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Biogenic components and trace elements in the sediments of river mouths and accumulation areas of the Curonian Lagoon (south-eastern Baltic Sea)

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Abstract Contribution of the Neman (Nemunas) River including the arms Atmata, Skirvytė and Razliv, as well as Matrosovka and Deima rivers to the general enrichment of the Curonian Lagoon with biogenic components and chemical elements is reflected in the composition of surface sediments accumulating in the river mouths. Other part of chemical substances are absorbed by the finest particles and reaches accumulation areas of the lagoon in the south-western and central parts. Results from the study have shown the prevalence of terrigenous and biogenic-terrigenous sediments in the mouths of small rivers. Highest average values of biogenic components (31.46–38.87% of $CaCO_3$; 0.63–1.03% of N; 0.07–0.1% of P) are observed in the accumulation areas of the coarse silt and fine silty mud. Same areas are characterized by the increased amounts of potentially hazardous metals (Cd, Cr, Cu, Ni, Pb, Zn). Results from this study have shown considerable enrichment of sediments with arsenic. In most places average contents of this metalloid are well above the geochemical background values, determined for the south-eastern part of the Baltic Sea. Absolutely highest values of arsenic (91–93 mg/kg) suggesting presence of serious contamination sources were determined in the south-eastern part of the lagoon, adjoining the mouth of small Matrosovka River.

Keywords • *biogenic components • trace elements • sediment pollution • coastal areas • river mouths • accumulation areas*

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INTRODUCTION

Lagoon is an invaluable component of the nature and ecosystem of the shores and gives comfortable possibilities for the surrounding inhabitants to develop fishery and tourism. They are the most productive of all the coastal components (Gonenc, Wolflin 2005). The Curonian Lagoon is one of the largest estuarine coastal freshwater lagoon in the Baltic Sea region. The total area of the lagoon is approximately 1,600 km^2. Total volume of water of the lagoon is approximately 6.2 km^3, and the average depth about 3.8 m (Pustelnikovas 1998). The lagoon lies along the Baltic Sea coast of Lithuania and the Kaliningrad District of Russia. Its greater part belongs to Russia (1171 km^2), whereas 413 km^2 is in the territory of Lithuania (Dubra 1978).

It is an open system, influenced by a discharge of the fresh Neman River (Nemunas) and other smaller channels and saline water of the Baltic Sea. Water salinity in the northern part of the lagoon may fluctuate between 0.1 and 7 psu. The Neman River with its arms in Atmata and Skirvytė as well as several smaller channels, located in the delta of the Curonian Lagoon are the main sources of sedimentary material. Atmata and Skirvytė together with smaller streams (Deima, Matrosovka and Razliv) endure in the lagoon 781,000 t of terrigenous material (Blazhchishin 1984). Sand (1.0–0.1 mm fractions) is deposited mainly in the coastal part of the delta, as well as in the northern, shallower part of the lagoon. Silty (0.1–0.01 mm fractions) material through the Klaipeda Strait is carried into the Baltic Sea, where it forms the extensive field of silts (Emelyanov *et al.* 2002). Another part of thin-dispersed sedimentary material reaches places of intensive accumulation, located in the south-western part close to the Zelenogradsk settlement and in the central part near the Nida city.

The Curonian Lagoon is heavily contaminated from agricultural and industrial sources. Concentrations of petrochemicals and heavy metals in the lagoon waters and sediments are very high (Pustelnikovas 1998). Sediment contamination spectrum is determined by the position of sedimentation region with respect to the dominant water streams and distance from the river mouths and other sources of pollution, including port areas and shipping activities (Galkus 2004).

The surface sediments of the Curonian Lagoon are investigated since 1931 (Pratje 1931; Gudelis 1959). Regularities of sediment composition and distribution, including their accumulation processes at the bottom, were described and sediment distribution maps compiled (Pustelnikovas 1983, 1998; Gulbinskas 1994). A very small depth, an active water dynamics and intensive sedimentation in the lagoon considerably affects the sediment distribution patterns and their changes over the time. Therefore, previously published material does not reflect the actual situation. Moreover, earlier published data were based on insufficiently accurate methods of samples collection and further analysis. The updated maps of recent sediment types including the distribution maps of biogenic substances in sediments were recently compiled for the southern part of the Curonian Lagoon (Emelyanov 2014). Still, there is a lack of detailed investigations of sediments, accumulating in the coastal areas of the Curonian Lagoon, which are located close to river mouths as well as in the places of intensive accumulation of finest particles of sediments.

The present article offers the results of quantitative analysis and distribution of potentially hazardous trace elements and relevant biogenic components in the sediments of river mouths and accumulation areas of the semi-enclosed coastal lagoon.

MATERIAL AND METHODS

Study area

Current study covers small areas (polygons) of the Curonian Lagoon, located at the outlets of main rivers entering the lagoon (polygons 1–5), as well as in the two areas (polygons 6–7) of intensive accumulation of silty and clayey sedimentary material (Fig. 1).

The first polygon is located at the mouth of small Atmata River – the largest deep-water arm of the Neman River delta. In the mouth of Atmata, there is a vast shallow Krokų Lanka Lake (depth of 2–3 m), which acts as a trap for Atmata sedimentary material. Bottom of the lake is covered with semi-liquid and very soft silty clayey mud (Pustelnikovas 1998) with the content of $CaCO_3$ reaching 5.8–33.0% and TOC reaching 0.45–4.2%.

Another investigated polygon includes the area of small Skirvytė River mouth, located at the state border between Lithuania and Russia. Differently from the Atmata, Skirvytė directly enters the lagoon in the southern part of Neman River delta. Further polygons situated at the mouths of small channels Razliv, Matrosovka and Deima. The streams enter the lagoon from the Kaliningrad District area.

Polygon VI is located in the most south-eastern corner of the lagoon, characterized by intensive accumulation of smallest fractions due to the greater depths and lower hydrodynamic processes. Similarly intensive accumulation of fines takes place in the last investigated area on the eastern coast of the Curonian Spit.

Sample collection

Sampling of surface sediments was carried out during the period of 2001–2003 by the scientists of the Atlantic Branch of Institute of Oceanology RAS. Bottom sediments of the uppermost layer (0–3 cm) collected at 46 stations. The sampling carried out mainly in the winter season using the manual box-corer.

Analyses

All samples were analyzed for the grain size composition and total content of the following chemical elements: Ag, Al, As, Ca, Cr, Co, Cr, Cu, Fe, K, Li, Mg, Mn, Na, Ni, Pb, Sb, Ti and Zn. Additionally, sediment samples were analyzed for the content of biogenic components: total organic carbon (TOC), nitrogen (N), phosphorus (P), biogenic silica (SiO_2), calcium carbonate ($CaCO_3$).

Sediment samples were subjected to water-mechanical particle size analysis. The smallest fractions (0.1–0.01 mm, <0.01 mm) were separated by suspension weighting method (Prokoptsev 1965), while coarser particles (>0.1 mm) were distinguished by sieving method. The types of bottom sediments are distinguished on the basis of the decimal grain size classification system in accordance with the dominant fraction and median diameter (Md) of particles (Bezrukov, Lisitzin 1960). Following types of sediments were distinguished: sand (prevalence of 1.0–0.1 mm fraction), coarse silt (prevalence of 0.1–0.01 mm fraction, dominant 0.1–0.05 mm fraction), fine silty mud (prevalence of 0.1–0.01 mm, dominant

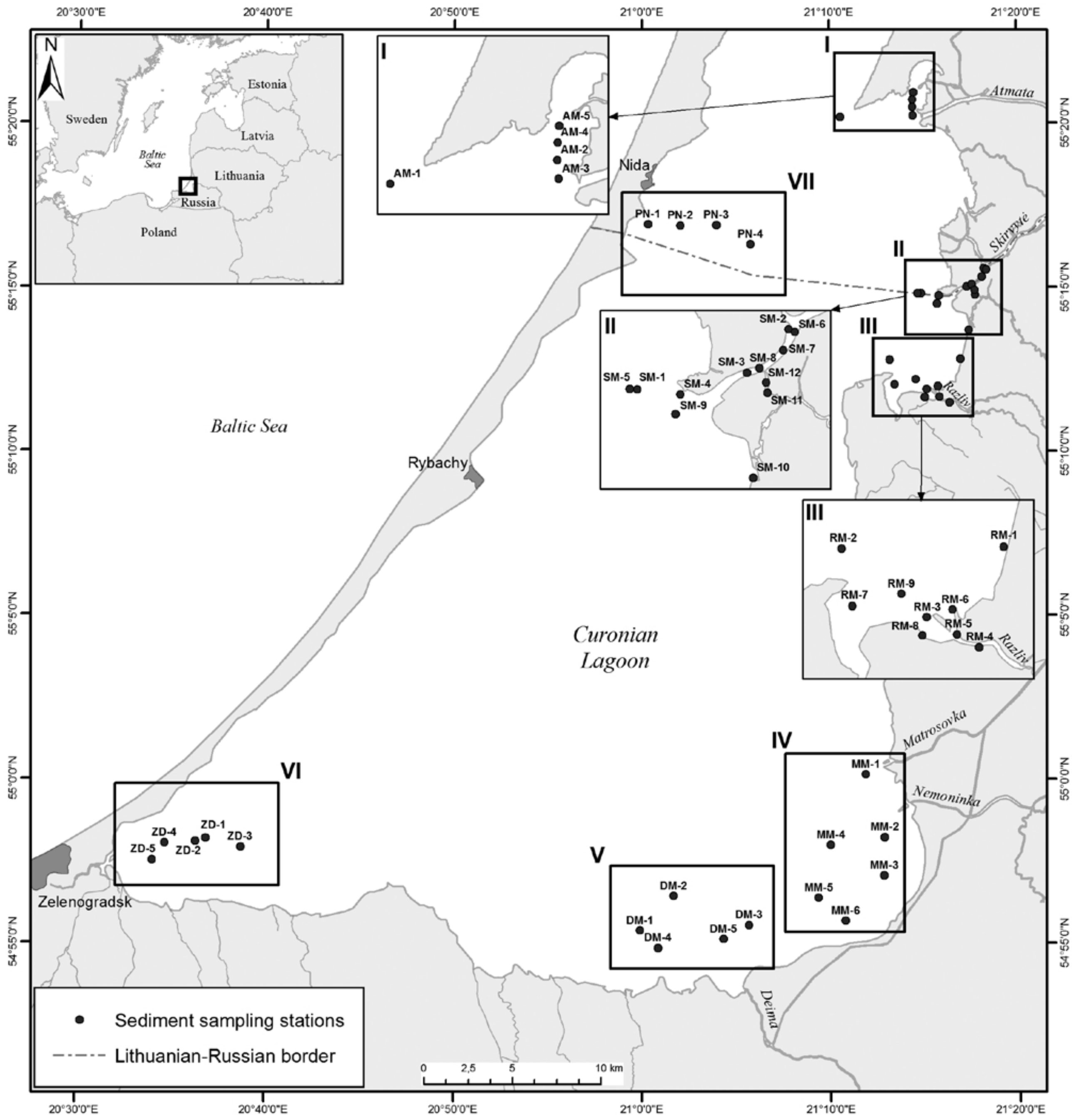

Fig. 1 Study area and sediment sampling stations

0.05–0.01 mm fraction) and silty clayey mud (50–70% of the <0.01 mm fraction).

Before the determination of chemical elements sediment samples were dried at 105°C temperature and powdered using the agate mortar. Method of atomic absorption investigation of rock-constituent elements (Fe, Mn, Ca, Mg, K, Na, Rb, Li, Cu, Zn, Cr, Ni, Co) lays in analyzing sample decomposition, then pulverizing of obtained solution to air-acetylene flame and measuring with the atomic absorption spectrometer *Vaian*. Optical density measuring for elements (Pb, As, Cd) was carried out under electro-thermal atomization of a sample in the graphite furnace AA–spectrometer *Quant-ZETA* is used. Determination of titanium (Ti) in sediments carried out by the method with photo-colorimeter ending with a use of the KFK-2MP (КФК-2МП).

Determination of P (gross) and N (gross) was carried out with a use of photometric method according to GOST 26261-4 (phosphorus) and GOST 26107-84 (nitrogen). Optical density measurement was made using photo-colorimeter KFK-2MP (КФК-2МП). Determination of SiO_2 in bottom sediments is made by sample's decomposition under water bath for two hours with a use of 5% Na_2CO_3 solution. Optical density measuring completed using the photo-colorimeter KFK-2MP (КФК-2МП).

Determination of TOC in the sediments was done by dry combustion with subsequent coulometric titration (express-analyzer of carbon – AN7529M). All the analyses performed in the Atlantic Geology Laboratory of ABIORAS (Kaliningrad), which have been widely used in studies of sediments from the Baltic Sea and other seas (Emelyanov 2005).

RESULTS

Sediment types and content of biogenic components

Most prevalent type of the sediments in the study area is fine silty mud. Silty mud covers the southern half of the central part of the lagoon, small areas close to the Neman River delta (opposite the mouths of Skirvytė and Razliv Rivers), as well as in the vicinity of Nida settlement. Sand is distributed in the coastal part of the Neman River delta and in the central part of the lagoon between the Neman River delta and the Curonian Spit. This is a common type of sediments accumulating along the coastline of the entire lagoon up to 2 m depth. Silty clayey mud occupies deepest and hydrodynamically less active areas in the south-western part of the lagoon.

Sediments of the lagoon contain 0.2–7.1% of TOC, which shows positive correlation with the amount of clayey fraction (<0.01 mm) and $CaCO_3$ content. This shows that most of the organic carbon in the lagoon is of biogenic origin. It has been observed that low-calcareous and calcareous sediments from the southern part of the lagoon are usually mixed with the terrigenous-calcareous sapropelitic (3–5% of TOC) or even sapropelic (5–7.1% of TOC) sediments.

The sediments of the Curonian Lagoon are rich in carbonates, mainly of biogenic origin. The amount of $CaCO_3$ locally can reach up to 59%. Elevated contents of $CaCO_3$ is typical for the southern part of the study area, where bottom is covered with muds. In the sands $CaCO_3$ content reaches 10–15%. Low-calcareous (10–30% of $CaCO_3$) and terrigenous (<10% $CaCO_3$) sediments are typical for the central part of the lagoon, as well as for the coastal areas.

The sand and mud of the Curonian Lagoon contains 0.1–0.3 and 0.5–1.33% of N, respectively, i.e., more than the marine sediments. This is obviously due to the younger age of the lagoon sediments (especially those near the Curonian Spit), which is explained by the more intensive accumulation rates of sediments and their higher enrichment with organic matter as compared with the marine sediments (Emelyanov 2014).

The amount of phosphorus ranging from 0.01 to 0.13%. Variable content of this element observed in each lithological type of sediments. Normally, its contents in sediments is 0.07–0.10%. In most of the samples comprised of fine silty mud, positive relation of phosphorus content and amount of clayey fraction was observed for the Atlantic Ocean (Emelyanov 1979) and the Baltic Sea (Emelyanov 1986).

Within this study the sediments downstream Atmata were investigated. Sand and fine silty mud prevails in this area. Coarse silt usually spreads from Neman River delta towards the deeper places of the lagoon, bordering the places of sand accumulation in the northern part. According to the new results, sand from the mentioned polygon contains from 1.83 to 4.33% of $CaCO_3$ and up to 0.5% of TOC. In the mineral composition of sand terrigenous feldspars are dominating. They are free from carbonates and contain 72–86% of SiO_{2bulk}. The mud is classified as low

Table 1 The Curonian Lagoon (polygon I – Atmata mouth)

	Station	AM-1	AM-2	AM-3	AM-4	AM-5
	Depth, m	1.0	2.0	4.0	3.0	3.0
	Sediment type	Sand	Sand	Sand	Coarse aleurite	Fine-aleuritic mud
Fraction, mm	Grain size distribution, %					
	>1.0	0.0	0.0	0.0	0.0	-
	1-0.5	0.8	0.8	0.1	0.2	-
	0.5-0.25	10.8	10.8	5.0	0.5	-
	0.25-0.1	86.5	86.5	92.8	10.2	-
	0.1-0.05	1.8	1.8	1.9	63.4	-
	0.05-0.01	0.1	0.1	0.2	20.0	-
	<0.01	0.0	0.0	0.0	5.6	-
%	Content of elements					
	$CaCO_3$	4.33	1.83	2.08	9.33	12.69
	TOC	0.35	0.19	0.15	3.44	4.23
	SiO_{2bulk}	72.0	86.0	80.0	61.0	-
	SiO_{2am}	-	-	-	-	3.20
	Al	2.11	1.85	1.32	4.60	-
	Fe	0.41	0.27	0.37	1.26	1.95
	Mn	0.02	0.02	0.02	0.06	0.07
	Ti	0.08	0.08	0.05	0.21	0.22
	N	0.05	0.03	0.03	0.38	0.49
	Ca	1.77	1.08	1.46	2.25	3.40
	Mg	0.15	0.15	0.14	0.46	0.52
	P	0.02	0.02	0.02	0.04	0.06
	K	0.88	0.55	0.62	1.24	1.75
	Na	0.49	0.32	0.42	0.46	0.75
	Li	2	3	3	5	
10^{-4}%	Cu	-	-	-	-	12
	Zn	32	23	68	132	52
	Cr	20	22	16	48	37
	Ni	14	12	7	20	19
	Co	8	10	9	8	7
	Pb	3	2	2	5	13
	As	23	36	21	22	14
	Sb	<0.2	<0.2	<0.2	<0.2	-
	Ag	<0.1	<0.1	<0.1	0.1	-
	Cd	0.10	0.10	0.10	0.2	0.3

calcareous and contain 12.69% of $CaCO_3$ and 4.23% of TOC (Table 1).

Sand prevails at the mouth of Skirvytė River and further from the coast. In the inland part of the river sand is medium grained (0.5–0.25 mm fractions prevail), at the mouth and further from the coast the sand fractions become smaller (0.25–0.1 mm). Similarly as at the mouth of previously mentioned Atmata River, the sand is purely clastic with very low content of carbonate (average content is 3.96% of $CaCO_3$) and organic carbon (up to 0.54%). The content of biogenic silica SiO_2 varies between 0.20 and 3.55%. The sediments from several stations stand out for one of the highest concentrations of biogenic substances, particularly nitrogen (0.23–0.97% of N) and phosphorus (0.10–0.12% of P) (Table 2).

Sandy sediments also prevail in the Razliv River mouth. Finer sediments represented by coarse silt are observed only at one investigated station (RM-9). Sand from this polygon contains 83–94% of SiO_{2bulk}, up to 10.67% of $CaCO_3$, up to 0.59% of TOC. Relatively high amount of biogenic silica (6.3%) is identified at one sampling station. The same station is characterized by relatively higher contents of N (1.0%) and P (0.11%) in comparison to other investigated places (Table 3).

Finer sediments represented by fine silty mud and silty clayey mud are observed in close vicinity to Matrosovka River mouth and to the south. The content of $CaCO_3$ in the fine silty mud reaches 14.9–15.98%, while in the clayey sediments $CaCO_3$ content increases up to 32.30%. Carbonate material is mostly of biogenic origin, represented by small shells of clams

Table 2 The Curonian Lagoon (polygon II – Skirvytė mouth)

Station		SM-1	SM-2	SM-3	SM-4	SM-5	SM-6	SM-7	SM-8	SM-9	SM-10	SM-11	SM-12
Depth, m		1.0	0.0	0.0	0.0	2.3	0.1	0.1	0.1	0.1	0.1	0.1	0.1
Sediment type		S	S	S	S	S	S	S	S	S	S	S	S
Grain size distribution,%													
Fractiom, mm	>1.0	0.0	0.3	1.7	0.2	-	-	-	-	-	-	-	-
	1-0.5	0.2	3.0	8.7	0.5	-	-	-	-	-	-	-	-
	0.5-0.25	13.5	63.7	75.9	27.7	-	-	-	-	-	-	-	-
	0.25-0.1	86.0	31.7	10.1	69.5	-	-	-	-	-	-	-	-
	0.1-0.05	0.3	0.0	0.0	0.0	-	-	-	-	-	-	-	-
	0.05-0.01	0.1	0.0	0.0	0.0	-	-	-	-	-	-	-	-
	<0.01	0.0	0.0	0.0	0.0	-	-	-	-	-	-	-	-
Content of elements													
%	$CaCO_3$	1.75	4.20	5.40	2.30	1.84	2.70	1.90	2.87	1.82	5.85	10.95	5.95
	TOC	0.16	0.24	0.54	0.14	0.09	-	-	-	-	-	-	-
	SiO_{2bulk}	77.0	-	-	-	-	-	-	-	-	-	-	-
	SiO_{2am}	-	0.20	0.65	0.20	0.42	0.63	0.58	0.60	0.37	3.55	3.30	1.66
	Al	1.85	-	-	-	-	-	-	-	-	-	-	-
	Fe	0.32	0.27	0.44	0.40	0.29	0.35	0.30	0.38	0.40	1.02	1.46	1.00
	Mn	0.03	0.01	0.03	0.02	0.02	0.02	0.01	0.02	0.01	0.04	0.06	0.03
	Ti	0.08	0.02	0.03	0.06	0.04	-	-	-	-	-	-	-
	N	0.03	0.08	0.16	0.10	0.02	0.03	0.03	0.03	0.03	0.97	0.55	0.23
	Ca	1.32	2.09	2.96	1.80	0.64	1.08	0.76	1.15	0.73	2.34	4.38	2.38
	Mg	0.15	0.25	0.30	0.15	0.08	0.26	0.21	0.27	0.15	0.45	1.25	0.54
	P	0.02	0.02	0.03	0.01	0.01	0.01	0.01	0.01	0.01	0.10	0.12	0.11
	K	0.48	1.05	0.95	1.24	0.94	0.90	0.86	0.70	0.65	1.00	1.35	1.04
	Na	0.39	0.80	0.70	0.70	0.52	0.51	0.33	0.41	0.30	0.53	0.44	0.40
10^{-4}%	Li	2	-	-	-	-	4	3	3	3	10	20	10
	Cu	-	6	4	5	4	4	4	4	4	20	48	18
	Zn	9	9	9	10	7	7	7	13	6	53	82	45
	Cr	15	17	19	17	12	10	12	10	10	25	40	22
	Ni	21	1	3	3	6	14	12	12	16	16	42	16
	Co	12	1	1	1	3	6	6	6	8	22	26	10
	Pb	3	4	5	4	7	5	5	4	5	10	15	23
	As	6	27	21	29	8	20	14	16	22	7	8	22
	Cd	0.10	0.10	0.10	0.10	0.20	0.10	0.10	0.10	0.10	0.50	0.30	0.20

(1–2 cm), dominated by the *Dreissena polymorpha*. Distribution of $CaCO_3$ further from the coast points to the intensive transport of sedimentary material, supplied by the river offsets and abrasion material. Muds are also characterized by relatively higher amounts of TOC (1.39–3.87%) in comparison to sands accumulating in the same area (0.46–0.56%) (Table 4).

Traces of coarse silt are locally observed at the mouth of Deima River. The sediments with dominating 0.05-0.01 mm fraction are of terrigenous origin and low-calcareous (26.3–26.4% of $CaCO_3$), noticeably enriched in organic matter (2.46–3.73% of TOC). Coarse silt also contains higher amounts of biogenic silica (up to 3.63%) and phosphorus (0.12%) (Table 5). However, the amount of biogenic components in the sediments from this polygon is considerably lower than in the other investigated river mouths.

South-western part of the lagoon is covered by fine silty and clayey mud. Accumulation of mud followed by the increased amounts of biogenic components. Sediments from this area are calcareous (35.10–43% $CaCO_3$), highly enriched in organic matter (4.84–7.10% of TOC) and amorphous silica (5.62–6.06%), which is represented by the skeletons of diatoms. This polygon is marked by highest sediment saturation with nitrogen (1.22%) among the investigated areas. Mud is also enriched with phosphorus, with the amount reaching 0.12–0.15% in particular stations (Table 6).

A slope section of the Curonian Spit at Nida settlement is covered by coarse silt in the shallower parts (up to

Table 3 The Curonian Lagoon (polygon III – Razliv mouth)

Station		RM-1	RM-2	RM-3	RM-4	RM-5	RM-6	RM-7	RM-8	RM-9
Depth, m		1.0	3.0	1.5	0.2	0.2	0.1	0.1	2.4	1.1
Sediment type		S	S	S	S	S	S	S	S	CA
Grain size distribution, %										
Fraction, mm	>1.0	0.0	0.7	-	-	-	-	-	0.7	-
	1-0.5	0.2	0.2	-	-	-	-	-	0.6	-
	0.5-0.25	8.3	0.6	-	-	-	-	-	45.2	-
	0.25-0.1	87.1	88.8	-	-	-	-	-	52.6	-
	0.1-0.05	3.9	4.9	-	-	-	-	-	0.9	-
	0.05-0.01	0.4	1.3	-	-	-	-	-	0.1	-
	<0.01	0.0	3.4	-	-	-	-	-	0.5	-
Content of elements										
%	$CaCO_3$	0.63	4.18	1.82	1.57	3.32	8.30	10.67	-	20.4
	TOC	0.20	0.59	-	-	-	-	-	-	0.67
	SiO_{2bulk}	94.0	83.0	-	-	-	-	-	-	-
	SiO_{2am}	-	-	1.05	0.68	0.39	6.3	4.18	-	-
	Al	2.11	3.17	-	-	-	-	-	-	-
	Fe	0.40	0.58	0.50	0.33	0.38	1.75	1.00	-	-
	Mn	0.01	0.04	0.01	0.01	0.02	0.04	0.04	-	-
	Ti	0.04	0.08	-	-	-	-	-	-	-
	N	-	-	0.57	0.03	0.03	1.00	0.68	-	0.16
	Ca	0.70	1.76	0.73	0.63	1.33	3.32	4.27	-	-
	Mg	0.10	0.24	0.13	0.13	0.12	0.66	1.85	-	-
	P	0.01	0.02	0.02	0.01	0.01	0.11	0.05	-	0.02
	K	0.50	0.80	0.72	0.74	0.79	1.21	1.68	-	-
	Na	0.30	0.40	0.54	0.36	0.94	0.57	0.54	-	-
	Rb	-	-	-	-	-	-	-	-	-
	Li	2	4	3	3	3	17	16	-	-
10^{-4}%	Cu	5	5	18	5	5	23	29	-	-
	Zn	24	14	84	24	15	71	57	-	-
	Cr	34	38	19	10	12	29	20	-	-
	Ni	11	15	26	12	10	40	16	-	-
	Co	5	16	12	10	6	20	18	-	-
	Pb	4	6	19	8	6	17	12	-	-
	As	45	17	20	19	21	7	20	-	-
	Sb	-	-	-	-	-	-	-	-	-
	Ag	0.60	0.70	-	-	-	-	-	-	-
	Cd	0.20	0.20	0.10	0.20	0.20	0.30	0.30	-	-

3 m in depth) and fine silty mud in deeper areas. Coarse silt contains up to 25.85% of $CaCO_3$, higher $CaCO_3$ contents (33.84–35.59%) is typical for the terrigenous-calcareous fine silty muds. The sediments noticeably enriched in organic matter (2.33–5.92% of TOC). The content of nitrogen in the sediments is high (1.0–1.18%) and most probably associated with the growing content of planktonic organic detritus (Table 7).

Distribution of major trace elements in the sediments

According to the results of current study, the sediments accumulating close to the mouth of Atmata River contain relatively low amount of the analyzed chemical ingredients. This is true both for the sands and muds observed in the area. Slightly higher concentrations are typical only for arsenic. In sands the concentration of As can reach up to 36 mg/kg, suggesting that sedimentary matter transported by the by the Atmata arm is saturated with this particular element. Increased concentrations of Cd observed in coarse silts and fine silty muds (0.2 and 0.3 mg/kg respectively) provides higher ability of cadmium to accumulate in the finer particles.

Higher concentrations of particular trace elements (Cd, Pb, Zn, Ni, Cr, Co) observed in sand sediments from three stations located in the Skirvytė River area.

Table 4 The Curonian Lagoon (polygon IV – Matrosovka mouth)

Station		MM-2	MM-5	MM-6	MM-1	MM-3	MM-4
Depth, m		2.2	3.0	3.0	3.0	2.3	3.6
Sediment type		S	S	S	FAM	FAM	APM
Grain size distribution, %							
Fraction, mm	>1.0	5.5	12.7	31.9	-	-	-
	1-0.5	1.3	13.0	1.0	-	-	-
	0.5-0.25	3.3	22.2	1.0	-	-	-
	0.25-0.1	40.1	30.5	11.4	-	-	-
	0.1-0.05	38.1	17.2	10.8	21.1	26.2	17.1
	0.05-0.01	9.4	2.1	18.6	62.2	44.4	32.3
	<0.01	2.2	2.3	25.3			
	>0.1	-	-	-	10.4	16.2	19.6
	0.01-0.005	-	-	-	3.1	4.4	18.5
	0.005-0.001	-	-	-	3.1	4.4	6.2
	<0.001	-	-	-	-	4.4	6.2
Content of elements							
%	$CaCO_3$	13.40	13.40	-	14.9	15.98	32.30
	TOC	0.56	0.46	-	1.39	1.98	3.87
	SiO_{2bulk}	-	-	-	-	-	-
	SiO_{2am}	0.48	0.55	-	0.40	1.33	3.63
	Al	-	-	-	-	-	-
	Fe	0.79	0.83	2.91	1.67	0.8	1.94
	Mn	0.03	0.02	0.12	0.06	0.04	0.07
	Ti	0.07	0.07	0.27	0.11	0.06	0.11
	N	0.08	0.08	0.67	0.1	0.16	0.50
	Ca	6.14	6.50	3.28	6.43	6.66	10.49
	Mg	0.30	0.30	0.52	0.7	0.6	0.54
	P	0.05	0.03	-	0.07	0.07	0.05
	K	1.26	1.14	1.44	1.95	1.89	1.22
	Na	0.58	0.65	2.26	0.69	0.70	0.43
	Rb	-	-	50	-	-	-
	Li	-	-	14	-	-	-
10^{-4}%	Cu	5	5	10	11	10	12
	Zn	19	13	92	45	40	50
	Cr	19	20	30	26	23	27
	Ni	8	3	60	12	8	11
	Co	5	3	14	5	3	4
	Pb	4	4	-	6	7	11
	As	29	93	-	**91**	34	17
	Cd	0.1	0.1	-	0.1	0.2	0.2

Those stations also characterized by high percent of biogenic compounds in the sediments. Considerable increases in chemical element concentrations compared to the sand from the other monitoring stations may suggest possible contribution of the anthropogenic contamination. Moreover, at many stations from this area high contents of As were observed, reaching up to 29 mg/kg.

The sediments from the Razliv River estuary characterized by slightly increased average concentrations of Ni (18.57 mg/kg), Pb (10.29 mg/kg) and Cd (0.21 mg/kg) in comparison to the surrounding areas. Average content of As (21.29 mg/kg) is comparable with the Atmata polygon (23.2 mg/kg).

The content of nearly all studied trace elements in the sediments of Matrosovka River mouth is comparable to their Clarke values, determined for the quartz sand. The average concentrations of most microelements are quite similar to the ones, determined in earlier mentioned polygons. The exception goes only for the As, which concentration in this particular area may reach up to 93 mg/kg and the average content reaching 52.8 mg/kg, which is the highest value among the all investigated polygons. The sediments accumulating in the southern part of the lagoon (estuary of Deima River) are characterized by slightly elevated average content of Fe (1.42%) and As (25 mg/kg).

Silty and clayey muds deposited in the area of intensive accumulation contain appreciably higher amounts of nearly all investigated chemical elements. The average content of As (30.8 mg/kg) is the second highest value from the investigated areas. The average concentration of Zn reaching 68.6 mg/kg, while in most of the polygons it rarely exceeds 45 mg/kg. The same goes for Cr average concentration (43 mg/kg), which is considerably higher than in the other areas.

Table 5 The Curonian Lagoon (polygon V – Deima mouth)

	Station	DM-1	DM-2	DM-3	DM-4	DM-5
	Depth, m	4.0	4.0	3.1	4.0	4.0
	Sediment type	S	S	S	CA	CA
Grain size distribution, %						
Fraction, mm	>1.0	4.0	-	-	7.1	-
	1-0.5	7.1	-	-	6.1	-
	0.5-0.25	7.6	-	-	9.5	-
	0.25-0.1	26.9	8.7	1.1	15.1	30.6
	0.1-0.05	12.4	22.8	0.8	28.7	28.0
	0.05-0.01	24.8	31.4	3.0	15.8	30.6
	<0.01	18.3	37.1	95.0	17.7	10.8
Content of elements						
%	$CaCO_3$	35.09	46.70	22.8	26.4	26.3
	TOC	4.90	2.56	0.57	2.46	3.73
	SiO_{2bulk}	39.5	-	-	-	-
	SiO_{2am}	-	1.64	0.28	2.45	1.23
	Al	4.70	-	-	-	-
	Fe	1.73	1.75	0.73	1.14	1.76
	Mn	0.08	0.06	0.01	0.05	0.08
	Ti	0.19	0.12	0.06	0.09	0.18
	N	-	0.38	0.03	0.15	0.54
	Ca	9.61	17.09	8.04	9.67	8.52
	Mg	0.94	0.48	0.20	0.33	0.46
	P	0.08	0.08	0.03	0.12	0.03
	K	0.74	1.00	0.94	1.14	1.10
	Na	0.50	0.45	0.67	0.63	0.50
10^{-4}%	Li	11	-	-	-	-
	Cu	16	14	9	11	13
	Zn	55	39	11	32	48
	Cr	34	25	13	23	25
	Ni	24	8	3	10	11
	Co	14	1	4	5	3
	Pb	4	11	7	6	13
	As	45	23	24	21	12
	Ag	0.60	-	-	-	-
	Cd	0.1	0.2	0.1	0.2	0.1

Table 6 The Curonian Lagoon (polygon VI – Zelenogradsk town)

	Station	ZD-1	ZD-2	ZD-3	ZD-4	ZD-5
	Depth, m	4.4	5.2	4.0	3.0	2.5
	Sediment type	FAM	FAM	APM	APM	APM
Grain size distribution, %						
Fraction, mm	>0.1	32.1	5.6	4.2	8.4	4.3
	0.1-0.05	4.4	4.3	6.6	1.7	3.9
	0.05-0.01	21.9	59.1	33.3	35.2	35.3
	0.01-0.005	16.6	7.7	27.9	27.3	14.1
	0.005-0.001	8.3	7.8	14.0	13.7	14.1
	<0.001	16.6	15.5	14.0	13.7	28.3
Content of elements						
%	$CaCO_3$	37.00	41.25	43.00	35.10	38.00
	TOC	4.84	7.10	5.45	6.64	6.00
	SiO_{2bulk}	-	5.4	-	-	-
	SiO_{2am}	5.62		5.85	5.65	6.06
	Al	-	-	-	-	-
	Fe	1.86	1.93	2.32	2.21	2.11
	Mn	0.06	0.08	0.08	0.08	0.07
	Ti	0.13	0.10	0.11	0.11	0.14
	N	0.82	0.36	0.38	0.38	1.22
	Ca	11.05	12.34	12.99	12.01	12.10
	Mg	0.51	0.91	0.86	0.80	0.80
	P	0.04	0.15	0.12	0.13	0.04
	K	0.92	1.20	1.34	1.23	1.03
	Na	0.42	0.55	0.90	0.63	0.30
10^{-4}%	Cu	16	19	19	16	21
	Zn	55	61	93	65	69
	Cr	33	48	47	49	38
	Ni	13	20	22	22	16
	Co	5	7	8	8	6
	Pb	13	6	6	10	16
	As	16	53	10	40	35
	Cd	0.2	0.2	0.3	0.3	0.3

The sediments from the Nida polygon are rich in Zn and Cr, which is confirmed by the highest average values (100 and 47.3 mg/kg respectively). Enrichment with arsenic is also obvious – the concentration in fine silty muds can reach up to 45 mg/kg, while average values reaching 23 mg/kg.

DISCUSSION

Composition of recent sediments is dependent on weathering of rocks, occurring in the source area, the properties of material originating from atmospheric deposition and material brought by rivers (Uścinowicz (Ed.) 2011). The chemistry of the sedimentary material transported and supplied by river waters into the lagoon is represented by the chemical composition of the sediments deposited in river mouths.

Apart from the main sources of terrigenous material represented by small rivers there is additional source of at least sandy fraction transport to the Curonian Lagoon from the Curonian Spit. Considerable amount of sandy material (enriched with quartz) is blown from the bared dunes by strong winds into the lagoon waters (Morkūnaitė *et al.* 2011). Finally, coastal abrasion and bottom erosion of the lagoon also provides particular amount of terrigenous material. Sand fraction (0.1–1 mm) comes into the lagoon mainly with the river offsets for which the lagoon acting as a natural reservoir. Coarser particles are deposited in the immediate vicinity of the river mouths, or at some distance from them, together with clayey and silty materials (Trimonis *et al.* 2010). Subsequently, as a result of wave processes and near bottom currents through multiple set offs (resuspension) of silty and clayey fractions <0.1 mm, they are deposited in hydrodynamically quieter areas of the lagoon in the south-western corner or in the central part, otherwise they are moving through the Klaipėda Strait barrier area towards the Gdansk Basin (Emelyanov *et al.* 2002).

Lagoons and semi-closed bays are particular areas where preliminary sedimentation of suspended solids rich in organic matter occurs (Uścinowicz (Ed.) 2011). Elevated contents of total organic carbon often occurs in rivers (Carman *et al.* 1996; Burska *et al.* 1999). Changes in TOC concentrations in the region affected by the river water inflow depend mainly on the amount and quality of the loading organic matter load, intensity of dynamic processes in the estuary area, as well as on the topography of the reservoir receiving the riverine waters. As reported earlier the amount of TOC in the Curonian Lagoon varied from 0.40 to 6.64 % (Emelyanov *et al*. 2002). Within this study higher values of TOC (up to 7.10%) were observed in the fine silty muds, accumulating in the deepest places in the south-western part of the lagoon. The sediments intensively accumulating in the central part of the lagoon contain from 2.33 to 5.92% of TOC, with average value reaching 4.5%. Biogenic origin of organic material is obvious – distribution of average $CaCO_3$ values is identical to the one of TOC. The average and median values of TOC and $CaCO_3$ from the different investigated polygons are presented below (Fig. 2).

Higher contents of TOC in the areas of intense accumulation of silty and clayey sediments were also recorded by other authors (Carman *et al*. 1996; Burska *et al*. 1999). Still, the amount of TOC accumulating in the study area is relatively lower if comparing to the neighbouring water basins. For instance, the highest amount of TOC (13.2%) deposited in the Szczecin

Table 7 The Curonian Lagoon (polygon VII – port of Nida town)

	Station	PN-1	PN-2	PN-3	PN-4
	Depth, m	4.0	5.0	3.0	3.0
	Sediment type	FAM	FAM	CA	CA
Grain size distribution, %					
Fraction, mm	>0.1	1.4	1.1	0.8	8.0
	0.1-0.05	9.5	13.4	43.9	66.9
	0.05-0.01	72.4	70.0	43.4	19.2
	0.01-0.005	9.4	8.1	11.9	2.1
	0.005-0.001	4.1	4.6	11.9	2.8
	<0.001	3.3	2.8	11.9	0.9
Content of elements					
%	$CaCO_3$	33.84	35.59	25.85	16.75
	TOC	5.92	5.60	4.00	2.33
	SiO_{2bulk}	40.0	40.0	46.0	52.0
	SiO_{2am}	-	-	-	-
	Al	3.39	3.34	3.06	3.17
	Fe	2.09	1.95	1.61	1.00
	Mn	0.08	0.10	0.08	0.04
	Ti	0.19	0.19	0.16	0.17
	N	1.00	1.03	0.89	1.180
	Ca	9.15	8.33	6.24	4.360
	Mg	0.82	0.76	0.62	0.510
	P	0.07	0.07	0.07	0.050
	K	1.13	1.27	1.38	1.470
	Na	0.56	0.55	0.63	0.670
10^{-4}%	Li	16	14	9	7
	Cu	-	-	-	-
	Zn	102	103	116	79
	Cr	66	55	43	25
	Ni	20	22	10	18
	Co	12	12	10	10
	Pb	5	7	3	5
	As	20	45	9	18
	Sb	<0.2	<0.2	<0.2	<0.2
	Ag	0.2	0.1	0.1	0.4
	Cd	0.1	0.1	0.1	0.1

Lagoon resulting from the intensive transport of organic reach sedimentary material by the Odra River. Huge amounts of TOC are produced during the process of photosynthesis owing to high concentrations of biogenic substances (Emeis *et al*. 2002). More than 10% of TOC is present in silty sediments of the north-western part of the Vistula Lagoon (Chechko, Blazhchishin 2002).

Earlier studies declared that P contents of 0.2–0.5% are typical for the sediments of almost all the Baltic Sea deeps as well as the Gulf of Riga and the Curonian Lagoon (Emelyanov 2014). This is not the case for this study, as maximum amounts of P in different parts of the study area varied from 0.11 to 0.15%. Highest average values of P is typical for the southern part of the lagoon in the mouth of Deima River (0.07% of P), same or higher amounts are observed in the fine-grained sediments from the areas of intensive accumulation. It is worth noting that elevated amounts of phosphorus observed in particular areas may be related to the natural properties of sedimentary material. This is confirmed by the increased concentrations of $CaCO_3$ average values observed in the same stations.

It was observed earlier that particular amount of biogenic components such as phosphorus and nitrogen, as well as some metals (Ca, Fe), can accumulate in the surface sediment layer as a result of early diagenesis of organic detritus (Emelyanov 1996).

Concentrations of trace elements in the sediments of rivers entering the lagoon may be determined by the sewage discharge from towns and countryside, industrial and agricultural activities taking place within their catchment areas. Amongst the group of the analyzed trace elements, arsenic in the sediments of the river mouths from the Curonian Lagoon catchment area occurs in high contents, reaching up to 93 mg/kg. High concentrations of As are characteristic of sediments of all the small rivers flowing into the lagoon both from the Lithuanian and Russian territories. In the sands from the Atmata River mouth located in the Lithuanian part of the lagoon As content varies from 21 to 36 mg/kg (mean value 23.2 mg/kg), similar or higher concentrations (from 20 to 45 mg/kg) are observed in fine silty muds covering the lagoon bottom in the central part (mean value 23 mg/kg). Higher concentrations of arsenic (40–53 mg/kg) are recorded in the muds intensively accumulating in the Russian part of the lagoon (mean 30.8 mg/kg), while highest average value (52.8 mg/kg) is observed in the estuary of small Matrosovka River located in the western coast.

It is worth noting, that sediments from the mentioned areas are not enriched neither with organic matter, nor with the other trace elements. Observed values at the mouths of small rivers entering the Curonian Lagoon are critically high in comparing with the sediments from the southern Baltic Sea, where As occurs at the level between <5 and 29 mg/kg with the mean value of 9.97 mg/kg (Fig. 3). The elevated levels of As recorded in the Bornholm Basin (up to 29 mg/kg) are often explained by the presence of chemical weapons dumping sites, which seems to be the cause of local contamination (Szczepańska, Uścinowicz 1994). The As content in the surface layer of the Gdansk Basin rarely exceed 20 mg/kg, i.e. the value corresponds to the geochemical background (Uścinowicz *et al.* 1998). Values of As observed within this study are well above than those, observed in the Ancylus Lake clays from the Gdansk Basin (2–9 mg/kg, maximum is 13 mg/kg) and can also suggest anthropogenic origin (Emelyanov, Kravtsov 2007).

It is obvious that extremely high concentrations of arsenic in the sediments of Curonian Lagoon are related to the human economic activity. The main sources of anthropogenic arsenic in nature are wood preservatives, pesticides and fertilizers as well as releases from smelters and metal industry (Loukola-Ruskeeniemi, Lahermo 2004). The Curonian Lagoon is surrounded by the agricultural and industrial areas, providing the pressure through loading with nutrients, organics and contaminants lost in the catchment area (Newton *et al.* 2013). Therefore, most probable source of arsenic in sediments of the investigated area

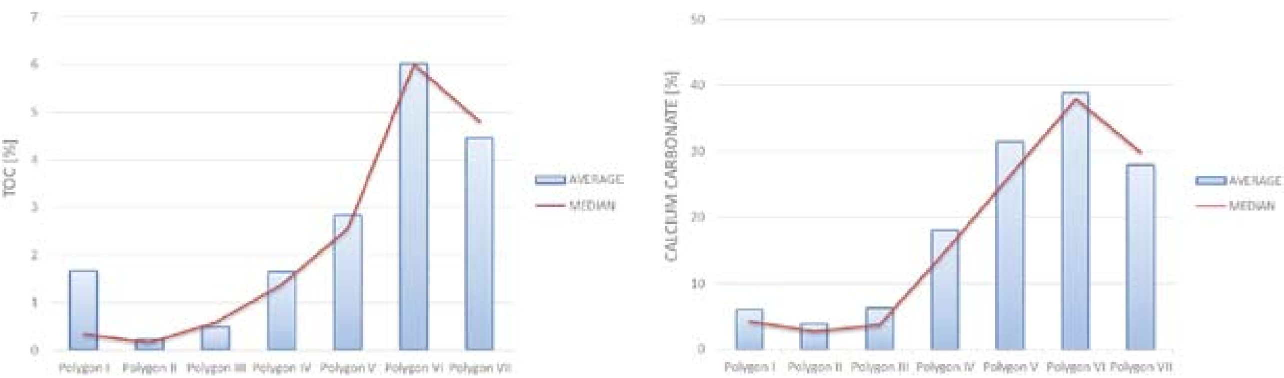

Fig. 2 Average and median values of TOC (a) and $CaCO_3$ (b) in the sediments of investigated polygons

is surface run-off from agricultural areas. However, further investigations of sediments chemical composition for more precise identification of contamination sources are of major importance.

CONCLUSIONS

Sediments accumulating in the mouths of small rivers falling into the basin of the Curonian Lagoon are of terrigenous and biogenic-terrigenous (calcareous) origin. In most cases calcareous substances are represented by mollusc shells and their detritus. Lowest average amounts of $CaCO_3$ are typical for sandy deposits, accumulating in the Neman River delta area (3.96–6.05%). Sediments accumulating in the deeper places of the lagoon are much richer in carbonates due to the prevalence of smallest fractions (< 0.01 mm) in sediment composition.

The areas covered with fine-grained sediments (primarily coarse silts and fine silty muds) are also enriched with other biogenic elements, such as phosphorus and nitrogen. Highest average values (0.63–1.03 of N; 0.07–0.1 of P) are observed in the western part of the lagoon, characterized by the intensive accumulation of sedimentary matter.

Distribution of the analyzed trace elements is quite chaotic through the study area. In most cases, obvious increases of potentially hazardous metals (Cd, Cr, Cu, Ni, Pb, Zn) is typical for the intensive accumulation zones of silty and clayey muds. The exception is attributable to the polygon located in the Razliv River mouth, where sands are enriched with Ni (mean value is 18.57 mg/kg), Pb (10.29 mg/kg) and Cd (0.21 mg/kg).

Results from this study have shown considerable enrichment of sediments with arsenic. In most places average contents of this metalloid are well above the geochemical background values, determined for

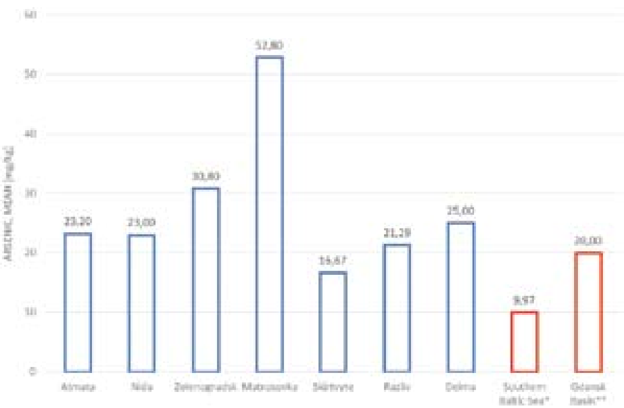

Fig. 3 Average values of arsenic in the sediments of study area (blue columns) and geochemical background values in the southern Baltic Sea (after Szczepańska, Uścinowicz 1994) and Gdansk Basin (after Uścinowicz *et al.* 1998) (red columns)

the south-eastern part of the Baltic Sea. Absolutely highest values of arsenic (91–93 mg/kg) suggesting presence of serious contamination sources were determined in the south-eastern part of the lagoon, adjoining the mouth of the small Matrosovka River. In other investigated polygons the amounts of arsenic in sediments are twice as low.

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