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Heavy metals and ^{137}Cs levels in macrophytes and temporal distribution in sediments – application for estuarine environment status assessment (Vistula Lagoon – southern Baltic)

by

Tamara Zalewska[1,*],
Paulina Brzeska-Roszczyk[2],
Beata Danowska[1],
Mariusz Pełechaty[3]

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[1]Institute of Meteorology and Water Management National Research Institute, ul. Waszyngtona 42, 81-342 Gdynia, Poland

[2]Gdynia Maritime University, Maritime Institute, Department of Aquatic Ecology, ul. Morska 81-87, 81-225 Gdynia, Poland

[3]Department of Hydrobiology, Institute of Environmental Biology, Faculty of Biology, Adam Mickiewicz University in Poznań, ul. Uniwersytetu Poznańskiego 6, 61-614 Poznań, Poland

* Corresponding author: *tamara.zalewska@imgw.pl*

Abstract

The paper presents the first data on the concentrations of heavy metals (Cd, Pb, Zn, Cu, Ni, Cr, Mn) and ^{137}Cs and their contamination ratios (CR) in the most abundant species of macrophytes in the Vistula Lagoon. No significant differences in the concentrations of heavy metals and ^{137}Cs between macrophyte taxa or the influence of rivers flowing into the Vistula Lagoon on heavy metal concentrations in the area were found. The concentrations of heavy metals in macrophyte taxa varied in the following ranges: Cd – 0.1–0.7 mg kg^{-1} d.w.; Pb – 0.5–5.0 mg kg^{-1} d.w.; Zn – 29–390 mg kg^{-1} d.w.; Cu – 2.5–8.3 mg kg^{-1} d.w.; Ni – 0.4–6.8 mg kg^{-1} d.w.; Cr – 0.5–2.8 mg kg^{-1} d.w.; Mn – 380–8500 mg kg^{-1} d.w. Since the 1990s, a decline or stable state of heavy metal concentrations in bottom sediments has been observed, reflecting changes in the environment of the Vistula Lagoon. The linear sedimentation rate in the Vistula Lagoon was 3.3 mm y^{-1}. The results presented in the paper can serve as a baseline for assessing changes in the environmental status of the Vistula Lagoon, which may occur as a result of future investments, including building a new navigable canal through the Vistula Spit.

Key words: heavy metals, ^{137}Cs, macrophytes, sediments, geochronology, Vistula Lagoon

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Introduction

The Vistula Lagoon is the second largest, semi-enclosed lagoon in the Baltic Sea (838 km^2). Due to its unique natural values, the lagoon was included in the network of Marine Protected Areas in the Baltic Sea (HELCOM 2016) and Natura 2000 sites in the European Union (Special Protection Area PLB290010 under the Birds Directive: Directive 2009/147/EC of the European Parliament and of the Council of 30 November 2009 on the conservation of wild birds; Special Area of Conservation PLH280007 under the Habitats Directive: Council Directive 92/43/EEC on the Conservation of natural habitats and of wild fauna and flora). At the same time, the Vistula Lagoon is exposed to various pressures, including those related to tourism and fishery, as well as pollution originating from land. A very important issue in terms of the environmental status of this unique basin is a plan to build a new navigable canal through the Vistula Spit, which will connect the lagoon waters with the Gulf of Gdańsk. The aim is to intensify shipping and increase its share in both the economic and tourism sector. Such a project may significantly affect the environmental conditions of the lagoon, e.g. through contamination with hazardous substances like heavy metals. Deepening of the shipping canal involves the necessity of disturbing sediments, which may result in the secondary release of pollutants present in sediments. The project is particularly relevant when considering obligations under EU legislation: the Water Framework Directive (Directive 2000/60/EC of the European Parliament and of the Council of 23 October 2000 establishing a framework for Community action in the field of water policy), which is the key directive on water protection and defines the principles of water policy; and the Marine Strategy Framework Directive (Directive 2008/56/EC of the European Parliament and of the Council of 17 June 2008 establishing a framework for Community action in the field of marine environmental policy). The Vistula Lagoon is a transitional water body and as such is subjected to all activities related to the implementation of both directives. It is therefore necessary to conduct research to increase knowledge about the distribution of pollutants in particular elements of the lagoon's environment, taking into account their potential sources. Such research should be aimed at supporting the decisions regarding management and protection of the Vistula Lagoon.

The research described in this paper was aimed at determining the concentrations of heavy metals in selected species of macrophytes (aquatic plants) that are most typical of the Vistula Lagoon. Macrophytes are among the most sensitive and reliable bioindicators, including contamination levels (Bojanowski 1973; Szefer & Skwarzec 1988; Haroon & Szaniawska 1995; Kruk-Dowgiałło & Pempkowiak 1995; Rainbow 1995; Leal et al. 1997; Ostapczuk et al. 1997; Malea & Haritonidis 2000; Szefer 2002a,b; Sawidis et al. 2003; Burger 2006; Żbikowski et al. 2007; Zalewska & Saniewski 2011; Zalewska 2012a,b; Zalewska & Suplińska 2012; Alquezar et al. 2013; Chakraborty et al. 2014; Zalewska 2015; Farias et al. 2018; Sinaei et al. 2018). This is mainly due to their ability to exchange elements with the surrounding environment. The exchange in macroalgae takes place through their thalli, uptaking elements directly from water, while in vascular plants mostly through their root system. In both cases, concentrations of contaminants in plants reflect levels in their nearest environment (water), which facilitates the interpretation of data, and additionally, the response of plants to changes in the environment is very fast. The most valuable feature is that concentrations of pollutants in plants reflect the condition of the basin.

On the basis of the obtained results, factors of heavy metal concentrations were calculated. They indicate the ability of individual species to accumulate heavy metals, which can be used to assess the environmental status and to predict changes in the environment.

The objective of the present paper was also to describe changes in heavy metal concentrations in the historical aspect, which was accomplished by analyzing changes in heavy metal concentrations in sediment cores in relation to the age of particular sediment layers. The sedimentation rate of bottom sediments in the Vistula Lagoon was also determined.

The results presented in the paper may serve as a basis for predicting the distribution of heavy metals also in other sea areas with similar environmental characteristics as the Vistula Lagoon. The results are also a baseline for potential changes in the environment caused by the canal in the Vistula Spit. This investment may affect directly and indirectly (by increasing pressure) hydrological, physicochemical and biological characteristics, which consequently may affect concentration levels and distribution of heavy metals in the Vistula Lagoon ecosystem.

Materials and methods

Study area

The Vistula Lagoon is a brackish water ecosystem with salinity ranging from 0.3 near estuaries up to 6.5

near the strait connecting with the Baltic Sea (Fig. 1). The basin is vast and shallow with an average depth of 2.7 m and a maximum depth of 5.2 m. The bottom is morphologically undifferentiated, muddy sediments prevail and cover deeper parts of the lagoon. Sandy shoals stretch along the coast to a maximum depth of 2 m (Chubarenko & Margoński 2008). These are the only areas with macrophytes. The vegetation consists mainly of *Phragmites australis* (Cav.) Trin. ex Steud. and *Scripus lacustris* L., rarely *Typha angustifolia* L., which on the open water side border on a belt of elodeids dominated by species of the *Potamogeton* L. genus, accompanied by *Myriophyllum spicatum* L. Shallow muddy bays are found along the western coast of the lagoon, usually densely covered by *Potamogeton* spp., *Ceratophyllum* spp., *Zannichellia palustris* L., *Chara* species and nymphaeids *Nuphar lutea* L., *Nymphaea alba* L., *Hydrocharis morsus-ranae* L. The submerged vegetation is often covered with thick "algal mats" of filamentous green algae *Cladophora* sp., *Oedogonium* sp. and *Rhizoclonium riparium* (Roth) Harvey (Pliński et al 1978; 1995; Brzeska et al. 2015). Since the 1970s, there has been a significant reduction in the area covered by elodeids and nymphaeids, presumably due to increasing trophic status (Pliński et al 1978; Pliński 1995).

Sampling and preparation for analyses

Macrophytes. Samples of macrophytes were collected in the Polish part of the Vistula Lagoon (Fig. 1) in July 2011. Laboratory analyses of taxonomic composition and biomass of macrophyte samples were conducted in accordance with the Polish standards (Kruk-Dowgiałło et al. 2010), which are based on the HELCOM guidelines (Bäck 1999). Each taxon was dried in a dryer at 60°C until constant weight was reached. Only species with biomass higher than 1 g d.w. were subjected to further analyses, i.e. for the content of ^{137}Cs and heavy metals.

The mineral incrustation of Charophyta specimens was not removed. In general, the incrustation can serve as an additional source of contaminants in plants (along with bioaccumulation), but species occurring in

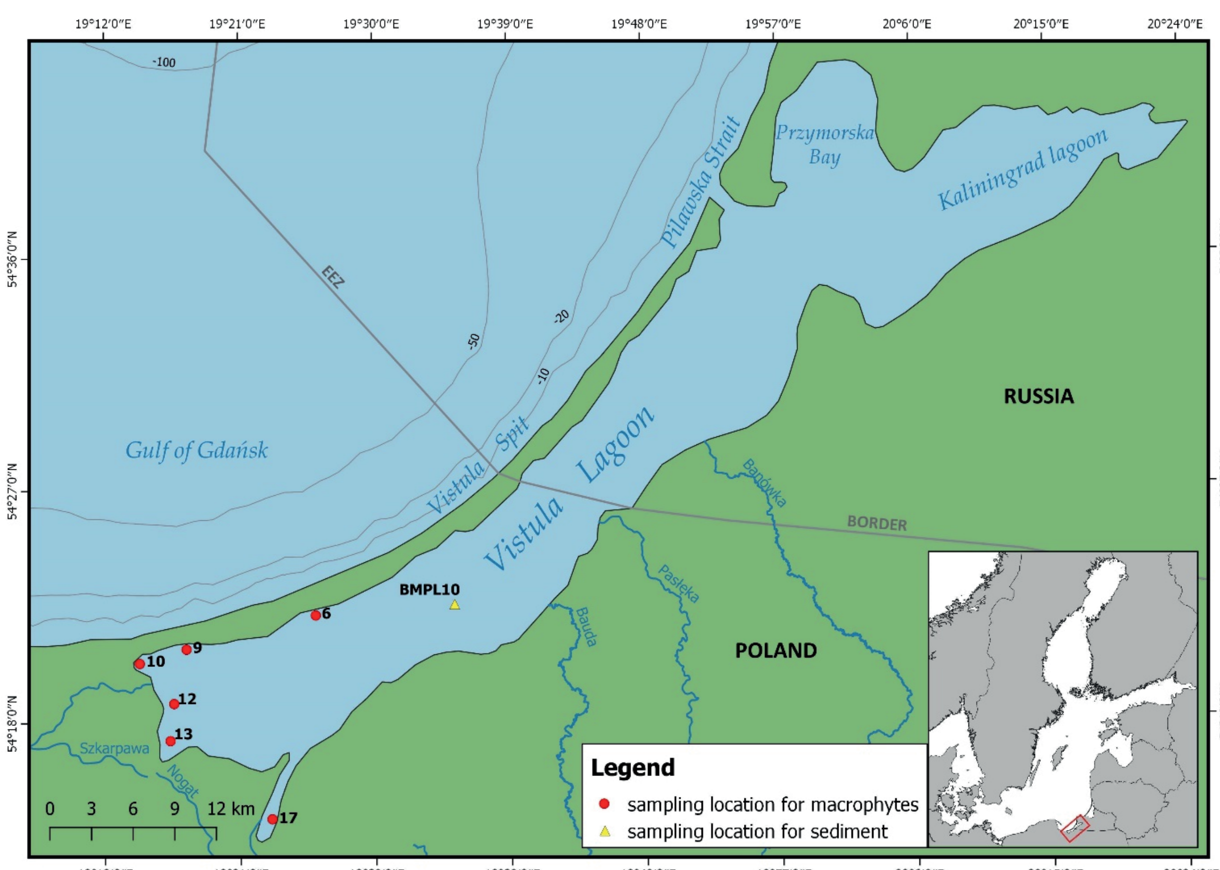

Figure 1

Sampling locations for the determination of contamination in the Vistula Lagoon (numbers – macrophytes, BMPL10 – sediments) saline environments are less encrusted compared to specimens from freshwater (Urbaniak 2010). Therefore, it was assumed that the effect of incrustation on the final results of this study can be neglected.

Sediments. Sediment samples were collected at one sampling location (Fig. 1) in July 2015, using a Niemistö corer with an inner diameter of 5 cm. Sediments in this area consist of silt and clay, with a predominantly fine fraction of less than 0.063 mm.

For the purpose of heavy metal determination, three parallel cores of about 40 cm length were collected; each of them was divided into 2 cm slices to a depth of 10 cm, and at greater depths, 2 cm slices were selected at every 5 cm of the core length. Eventually, the three parallel cores were divided into the following samples: 0–2 cm, 2–4 cm, 4–6 cm, 6–8 cm, 8–10 cm, 15–17 cm, 22–24 cm, 29–31 cm and 36–38 cm. The selection of sediment layers and the number of samples sufficient to assess heavy metal distribution in the sediment core was based on the experience gained from the Baltic Sea monitoring. Wet sediment samples were preserved by deep-freezing on board the vessel and then freeze-dried, homogenized and stored for further analysis in the laboratory. Analyzes of metals in sediments were carried out as part of the State Environmental Monitoring coordinated by the Chief Inspectorate for Environmental Protection financed from the National Fund for Environmental Protection and Water Management.

Three additional parallel sediment cores were collected at the sampling location to determine the age of the sediment. The cores were divided into 2 cm wide slices to a depth of 50 cm. The corresponding slices/layers from the three parallel cores were integrated to produce a single analytical sample. These samples were initially deep-frozen on board the ship and freeze-dried and homogenized in the laboratory prior to analysis.

Contamination analyses

Radionuclides. Dry biomass of selected macrophyte species was incinerated at 450°C. Homogenized samples of vegetation and sediments were placed in plastic containers of identical geometry as those used for calibration and in this form they were ready for measurements using the gamma spectrometric method. Activity concentrations of ^{137}Cs, ^{210}Pb, ^{214}Pb and ^{214}Bi were measured using a spectrometric system: an HPGe detector with a relative efficiency of 40% and a resolution of 1.8 keV for a peak of 1332 keV of ^{60}Co. The detector was coupled to an 8192-channel computer analyzer. The analysis of radioactivity spectra, registered in 8000 channels, was carried out by the Genie-2000 software. The reliability and accuracy of the applied method was verified by proficiency tests in the HELCOM – IAEA – 446 Proficiency Test Determination of Radionuclides in Fucus Sample. The IAEA ^{137}Cs concentration recommended value was 18.8 ± 0.4 Bq kg^{-1} d.w. Repeated analysis provided a result of 18.8 ± 0.8 Bq kg^{-1} d.w. (IAEA 2013; Pham et al. 2014).

Heavy metals. Concentrations of heavy metals were measured in mineralizates of plants (Cd, Pb, Zn, Cu, Ni, Cr and Mn) and sediments (Cd, Pb, Zn, Cu, Hg and Al), obtained by treating the samples with concentrated acids HNO_3 and HF. Laboratory vessels used in metal analyses were prepared by treatment with dilute nitric acid. Mineralization was carried out in Teflon vessels using a Milestone UltraWave microwave mineralizer. The concentrations of metals in plants were measured applying atomic absorption spectrometry (AAS) using a Shimadzu AA-6601F flame atomic absorption spectrometer. Concentrations of metals in the sediments were determined by the flame atomic absorption method (FAAS) and the electrothermal method (GF-AAS) using Thermo Scientific spectrometers.

On the basis of the obtained results, the concentration factors of heavy metals were calculated. They indicate the ability of individual macrophyte species to accumulate heavy metals, which can be used to assess the environmental status and to predict changes in the environment.

The content of mercury in the sediments was determined using cold vapor atomic absorption spectrometry in an AMA 254 mercury analyzer. A sample (ca. 100 mg) was placed in a combustion chamber of the analyzer, where it was dried and burned in an oxygen flame at 600°C. The released mercury was collected in a gold amalgam catalyst. After the sample decomposition was completed, the temperature was stabilized at 120°C and the content of mercury was measured with a detection limit of 0.05 ng.

The accuracy and precision of measurements were controlled using a certified reference material (Table 1), analyzed parallel to sediment samples.

Models for ^{210}Pb sediment dating

^{210}Pb identified in sediment samples originates from two sources. A certain fraction is the result of radium (^{226}Ra) radioactive decay and this is called supported ^{210}Pb ($^{210}Pb_{supp}$). Its activity along the vertical sediment profile practically does not change. The other source of ^{210}Pb deposited in marine sediments is atmospheric fallout. The activity of ^{210}Pb unsupported

Table 1

Results of the analysis of the certified reference materials

		Cd	Pb	Zn	Cu	Ni	Cr	Mn	Hg
		mg kg^{-1} d.w.							
BCR-414 (biological material)	Certified	0.383 ± 0.014	3.97 ± 0.19	111.6 ± 2.5	29.5 ± 1.3	18.8 ± 0.8	23.8 ± 1.2	299 ± 13	-
	Measured	0.37	3.84	109.59	28.9	19.2	22.6	287	-
	RSD	0.098	0.029	0.035	0.009	0.113	0.087	0.017	0.098
	LOD	0.0015	0.03	0.012	0.012	0.015	0.02	0.01	0.0015
	LOQ	0.005	0.10	0.035	0.035	0.045	0.06	0.03	0.005
MESS-4 (sediment)	Certified	0.28 ± 0.04	21.5 ± 1.2	147 ± 6	32.9 ± 1.8	-	-	-	0.08 ± 0.06
	Measured	0.26	20.1	143	31.8	-		-	0.08
	RSD	0.056	0.022	0.012	0.037				0.006
	LOD	0.0015	0.07	0.007	0.012				0.001
	LOQ	0.005	0.20	0.020	0.035				0.003

or excess ($^{210}Pb_{ex}$), originating from atmospheric deposition, decreases with the sediment depth. This activity constitutes the basis for determining sediment accumulation rates: the mass accumulation rate (MAR) and the linear accumulation rate (LAR) and for determining the age of particular sediment layers. The $^{210}Pb_{ex}$ activity concentration is determined from the total activity of this isotope ($^{210}Pb_{tot}$) in the analyzed layer by subtracting the activity of one of the products of ^{226}Ra decay, e.g. ^{214}Bi or ^{214}Pb. In the present study, sedimentation rates and sediment age along the vertical profiles were determined using two models: the Constant Rate of Supply (RSC) model and the Constant Flux Constant Sedimentation Rate (CF:CS) model (Appleby & Olfield 1992; Appleby 1997; Boer et al. 2006; Diaz-Asencio et al. 2009; Szmytkiewicz & Zalewska 2014).

In order to verify the results of age determination by the ^{210}Pb method, it is necessary to apply an additional tag whose concentration changes in the marine environment can be easily documented in relation to specific events. In the case of the Baltic Sea, the most obvious tag is the totally anthropogenic isotope of cesium – ^{137}Cs. When verifying the age determination method based on ^{137}Cs, it is assumed that the described historical events (e.g. testing of nuclear weapons performed since 1945 with maximum deposition recorded in 1963 and the accident at the Chernobyl power plant, Ukraine, in 1986) should be well marked as an increase in the curve of isotope changes along the sediment core. At the same time, the results have to be interpreted with caution, taking into account the complexity and the large number of processes affecting the final result – the presentation of ^{137}Cs distribution in the sediment vertical profile. Therefore, the isotope could be useful for verifying sediment chronology when post-depositional processes are not affecting this radionuclide (Diaz-Asencio et al. 2009).

Results and discussion

Concentrations of heavy metals and ^{137}Cs in macrophytes

Green algae *Cladophora glomerata* (noted at four sampling locations) and *C. fracta* (at one sampling location) were the most abundant taxa in this study. In the discussion of the results, both species were combined into the group of *Cladophora* spp., where average concentrations of Cd varied from 0.42 to 0.93 mg kg^{-1} d.w. The lowest value was determined in the mouth of the Nogat River and the Szkarpawa River (location 13), while the highest concentration occurred near the Vistula Spit (location 9; Table 2, Fig. 2). Similar characteristics were observed for Zn and Ni, with concentration ranges of 242.3–645.4 mg kg^{-1} d.w. and 3.2–9.6 mg kg^{-1} d.w., respectively. Pb and Cu were rather evenly distributed among the locations (Fig. 2) and ranged from 4.6 to 5.5 mg kg^{-1} d.w. and from 6.4 to 8.0 mg kg^{-1} d.w., respectively (Table 2, Fig. 2). The largest statistically significant differences between the locations were recorded for Cr: 0.08–0.86 mg kg^{-1} d.w. and Mn: 1553–10315 mg kg^{-1} d.w, with the highest concentrations at location 13. A similar situation was

Table 2

Concentrations of heavy metals and ^{137}Cs in macroalgae and vascular plants at the sampling locations in the Vistula Lagoon in 2011

Species	Sampling location (number of samples)		Cd	Pb	Zn	Cu	Ni	Cr	Mn	^{137}Cs
			mg kg^{-1} d.w.							Bq kg^{-1} d.w.
Cladophora spp. (*C. glomerata, C. fracta*)	9 (1)	value	0.93	5.5	645.4	8.0	9.6	0.08	2706	< 6.0
	10 (5)	mean*	0.84	4.2	430.1	5.0	4.7	0.18	3605	3.0 ± 1.4
		min.	0.41	2.9	327.3	3.5	4.2	0.11	2208	
		max	1.24	5.2	651.8	8.2	5.5	0.28	5485	
	12 (5)	mean*	0.67	3.2	307.7	6.7	3.2	0.60	1553	6.4 ± 3.5
		min.	0.30	1.4	107.3	1.7	0.1	0.12	996.8	
		max	1.23	5.7	653.0	12.0	8.9	1.08	2182	
	13 (4)	mean*	0.42	4.6	242.3	6.4	3.2	0.86	10315	5.3 ± 2.1
		min.	0.35	2.4	186.2	5.9	1.0	0.72	5063	
		max	0.53	6.6	293.2	7.0	5.5	0.99	12840	
Enteromorpha clathrata	12 (1)	value	0.65	1.8	390.5	3.2	0.41	0.51	383.4	< 3.0
Potamogeton sp. (*P. pussilus, P. pectinatus, P. compressus, P. perfoliatus*)	6 (1)	value	0.38	1.2	175.4	4.9	1.4	0.93	673.4	< 5.2
	10 (2)	mean*	0.20	1.0	187.8	3.7	1.0	0.70	784.6	< 2.1
		min.	0.12	0.69	108.2	2.9	0.15	0.65	782.8	
		max	0.28	1.3	267.5	4.4	1.8	0.75	786.5	
	12 (2)	mean*	0.20	0.56	136.2	4.2	1.7	0.89	1255	< 5.5
		min.	0.16	0.09	67.8	3.1	1.2	0.56	634.7	
		max	0.24	1.02	204.6	5.3	2.3	1.22	1874	
	13 (1)	value	0.39	1.7	177.4	4.3	2.3	0.85	3950	< 5.2
	17 (1)	value	0.25	3.5	130.4	5.4	5.5	1.13	10035	7.3 ± 1.6
Chara sp. (*Ch. aspera, Ch. contraria, Ch. globularis*)	10 (6)	mean*	0.25	0.58	195.6	2.7	3.3	0.75	1697	0.9 ± 0.5
		min.	0.22	0.20	157.3	1.2	0.81	0.36	965.6	
		max	0.28	1.3	242.1	8.7	14.4	2.34	2709	
	12 (1)	value	0.13	0.28	58.5	1.3	0.76	0.30	694.9	< 0.7
Sparganium emersum	6 (1)	value	0.39	1.1	61.5	4.7	1.7	1.45	919.5	< 2.0
	12 (1)	value	0.10	0.72	52.2	1.2	0.69	0.39	463.7	< 2.0
Ceratophyllum demersum	17 (2)	mean*	0.39	5.0	179.2	8.3	4.1	2.79	8543	7.9 ± 3.5
		min.	0.26	4.7	116.1	7.2	2.6	2.39	8259	
		max	0.52	5.4	242.2	9.4	5.5	3.19	8828	
Zanichellia palustris	12 (1)	value	0.63	1.8	391.8	6.3	1.1	2.22	772.3	< 7
Scirpus lacustris	10 (1)	value	0.05	1.9	28.6	5.0	6.8	1.79	738.4	4.9 ± 0.6

*mean value based on the number of samples given in brackets

observed in the case of the second taxon, representing vascular plants in the Vistula Lagoon, *Potamogeton* spp., for which the highest concentrations of Cr (1.13 mg kg^{-1} d.w.) and Mn (100355 mg kg^{-1} d.w.) were detected in Elbląg Bay (location 17). Concentrations of other metals in *Potamogeton* spp. were at slightly lower levels than in *Cladophora* spp. (Fig. 2). Values of Cd concentrations were in a narrow range from 0.2 to 0.38 mg kg^{-1} d.w. (Table 2, Fig. 2). The lowest concentration of Pb was recorded at location 12: 0.56 mg kg^{-1} d.w. and the highest one at location 17: 3.5 mg kg^{-1} d.w. Concentrations of Zn and Cu were more evenly distributed (Fig. 2), while the content of Ni in *Potamogeton* spp. varied from 1.0 mg kg^{-1} d.w. at location 10 to 5.5 mg kg^{-1} d.w. at location 17 (Table 2, Fig. 2).

The Kruskal–Wallis test showed statistically significant differences between the locations only in the case of *Cladophora* spp. and only for Cr ($p = 0.0005$) and Mn ($p = 0.0136$), which was not observed for *Potamogeton* spp.

When comparing the average concentrations of heavy metals (calculated on the basis of all data for a particular taxon; Table 2, Fig. 3), one should consider the way the elements are exchanged with the environment, which is one of the key factors affecting the bioaccumulation. In the case of macroalgae, which in the Vistula Lagoon were represented by green algae *Cladophora* spp., *E. clathrata* and *Chara* spp., bioaccumulation is performed through the entire surface of a thallus, while in vascular plants (which included all other species), the roots also play an

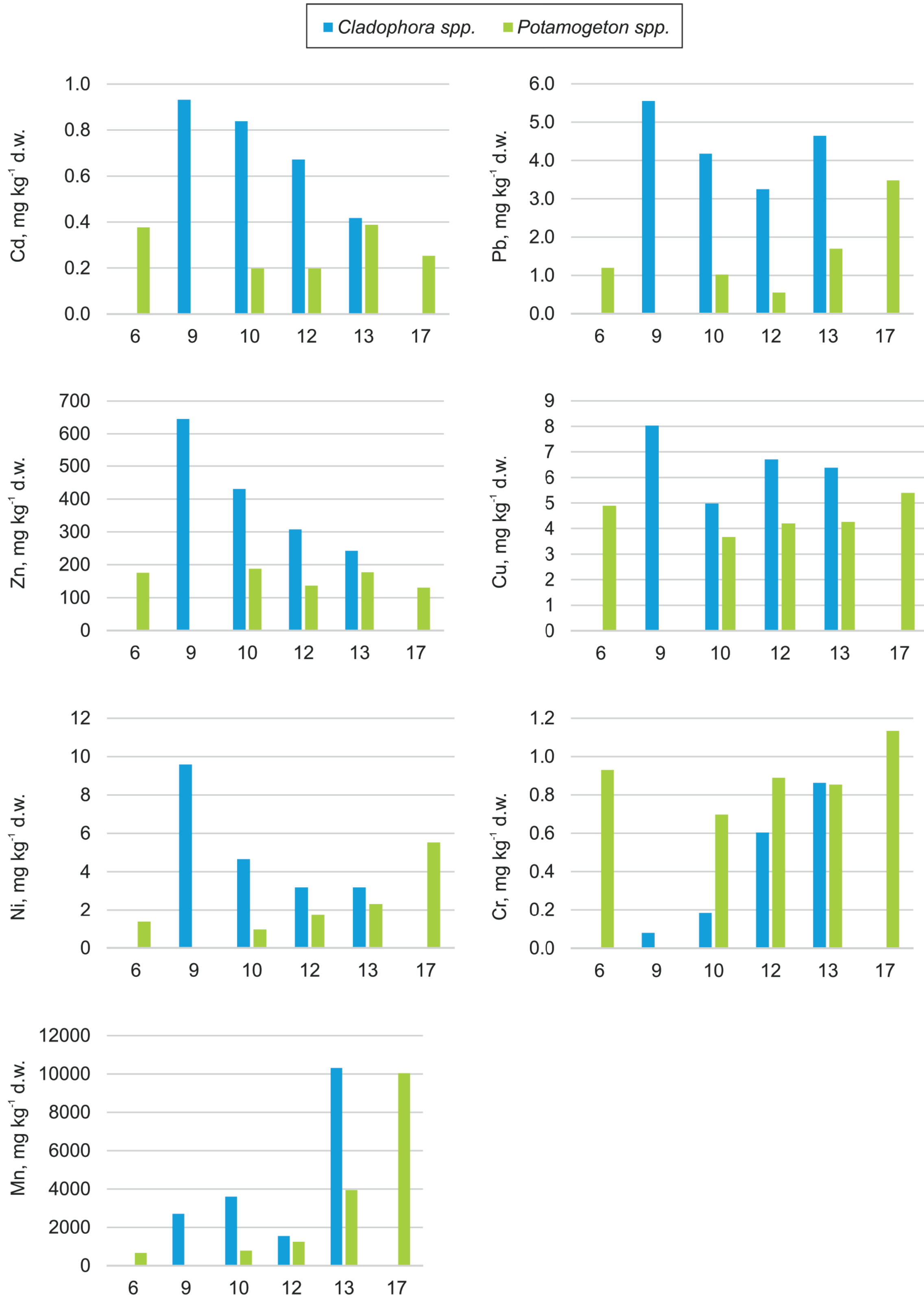

Figure 2

Average concentrations of heavy metals in two groups of the most abundant macrophytes in the Vistula Lagoon. Numbers refer to the numbers of the sampling locations

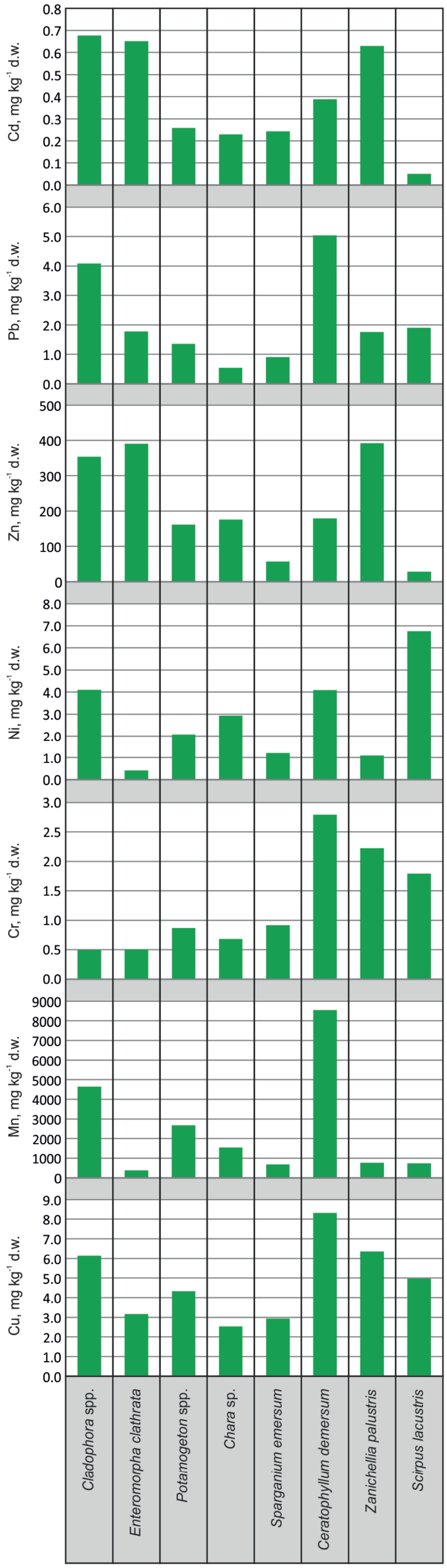

Figure 3

Average concentrations of heavy metals in macrophytes collected in the Vistula Lagoon

important role. The previous research conducted in the marine environment showed that the efficiency of the foliar uptake is greater when the exchange surface is well developed (Zalewska 2015). In *Cladophora* spp. and *E. clathrata*, concentrations of Cd, Zn and Cr were at a very similar level, while concentrations of Pb, Ni, Mn and Cu were higher in *Cladophora* spp. (Fig. 3). Cd and Zn concentrations in vascular plants were slightly lower than those recorded in green algae, respectively 0.2–0.4 mg kg^{-1} d.w. and 30–180 mg kg^{-1} d.w., with the exception of *Z. palustris*, for which these values were similar. Pb concentrations in vascular plants were in the range of 1–2 mg kg^{-1} d.w. with the exception of *C. demersum* (5 mg kg^{-1} d.w.), where the content was slightly higher than in *Cladophora* spp. (4 mg kg^{-1} d.w.). The highest Ni concentration reached 6.8 mg kg^{-1} d.w. and was characteristic for *S. lacustris*, while for other species it was below 4 mg kg^{-1} d.w. The highest mean concentration of Mn was recorded in *C. demersum* tissues (8500 mg kg^{-1} d.w.) and at the same time this species occurred at location 17, where very high concentration of Mn was also observed in *P. pectinatus*. Considerably higher concentrations of Cr compared to green algae were recorded in three species of vascular plants: *C. demersum*, *Z. palustris*, *S. lacustris*, for which also elevated levels of Cu were detected.

Concentrations of Cd, Pb, Cu and Ni in *Cladophora* spp. and *Potamogeton* spp. from the Vistula Lagoon were at the levels comparable to the values characteristic for the same groups of plants in Puck Bay (Zalewska 2015). However, noticeable differences were found in the concentrations of Zn, which in green algae were 19.5 mg kg^{-1} d.w. in Puck Bay and 350 mg kg^{-1} d.w. in the Vistula Lagoon, whereas in *Potamogeton* spp. – 44 mg kg^{-1} d.w. and 162 mg kg^{-1} d.w., respectively. The markedly elevated levels in both taxa from the Vistula Lagoon were also significantly higher in the case of Mn, with average concentrations of 4650 mg kg^{-1} d.w and 2677 mg kg^{-1} d.w. in *Cladophora* spp. and *Potamogeton* spp., respectively, whereas in Puck Bay they remained at the level of 100 mg kg^{-1} d.w. and 285 mg kg^{-1} d.w.

The ranges of heavy metal concentrations in macrophytes collected in the Vistula Lagoon in 2011 (Cd: 0.1–1.2 mg kg^{-1} d.w.; Pb: 0.1–5.7 mg kg^{-1} d.w.; Zn: 29–653 mg kg^{-1} d.w.; Cu: 1.2–12.0 mg kg^{-1} d.w.; Ni: 0.1–14.4 mg kg^{-1} d.w.; Cr: 0.1–3.2 mg kg^{-1} d.w.) are comparable to the concentration ranges of the same metals determined in the vegetation from the Puck Lagoon in 1989 and 1990 (Cd: 0.4–8.0 mg kg^{-1} d.w.; Pb: 0.2–7.5 mg kg^{-1} d.w.; Zn: 170–910 mg kg^{-1} d.w.; Cu: 5.5–14.0 mg kg^{-1} d.w.; Ni: 1.3–11.0 mg kg^{-1} d.w.; Cr: 0.7–6.7 mg kg^{-1} d.w.; Mn: 100–1400 mg kg^{-1} d.w.; Kruk-Dowgiałło & Pempkowiak 1995). The exception is Mn: 380–10035 mg kg^{-1} d.w., the concentration of which in the Puck Lagoon remained in the range of 100–1400 mg kg^{-1} d.w.

Based on the concentrations of heavy metals in water of the Vistula Lagoon (data from the State Environmental Monitoring in Poland in 2012, 2014 and 2016), the concentration ratios (*CR*) were determined by referring the concentrations of heavy metals in a particular taxon to the values in water (eq. 1, Table 3).

$$CR(l\ kg^{-1}\ d.w.) = \frac{metal\ concentration\ in\ plant\ (\mathrm{mg\ kg^{-1}\ d.w.})}{metal\ concentration\ in\ water\ (\mathrm{mg\ l^{-1}})} \qquad (1)$$

As in the case of aquatic vegetation, the elements are accumulated directly from water through thalli or the root system, values of concentration ratios express the bioaccumulation ability of selected species. In the case of Cd and Zn in most of the studied species, the CR values were at the level of 10^4; only for *S. lacustris* the CR values were 2×10^3 and 4.8×10^3, respectively. The CR values for Pb were in the order of 10^3, with a maximum value of 1×10^4 characteristic for *C. demersum*. The CR values for Cu varied between 7.9×10^2 and 1.9×10^3 and their range was similar to that observed for Ni (1.4×10^2–2.3×10^3). In the case of Cr, all CR values were at the level of 10^3. No significant differences were found in CR values among particular taxa, given that they represent both green algae and vascular plants. In the case of Cd, Pb and Ni, the CR values were similar to those noted in the waters of the Baltic Sea (Zalewska 2015). In addition, the CR values for the vegetation from the Vistula Lagoon were more comparable with the values recommended for the marine environment than for the freshwater environment (IAEA 2014). The average CR values recommended for predicting the transfer of metals to macroalgae in the marine environment were at the levels of 8–9 × 10^2 for Cd, Pb, Ni and 1.4×10^3 for Cu, while for the freshwater environment they were one order of magnitude lower, except for Cd (IAEA 2014). This can lead to the conclusion that in terms of bioaccumulation of heavy metals in vegetation, the Vistula Lagoon environment may be more similar to the southern Baltic environment than to freshwater reservoirs, although the average salinity in the Vistula Lagoon is 3, i.e. twice as low as in the Gulf of Gdańsk. Taking this into account, it can be expected that the bioaccumulation efficiency should be higher in the Vistula Lagoon, because it theoretically increases with lower salinity. However, this relationship was not observed.

The content of cesium ^{137}Cs in the vegetation was relatively low, often not exceeding the detection limit of the applied method (Table 2). The ^{137}Cs activity in *Cladophora* spp. remained in the range of 3–6.4 Bq kg^{-1} d.w., while in the same year (2011) the values characteristic for *C. glomerata* in the Orłowo Cliff area (Puck Bay) varied from 11 to 33 Bq kg^{-1} d.w. (Zalewska 2015). Concentrations of ^{137}Cs at the level of 7 Bq kg^{-1} d.w. were observed in *Potamogeton* spp. and *C. demersum* in Elbląg Bay (location17).

Concentrations of heavy metals in sediments

Based on changes in $^{210}Pb_{ex}$ activity concentrations along the depth profile in bottom sediments (Fig. 4), a

Table 3

Concentration ratios for heavy metals in macrophytes studied in the Vistula lagoon in 2011

Concentration Ratio (*CR*) l kg^{-1} d.w.	Cd	Pb	Zn	Cu	Ni	Cr	Hg
Cladophora spp.	2.7×10^4	8.2×10^3	6.0×10^4	1.9×10^3	1.4×10^3	1.0×10^3	-
Enteromorpha clathrata	2.6×10^4	3.6×10^3	6.6×10^4	9.9×10^2	1.4×10^2	1.0×10^3	-
Potamogeton spp.	1.0×10^4	2.7×10^3	2.7×10^4	1.4×10^3	6.9×10^2	1.7×10^3	-
Chara spp.	9.2×10^3	1.1×10^3	3.0×10^4	7.9×10^2	9.8×10^2	1.4×10^3	-
Sparganium emersum	9.7×10^3	1.8×10^3	9.6×10^3	9.2×10^2	4.0×10^2	1.8×10^3	-
Ceratophyllum demersum	1.6×10^4	1.0×10^4	3.0×10^4	2.6×10^3	1.4×10^3	5.6×10^3	-
Zanichellia palustris	2.5×10^4	3.5×10^3	6.6×10^4	2.0×10^3	3.6×10^2	4.4×10^3	-
Scirpus lacustris	2.0×10^3	3.8×10^3	4.8×10^3	1.6×10^3	2.3×10^3	3.6×10^3	-
Concentration in water* (mg l^{-1})	2.5×10^{-5}	5.0×10^{-4}	5.9×10^{-3}	3.2×10^{-3}	3.0×10^{-3}	5.0×10^{-4}	3.1×10^{-6}
Distribution Coefficient K_d (l kg^{-1} d.w.)	2.12×10^4	2.46×10^4	1.60×10^4	5.25×10^3	-	-	3.87×10^4

*data from the State Environmental Monitoring in Poland in 2012, 2014 and 2016

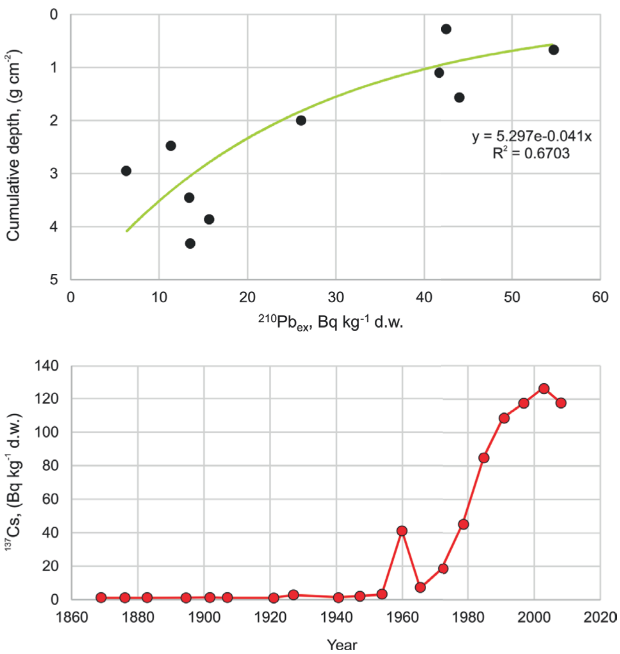

Figure 4

$^{210}Pb_{ex}$ concentrations along the depth profile in bottom sediments and deposition history of ^{137}Cs in the sediments of the Vistula Lagoon

linear sedimentation rate was determined at 3.3 mm y^{-1} applying the CF:CS model. This value is slightly higher than that observed in the Gulf of Gdańsk (1.8 mm y^{-1}; Zalewska et al. 2015) and may be related to the difference in the weight of matter suspended in the waters of both basins. Such a linear sedimentation rate was reflected in the mass accumulation rate (0.074 g cm^{-2} y^{-1}), which was also higher than that observed in the Gulf of Gdańsk (0.032 g cm^{-2} y^{-1}; Zalewska et al. 2015).

The determination of the age of sediment layers, verified by ^{137}Cs distribution analysis in the vertical profile (Fig. 4), made it possible to trace changes in metal concentrations in the historical aspect. The deepest layer at a depth of 30 cm was formed around 1920. The concentrations of metals in this layer were as follows: Cd – 0.44 mg kg^{-1} d.w., Pb – 19.9 mg kg^{-1} d.w., Hg – 0.08 mg kg^{-1} d.w., Zn – 103 mg kg^{-1} d.w. and Cu – 27.4 mg kg^{-1} d.w. (Fig. 5). Metal concentrations were normalized to 5% Al content in order to avoid discrepancies resulting from differences in the composition of sediments. The content of Al in the layers of sediments ranged from 4.29% to 5.33%, with slightly higher concentrations characteristic for the deepest layers. Concentrations of Cd and Hg increased from 1923 to 1968 by 24% and 19%, respectively. Subsequently, a significant increase was observed until the maximum concentrations were reached in 1988 for Cd (0.75 mg kg^{-1} d.w.) and in 2000 for Hg (0.14 mg kg^{-1} d.w.). In the following years, the Cd concentration declined to 0.61 mg kg^{-1} d.w. in 2011, while the Hg concentration was relatively stable. A slightly different pattern was found in the content of Pb, Cu and Zn, which remained practically unchangeable in the period of 1923–1968. After 1968, the content of Zn insignificantly increased to the maximum value of 120 mg kg^{-1} d.w. in 1988, while in the following years the concentration was at a similar level. In the case of Cu, a decline in its concentration was observed until 2000. Later on, the Cu content reached a fairly stable value of ca. 19 mg kg^{-1} d.w. A well-marked drop in Pb concentrations occurred after 1987 and was related to the introduction of unleaded petrol in Poland in 1986. In 2011, the concentration of lead was 14.2 mg kg^{-1} d.w.

Taking into account the age of particular sediment layers, the content of heavy metals in the layer dated at 1994 was compared with concentrations of metals in the surface layer of sediments collected in 1995 in the Vistula Lagoon (Szefer et al. 1999). For the purpose of comparison, data from the location nearest to the location under study were selected. Concentrations of Cd, Zn and Cu were similar. Their content was as follows: Cd – 0.70 mg kg^{-1} d.w. (this study) and 0.69 mg kg^{-1} d.w. (Szefer et al. 1999), Zn – 112 mg kg^{-1} d.w.

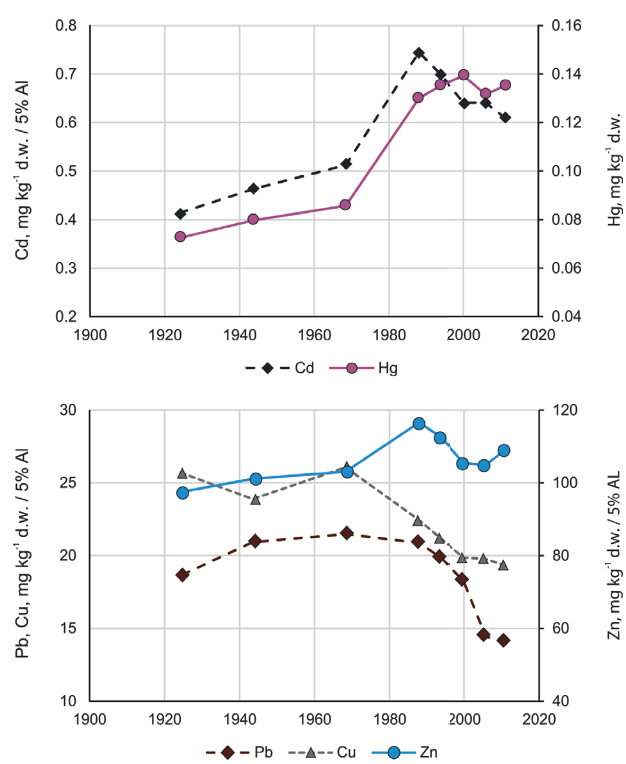

Figure 5

Deposition history of selected heavy metals in the sediments of the Vistula Lagoon and 114 mg kg^{-1} d.w., Cu – 21.2 mg kg^{-1} d.w. and 20.6 mg kg^{-1} d.w. A significant difference was found in the concentration of lead – 19.9 mg kg^{-1} d.w. in the sediment layer dated at 1994, while 30.2 mg kg^{-1} d.w. according to Szefer et al. (1999). Very similar values were recorded in another study conducted by Glasby et al. (1998) in the Vistula Lagoon. They showed that metal concentrations were as follows: Cd – 0.9 mg kg^{-1} d.w., Pb – 33 mg kg^{-1} d.w., Zn – 111 mg kg^{-1} d.w. and Cu – 23 mg kg^{-1} d.w.

Based on the values of metal concentrations in the sediments and waters of the Vistula Lagoon, distribution coefficients K_d (eq. 2, Table 3) were determined and compared with the values recommended by the IAEA (2004) for modelling and forecasting. The K_d values for Cd, Pb, Zn and Hg were similar and ranged from 1.6 × 10^4 to 3.9 × 10^4. Only in the case of Cu, the K_d value was 5.2 × 10^3. The K_d values calculated for Cd and Zn were close to the literature values of 3 × 10^4 and 7 × 10^4 (IAEA 2004). In the case of Pb, the literature value was an order of magnitude higher (1 × 10^5) as opposed to Hg, for which K_d was an order of magnitude lower than that calculated for the Vistula Lagoon (4 × 10^3).

$$K_d(l\ kg^{-1}) = \frac{\textit{metal concentration in sediment}\ (\text{mg kg}^{-1}\ \text{d.w.})}{\textit{metal concentration in water}\ (\text{mg l}^{-1})} \quad (2)$$

Status of the Vistula Lagoon environment

The Vistula Lagoon, a basin very sensitive to human pressure, is one of the transitional water bodies in the Polish sector of the Baltic Sea, whose environmental status is assessed in accordance with the recommendations of the Water Framework Directive and the Marine Strategy Framework Directive.

The environmental status of the Vistula Lagoon was assessed based on the obtained data on heavy metal concentrations in macrophytes and bottom sediments. The concentrations were referred to the threshold values defining the borderline between good and inadequate status, which basically means that concentrations of heavy metals below the specified value should not negatively affect the functioning of the ecosystem.

Taking into account the average concentrations of Cd (0.4 mg kg^{-1} d.w.), Pb (2.6 mg kg^{-1} d.w.) and Ni (3.2 mg kg^{-1} d.w.) calculated for all the studied macrophyte taxa and environmental quality standards (EQS) determined for plants (Pb – 26 mg kg^{-1} d.w., Cd –33 mg kg^{-1} d.w., Ni – 32 mg kg^{-1} d.w.; Zalewska & Danowska 2017), the environmental status of the Vistula Lagoon was assessed as good. In order to assess the status based on heavy metal levels in sediments, the most recent data were used, i.e. concentrations detected in the first sediment layer: Cd – 0.53 mg kg^{-1} d.w., Pb – 12.3 mg kg^{-1} d.w., Hg – 0.12 mg kg^{-1} d.w., Zn – 94.2 mg kg^{-1} d.w. Comparison of these data with the threshold values, determined through geochronological analyses carried out in the Gulf of Gdańsk (Cd – 0.3 mg kg^{-1} d.w., Pb – 30 mg kg^{-1} d.w., Hg – 0.05 mg kg^{-1} d.w., Zn – 110 mg kg^{-1} d.w.; Zalewska et al. 2015), revealed that only Pb and Zn indicated good environmental status of the sediments, while Hg and Cd indicated that the environmental status of the Vistula Lagoon is bad.

Conclusions

- The study showed no significant differences in the concentrations of heavy metals and ^{137}Cs between the taxa of macrophytes in the Vistula Lagoon. The concentrations of heavy metals in the macrophyte taxa varied in the following ranges: Cd – 0.1–0.7 mg kg^{-1} d.w., Pb – 0.5–5.0 mg kg^{-1} d.w.; Zn – 29–390 mg kg^{-1} d.w.; Cu – 2.5–8.3 mg kg^{-1} d.w.; Ni – 0.4–6.8 mg kg^{-1} d.w.; Cr – 0.5–2.8 mg kg^{-1} d.w.; Mn – 380–8500 mg kg^{-1} d.w. The lowest activity concentration of ^{137}Cs was below the determination limit of 0.7 Bq kg^{-1} d.w. and the highest one reached 7.9 Bq kg^{-1} d.w.

- No effect of rivers flowing into the Vistula Lagoon on the concentrations of heavy metals in the area was observed.

- The linear sedimentation rate in the Vistula Lagoon was 3.3 mm y^{-1}.

- Since the 1990s, a decline or stable state of heavy metal concentrations in bottom sediments has been observed, reflecting changes in the environment of the Vistula Lagoon.

- The environmental status of the Vistula Lagoon can be considered good in terms of heavy metal contamination, except for Hg and Cd, whose concentrations in the sediments slightly exceed the threshold value.

- Concentration ratios (CR) calculated for macrophytes and distribution coefficients (K_d) determined for heavy metals in the bottom sediments can serve as a baseline for predicting the future distribution of heavy metals in these two aspects of the Vistula Lagoon ecosystem, especially when considering the plans of connecting the Polish part of the lagoon with the Baltic Sea.

- The results presented in the paper can serve as a baseline for assessing changes in the environmental status of the Vistula Lagoon, which may occur as a result of building a new navigable canal through the Vistula Spit.

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