified according to the "leaps" of metal concentrations not consistent with or inadequate to the mentioned dependence (Kersten, Forstner, 1991; Ebbing et al., 2002; Loring, 1991; Schropp et al., 1990; Summers et al., 1996).

INTERRELATIONS BETWEEN THE CONCENTRATIONS OF METALS AND ORGANIC CARBON AND LITHOLOGICAL COMPOSITION OF SEDIMENTS

In 2004, Al and V were analysed together with Cr, Pb, Cd, Ni, Cu, Zn, and Hg (which also were analysed in 1996) in the bottom sediments of the Šventoji River and the Šventoji Port. Metal concentrations in bottom sediments depend on the lithological composition of soils and the content of organic matter in them. In finer sediments with a higher content of organic matter, the concentrations of metals are also higher (Table 1). The maximal concentrations of metals in 1996 and 2004 were recorded in organics-rich silty-clayey mud, and the minimal concentrations were found in the sedimentation zones of gravel and coarse-grained sand.

Average and strong inverse correlations between the concentrations of the most analysed metals and the content of coarse-grained fraction in surface bottom sediments were determined. The absolute values of the correlation coefficients (r) between the content of coarse-grained fraction and the concentrations of lead and mercury exceeded 0.5, and between the content of coarse-grained fraction and aluminium, copper, nickel, vanadium and chromium they exceeded 0.7 (Table 2). The concentration of cadmium was poorly related with sand fraction: the correlation coefficient between these two indices was only 0.32. The correlation between the concentrations of the most analysed metals, except cadmium, lead and mercury, and silt fraction was average. Metal concentrations were in a strongest direct correlation with the content of fine-grained clayey fraction. The correlation coefficients between these indices exceeded 0.7. Only the concentration of cadmium in a bottom sediments was in weak correlation with the content of fine-grained fraction ($r = 0.29$).

A strong correlation ($r > 0.7$) was determined between the content of organic carbon and Zn, Ni, Cr, Hg, Cu, and V. A medium correlation was characteristic of lead and organic carbon concentrations ($r = 0.7$). The correlation coefficient between cadmium and total organic carbon in the port bottom sediments was only 0.27 and showed a weak relation between these variables.

The concentrations of aluminium were in a strong linear correlation (>75% variation explained) with Ni, Cr and V concentrations (Table 3), in a medium linear correlation with copper and zinc concentrations (56% and 61% of variation explained), and in weak linear correlation with mercury, lead and cadmium (18–45% of variation explained). Similar correlation links were characteristic of Al versus Ni, Cr, Hg and Cd in the bottom sediments of the Gulf of Mexico (Summers et al., 1996) and Klaipėda Port (Jokšas et al., 2003). The different correlation links of Al versus Pb, Cu and Zn concentrations in the bottom sediments of the Šventoji Port may be predetermined by local features of the water area and by geographical differences.

Table 1. **Concentration of metals and total organic carbon in the bottom sediments of Šventoji Port**
1 lentelė. **Metalų ir bendros organinės anglies koncentracija Šventosios uosto dugno nuosėdose**

Sediments types	Parameters of concentration	Al, %	Cu, mg/kg	Pb, mg/kg	Zn, mg/kg	Ni, mg/kg	Cd, mg/kg	Cr, mg/kg	Hg, μg/kg	V, mg/kg	TOC, %
Sand (29 samples) (with gravel coarse-grained medium grained muddy medium-grained fine grained silty muddy fine-grained)	Average	2.12	5.10	6.70	30.4	4.21	<0.4	7.28	16.9	11.7	0.71
	SD	0.31	3.20	5.00	15.4	2.19	0.04	5.29	7.66	4.67	0.72
	Min	1.46	<2.00	<5.00	5.0	<2.00	<0.4	1.00	<10.0	5.00	0.03
	Max	2.78	16.0	30.0	72.0	10.0	0.40	19.0	39.0	19.0	3.20
Silt (15 samples) (muddy sandy silt)	Average	2.30	6.11	7.29	57.4	7.43	0.40	14.4	23.6	16.9	1.69
	SD	0.27	4.11	2.95	15.3	2.44	0.11	4.80	7.37	4.45	0.55
	Min	1.99	<2.00	<5.00	30.0	2.00	<0.4	6.00	10.0	10.0	0.65
	Max	2.68	18.0	13.0	86.0	12.0	0.60	21.0	37.0	23.0	2.97
Mud (27 samples) (silty-clayey mud)	Average	3.31	24.7	32.8	158	15.4	0.51	33.8	87.7	35.4	4.94
	SD	0.65	10.2	16.7	59.0	5.72	0.42	13.0	43.3	12.9	1.81
	Min	2.10	8.00	10.0	45.0	5.00	<0.4	7.00	32.0	14.0	1.70
	Max	4.46	49.0	75.0	276	25.0	1.90	54.0	201	57.0	8.68

SD: standard deviation; Min–max: minimum–maximum levels.

Table 2. **Correlation coefficients of metals, total organic carbon and bottom sediment fractions**

2 lentelė. **Metalų bendros organinės anglies ir dugno nuosėdų frakcijų kiekio koreliacijos koeficientai**

Parameters	F1	F2	F3	Al	Cu	Pb	Zn	Ni	Cd	Cr	Hg	V	TOC, %
F1	1.00												
F2	–0.94	1.00											
F3	–0.81	0.57	1.00										
Al	–0.74	0.60	0.78	1.00									
Cu	–0.72	0.55	0.82	0.75	1.00								
Pb	–0.63	0.48	0.75	0.61	0.79	1.00							
Zn	–0.75	0.57	0.87	0.78	0.92	0.83	1.00						
Ni	–0.77	0.61	0.82	0.87	0.82	0.66	0.90	1.00					
Cd	–0.32	0.32	0.29	0.43	0.25	0.36	0.39	0.38	1.00				
Cr	–0.78	0.63	0.82	0.88	0.82	0.66	0.90	0.98	0.38	1.00			
Hg	–0.68	0.49	0.83	0.67	0.85	0.81	0.93	0.83	0.24	0.82	1.00		
V	–0.76	0.61	0.80	0.91	0.79	0.63	0.85	0.97	0.37	0.98	0.78	1.00	
TOC, %	–0.79	0.59	0.91	0.80	0.89	0.70	0.94	0.91	0.27	0.90	0.90	0.86	1.00

F1 – content (%) of fracion (>0.1 mm) in the bottom sediments,
F2 – content (%) of fracion (0.1–0.063 mm) in the bottom sediments,
F3 – content (%) of fracion (<0.063 mm) in the bottom sediments.

F1 – frakcijos (>0.1 mm) kiekis (%) dugno nuosėdose,
F2 – frakcijos (0.1–0.063 mm) kiekis (%) dugno nuosėdose,
F3 – frakcijos (<0.063 mm) kiekis (%) dugno nuosėdose.

Table 3. **Determination coefficients of aluminium versus concentrations of different metals in the bottom sediments of Šventoji Port (%)**

3 lentelė. **Šventosios uosto dugno nuosėdų aliuminio ir įvairių metalų koncentracijų determinacijos koeficientai (%)**

Metals	Cu	Pb	Zn	Ni	Cd	Cr	Hg	V
$d_{xy,\%}$	56	40	61	76	18	77	45	83

The correlation between Al and V, Cr and Ni was also strong: $r > 0.9$. These metals are ranged according to an increasing atomic weight in line 4 of period IV of the Mendeleyev periodic table. Cu and Zn, which are in a medium correlation with Al, are ranged side by side in line 5 of period IV. The correlation coefficient of their concentrations is $r = 0.92$.

The correlation between mercury and lead, weakly related with Al concentration, is strong ($r = 0.81$). According to atomic weight, they are ranged in line 9 of period VI of the Mendeleyev periodic table. Cadmium, which is weakly correlated with the analysed variables, is ranged in a different place of the Mendeleyev table: line 7 of period V.

These observations show that the distribution pattern of metals in the bottom sediments of the Šventoji Port depends not only on the lithological composition and content of organic matter, but also on the chemical properties of metals. Moreover, close links between some metals (Cr, V and Ni) and their close link with aluminium concentration may be related with a similar sedimentation and migration patterns of these metals. Cu and Zn; Hg and Pb, and Cd classified in other groups according to chemical properties and correlation links may have other sources and slightly different migration patterns in bottom sediments than the above-mentioned metals.

SPECIFIC DISTRIBUTION PATTERNS OF METALS

Aluminium (Al). The concentration of aluminium in the bottom sediments of the Šventoji River and the Šventoji Port increases with an increasing content of fine-grained fraction. In silt, aluminium concentration is on the average 1.1 times higher than in sandy sediments, and in mud it is 1.6 times higher.

In the highly heterogeneous bottom sediments of the river channel (Fig. 1), the distribution pattern of aluminium concentrations is rather monotonous, ranging within the limits of 2–3% (Fig. 2). In the approaches to the river mouth, where the Baltic Sea coastal sand plays the most important role in sedimentation, the concentration of aluminium in sediments reduces and reaches only less than 2%. Only in the northern edge of the mentioned sector and in the mouth itself, the concentration of aluminium increases to 2.5 and 2.2%, respectively. The highest concentration of aluminium in silty-clayey mud covering the coarse-grained alluvial sediments of the Šventoji River branch to the Eastern basin reaches 3.57% at the eastern embankment of the Eastern basin. The concentration of aluminium in the silty-clayey mud of the southern part of the Eastern basin ranges within the limits of 3–4%. In the coarse-grained sediments of the basin, the concentration of aluminium decreases: to 2–3% in silty medium-grained sand and to <2% in medium-grained sand (Figs. 1, 2).

The Western basin, once part of the river channel, today is a peripheral water area in terms of water circulation. It accumulates the finest sedimentary material (fraction <0.063 mm accounts for >70%) which together with plankton detritus forms areas of spongy mud (Galkus et al., 2006). In the silty-clayey mud covering almost the whole Western basin, the concentration of aluminium does not fall below 3% and in many places of the central and southern parts of the basin exceeds 4%. Only in the approaches to the river mouth and at the southern edge of the basin, the concentration of aluminium is lower and ranges within the limits of 2–3%.

The concentration of Cu, Pb, Zn, Ni, Cd, Cr, Hg and V in the finest (silty-clayey mud) bottom sediments of the Šventoji Port water is on the average 2–5 times higher than in the coarse-grained (sands) sediments. The dynamics of V, Cr and Ni concentrations, closely related with the shifts of Al concentrations in bottom sediments, follows a similar pattern.

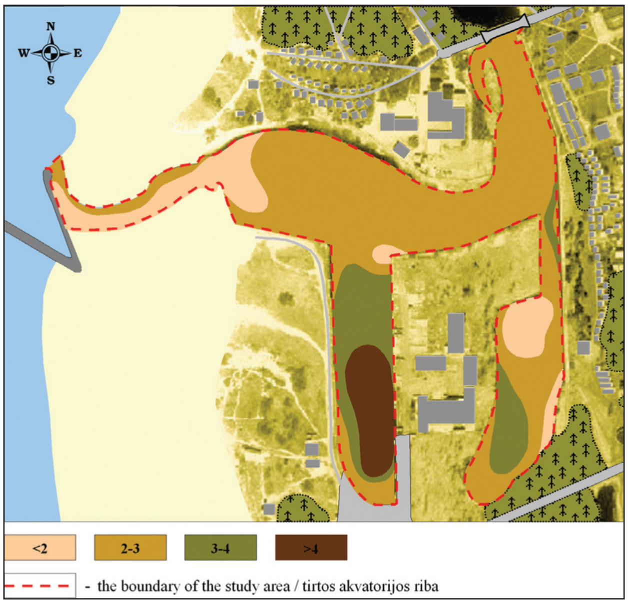

Fig. 2. Aluminium (Al) concentration (%) in the bottom sediments (0–5 cm) of Šventoji Port

2 pav. Aliuminio (Al) koncentracija (%) Šventosios uosto paviršinėse (0–5 cm) dugno nuosėdose

Vanadium (V). The concentration of vanadium in the bottom sediments of the Šventoji River ranges within <10–30 mg/kg (Fig. 3). Higher concentrations (20–30 mg/kg) are characteristic of the Šventoji reach up to its sharp turn to the west towards the sea and a small area of silty sediments north of the water area overgrown with water plants where an intensive accumulation takes place (Galkus et al., 2006). In the mud sediments of the Western basin, V concentrations range from 33.0 to 57.0 mg/kg. The highest concentrations are characteristic of the southern part of the basin. The mud of the eastern basin contains smaller concentrations of V. The highest concentration reaches 44.0 mg/kg. In the coarse-grained sediments, V concentrations decline and do not exceed 20 mg/kg.

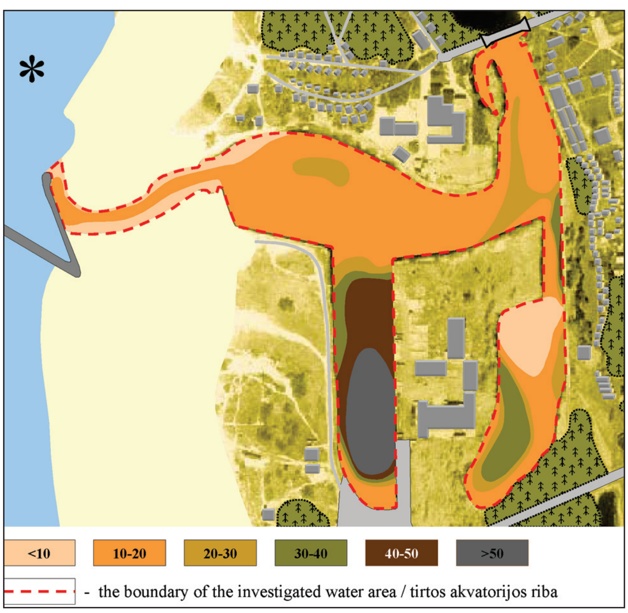

Fig. 3. Vanadium (V) concentration (mg/kg) in the bottom sediments (0–5 cm) of Šventoji Port

3 pav. Vanadžio (V) koncentracija (mg/kg) Šventosios uosto paviršinėse (0–5 cm) dugno nuosėdose

The distribution of the V/Al ratio in the bottom sediments shows the areas of highest vanadium loads (V/Al > 11): the Western basin; the southern part of the Eastern basin, and the area of the branch to the Eastern basin. North of the Western basin forming an alluvial shoal and at the old jetty in the approaches to the Šventoji mouth, the values of V/Al ratio in the bottom sediments (V/Al 9–11) are higher than in the remaining part of the investigated river segment. Slightly higher values of the V/Al ratio (V/Al: 7–9) occur in the area extending along the deep western part of the Šventoji River almost up to the mouth of the Western basin and in the wide area extending along the fairway from the approaches to the Šventoji mouth to the old jetty (Fig. 4).

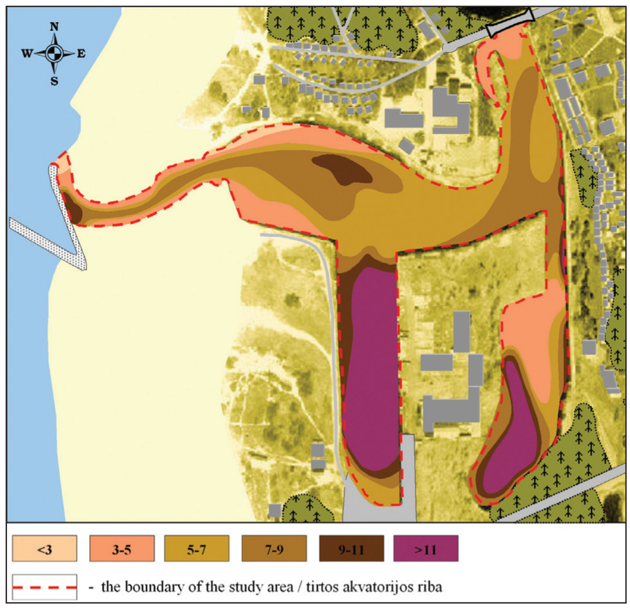

Fig. 4. Vanadium (V) concentration normalized relatively to aluminium in the surface bottom sediments (0–5 cm) of Šventoji Port

4 pav. Vanadžio (V) ir aliuminio koncentracijos santykis Šventosios uosto paviršinėse (0–5 cm) dugno nuosėdose

Chromium (Cr). The highest concentrations of chromium have accumulated in the silty-clayey mud of the Eastern and Western basins where they reach 47.0 mg/kg and 54.0 mg/kg, respectively. The concentration of Cr in the mud of the branch to the Eastern basin approaches 44.0 mg/kg. In the area of silty-clayey mud near the Western shoal in the Šventoji River, the concentration of chromium is lower and reaches 28.0 mg/kg. In the remaining part of the port water area where coarser fractions are dominant in bottom sediments, the concentration of chromium does not exceed 20 mg/kg (Fig. 5).

According to the values of the Cr/Al ratio, the highest loads (Cr/Al: >11) are characteristic of the area at the eastern embankment of the branch to the Eastern basin, the southern part of the Eastern basin and a wide area extending along the eastern embankment of the Western basin. Small chromium anomalies (Cr/Al: 8–11) were recorded in the sediments of the Šventoji channel (Fig. 6).

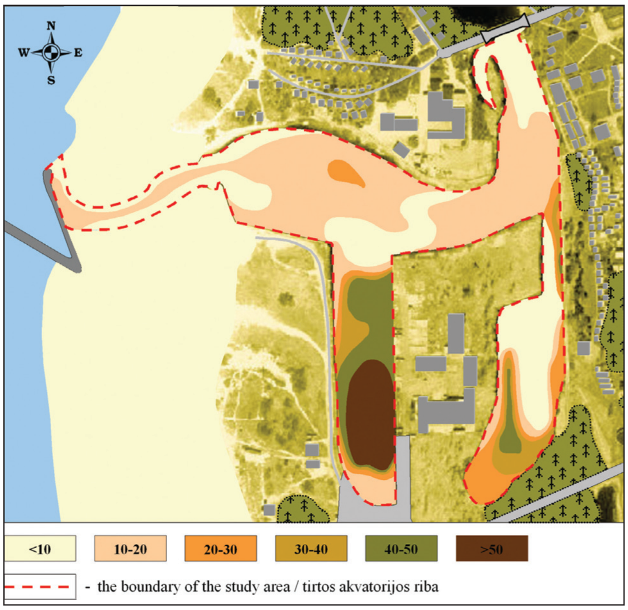

Fig. 5. Chromium (Cr) concentration (mg/kg) in the bottom sediments (0–5 cm) of Šventoji Port

5 pav. Chromo (Cr) koncentracija (mg/kg) Šventosios uosto paviršinėse (0–5 cm) dugno nuosėdose

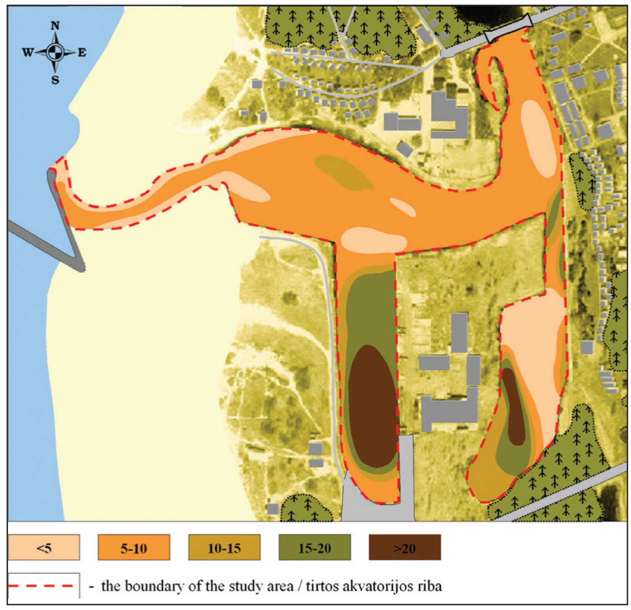

Fig. 7. Nickel (Ni) concentration (mg/kg) in the bottom sediments (0–5 cm) of Šventoji Port

7 pav. Nikelio (Ni) koncentracija (mg/kg) Šventosios uosto paviršinėse (0–5 cm) dugno nuosėdose

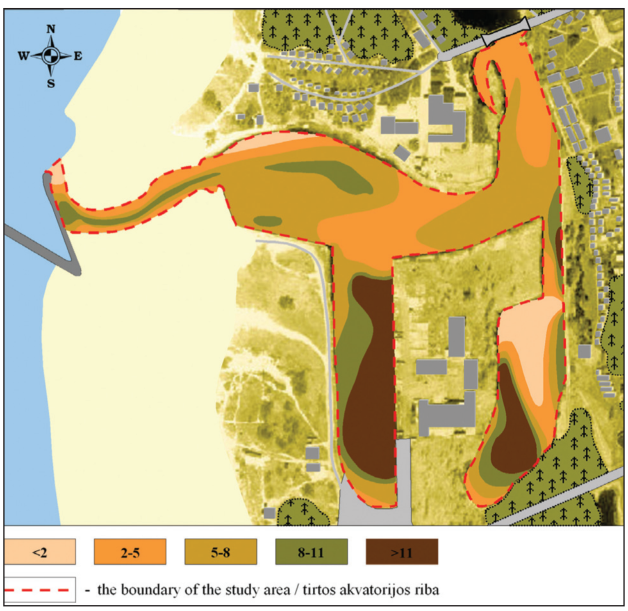

Fig. 6. Chromium (Cr) concentration normalized relatively to aluminium in the surface bottom sediments (0–5 cm) of Šventoji Port

6 pav. Chromo (Cr) ir aliuminio koncentracijos santykis Šventosios uosto paviršinėse (0–5 cm) dugno nuosėdose

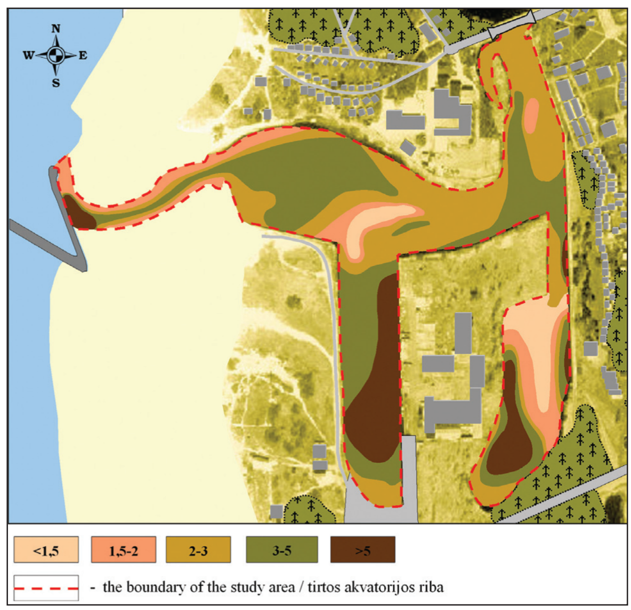

Fig. 8. Nickel (Ni) concentration normalized relatively to aluminium in the surface bottom sediments (0–5 cm) of Šventoji Port

8 pav. Nikelio (Ni) ir aliuminio koncentracijos santykis Šventosios uosto paviršinėse (0–5 cm) dugno nuosėdose

Nickel (Ni). The highest nickel concentrations were found in the mud of the Eastern and Western basins (25.0 mg/kg and 24.0 mg/kg, respectively). In the locally accumulated Šventoji mud north of the Western alluvial shoal, the concentration of Ni ranges within the limits of 10–15 mg/kg. In sand and silty sediments, Ni concentration usually does not exceed 10 mg/kg (Fig. 7). Only northward of the Westetrn alluvial shoal, the concentration of Ni in muddy sandy silt of the Šventoji River equals to 12.0 mg/kg.

The distribution pattern of the Ni/Al ratio shows the following main areas of anomalies: the area at the eastern embankment of the branch to the Eastern basin, mud accumulation areas in the Eastern basin, the southern part of the Western basin, and the area at the old jetty in the approaches to the Šventoji mouth (Fig. 8).

Zinc (Zn). The concentration of Zn in the inequigranular sand of the Šventoji River, increasing almost two-fold around the forming alluvial shoals in the areas predominated by finer sediments and in the approaches to the mouth at the old jetty, ranges within 10–50 mg/kg (Fig. 9). The highest concentrations of Zn (up to 232 mg/kg) in the mud of the Western basin occur in its southern part. The maximal Zn concentration (276 mg/kg) was recorded in the mud of the Eastern basin. In the coarser bottom sediments of the central and northern part of the basin, the value of Zn concentration does not exceed 50 mg/kg.

The distribution pattern of the values of the Zn/Al ratio shows that the highest loads (Zn/Al >70) at present occur in the areas of mud accumulation of the Eastern basin (Fig. 10). The loads are smaller in the Šventoji branch to the Eastern basin and in the Western basin. Areas of higher loads are situated in the fairway of the Šventoji River up to the Eastern basin, north and south of the Western alluvial shoal and at the old jetty in the approaches to the river mouth.

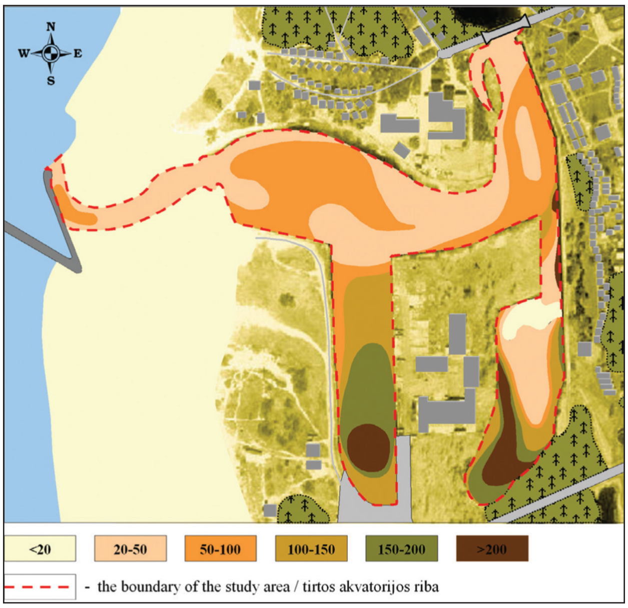

Fig. 9. Zinc (Zn) concentration (mg/kg) in the bottom sediments (0–5 cm) of Šventoji Port

9 pav. Cinko (Zn) koncentracija (mg/kg) Šventosios uosto paviršinėse (0–5 cm) dugno nuosėdose

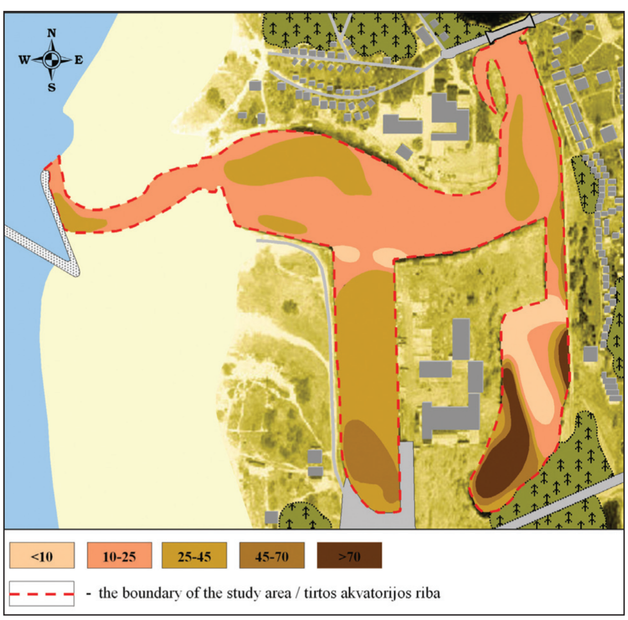

Fig. 10. Zinc (Zn) concentration normalized relatively to aluminium in the surface bottom sediments (0–5 cm) of Šventoji Port

10 pav. Zinko (Zn) ir aliuminio koncentracijos santykis Šventosios uosto paviršinėse (0–5 cm) dugno nuosėdose

Copper (Cu). The concentration of copper in the inequigranular bottom sediments of the Šventoji River is below 10 mg/kg, except the approaches to the mouth area where it ranges within 10–20 mg/kg (Fig. 11). Highest concentrations of copper were found in the mud at the eastern embankment of the Šventoji River branch to the Eastern basin (49.0 mg/kg). The concentration of copper in the silty-clayey mud in the southern parts of the Eastern and Western basins ranges within 30–40 mg/kg. In the coarse-grained sediments of the Eastern basin, the concentrations are considerably lower and not exceed 10 mg/kg.

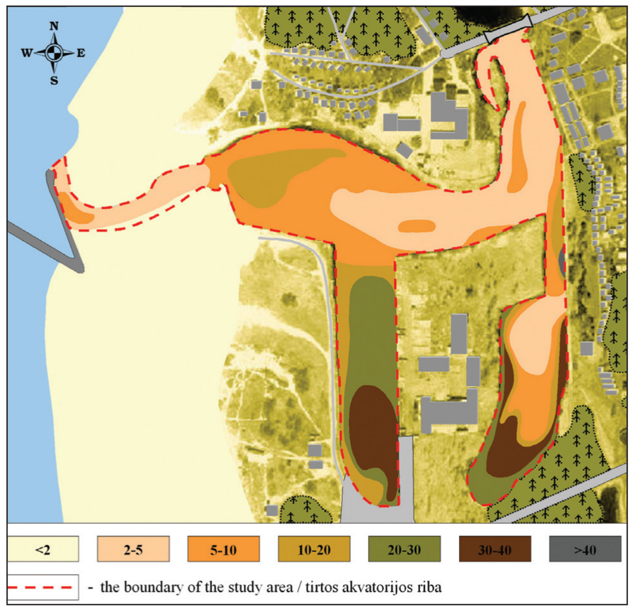

Fig. 11. Copper (Cu) concentration (mg/kg) in the bottom sediments (0–5 cm) of Šventoji Port

11 pav. Vario (Cu) koncentracija (mg/kg) Šventosios uosto paviršinėse (0–5 cm) dugno nuosėdose

The distribution pattern of the Cu/Al ratio shows that at present, the greatest loads are characteristic of the area at the embankment in the eastern part of the Eastern basin. The load is slightly smaller in the Šventoji branch to the Eastern basin and in the southern parts of the Eastern and Western basins. Areas distinguished by a higher load are situated in the central part of the Western basin and approaches to the mouth area (Fig. 12).

Mercury (Hg). Hg concentrations in sediments of the Šventoji fairway up to the mouth of the Eastern basin, a wide water area around the second forming shoals and at the old jetty in the approaches to the mouth, range from <20 to 50 μg/kg. In the mud of the Western basin, the concentration increases twice on the average. The highest concentration (up to 107 μg/kg) was found in the southernmost part of the Western basin. In the mud of the Eastern basin, Hg concentration is higher 1.5 times on the average, reaching its maximal value (201 μg/kg) in the western nearshore.

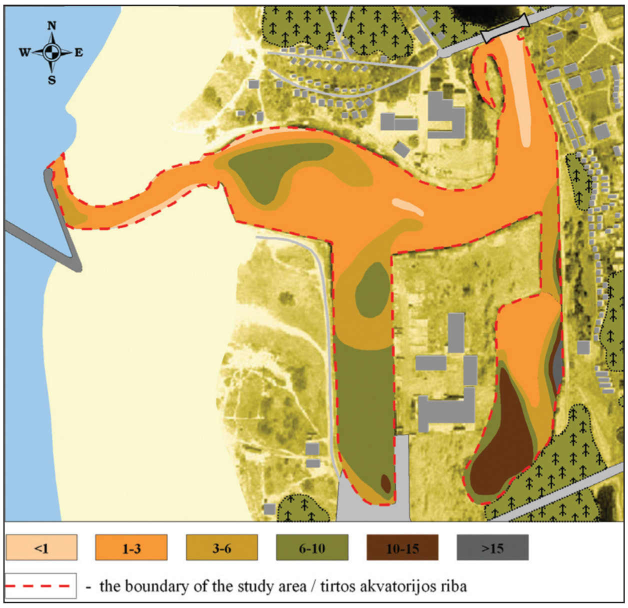

Fig. 12. Copper (Cu) concentration normalized relatively to aluminium in the surface bottom sediments (0–5 cm) of Šventoji Port

12 pav. Vario (Cu) ir aliuminio koncentracijos santykis Šventosios uosto paviršinėse (0–5 cm) dugno nuosėdose

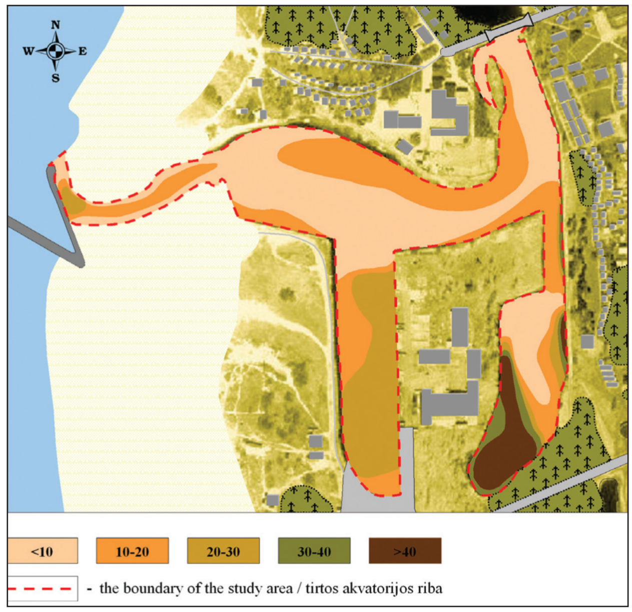

Fig. 14. Mercury (Hg) concentration relatively normalized to aluminium in the surface bottom sediments (0–5 cm) of Šventoji Port

14 pav. Gyvsidabrio (Hg) ir aliuminio koncentracijos santykis Šventosios uosto paviršinėse (0–5 cm) dugno nuosėdose

The concentration of Hg in the sand accumulated in the northern edge of the basin does not reach 20 μg/kg (Fig. 13).

The Hg/Al ratio shows that the highest loads of Hg occur in the mud areas of the Eastern basin (Fig. 14).

Lead (Pb). In the inequigranular sands and silt of the Šventoji River, Pb concentration does not reach 10 mg/kg, except local leaps of concentrations up to 20 mg/kg in the area extending along the Šventoji fairway up to the western bend of the river and in the area north of the West shoal (Fig. 15). The highest lead concentrations were found in the mud of the Eastern and Western basins: 75.0 mg/kg and 60.0 mg/kg, respectively.

The distribution pattern of the Pb/Al ratio shows that the areas of highest values (Pb/Al > 20) are located in the southern parts of the eastern and Western basins. Areas standing out for slightly higher Pb/Al values (Pb/Al: 3–5) can be distinguished in the Šventoji River sediments at the western nearshore north of the East shoal, north of the West shoal and at the old jetty in the approaches to the mouth (Fig. 16).

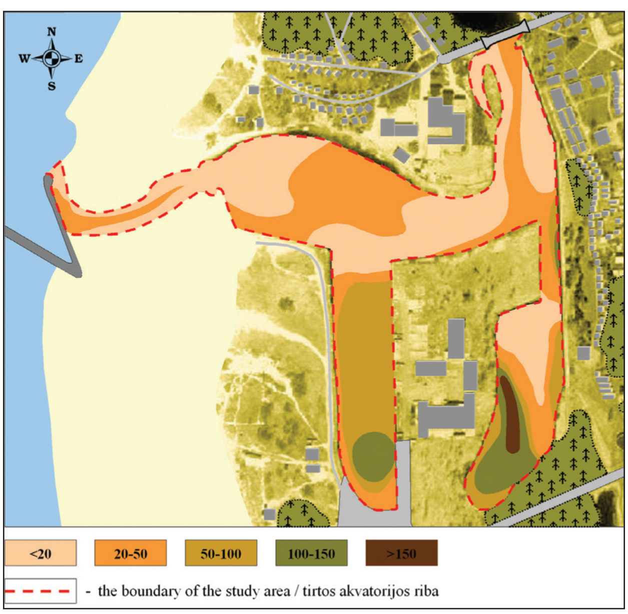

Fig. 13. Mercury (Hg) concentration (μg/kg) in the bottom sediments (0–5 cm) of Šventoji Port

13 pav. Gyvsidabrio (Hg) koncentracija (μg/kg) Šventosios uosto paviršinėse (0–5 cm) dugno nuosėdose

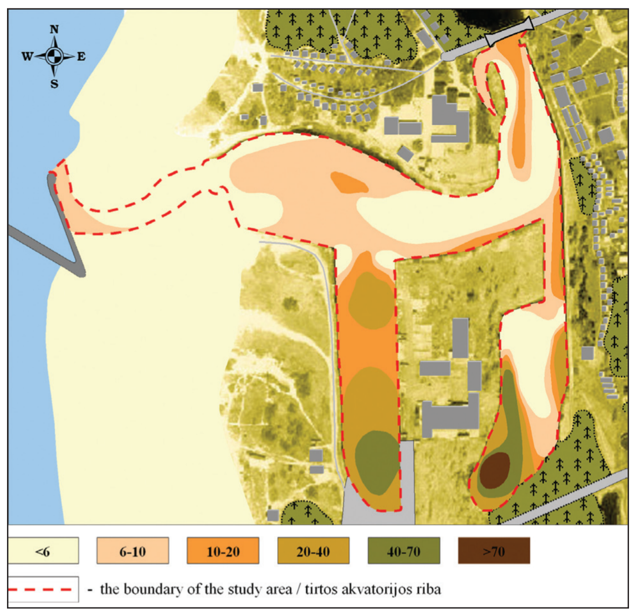

Fig. 15. Lead (Pb) concentration (mg/kg) in the bottom sediments (0–5 cm) of Šventoji Port

15 pav. Švino (Pb) koncentracija (mg/kg) Šventosios uosto paviršinėse (0–5 cm) dugno nuosėdose

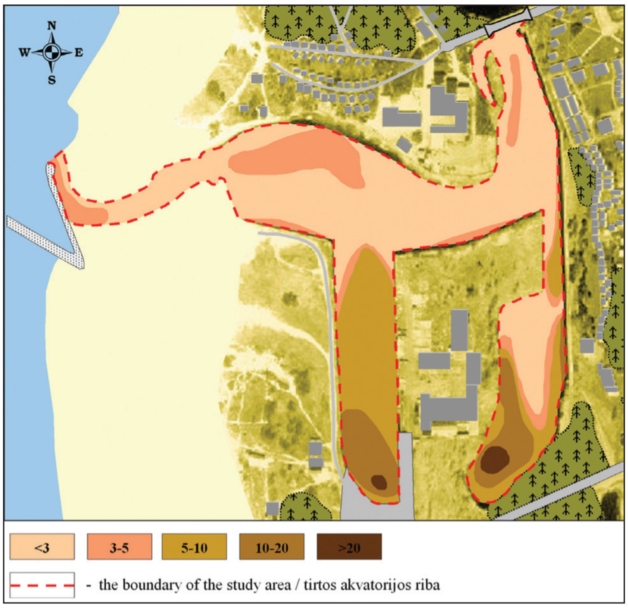

Fig. 16. Lead (Pb) concentration normalized relatively to aluminium in the surface bottom sediments (0–5 cm) of Šventoji Port

16 pav. Švino (Pb) ir aliuminio koncentracijos santykis Šventosios uosto paviršinėse (0–5 cm) dugno nuosėdose

Cadmium (Cd). The distribution pattern of Cd concentrations in the sediments of the Šventoji Port is rather monotonous (Fig. 17). Almost over the whole water area, Cd concentrations do not exceed 0.5 mg/kg. A local increase of Cd concentration up to 0.6 mg/kg occurs in the Šventoji River sediments at the western embankment north of the Eastern and the Western shoal. Only in the southern part of the Western basin and at the right embankment of the Eastern basin, Cd concentration in mud increases more than twice.

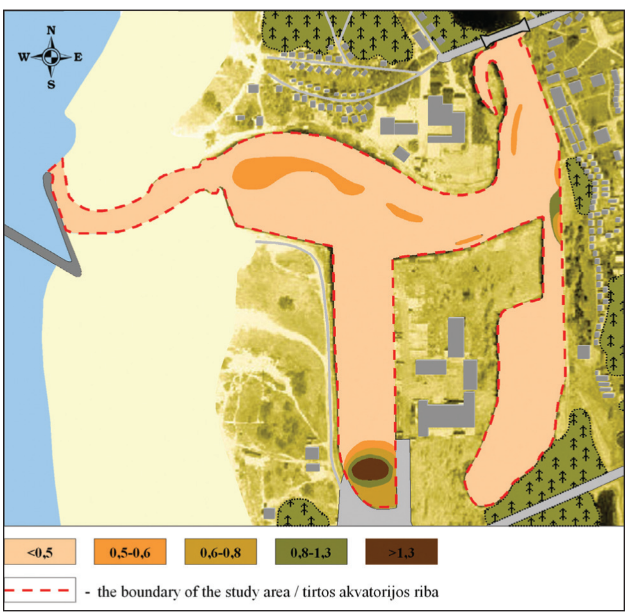

Fig. 17. Cadmium (Cd) concentration (mg/kg) in the bottom sediments (0–5 cm) of Šventoji Port

17 pav. Kadmio (Cd) koncentracija (mg/kg) Šventosios uosto paviršinėse (0–5 cm) dugno nuosėdose

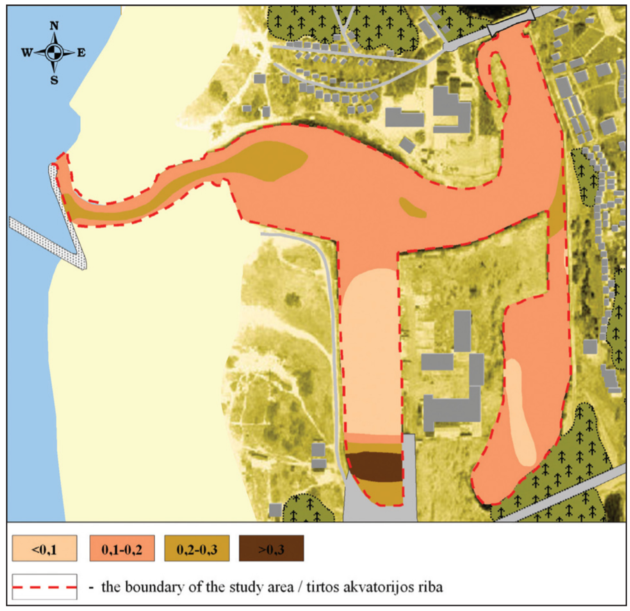

Fig. 18. Cadmium (Cd) concentration relatively normalized to aluminium in the surface bottom sediments (0–5 cm) of Šventoji Port

18 pav. Kadmio (Cd) ir aliuminio koncentracijos santykis Šventosios uosto paviršinėse (0–5 cm) dugno nuosėdose

The distribution pattern of Cd/Al values considerably differs from the one characteristic of other metals. According to it, the strongest Cd concentration anomalies (Cd/Al > 0.3) occur in the southern part of the Western basin and in the branch to the Eastern basin (Fig. 18). Somewhat smaller loads (Cd/Al: 0.2–0.3) are recorded in small areas in the northern corner of the Eastern basin, north-east of the Western alluvial shoal and in the water area between the approaches to the river mouth and the old jetty in the mouth area.

In spite of the general rather high variability of metal concentrations, there are certain zones of bottom surface sediments of the Šventoji Port waters tending to higher or lower concentrations. As the formation of material and the chemical composition of bottom sediments mainly depends on the sedimentation environment, we compared the average values of the study indices in three previously distinguished (Galkus et al., 2006) sedimentation zones of the Šventoji Port waters: the sedimentation zone of marine sand (A), the sedimentation zone of alluvium (B), and the sedimentation zone of thin-dispersed material (C) (Fig. 19). Analysis of the average concentrations of the metals showed that they were highest in zone C and lowest in zone A (Fig. 20). The most asymmetric distribution pattern was characteristic of Pb and the most even distribution pattern of Cd concentrations.

Analysis of the average normalized values of metals with aluminium showed the relative distribution trend of geochemical anomaly in sedimentation zones. The highest load of all metals, except cadmium, is characteristic of zone C. Though the lowest average metal concentrations are characteristic of zone A, the lowest geochemical anomaly of Ni, Cu and Hg was determined in zone B and of Cd in zone C. The highest load of cadmium is characteristic of zone A.

For identification of the areas of a higher summary geochemical anomaly of metals, data on Me/Al values were summarized distinguishing the areas in each sedimentation zone where

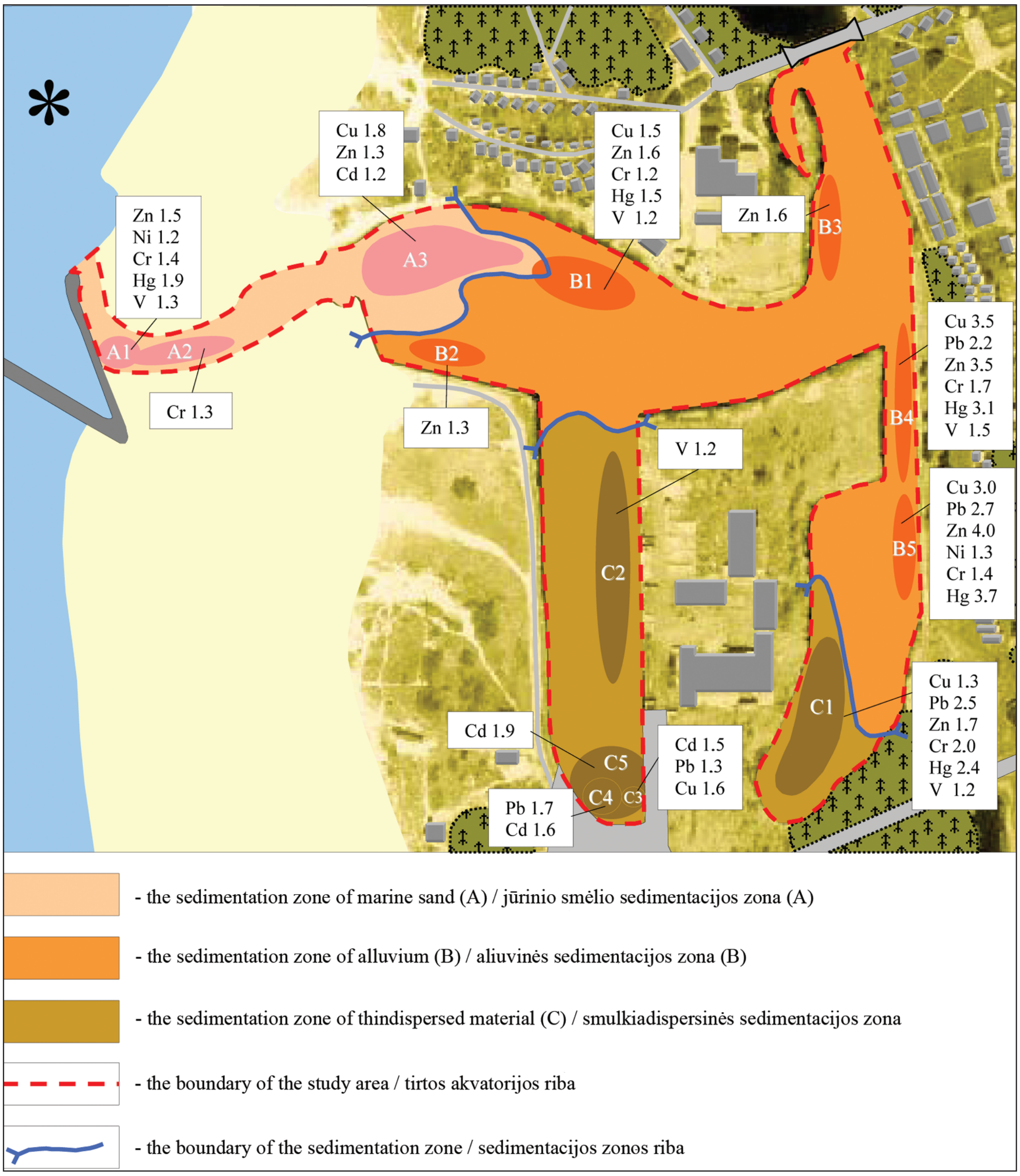

Fig. 19. Šventoji Port marine sand (*A*), alluvial (*B*) and thin-dispersed material (*C*) sedimentary zones and areas of geochemical anomaly. In footnotes: the highest load coefficient values (maximal of Me/Al ratio value in an area with the background value for the zone)

Fig. 19. Šventosios uosto jūrinio smėlio (*A*), aliuvinės (*B*) bei smulkiadispersinės medžiagos (*C*) sedimentacijos zonos ir geocheminių anomalijų arealai. Išnašose: didžiausios apkrovos koeficientų reikšmės (arealo maksimalios Me/Al reikšmės santykis su zonos fonine verte)

the ratio of metal and aluminium concentrations more than 1.1 times exceeded the background Me/Al values calculated for each zone (Table 4, Fig. 19). The background values of Me/Al for each zone (Table 4) were calculated according to the consistent rejection method of values not included within the X ± 2S interval (S – standard deviation, X – arithmetic mean) (Zinkutė, 2002). The coefficients of the highest geochemical anomaly for the areas were derived dividing their maximal value of Me/Al by the background value for the zone.

The studied water area includes 13 areas of a higher load (Fig. 19). Sedimentation zone A, where marine sand plays the most important role in sedimentation, includes three areas. In this sedimentation zone, the most toxic metals Hg and Cu exceed the background values of Me/Al more than 1.5 times. The Al area of sedimentation zone A in the mouth front bend of the Šventoji River at the old jetty, with a local trough in the river channel, endures a relatively highest load (Fig. 19).

The sedimentation zone B, where inequigranular alluvial material plays the most important role in formation of bottom sediments, five areas of a higher geochemical anomaly can be distinguished. The geochemical anomalies in areas B2 and B3 are constituted of only one metal – zinc: the Zn/Al values maximally exceed the background values 1.3 and 1.6 times, respectively. Zinc accumulates in the areas with muddy sandy silt

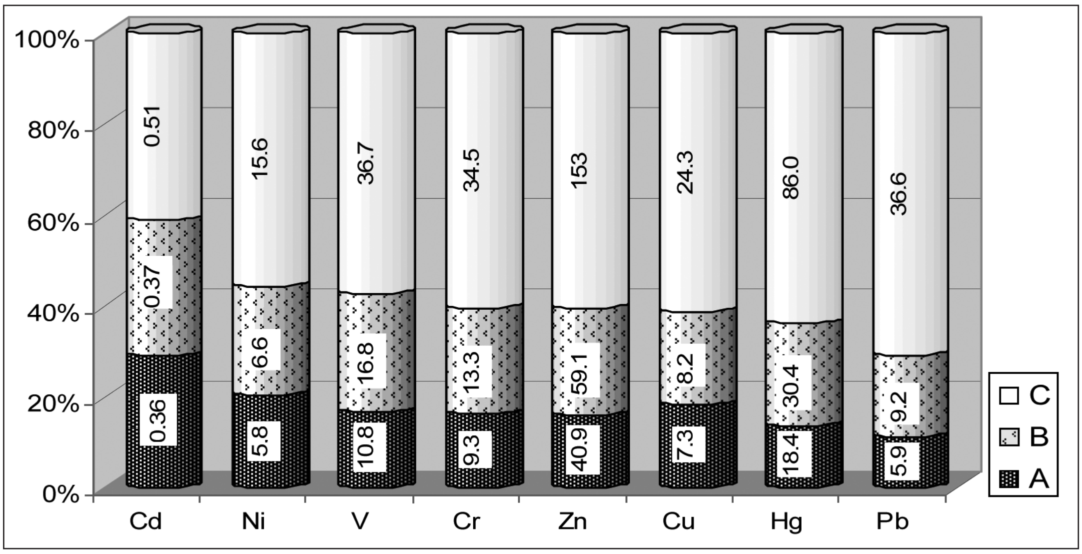

Fig. 20. Distribution of the average concentrations of metals (mg/kg, Hg – μg/kg) and their ratio with Al (%) in the Šventoji Port marine sand (*A*), alluvium (*B*) and thin-dispersed material (*C*) sedimentation zones

Fig. 20. Metalų vidurkinių koncentracijų (mg/kg, Hg – μg/kg) ir jų santykio su Al (%) reikšmių pasiskirstymas Šventosios uosto jūrinio smėlio (*A*), aliuvinės (*B*) bei smulkiadispersinės medžiagos (*C*) sedimentacijos zonose

Table 4. **Background values of Me / Al in different sedimentation zones of Šventoji Port**
4 lentelė. **Metalų ir aliuminio santykio (Me / Al) foninės reikšmės įvairiose Šventosios uosto sedimentacinėse zonose**

Zones	Cu/Al	Pb/Al	Zn/Al	Ni/Al	Cd/Al	Cr/Al	Hg/Al	V/Al
Sedimentation	2.82 ± 2.02	2.82 ± 2.02	22.4 ± 1.48	2.95 ± 1.48	0.20 ± 0.05	4.99 ± 2.19	10.2 ± 1.67	5.75 ± 1.48
zone A	n* = 8	n = 8	n = 8	n = 5	n = 8	n = 8	n = 8	n = 8
Sedimentation	2.45 ± 1.44	3.02 ± 1.51	18.6 ± 1.65	2.88 ± 1.34	0.15 ± 0.05	5.37 ± 2.04	10.0 ± 1.62	7.07 ± 1.26
zone B	n = 34	n = 41	n = 39	n = 33	n = 38	n = 37	n = 38	n = 37
Sedimentation	7.24 ± 1.38	10.2 ± 1.70	45.8 ± 1.38	5.13 ± 1.12	0.11 ± 0.05	10.7 ± 1.23	24.6 ± 1.52	10.2 ± 1.05
zone C	n = 18	n = 17	n = 18	n = 14	n = 13	n = 16	n = 18	n = 11

n* – the number of samples from which the background value was calculated.
n* – ėminių skaičius, pagal kuriuos apskaičiuota foninė reikšmė.

sediments. In area B1 situated in the shadow side (in respect to the water flow) of shoals, the spectrum of metals constituting the geochemical anomaly expands: Hg, Cr, Cu, and V anomalies can be observed there along with the zinc load. The geochemical anomaly is even higher in areas B4 and B5 predominated by silty-clayey mud sediments. They are marked by an extra load of lead and nickel, whereas the coefficients of the highest geochemical anomalies of Zn, Hg and Cu increase approximately two-fold and of V and Cr 1.2 times (Fig. 19).

The sedimentation zone C, where bottom sediment formation is predetermined by fine thindispersed material travelling in suspensions and plankton detritus, includes five areas of a higher geochemical anomaly. The coefficients of the highest geochemical anomaly of many metals (V, Cu, Zn, Cr, Hg, and Pb) in area C1 in the eastern part of the basin exceed 1.1. The area C2 of the geochemical anomaly of vanadium is distinguished in the Western basin. It should be noted that the small areas C3, C4 and C5 of sedimentation zone C are distinguished by Cd anomalies. The highest geochemical anomaly coefficients in them reach 1.5–1.9. Among all other investigated metals, only Pb and Cu occur in these areas in anomalous concentrations (Fig. 19).

Summarizing the data on metal anomalies, we can conclude that the Šventoji River is one of the main sources of metals getting into the Šventoji Port waters and forming areas of a higher geochemical anomaly. When the Šventoji water reaches the port waters, zinc is the first of the study metals to form areas of technogenic anomalies (Fig. 19). Its average concentration in all sedimentation zones is highest in comparison with other metals. The highest values of geochemical anomaly coefficients in all anomalous areas of the alluvial sedimentation zone are characteristic of zinc. Zn is distinguished for a good accumulation in plants and leaves. About 30–40% of zinc is transported by air and water (Kadūnas, 1998). The highest summary geochemical anomaly of metals (except Cd) falls on the area of the branch to the eastern basin in the sedimentation zone B and on the sedimentation zone of thin-dispersed material C. The river runoff and anthropogenic activity in the littoral part of the basin may be responsible for the input of various metals into the port waters where small depths and inhibited water circulation facilitate the accumulation of fine-grained bottom sediments saturated with metals. The geochemical anomaly in the Western basin, though accumulating fine-grained sediments yet with a better circulation, is lower than in the Eastern basin.

CONCLUSIONS

1. The dispersion and accumulation of metals in the bottom sediments of the Šventoji Port depend on specific features of the sedimentation environment. The concentrations of metals Cr, Pb, V, Ni, Cu, Zn, and Hg in bottom sediments are mainly associated with the content of fraction in the sediments: the grain size composition of sediments does not play a marked role in the dynamics of Cd concentrations.

2. In the bottom sediments of the Šventoji Port, the sources and migration paths of metals, interrelated by chemical properties and close correlation links, are comparable. More than 75% of Al concentration variations in bottom sediments is related with Ni, Cr, and V, with Cu and Zn on the average 56% and 61% and weakly (18–45% Al variation) is explained by Hg, Pb and Cd concentrations.

3. The highest average concentrations of all metals were determined in the sedimentation zone of thindispersed material (C). The lowest concentrations were determined in the sedimentation zone of marine sand (A). Normalization of the concentrations of the studied metals with Al showed that the highest average geochemical anomaly of all metals, except cadmium, is characteristic of sedimentation zone C, the lowest of Ni, Cu and Hg of the sedimentation zone of alluvial sedimentation, Cd – of zone C, Cr, Pb and Zn – of zone A. The highest average load of cadmium is characteristic of zone A.

4. Based on the normalization of the metals studied (Cr, Pb, Cd, Ni, Cu, Zn, Hg, and V) with Al concentration, thirteen areas of elevated geochemical anomaly were distinguished in the bottom sediments of the Šventoji Port. The values of Me/Al in these zones more than 1.1 times exceed the calculated background values.

5. The highest geochemical anomaly of all metals except Cd falls on the area in the branch to the Eastern basin in the alluvial sedimentation zone (B) and on the sedimentation zone of thindispersed material (C) of the Eastern basin. The highest load of technogenic Cd falls on the southern part of the Western basin in sedimentation zone C.

6. Among the studied metals (Cr, Pb, Cd, Ni, Cu, Zn, Hg, and V), the concentration of zinc is the highest. The concentration of Zn and its migration and accumulation properties predetermine that zinc is the first to form geochemical anomalies in the river–sea section. The values of the Zn geochemical anomaly coefficient in the alluvial sedimentation zone are highest.

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METALAI ŠVENTOSIOS UOSTO AKVATORIJOS (LIETUVA) DUGNO NUOSĖDOSE

Santrauka

Straipsnis parašytas remiantis kompleksinių lauko tyrimų rezultatais. 2004 m. birželį Šventosios uosto akvatorijoje buvo atlikti geodeziniai, batimetriniai ir vandens rodiklių (skaidrumas, spalva, drumzlių koncentracija) matavimai, taip pat vandens ir suspenduotų nešmenų debitų Šventosios upėje matavimai, paimti paviršinių dugno nuosėdų ėminiai. Šventosios uosto sedimentacinių procesų charakterį nulemiantys svarbiausi geografiniai veiksniai ir paviršinių dugno nuosėdų litologinės sudėties formavimosi specifinėse sedimentogenetinėse zonose ypatumai jau aprašyti (Galkus, Jokšas, Stakėnienė, 2006). Šiame straipsnyje analizuojami metalų koncentracijų pasiskirstymo paviršinėse (0–5 cm) Šventosios uosto akvatorijos dugno nuosėdose ypatumai, atskleidžiamos jų erdvinės kaitos priežastys. Didžiausias dėmesys skiriamas sunkiųjų metalų analizei, jų technogeninės sudėtinės dalies nustatymui bei didesnę technogeninę apkrovą patiriančių Šventosios uosto dugno nuosėdų arealų išskyrimui. Zn, Pb, Cr, V, Ni, Cd, Pb, Al, Hg koncentracija buvo nustatyta induktyviai sužadintos plazmos masių spektrometrijos metodu. Suardžius karbonatus druskos rūgštimi, organinė anglis išskirta aukštatemperatūrinio oksidavimo metodu. Gauti duomenys analizuoti ir apibendrinti GIS (Geografinės informacinės sistemos) metodais, atlikta duomenų statistinė analizė.

Nustatyta, kad metalų sklaida ir akumuliacija Šventosios uosto dugno nuosėdose priklauso nuo sedimentacinės aplinkos ypatumų. Cr, Pb, V, Ni, Cu, Zn ir Hg koncentracija dugno nuosėdose labiausiai susijusi su < 0,063 mm frakcijos kiekiu. Cd koncentracijos kaitai nuosėdų granuliometrinė sudėtis pastebimos įtakos neturi. Cheminėmis savybėmis bei artimais koreliacijos ryšiais tarpusavyje susijusių metalų šaltiniai bei migracijos keliai Šventosios uoste yra panašūs.

Didžiausios vidurkinės visų tirtų metalų koncentracijos yra Šventosios uosto smulkiadispersinės medžiagos sedimentacijos zonoje (C), mažiausios – jūrinio smėlio sedimentacijos zonoje (A). Normalizavus tirtųjų metalų koncentracijas su Al, nustatyta, kad didžiausia vidurkinė apkrova visais metalais, išskyrus kadmį, tenka C sedimentacijos zonai, o mažiausia Ni, Cu, Hg – aliuvinės sedimentacijos zonai (B), Cd – C zonai ir Cr, Pb, Zn – A zonai. Šventosios uosto akvatorijoje išskirta 13 padidintos technogeninės apkrovos arealų, kuriuose bet kurio metalo koncentracijos santykio su Al koncentracija reikšmės 1,1 karto ir daugiau viršija apskaičiuotas fonines vertes. Didžiausia technogeninė apkrova visais metalais, išskyrus Cd, tenka protakos į Rytinį baseiną rajonui aliuvinės sedimentacijos zonoje ir Rytinio baseino smulkiadispersinės medžiagos sedimentacijos zonai. Didžiausia apkrova technogeniniu Cd tenka smulkiadispersinės medžiagos sedimentacijos zonoje esančio Vakarinio baseino pietinei daliai.

Nustatyta, kad iš tirtųjų metalų cinko koncentracija Šventosios uosto dugno nuosėdose yra didžiausia. Zn kiekis, migracinės ir akumuliacinės savybės lemia, kad jis pirmasis pradeda formuoti geochemines anomalijas pjūvyje upė–jūra, šio metalo technogeninės apkrovos koeficiento reikšmės yra didžiausios visoje aliuvinės sedimentacijos zonoje.

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МЕТАЛЛЫ В ДОННЫХ ОСАДКАХ АКВАТОРИИ ПОРТА ШВЯНТОЙИ (ЛИТВА)

Резюме

В основе статьи лежат результаты комплексного полевого исследования. В июне 2004 г. в акватории порта Швянтойи (Литва) производились геодезические и батиметрические работы, исследования показателей воды (прозрачность, цветность, концентрация взвеси), отбор проб поверхностного слоя донных осадков, измерения расходов воды и взвешенных наносов в реке Швянтойи. Наиболее значительные географические факторы, определяющие характер процессов осадконакопления, а также особенности формирования литологического состава донных отложений в специфических зонах седиментации описаны ранее (Galkus, Jokšas, Stakėnienė, 2006). В настоящей статье исследуются особенности и выявляются причины распределения концентраций металлов в поверхностном слое (0–5 см) донных осадков. Наибольшее внимание уделено анализу тяжелых металлов, установлению их техногенной компоненты и выявлению подверженных повышенной нагрузке ареалов поверхностных донных осадков порта Швянтойи.

Определение металлов Zn, Pb, Cr, V, Ni, Cu, Cd, Al и Hg проведено методом масс-спектрометрии индукционно возбужденной плазмы. После деструкции карбонатов соляной кислотой органический уголь определен методом высокотемпературного окисления. Данные анализировались и обобщались методами ГИС (географических информационных систем), проведен статистический анализ.

Установлено, что распределение и аккумуляция металлов в донных осадках порта Швянтойи зависят от особенностей среды осадконакопления. Концентрация Zn, Pb, Cr, V, Ni, Cu и Hg в донных осадках наиболее тесно связана с количеством фракции <0,063 мм. На динамику концентрации Cd гранулометрический состав донных осадков ощутимого влияния не оказывает. В порту Швянтойи источники и пути миграции тесно взаимосвязанных корреляционными связями металлов весьма схожие. Наивысшие средние концентрации всех изученных металлов выявлены в зоне седиментации тонкодисперсного осадочного материала (С), наименьшие – в зоне седиментации морского песка (А). Установлено, что после нормализации исследованных металлов алюминием наибольшей техногенной нагрузке всеми металлами, кроме Cd, подвергается зона С, наименьшей – зона аллювиальной седиментации (В) (Ni, Cu, Hg), зона С (Cd) и зона А (Cr, Pb, Zn). В акватории порта Швянтойи выделено 13 ареалов с повышенной техногенной нагрузкой, в которых значения соотношения концентрации любого металла с концентрацией Al 1,1× и более превышают фоновое значение. Наибольшая техногенная нагрузка всеми металлами, кроме Cd, падает на район протоки в Восточный бассейн (зона аллювиальной седиментации) и на всю находящуюся в этом бассейне зону седиментации тонкодисперсного осадочного вещества.

Установлено, что из всех изученных в донных осадках порта Швянтойи металлов наивысших значений концентрации достигает цинк. Относительно большое количество этого металла, как и специфические его свойства в процессах миграции и аккумуляции, предопределяют, что на пути следования река–море геохимические аномалии, во-первых, формирует именно цинк. Во всей зоне аллювиальной седиментации значения коэффициента техногенной нагрузки для цинка по сравнению с остальными металлами остаются максимальными.