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Heavy metal contamination of the Curonian Lagoon bottom sediments (Lithuanian waters area)

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Abstract Samples of surface (0–3 cm) bottom sediments of the Lithuanian aquatic area of the Curonian Lagoon and Nemunas River delta were taken from 41 sites in 2013 and 2014. Sediment parameters, such as the percentage of particles, concentration of organic carbon and heavy metals (Cu, Zn, Ni, Pb, Cr, Cd, and Hg), were determined. The heavy metal contamination of the surface layer of bottom sediments was determined using the Nemerov's pollution index applied to soil. The spatial distribution of contamination indices and the dependence of contamination dynamics on sedimentation factors were analysed. It was determined that heavy metal concentrations had a tendency to increase as sediment particles became finer and as C_{org} concentration increased. A greater amount of pollutants got into the aquatic area of the Curonian Lagoon that was closer to the Klaipėda harbour than into other lagoon zones. The heavy metals for the integral pollution index for the Curonian Lagoon and Nemunas River delta surface bottom sediments could be arranged in the following order: Cd>Pb>Cu>Ni>Hg>Cr>Zn.

Keywords *• bottom sediments • contaminants • heavy metals • pollution • mud • sand • sedimentation*

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INTRODUCTION

The Lithuanian aquatic area of the Curonian Lagoon (35% of the total area of the lagoon) is a shallow flow-through freshwater body. The water from an area of more than 100 thousand sq. km (98% of this area belongs to the Nemunas River basin) flows into the Baltic Sea through this lagoon (Jokšas *et al.* 2003). Some water runs into the Curonian Lagoon from the Baltic Sea. The Nemunas River, with its main delta arms the Atmata and the Skirvytė as well as some smaller channels, are the main sources of water and sedimentary material to the lagoon (Emelyanov et al., 2015). Before water flows into the Baltic Sea, a large portion of heterogeneous sedimentary material mobilized from the land migrates with the surface waters of the Curonian Lagoon and gradually gets deposited on the bottom of the lagoon (Galkus, Jokšas 1997; Jokšas *et al.* 2003).

The Klaipėda Strait is the only connection between the Curonian Lagoon and the Baltic Sea. This narrow passage is also an aquatic area of the Klaipėda harbour. Sea harbours are always important sources of environmental pollution (Bird 1971; Denton *et al.* 2005; Mzimela *et al.* 2014). The rates of contamination in the Klaipėda Harbour water are considerably higher than in saline marine or fresh lagoon waters (Stakeniene *et al.* 2011). The prevailing northwest water flow sometimes changes direction and turns back toward the lagoon (Galkus 2007); therefore, a certain amount of contamination is likely to get transferred from the Klaipėda harbour to the lagoon. The lagoon ward movement of water and dissolved substances is becoming more frequent because of the permanent deepening of the aquatic harbour area and the gliding of the saline-fresh water zone barrier towards the lagoon (Jokšas, Galkus 2000). The annual water exchange via the Klaipėda strait is as follows: 6.1 km^3 inflow from the Baltic Sea to the Curonian Lagoon and 27.65 km^3 runoff from the Curonian Lagoon to the Baltic Sea (Jakimavičius, Kovalenkovienė 2010).

The multi-factorial average contamination of the aquatic environment a certain period is integrally reflected in the bottom sediments. Contamination dynamics are perfectly represented by the changes in the spectrum and concentration of heavy metals (Pekey *et al*. 2004; Nasr *et al.* 2006). The bottom sediments of the Curonian Lagoon have been investigated since 1931 (Pratje 1931). The distribution and composition of sediments have been the object of investigation by a number of researchers (Gudelis 1959; Pustelnikovas 1983; Gudelis, Pustelnikovas 1983; Gulbinskas 1995; Galkus, Jokšas 1997; Jokšas *et al.* 1998; Pustelnikovas 1998; Galkus, Jokšas 1999; Jokšas 1999; Petkus *et al.* 1999; Emelyanov 2002; Trimonis *et al.* 2003; Jokšas *et al.* 2003; Galkus 2004; Emelyanov *et al.* 2015, etc.). The authors of this article already investigated heavy metal contamination of bottom sediments in the Klaipėda strait and in contaminated aquatic areas of the harbour; these studies revealed a direct dependence of sediment contamination level on the distance from the source of pollution and on water circulation peculiarities (Galkus *et al*. 2012a; Galkus *et al*. 2012b). This article describes contamination of the surface (0–3 cm) layer of bottom sediments of the Curonian Lagoon and is a continuation of earlier investigations carried out in the Klaipėda harbour (Galkus *et al*. 2012b).

The aim of this study is to determine the scope of the contamination and the spectrum of contaminants in the bottom sediments of the Curonian Lagoon and to complete an analysis of the spatial change of indices to assess possibilities for heavy metals to get into the Curonian Lagoon both with river water and from the Klaipėda harbour. The article presents an analysis of heavy metal (Zn, Cr, Cd, Ni, Cu, Pb, and Hg) contamination of the surface (0–3 cm) layer of bottom sediments of the Lithuanian part of the Curonian Lagoon, Nemunas delta, and lower reaches of the Minija River and identifies bottom areas most receptive to contaminants.

MATERIAL AND METHODS

Sample collection and analyses

The investigation was carried out using 41 stations in July 2013 (Nemunas River delta and Minija River) and August 2014 (Lithuanian part of the Curonian Lagoon up to the southern boundary of the Klaipėda harbour). The location of sampling stations and depth measurement points were determined (with a 1 m accuracy) by the positioning equipment available aboard vessels. The samples were collected from the surface (0–3 cm) layer of the bottom sediments (Fig. 1). A Van Veen grab sampler was used for sampling. Sediment samples were placed in plastic bags. Later sediments were freeze dried and sieved. The percentage of particles of <0.063 mm in diameter was determined following the Lithuanian standards of classification (Lithuanian standards 2002) and traditional methods (Gaigalas 1995; Петелин 1967). Sand and mud (silt) (<0,063 mm) fractions were distinguished and sediment classification made by the following LAND 46A-2002 (Lithuanian standards 2002).

For metal (Zn, Cr, Cd, Ni, Cu, Pb) analysis total digestion method was used. The split freeze dried sediments (0.25 g) were heated using microwave in acid mixture (HNO_3-$HClO_4$-HF) to fuming and taken to dryness. The residue was dissolved in HCl (Loring, Rantala 1992). The content of trace elements in the bottom sediments WAS analysed by ICP-MS (Perkin Elmer NexION 300) method (Montaser 1998).

For Hg analysis sample splits of 0.5 g was leached in Agua Regia (Sunderland *et al*. 2004; Jagtap, Maher 2015). The content of Hg in the bottom sediments WAS analysed by HPLC-ICP-MS (Perkin Elmer NexION 300) method. For results control the NIST Standard reference material 2702 (Inorganics in Marine Sediments) was used.

Total carbon content in dried sediments was measured by high-temperature catalytic oxidation at 910 °C by a liquiTOC analyzer. Detection is based on the IR-detector where combustion gases (carbon dioxide) are purged by synthetic air as carrier gas. The mass is converted to percent carbon based on the dry sample weight. The total organic carbon content was subtracted from the total carbon content to determine the total inorganic carbon content of a given sample (Tiessen, Moir 1993). For the validation of TC determination Calcium Carbonate Purity Reference Material AOKE-00110 were used. One reagent blank and one commercial certified reference material has been added in every bathch (10 vessels).

Contamination assessment

As there is no and cannot be a single universal method for estimating heavy metal contamination, an appropriate method should be chosen to serve the purpose of the investigation (Mali *et al*. 2015). In our work, the values of the maximum concentration limit (MCL) were taken as a starting point for the estimation of sediment contamination. As MCLs used in Lithuania are different for bottom sediments of different composition (Lithuanian standards 2002),

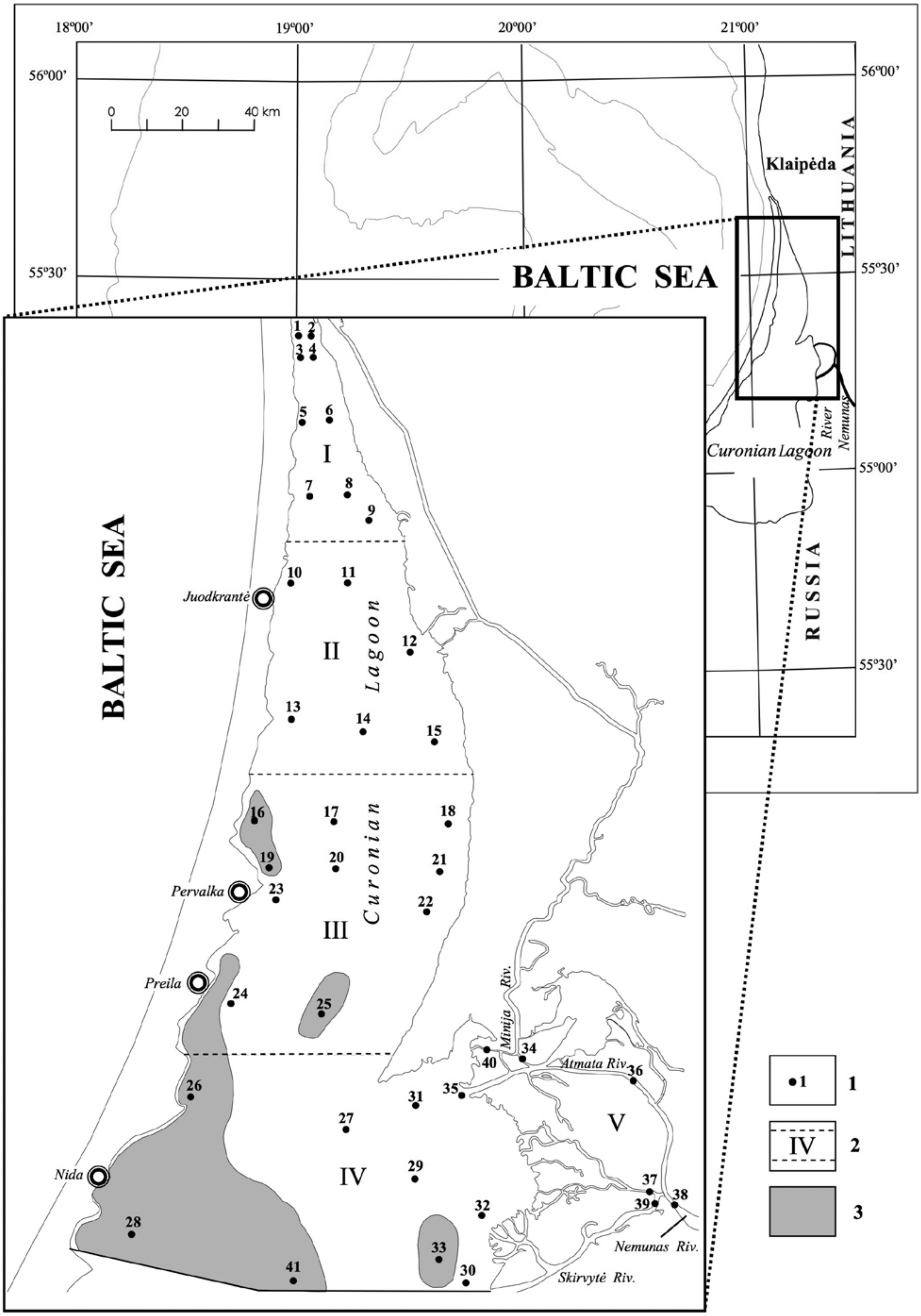

Fig. 1 Stations (1), zones of sedimentation (2) and mud accumulation areas (3) in the Lithuanian aquatic area of the Curonian Lagoon and Nemunas Delta zone

we applied the same classification as was used in the investigations of the Klaipėda strait (Galkus *et al.* 2012b), where uniform MCLs are used for all bottom sediments (Bakke *et al.* 2010). The Norwegian classification system is used to estimate the contamination of marine and lagoon sediments and focuses on toxicity to aquatic organisms as well as the impact to human health (Bakke *et al.* 2010; Galkus *et al.* 2012b). The values of this classification system for the upper limits of background levels (Class I) were called the MCLs in this study.

The pollution index used in this work was chosen according to the following criteria: all heavy metals are integrated into a single value (Galkus *et al.* 2012b). The contamination level was determined by calculating Nemerov's pollution index (PI) applied to soil (Gouzhang *et al.* 2006), which reflects the effect of each investigated heavy metal on bottom sediments and highlights the influence of contaminants (heavy metals) on the quality of the environment (Galkus *et al.* 2012b). The impact of a metal on the environment (single-factor index PI_i) was estimated by the ratio between its measured value (C_i) and MCL value (L_i): $PI_i = C_i/L_i$ (Galkus *et al.* 2012b).

The integral pollution index for bottom sediments of every site was derived using the following formula (Gouzhang *et al.* 2006):

$$PI = \sqrt{\frac{[AVG(PI_i)]^2 + [MAX(PI_i)]^2}{2}},$$

where PI stands for the synthetic contamination index of bottom sediments, $AVG(PI_i)$ represents the mean of various pollutant indices in bottom sediments, and $MAX(PI_i)$ is the maximum pollution index of a single-factor pollutant in the bottom sediments. The level of contamination was evaluated using a classification analogous to single-factor (PI_i) and multi-factor (PI) impacts: $PI(PI_i) \leq 0.7$ are clean sediments (heavy metal content in sediments stands under the warning limit); $0.7 < PI(PI_i) \leq 1$ are sediments in the warning condition (heavy metal concentration in sediments has already been in the warning condition, but it does not exceed the environment quality standard); $1 < PI(PI_i) \leq 2$ are lightly contaminated sediments; $2 < PI(PI_i) \leq 3$ are moderately contaminated sediments; and $PI(PI_i) > 3$ are heavily contaminated sediments (Gouzhang *et al.* 2006; Galkus *et al.* 2012b). The spatial structure of the data obtained in the bottom sediments in the study area of the Curonian Lagoon was analysed using GIS methods. To facilitate the estimation of contamination of bottom sediments deposited at different distances from potential sources of pollution, the Lithuanian part of the Curonian Lagoon was divided into five sedimentation zones (see Fig. 1). To measure correlation between concentrations of heavy metals (Zn, Cr, Cd, Ni, Cu, Pb, and Hg), the total organic carbon, and the content of fine-grained particles (<0.063) in bottom sediments, the Pearson's correlation coefficient (r) was calculated (Galkus *et al.* 2012b). Each data set was checked for normality using the Kolmogorov-Smirnov test and evaluated using histograms (Kruopis 1993). The linkage between two variables using significant correlation coefficients (significance level $p<0.01$) was estimated using the commonly accepted gradation: when $r \leq 0.5$, the link between variables is poor; when $0.5<r<0.7$, it is relatively fair; and when $r \geq 0.7$, it is strong (Chertko 1987; Galkus *et al.* 2012b).

RESULTS

Sediment types

The Lithuanian part of the Curronian Lagoon and the Nemunas River delta are characterised by bottom sediments with a prevalence of sand. (Trimonis *et al.* 2003; Žaromskis 2013). Most of the samples collected during this investigation were composed of fine-grained sand (0.25–0,1 mm >50%). Medium-grained sand (0.5–0.25 mm >50%) was detected in only certain parts of the Nemunas delta (stations 34, 38, and 39) (Fig. 1). Extra-fine-grained sand (0.1–0.063 mm >50%) samples were taken near mud accumulation areas (stations 5, 24, and 29); these samples contained admixture of mud (see Fig. 1). Other sand samples were composed of fine-grained sand.

Mud sediments are composed of sandy mud (2–0.63 mm 30–50%) and silty mud (0.63–0.002 mm 50–70%; < 0.063 mm >70%). Mud admixtures were often in the zones dominated by sandy sediments, particularly in the transition zones between sand and mud sediments. During our investigations, accumulations of mud were found in the southern-south-western parts of the Lithuanian aquatic area of the Curonian Lagoon (Fig. 1).

Distribution of heavy metals in the sediments

The greatest heavy metal concentrations were detected in the mud sediments (Table 1). The concentrations of most of heavy metals (Cu, Hg, Ni, and Pb) in the silty mud (<0.063 mm 53%, C_{org} 4.5%) were 2.4, 1.6, 1.5, 1.3, and 1.2 times higher than in the slightly coarser sandy mud (<0.063 mm 43.6%, C_{org} 4.3%), respectively. The concentration of Cd was essentially the same in both types of mud, and the Zn concentration was 1.2 times higher in sandy mud. The average concentrations of all the heavy metals (Cu, Hg, Ni, Cd, Zn, Cr, and Pb) in the mud sediments were 7.4, 6.7, 6.0, 5.3, 5.1, 4.1, and 3.7 times higher than in sand, respectively (Table 1).

In sand sediments, the average concentrations of all heavy metals consistently increased in the following order: medium-grained sand, fine-grained sand,

Table 1 Indices of the surface layer of the bottom sediments and the maximum concentration limit (MCL) for heavy metals in the Curonian Lagoon (Lithuanian area) and Nemunas Delta zone

Sampling site	*Sediment subtype. **Parameter	Fraction <0.063 mm,%	C_{org},%	Heavy metal concentration, ppm						
				Cu	Pb	Zn	Ni	Cr	Cd	Hg
Sand										
1	f-g	3.2	0.2	2.9	8.8	20.1	3	9.2	0.1	0.02
2	f-g	5.9	0.3	5.1	9.1	31.6	4.1	10.1	0.2	0.03
3	f-g	1.3	0.4	2.5	7.4	11.2	2.6	8.3	<0.10	0.02
4	f-g	0.8	0.2	3.1	6.9	25.3	4.5	9.1	<0.10	0.02
5	f-g	10.1	1	9.4	10.7	30.7	7	13.3	0.1	0.01
6	f-g	5.5	0.5	8.2	7.7	23.3	5.5	7.5	0.1	0.01
7	f-g	4	0.3	2.1	7.1	7.5	1.9	5.3	<0.10	0.01
8	f-g	8.5	0.4	1.5	6.4	13.8	2.8	6.1	0.1	0.01
9	f-g	9.4	0.6	9.9	10.5	29.6	7.1	11	0.2	0.02
10	f-g	7.7	0.4	1.8	6.8	8.8	1.6	7.1	0.1	0.02
11	f-g	2.3	0.1	1.5	5.6	7.3	1.4	4.3	<0.10	<0.01
12	f-g	8.6	0.8	12.3	11.8	33	7.2	14	0.2	0.01
13	f-g	8.9	0.7	2.8	6.9	9.9	1.9	4.5	<0.10	0.01
14	f-g	3.3	0.1	2	4.3	6.8	1.2	3.3	<0.10	<0.01
15	f-g	0.9	0.2	1.4	5.9	5.6	2	2.6	<0.10	0.01
17	f-g	6.1	0.2	3.1	5.1	8.4	2.1	5.2	<0.10	0.01
18	f-g	0.9	0.4	5.2	7.3	10.3	3.4	7.1	0.1	0.01
20	f-g	3.3	0.2	2.5	4.9	7.6	3.6	2.6	<0.10	0.01
21	f-g	9	0.3	5.6	4.5	6.4	5.6	4.2	0.1	0.01
22	f-g	6.2	0.3	2.8	3.8	11.1	6.7	7.1	<0.10	0.03
23	f-g	4.1	0.2	0.9	6.5	9.5	1	4	<0.10	<0.01
24	e-f-g	9.2	0.8	9.8	18.9	20.5	4.5	7.6	0.1	0.01
27	f-g	2.1	0.4	8.7	11.6	20.1	8.4	18	0.1	0.01
29	e-f-g	9.9	1.1	4.1	10	26.4	3.9	16	0.2	0.03
30	f-g	0.7	0.1	1.1	5.6	7.6	1.2	3.5	<0.10	<0.01
31	f-g	1	0.6	4.1	10.8	24.2	4.6	13	0.1	0.01
32	f-g	0.8	0.2	2.2	6.5	9.5	1.6	4.6	<0.10	0.01
34	m-g	0.6	0.8	3.6	8.2	20	3.7	8.8	0.1	0.01
35	f-g	5.7	0.1	1.2	5.7	7.5	0.9	2.3	<0.10	<0.01
36	f-g	3.3	0.5	1.3	6.1	9.1	1.1	2.2	<0.10	<0.01
37	f-g	7.5	0.1	2	6.6	12	1.3	4.5	0.1	<0.01
38	m-g	0.5	0.1	1	6.4	8.3	1	2	<0.10	<0.01
39	m-g	0.8	0.05	1.1	6	9.5	0.8	2.1	<0.10	<0.01
40	f-g	6.9	0.1	4.2	8.3	20.3	3.7	8	0.2	<0.01
Mud										
16	sandy	45.4	5	17.6	23.1	81.1	14.6	30.5	**0.8**	0.12
19	sandy	48.5	6.3	22.3	28.4	105	20.1	32	**0.9**	0.1
25	silty	50.4	2.9	18.2	14.9	45.5	8.4	22.4	**0.4**	0.05
26	silty	55.6	5.4	**45.6**	**35.8**	73	26.5	33.2	**0.7**	0.14
28	silty	53.1	5.3	**65.7**	**44.7**	92.3	**39.6**	40	**0.8**	**0.21**
33	sandy	49.7	3	12.6	17.8	60.1	9.8	25.5	**0.6**	0.08
41	sandy	30.8	2.8	20.1	**31.3**	77.5	19.9	19	**0.3**	0.03
Sand 34 stations	AVG	4.68	0.38	3.9	7.6	15.1	3.3	7.0	0.12	0.015
	STDV	3.33	0.28	3.07	2.89	8.59	2.16	4.18		
	MIN	0.5	0.05	0.9	3.8	5.6	0.8	2.0	<0.1	<0.01
	MAX	10.1	1.1	12.3	18.9	33	8.4	18.0	0.20	0.03
Mud 7 stations	AVG	47.64	4.39	28.9	28.0	76.4	19.8	28.7	0.64	0.10
	STDV	8.11	1.45	19.42	10.41	19.69	10.75	7.14	0.22	0.06
	MIN	30.8	2.8	12.6	14.9	45.5	8.4	19.0	0.30	0.03
	MAX	55.6	6.3	65.7	44.7	105	39.6	40.0	0.90	0.21
	MCL			35.00	30.00	150.00	30.00	70.00	0.25	0.15

*Sediment subtype: e-f-g - extra-fine-grained; f-g – fine grained; m-g – medium-grained
** Parameter: AVG – average value; STDV – standard deviation; MIN – minimum value; MAX – maximum value

and extra-fine-grained sand (Hg 0.007, 0.012, and 0.017 mg/kg; Cu 1.9, 3.6, and 7.8 mg/kg; Pb 6.9, 7.1, and 13.2 mg/kg; Zn 12.6, 14.2, and 25.9 mg/kg; Ni 1.8, 3.3, and 5.1 mg/kg; Cd 0.07, 0.09, and 0.13 mg/kg; Cr 4.3, 6.7, and 12.3 mg/kg, respectively). The average C_{org} concentration was approximately the same (0.3%) in medium-grained and fine-grained sand sediments, and it increased up to 1% in extra-fine-grained sand sediments.

The average heavy metal concentrations in each sedimentation zone (see Fig. 1) mostly depended on the presence or absence of mud sediments in each zone. Southern sedimentation zones III and IV had mud sediments and were clear leaders in average heavy metal concentrations in bottom sediments. The average integral pollution index generally increased in delta-front zone IV (up to 1.01 – lightly contaminated sediments), followed by zone III (0.82 – sediments in the warning condition) (Fig. 2). PI_{Zn} was the least significant single factor for sediment contamination (0.17). PI_{Cr} (0.18) was slightly more significant. Other PI_i values were as follows: PI_{Hg}=0.19, PI_{Ni}=0.20, PI_{Cu}=0.23, PI_{Pb}=0.37, and PI_{Cd}=0.77. Upon assessment of mud sediments only, PI values were found to increase significantly and ranged from 0.96 (sediments in the warning condition) to 2.66 (moderately contaminated sediments). Among single-factor index values, PI_{Cd} is the most important factor for mud sediments and exceeded 1 in all cases (1.2–3.6) (Fig. 3). PI_i >1 was detected in three cases: PI_{Pb} (1.04–1.5), PI_{Cu} (1.3–1.9) and PI_{Hg} (1.4).

The results of the investigation of mud sediments receptive to contaminants significantly affects the average values of indices of heavy metal contamination in bottom sediments of a particular area. For this reason, in order to evaluate the possible impact of waters coming from the Klaipėda harbour and the Nemunas River on the heavy metal contamination of the Curonian Lagoon, our comparative analysis of sedimentation zones located at different distances from potential sources of pollution was based exclusively on the results of analytical analysis of sand samples (Table 2).

The highest average concentrations of heavy metals were in zone I, the closest zone to the Klaipėda harbour. The exceptions were Pb and Cr, where the maximum concentrations were detected in the delta-front zone IV. The minimum average concentrations of most metals (Hg, Cu, Ni, Cd, and Cr) were found in the Nemunas delta sediments. The Pb concentration was lowest in zone II; Zn and Cd concentrations were lowest in zone III. Zone I was ranked first according to the distribution of single-factor average values and integral pollution index values (Table 2). The calculated integral pollution index values showed that all examined sand sediments could be assigned to the category of clean sediments according to contamination by all heavy metals (PI≤0.7). Relatively high contamination levels were detected in zone I, followed by zone IV (Fig. 1, Table 2). The maximum single PI values (≥0.6) were calculated for stations 2, 5, and 9 in northern zone I and in station 12 in zone II. An insignificantly lower PI value (0.59) was detected in station 40, i.e., in the Upaitė River branch connecting the Minija River with the Curonian Lagoon (seeFig 1 and 2). In the majority of sand sediments, PI values ranged from 0.21–0.34. Similar to mud, the highest PI_i values were found for PI_{Cd} (up to 0.8) in

Table 2 Average heavy metal concentrations (ppm) and the average values of single factors (PI_i) and integral pollution indices *PI* in the sand of different sedimentation zones in the Lithuanian area of the Curonian Lagoon

Content of element	Zone of sedimentation				
	I	II	III	IV	V
Hg	0.017	0.010	0.012	0.012	0.006
Cu	4.97	3.63	4.27	3.57	2.20
Pb	8.29	6.88	7.29	8.37	6.93
Zn	21.5	11.9	8.88	15.9	13.2
Ni	4.28	2.55	3.73	3.43	1.93
Cd	0.11	0.083	0.067	0.092	0.092
Cr	8.88	5.97	5.03	9.57	4.60
PI_{Hg}	0.11	0.07	0.08	0.08	0.04
PI_{Cu}	0.14	0.10	0.12	0.10	0.06
PI_{Pb}	0.28	0.23	0.24	0.28	0.23
PI_{Zn}	0.14	0.08	0.06	0.10	0.09
PI_{Ni}	0.14	0.09	0.12	0.11	0.06
PI_{Cd}	0.44	0.33	0.27	0.37	0.37
PI_{Cr}	0.13	0.09	0.07	0.14	0.06
PI	*0.39*	*0.30*	*0.29*	*0.32*	*0.29*

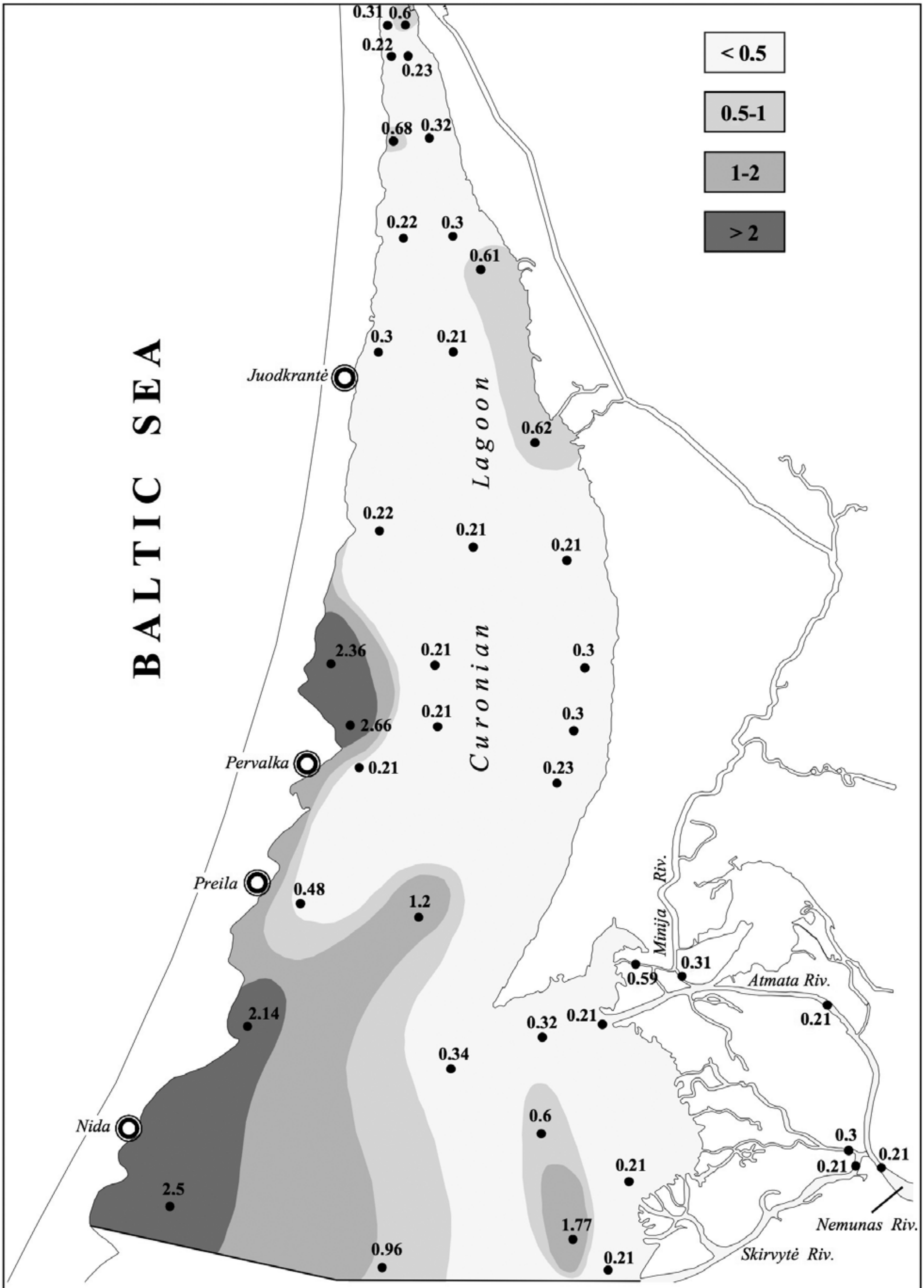

Fig. 2 The integral pollution index (PI) values for bottom sediments of the Lithuanian aquatic area of the Curonian Lagoon and Nemunas Delta zone

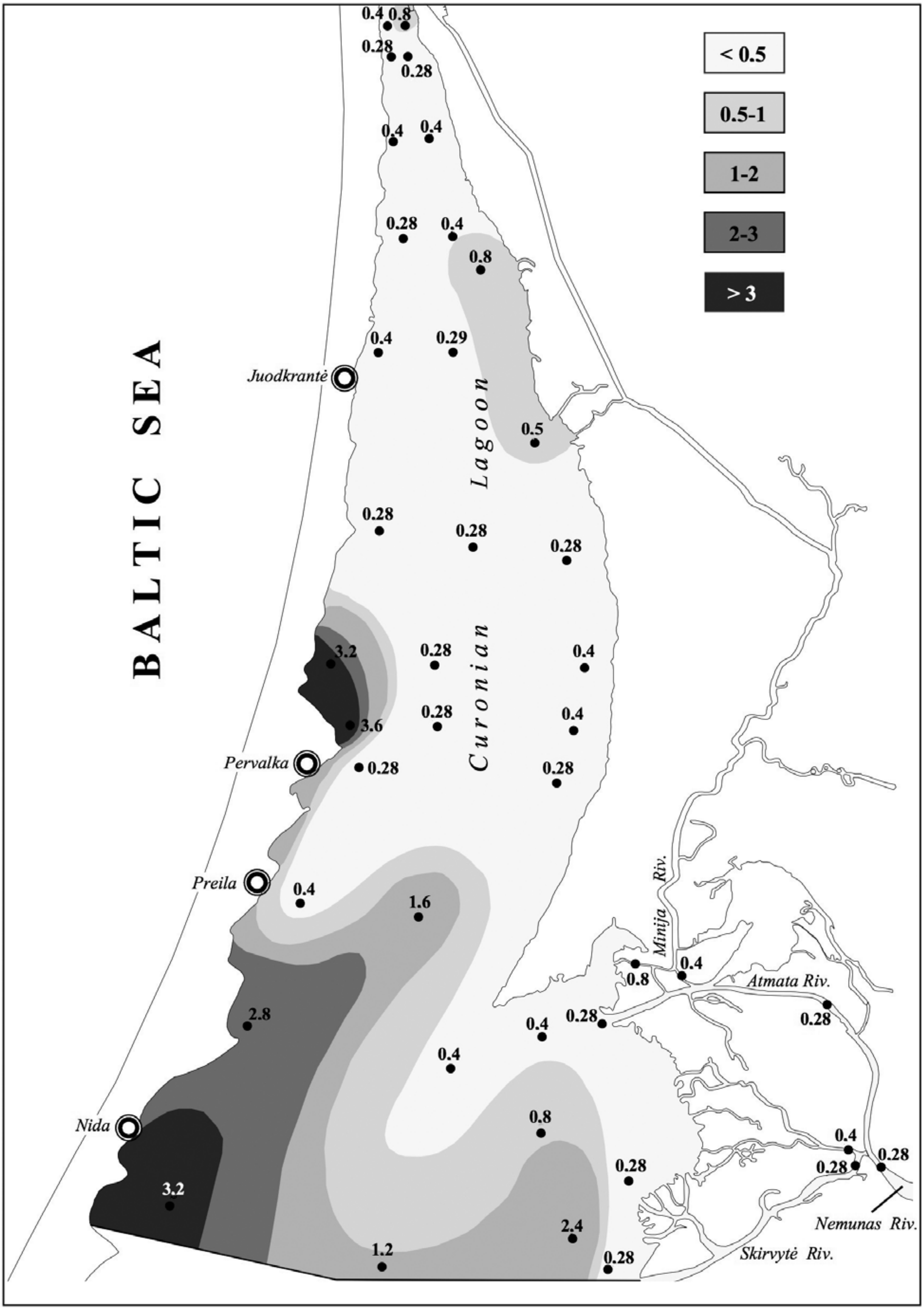

Fig. 3 The single factor PI_{Cd} values for bottom sediments of the Lithuanian aquatic area of the Curonian Lagoon and Nemunas Delta zone

stations 2; 9; 12; 29; and 40 (0.4 for all sand sediments, on average). The least significant single factor for general contamination of sand sediments was PI_{Hg} (0.08). The significance of single factors increased in the following order: PI_{Zn}=0.09, PI_{Cr}=0.098, PI_{Ni}=0.103, PI_{Cu}=0.104, PI_{Pb}=0.25, PI_{Cd}=0.36.

The values of correlation (r) between sand sediment indices showed the poor dependence of all heavy metals on the size fraction <0.063 mm, or the r value was below the significant level. Correlations between most of the investigated heavy metals (except for Hg and Cd) and the amount of C_{org} showed a relatively fair relationship. Inter-correlations between concentrations of heavy metals were highly diverse. Strong correlations were found between Cu and Pb, Zn and Ni, Zn with Ni and Cr, and Cr and Cd. Relatively fair correlations were found between Cu with Cd and Cr, Pb and Zn, Ni and Cr, and Cd and Zn. The lowest correlations with metal concentrations were detected for Hg; it was only with Cd that the r value exceeded the significant level (poor dependence) (Table 3).

The r values calculated for mud sediments were less representative than for the wider distributed sands. The correlation coefficient values for mud were highly variable. It was only Cr that maintained a relatively fair or strong correlation with all concentrations of heavy metals, the size-fraction <0.063 mm, and C_{org} concentration.

The r values calculated for both sand and mud sediments jointly demonstrated the close interrelationship between the indicated indices. Concentrations of heavy metals were found to be strongly related both with the size-fraction <0.063 mm and the C_{org} concentration. Furthermore, the interrelationship between concentrations of separate metals was also strong (Table 4).

DISCUSSION

Sediment formation

The composition of bottom sediments of the Curonian Lagoon depends on the sedimentation conditions, water and particulate material circulation patterns being the most important ones (Galkus 2004). The sediment arriving from rivers and washed from the banks differentially accumulates on the bottom of the study area. Sand particulates abundantly accumulate in the Nemunas delta branches and the delta front, whereas finer sedimentary matter spreads westward and south-westward to the Curonian Lagoon (Galkus,

Table 3 Pearson's correlation coefficients (r) calculated for heavy metal (Hg, Cu, Pb, Zn, Ni, Cd, Cr) concentrations, organic carbon (C_{org}), and the amount of sediments in the size-fraction <0.063 mm (FR) in the sandy bottom sediments of the Lithuanian part of the Curonian Lagoon and Nemunas Delta zone

	FR	C_{org}	Hg	Cu	Pb	Zn	Ni	Cd	Cr
FR	1.00								
C_{org}	0.50	1.00							
Hg			1.00						
Cu	0.48	0.62		1.00					
Pb		0.64		0.73	1.00				
Zn		0.62		0.75	0.68	1.00			
Ni		0.53		0.85	0.50	0.71	1.00		
Cd	0.47		0.44	0.51		0.69		1.00	
Cr		0.65		0.69	0.65	0.82	0.77	0.54	1.00

Significance level p<0.01, significant r>0.436

Table 4 Pearson's correlation coefficients (r) calculated for heavy metal (Hg, Cu, Pb, Zn, Ni, Cd, Cr) concentrations, organic carbon (C_{org}), and the amount of the size-fraction <0.063 mm (FR) in the bottom sediments of the Lithuanian part of the Curonian Lagoon and Nemunas Delta zone

FR	C_{org}	Hg	Cu	Pb	Zn	Ni	Cd	Cr
1,00								
0,94	1.00							
0,87	0.90	1.00						
0,81	0.83	0.91	1.00					
0,85	0.90	0.87	0.93	1.00				
0,89	0.95	0.84	0.81	0.91	1.00			
0,82	0.87	0.91	0.97	0.95	0.88	1.00		
0,93	0.97	0.92	0.80	0.85	0.94	0.84	1.00	
0,90	0.93	0.89	0.86	0.90	0.94	0.90	0.92	1.00

Significance level p<0.01, significant r>0.398

Jokšas 1997). The Curonian Lagoon produces over 600 thousand tonnes of plankton per year, one-third of which is carried to the Baltic Sea. The remaining material, a little decomposed, gets deposited onto the bottom of the lagoon and participates in the formation of mud sediments and increases C_{org} concentrations (Jokšas *et al.* 1998). An area of intensive sedimentation of fine-grained matter forming mud sediments was detected by the Nemunas delta. In the western part of the delta-front aquatic area of the lagoon, mud sediments accumulated over a wide bottom area. Areas with mud sediments were also found further north (see Fig. 1). Fine-grained sediment matter together with northward moving water reach the Lithuanian part of the lagoon from the southern region of the lagoon after having suffered marked compositional changes (Galkus, Jokšas 1997). With the Curonian Lagoon narrowing towards the Klaipėda strait, water stream velocity increases, and the effects of waves reach the bottom in shallow waters (Dailidienė *et al.* 2004; Gailiušis *et al*. 2005; Galkus 2007). Due to intensive hydrodynamic factors, mud sediments do not form in the northern part of the Lithuanian lagoon. Small areas of mud sediments can only be detected in some inlets of the Curonian Spit (Jokšas *et al.*1998).

Sediment properties

The high absorption potential of the mud sediments was attributed to the greater quantity of the fine particles (fraction <0.063 mm 10.2 times greater) and higher C_{org} concentration (11.6 times) compared to sand (Table 1). Similar to the Klaipėda harbour (Galkus *et al.* 2012b), the concentrations of heavy metals in the lagoon had a tendency to increase as the sediments became siltier and the sediment particles finer. A strong correlation between decreasing particle size and increasing heavy metal concentration is well documented by Lithuanian (Jokšas 1999; Galkus *et al.* 2012b) and foreign scientists (Matthai, Birch 2001; Guerra-Garcia *et al.* 2005). Organic matter also plays an important role in metal binding in sediments (Stephens *et al.* 2001; Galkus *et al.* 2012b; Mzimela *et al*. 2014).

When investigating only sand sediments, the correlation was not as clear due to the decreasing relative fluctuation amplitudes of the size-fraction <0.063 mm and C_{org} concentration (Table 3). When investigating the different composition of sediments jointly, the correlation between heavy metal concentrations and the size-fraction <0.063 mm as well as C_{org} concentrations became obvious; interdependence between metal concentrations were all strong (Table 4). The close correlations between most metal concentrations (in most cases $r \geq 0.9$) suggest similar migration and deposition rates for these contaminants. As a mixture of fine mineral and organic particles spread, they become a very receptive medium for the absorption of heavy metals from water (Galkus, Jokšas 1997; Zoumis *et al.* 2001; Stephens *et al.* 2001; Fernandes *et al.* 2008). Heavy metals can get into the finely dispersing sediment matter, even far from the place of settlement, i.e., when such matter is still migrating with river waters and in the southern part of the lagoon.

Sediment contamination by heavy metals

Compared with earlier investigations (Table 5), the average concentrations of heavy metals detected

Table 5 Average content of heavy metals (ppm) in the Lithuanian area of the Curonian Lagoon measured in the current and earlier investigations and in the Klaipėda strait according to earlier investigations based on the same research methods

Sediment	Layer, cm	Elements							Period of sampling	Source
		Cu	Pb	Zn	Ni	Cr	Cd	Hg		
Curonian Lagoon										
Sand	0-10	2	10	-	8.5	20.5	-	-	1974–1996	Pustelnikovas, 1998
Mud	0-10	9	51.5	-	24	55.5	-	-		
Sand	0-3	4.2	8	10.9	-	5.9	0.3	-	1996	Jokšas, 1999
Mud	0-3	16.8	20.8	65	-	36.2	0.8	-		
Sand[1]	0-3	-	2	41	11	19	0.1	-	2001–2003	Emelyanov et al. 2015
Sand[2]	0-3	11	7.5	21.4	13.6	17.4	0.17	-		
Mud[3]	0-3	-	6	102	21	60	0.1	-		
Sand	0-3	3.9	7.6	15.1	3.3	7	0.12	0.015	2013–2014	Current article
Mud	0-3	28.9	28	76.4	19.8	28.7	0.64	0.1		
Klaipėda strait (Klaipėda harbour)										
Sand	0-10	5.5	10.9	28.6	5.3	14.0	<0.4	0.02	2008–2009	Galkus et al. 2012b
Mud	0-10	21.9	22	109	12.1	31.5	0.84	0.07		
Klaipėda harbour semi-closed bays										
Sand	0-10	19.9	18.1	83.8	7.15	15.8	0,4	0.07	2008–2009	Galkus et.al. 2012a
Mud	0-10	63.7	42.7	239	22.8	33.3	1.03	0.15		

1 – Atmata mouth, 2 – Skirvytė mouth, 3 – area by Nida harbour

during the current investigation did not show any significant changes in the heavy metal contamination of the bottom sediments. Notable exceptions are Ni, the average concentration of which did not reach those detected earlier in sand and mud, and Cu, the average concentration of which exceeded that detected earlier in mud (Table 5).

Compared with the authors' investigation of contamination of bottom sediments in the Klaipėda strait (Klaipėda harbour aquatic area) using analogous methods of investigation (Galkus *et al.* 2012b), a decrease in heavy metal concentrations in the Curonian Lagoon was more noticeable in sand sediments. The average concentrations of Cu, Pb, Ni, and Hg in the mud of the lagoon were even higher that in the mud sediments of the Klaipėda strait although differences were not very clear. Higher concentrations of some heavy metals in the lagoon could be due to the fact that mud has been accumulating and degrading in the lagoon for a long time, whereas the harbour is undergoing permanent cleaning and deepening procedures (Galkus *et al.* 2012b). Concentration differences in the different bottom sediments can be seen by comparing the lagoon with the semi-closed bays of the Klaipėda harbour, where high contamination of the surface layer of bottom sediments with heavy metals is mainly predetermined by a short distance from the pollution sources and slow renewal of water (Galkus *et al.* 2012a; Kriaučiūnienė, Gailiušis 2016).

The integral pollution index PI reflects the impact of all heavy metals jointly and each separately on the quality of the aquatic environment (Guozhang *et al.* 2006). When evaluating contamination of the surface layer of bottom sediments of the Curonian Lagoon according to this index, an obvious effect of higher contamination of mud sediments of the lagoon on the distribution of general sediment contamination indices can be seen (see Figs 1 and 2). The PI values calculated for mud accumulation areas (0.96–2.66) were rather high and close to the values calculated for similar sediments in the Klaipėda harbour (except for more contaminated semi-closed bays). Mud sediments were lightly and moderately contaminated both in the harbour and the lagoon. Only in rare cases were the PI values in the Klaipėda harbour still higher and exceeding 3 (heavily contaminated sediments) (Galkus *et al.* 2012b). Compared with the southern zone of the Klaipėda harbour (PI=0.87, sediments in the warning condition), the PI values calculated for sand sediments of the northern sedimentation zone I of the lagoon decreased twofold (PI=0.39 on average) (Table 2) (Galkus *et al.* 2012b). There were no clean sediments in the Klaipėda harbour aquatic area (Galkus *et al.* 2012b); however, just behind the southern limit of the harbour, the sand sediments of the lagoon already met the requirements for the category of clean sediments. Nevertheless, the average PI value in sedimentation zone I was the highest compared with sand sediments of zones further southward (Table 2), though the shallow and hydrodynamically active zone did not provide favourable conditions for deposition of contaminants in bottom sediments. This circumstance demonstrates that more contaminants get into zone I than other zones. The permeability of the Klaipėda strait increased by 10% compared to 1996 (Kriaučiūnienė, Gailiušis 2016), and the water circulation between the Klaipėda harbour aquatic area and the Curonian Lagoon became more active. Contaminants can possibly get into the lagoon not only from a more contaminated Klaipėda harbour aquatic area (Galkus *et al.* 2012b) but also from the inlets of eastern coasts of the lagoon, near the mouths of which PI values significantly increase (Fig. 2). Increases in heavy metal concentrations in the surface layer of bottom sediments of this coastal region were recorded earlier as well (Galkus 2004). In other sedimentation zones, the average PI values fluctuated insignificantly. A slight increase in PI value in the delta-front zone IV that is close to a very clean Nemunas delta zone V should be related to the closeness of the contaminated mud areas rather than with the Nemunas River runoff. According to the assessment criteria used in this article, sand sediments of the Curonian Lagoon and Nemunas delta did not overstep the limits of the category of clean sediments.

As in the Klaipėda harbour (Galkus *et al.* 2012b), PI_{Cd} was the main single factor for the integral pollution index in the Curonian Lagoon. The regularities in distribution of PI_{Cd} values Curonian Lagoon were close to the distribution of total PI values (Fig. 3). Cd concentrations increase in different media in the Curonian Lagoon, Klaipėda harbour and Baltic Sea, which was also recorded by other researchers (Garnaga 2011). The Cd concentration increases in the mud sediments accumulating in the Curonian Lagoon were detected in earlier works of the authors of this article (Galkus 2004). Increases in Cd contamination of the bottom sediments were also recorded in other aquatic areas, sometimes far from Lithuania: in the Szczecin Lagoon and Gdansk Basin, Poland (Glasby *et al.* 2004), in the Wadi Al-Arab Dam, Jordan (Ghrefat, Yusuf 2006), in the Pulicat Lake, India (Seralathan Kamala-Kannan *et al.* 2008), etc. Increases in concentrations of heavy metals, including Cd, are not universally characteristic of bottom sediments of all harbours and adjacent aquatic areas. Some authors have even detected a decreasing tendency (Mason *et al.* 2004; Alve *et al.* 2009).

According to the single-factor index values, the heavy metals for the integral PI index for the Curonian Lagoon can be arranged in the following order Cd>Pb>Cu>Ni>Hg>Cr>Zn. For a relatively clean zone of the Klaipėda harbour, this order was Cd>Pb>Cr>Ni>Zn>Hg>Cu; for a more contaminated zone, it was Cd>Pb>Zn>Cu>Cr>Hg>Ni (Galkus *et al.* 2012b). In all cases, PI_{Cd} and PI_{Pb} were the main single factors. Compared with the Klaipėda harbour, the values of other single factors were distributed unevenly. The PI_{Cu} value for the integral PI index for the Curonian Lagoon was even higher than for contaminated sediments of the Klaipėda harbour. PI_{Zn} and PI_{Cr} were more significant in the harbour, while PI_{Hg} impact on the total PI was slightly higher in the Curonian Lagoon.

CONCLUSIONS

In the surface layer (0–3 cm) of bottom sediments of the Curonian Lagoon, the concentrations of heavy metals had a tendency to increase as sediment particles became finer, as sediments became siltier and with increasing C_{org} concentrations. Compared with earlier investigations, the heavy metal concentrations described in the current work did not demonstrate significant changes in the heavy metal contamination of bottom sediments during the last 40 years. An insignificant decrease in Ni concentration and increase in Cu concentration were detected in mud sediments. Compared with the Klaipėda strait (harbour's aquatic area without semi-closed bays), the heavy metal contamination of the Curonian Lagoon sand sediments was several times lower. The PI values showed that sand sediments of the Lithuanian part of the Curonian Lagoon and Nemunas River delta fell into the category of clean sediments.

The average concentrations of Cu, Pb, Ni, and Hg in the mud sediments of the Curonian Lagoon insignificantly exceeded those in the Klaipėda strait, whereas Zn, Cr and Cd concentrations were lower in the lagoon. The mud sediments of the lagoon were lightly and moderately contaminated with heavy metals.

An analysis of the spatial dynamics of sand sediment contamination indices showed that more contaminants got into the area of the Curonian Lagoon that was closer to the Klaipėda harbour than into the other zones of the lagoon. Contaminants can possibly get into the lagoon from the more contaminated aquatic area of the Klaipėda harbour and from the inlets of the eastern coasts of the lagoon. No traits of heavy metal movement from the Nemunas delta area were detected in the sand sediments.

PI_{Cd} was the main single-factor index for the integral pollution index of the bottom sediments of the Lithuanian part of the Curonian Lagoon and Nemunas delta. The regularities of the distribution of PI_{Cd} values in the investigated area of the lagoon were close to the distribution of total PI values. According to the single-factor index values, the heavy metals for the integral PI index for the Curonian Lagoon can be arranged in the following order Cd>Pb>Cu>Ni>Hg>Cr>Zn.

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