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ORIGINAL PAPER

DISTRIBUTION AND POTENTIAL RISK ASSESSMENT OF HEAVY METALS AND ARSENIC IN SEDIMENTS OF A DAM RESERVOIR: A CASE STUDY OF THE ŁOJE RETENTION RESERVOIR, NE POLAND*

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Abstract

The objective of this study has been to determine the potential toxicological effect of heavy metals accumulated in bottom sediments of a lowland retention reservoir located in north-eastern Poland. For this purpose the ecotoxicological (TEL, PEL) criteria were selected to quantify sediment contamination. The potential ecological risk was assessed. The analyzed reservoir was characterized by significant spatial variations in heavy metals (Cd, Cr, Cu, Ni, Pb, Zn) and As related to organic carbon ($C_{org.}$) concentrations in bottom sediments during its 30-year-long life. Thus, the highest organic matter content and heavy metals accumulation was reported in the reservoir's riverine and transitional zones, whereas predominance of mineral fractions did not favor metal content in the lacustrine zone. One kilogram of sediment dry matter contained 39.9 mg Zn, 20.5 mg Pb, 11.0 mg Cr, 9.1 mg Ni, 7.2 mg Cu, 1.29 mg As and 0.39 mg Cd on average. Nonmetric multidimentional scaling (NMS) revealed distinct elemental fingerprints in each sampling site, while two-way hierarchical agglomerative cluster analysis (TWHCA) provided insight to metal association and pollution distribution. It emerged that the riverine and transitional zones of the reservoir underwent progressively increasing contamination of the sediments, mainly due to a high content of Cd, Cr, Cu, Ni and Pb, although only three of these metals, i.e. Cd, Ni and Pb, reached levels that could produce an adverse effect on aquatic organisms. The application of geochemical and ecotoxicological indices is a helpful tool for the risk assessment of environmental pollution, as well as a tool supporting decision-making for water source security.

Keywords: dam reservoir, bottom sediments, heavy metals, geoaccumulation index, potential risk assessment

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INTRODUCTION

Bottom sediments constitute an integral part of aquatic ecosystems as they provide a habitat for numerous aquatic organisms (Brils 2008) as well as they are a storage site for various kinds of substances, including heavy metals (Glińska-Lewczuk et al. 2009, Smal et al. 2015). Aquatic ecosystems act as both carriers and absorbers of contaminating substances, and bottom sediments are potent indicators of environmental pollution (Vandecasteel et al. 2004, Brils 2008). They play an important role in assessment of water ecosystems and identification of substances that reach such water bodies (Wang et al. 2012), providing a record of catchment inputs into aquatic ecosystems (Karlik, Szpakowska 2001).

In contrast to urbanized and industrial areas, sediments from reservoirs located at the agriculture-forest or non-industrial catchment contain metals at the geochemical background level or are only slightly contaminated (Bojakowska, Sokołowska 1998). Nevertheless, fertilizers containing heavy metals, including As, Cd, Hg, Ni, Pb and Zn, as well as fertilization regimes in rural areas can significantly affect the soil, water quality, and bottom sediments (Karlik, Szpakowska 2001, Szejba et al. 2016). Heavy metals and other pollutants can be immobilized in bottom sediments for very long periods of time due to a long biological half-life and high potential for accumulation, which is why they pose a considerable environmental threat (Klavins et al. 2000). Binding of heavy metals by sediments depends on their grain-size distribution (clay fraction), organic matter content and also Fe, Al and Mn oxides (Çevik et al. 2009, Glińska-Lewczuk et al. 2009, Masoud et al. 2011, Yu et al. 2012, Venkatramanan et al. 2015, Kuriata-Potasznik et al. 2016). Carbonate fractions have a high capacity for binding cadmium (Korfali, Davies 2004), lead (Loska et al. 2003, Korfali, Davies 2004) and copper (Pertsmeli, Voutsa 2007). Metals bound by co-precipitation with hydrated oxides of iron are less readily released. The above also applies to metals bound to organic matter which can be brought to water reservoirs in the decomposition process. According to Akhurst et al. (2012) concentration of particulate-bound elements results from the hydrodynamic conditions governing the sediment deposition. Changes in water flow velocity has been reported as a key factor contributing to varied heavy metal contents in sediments of dam reservoirs (Gierszewski 2008). Nevertheless, few studies have focused on the spatial distribution and pollution assessment of heavy metals in sediments in man-made water reservoirs.

Nowadays assessment of the chemical quality of bottom sediments indicates both the state of the aquatic environment as well as it serves as the basis for development of sediment management strategies in the environment (Dmitruk et al. 2013). The need of evaluation of bottom sediment quality in terms of pollution with heavy metals resulted in the development of diverse geochemical and ecotoxicological criteria. Apart of geochemical classification by Bojakowska, Sokołowska (1998), sediment quality is evaluated using the index of geoaccumulation and the enrichment factor (EF) (Çevik et al. 2009, Wang et al. 2012). Among ecotoxicological criteria, such indices as the Threshold Effects Level (TEL), and Probable Effects Level (PEL) are applied based on a particular contaminant's impact on aquatic organisms (Smith et al. 1996).

The objective of this study was (i) to investigate the contamination levels and spatial distribution of heavy metals in bottom sediment of a retention reservoir in conditions of mid-eastern Europe (NE Poland); (ii) to assess bottom sediment quality, considering metal concentration according to the geochemical and ecotoxicological criteria; (iii) to assess the potential ecological risk.

MATERIAL AND METHODS

Bottom sediments were sampled from the Łoje retention reservoir with an area of 4.56 ha and capacity of $5 \cdot 10^4$ m^3. The reservoir is situated in the North Masovian Plain in NE Poland. It is one of the many reservoirs developed in Poland under the framework of the 'small retention' programme in the 1970s. The Łoje reservoir was created in 1975, by damming water flowing in the Łojówka stream and flooding a waterlogged meadow. The average water inflow to the reservoir amounts to 9.0 dm^3 s^{-1} (0.1 dm^3 s^{-1} - 21.0 dm^3 s^{-1}). The catchment basin of the Łojówka stream has an area of 6.0 km^2, and it is occupied by grasslands and arable land. The reservoir's direct catchment has an area of 1.2 km^2, 80% of which is covered by forests. The remaining 20% is composed of extensively used meadows in the upper part of the reservoir.

Three zones: I – upper part, the riverine zone overgrown by rush plants, II and III – middle part, the transitional zone with submerged vegetation, and IV – lower part, the lacustrine zone free from aquatic plants can be distinguished in the Łoje reservoir (Figure 1). The reservoir is shallow on the side of the riverine zone (0.8 m). The transitional zone has an average depth of 1.5 m, whereas in the lacustrine zone the maximum depth of 5 m was identified in the area of the dam. Water is released via an outlet tower equipped with two stoplogs, which reach to the bottom of the reservoir.

Sediment samples were collected from four transect, subject to landform at the bottom of the reservoir and sediment thickness. Information on the lithology and thickness of sediments contained in the publication Cymes and Szymczyk (2006). The sediment material was taken in triplicates from 10 sampling sites (transects I and II – 3 samples each; transects III and IV – 2 samples each). To obtain undisturbed core samples we applied a sampler of the Beeker type (Eijkelkamp). At each sampling point, 3-4 hydrated sediment cores were taken from the bottom. The samples were then air-dried, passed through a 2 mm sieve, homogenized in an agate mortar, and subjected further laboratory analyses. Organic carbon content was calculated on

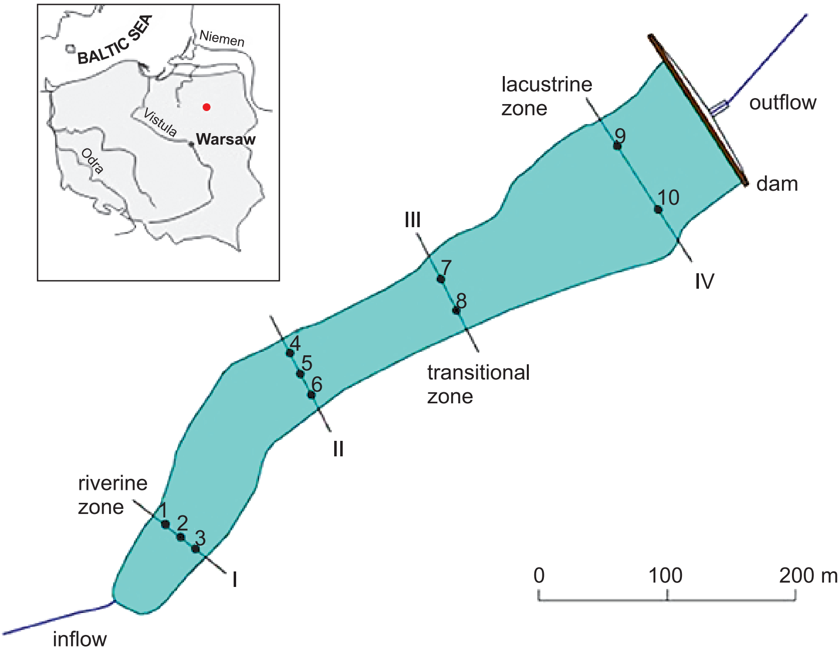

Fig. 1. Distribution of sampling sites in the Łoje Reservoir: I-IV – transects (I – riverine zone, II and III – transitional zone, IV – lacustrine zone); 1-10 – sediment sampling sites

the basis of the determined weight organic matter. A sample of sediments was dried at 105°C, cooled and weighed. For 6 h then was burned at 600°C, cooled and weighed. The content of the organic matter is calculated using the equation:

$$Wso = \frac{m_1 - m_2}{m_1 - m_0} \cdot 100,$$

where: m_0 – weight dishes,

m_1 – weight dishes and the sample after drying at 105°C,

m_2 – weight dishes and samples after burning at 600°C.

The percentage of organic carbon determined from the equation:

$$C_{org.} = \frac{Wso}{1.724}.$$

The limit of detection for organic carbon was 0.01 mg kg^{-1}.

The concentrations of heavy metals – cadmium, chromium, copper, nickel, lead, zinc, and arsenic – were measured with the use of an Atomic Absorption Spectrophotometer (AAS-6800 Schimadzu) and the air-acetylene flame. The BGC-D2 (deuterium background correction) was used. Internal standards were applied for analytical quality control. A peak search was performed in the vicinity of the expected analytical lines: Cr – 357.9 nm, Cu – 324.8 nm, Ni – 232.0 nm, Cd – 228.8 nm, Pb – 217.0 nm, Zn – 139 nm and As – 193.7 nm. We used the certified reference material (Sewage Sludge Certified Reference Material specified by the ISO Guides 34, 35 and ISO 17025, ACLASS Company). The limit of detection for heavy metals was: Cr – 0.1 mg kg^{-1}, Cu – 0.1 mg kg^{-1}, Ni – 0.005 mg kg^{-1}, Cd – 0.003 mg kg^{-1}, Pb – 0.1 mg kg^{-1}, Zn – 0.1 mg kg^{-1} and As – 0.003 mg kg^{-1}.

The sediment content of an element is assessed relative to its average content in parent material and expressed on a seven-grade scale. The potential effect of bottom sediment concentrations on the Łoje reservoir's ecosystem was evaluated with the ecotoxicological assessment criteria, by calculating the TEL (Threshold Effects Level), PEL (Probable Effects Level) indices (Smith et al. 1996). We applied also the potential ecological risk index (PER), originally proposed by Håkanson (1980). The sum of the potential risk of individual heavy metal (RI) was calculated with the use of the following formulas:

$$C_f^i = \frac{C_D^i}{C_B^i},$$

$$E_r^i = T_r^i \cdot C_f^i,$$

$$RI = \sum_{i=1}^{m} E_r^i,$$

where: C_f^i – is the contamination factor,
C_D^i – is the present concentration of heavy metals in sediments,
C_B^i – is the pre-industrial record of heavy metal concentration in sediments,
E_r^i – is the potential risk of individual heavy metal,
T_r^i – is the toxic-response factor for a given heavy metal,
RI – is the sum of the potential risk of individual heavy metal.

Based on the Håkanson's approach, the toxic-response factors for Cr, Cu, Zn, Pb, Cd, and As are 2, 5, 1, 5, 30 and 10, respectively. Håkanson defined five categories of E_r^i, and four degrees of potential ecological risk, as shown in Table 1.

Multiresponse Permutation Procedures (MRPP, PC-ORD 6.0) were used to describe spatial distribution of heavy metal contents across the studied dam reservoir. This procedure is useful for testing differences among groups of parameters in non-normal data distribution (McCune, Grace 2002). Significantly different groups determined by the MRPP were further analyzed using the multivariate analyses of Nonmetric Multidimensional Scaling – NMS (McCune, Mefford 2011). MRPPs only state that a significant difference is present, while NMS is useful for visually representing these differences. NMS as an unconstrained ordination, was used in this study to reveal variance structure among metal species, irrespective of any environmental

Table 1

Indices and corresponding degrees of potential ecological risk (HÅKANSON 1980)

E_r^i value	Grades of ecological risk of a single metal	RI value	Grades of potential ecological risk of the environment
$E_r^i < 40$	low risk	RI < 150	low risk
$40 \leq E_r^i \leq 80$	moderate risk	150 ≤ RI ≤ 300	moderate risk
$80 \leq E_r^i \leq 160$	considerable risk	300 ≤ RI ≤ 600	considerable risk
$160 \leq E_r^i \leq 320$	high risk	RI ≥ 600	very high risk
$E_r^i \geq 320$	very high risk	-	-

gradients. NMS is appropriate for sparse data matrices because it uses rank-distances to avoid assumptions of linearity (MCCUNE, GRACE 2002).

The pattern and order in the site data and heavy metals content were performed by two-way hierarchical cluster analysis (TWHCA) with the use of Ward's Linkage method and Euclidean (Pythagorean) distance measure. The data were log(x+1) transformed prior to analyses. All the statistical procedures have been carried out in PC-ORD 6.0.

RESULTS AND DISCUSSION

The water velocity decreased at the reservoir's inlet, which created favorable conditions for deposition of the transported material. Mineral and organic matter supplied was more readily deposited in the upper part of the reservoir. The sediment layer thickness gradually decreased towards the spillway (transects III and IV).

The studied metals accumulated in bottom sediments showed significant spatial distribution across the dam reservoir (Table 2) as well as the correlations of the particular elements. Subject to the thickness and type of bottom sediments, one kilogram of sediment dry matter contained 0.11 - 0.79 mg Cd, 3.4 - 33.0 mg Cr, 1.2 - 19.0 mg Cu, 2.1 - 22.7 mg Ni, 6.3 41.0 mg Pb, 17.6 - 127.2 mg Zn and 0.82 - 3.04 mg As. One kilogram of sediment dry matter contained 9.71 g of organic carbon on average.

MRPP allowed to identify the differences in the heavy metal content in the three distinguished zones: riverine, transitional and lacustrine (Table 3). MRPP analysis revealed significant differences in metal composition concentrations between samples in transects combined. The pairwise comparisons of heavy metals concentrations in bottom sediments within the three zones showed no statistically significant differences between riverine and transitional zones (A = -0.09; p =0.219). However, the comparison of riverine vs. lacustrine zone as well as transitional versus lacustrine zone showed statis-

Table 2

Properties and concentrations of heavy metals in the bottom sediments of the Łoje Reservoir

Index	Riverine zone				Transitional zone							Lacustrine zone			Reservoir (average)
	transect I				transect II				transect III			transect IV			
	1	2	3	average	4	5	6	average	7	8	average	9	10	average	
Dry matter (%)	25.20	20.76	23.19	23.05	14.75	19.18	16.60	16.84	35.52	35.20	35.36	77.85	79.91	78.88	38.53
Reaction (pH)	7.16	6.76	6.83	-	5.69	6.38	5.69	-	7.46	6.58	-	7.70	7.16	-	-
Organic carbon $C_{org.}$ (%)	1.72	23.75	6.62	10.70	21.48	11.00	2.31	11.60	4.00	29.01	16.51	0.06	0.03	0.05	9.71
Cd (mg kg^{-1})	0.45	0.61	0.18	0.41	0.50	0.51	0.16	0.39	0.38	0.79	0.59	0.11	0.26	0.19	0.39
Cr (mg kg^{-1})	11.7	16.0	6.6	11.4	33.0	14.7	8.5	18.7	11.7	9.5	10.6	3.4	3.4	3.4	11.0
Cu (mg kg^{-1})	19.0	11.1	4.1	11.4	6.0	10.9	4.0	7.0	10.2	7.8	9.0	1.3	1.2	1.3	7.2
Ni (mg kg^{-1})	22.7	15.0	4.0	13.9	10.0	12.7	4.3	9.0	13.0	9.5	11.3	2.3	2.1	2.2	9.1
Pb (mg kg^{-1})	17.7	19.5	11.6	16.3	16.0	36.7	17.5	23.4	30.5	41.0	35.8	7.2	6.3	6.8	20.5
Zn (mg kg^{-1})	88.0	63.3	13.4	54.9	18.2	72.0	27.4	39.2	62.9	53.1	58.0	6.5	8.7	7.6	39.9
As (mg kg^{-1})	1.68	1.36	0.84	1.30	3.04	1.02	0.82	1.63	1.82	0.84	1.33	0.96	0.88	0.92	1.29

Table 3

MRPP results testing for differences between the three transects (pairwise comparisons)
Denotations: T - test statistic, A - a chance-corrected within-group agreement;
p values were not corrected for multiple comparisons

Zones compared	T	A	*p*
Riverine vs. transitional	0.884	-0.090	0.219
Riverine vs. lacustrine	-1.510	0.226	* <0.001
Transitional vs. lacustrine	-1.974	0.219	*0.048

tically significant differences (A = 0.22, $p < 0.001$; A = 0.22; $p = 0.048$, respectively).

Strong differences in metal composition among all 10 sampling sites across Łoje reservoir were indicated by NMS ordination (Figure 2a), final stress = 0.19. The degree of separation of metals varied among transects and zones. Clear separation was observed between lacustrine versus riverine and transitional zones, while there was some overlap between the riverine and transitional zones. Organic carbon content, particularly in the riverine and transitional zones (compare with Figure 2b), showed a significant positive correlation with the concentration of Cu, Zn and Pb in bottom sediments.

The analysis of two-way Hierarchical Cluster Analysis (Figure 2b) showed that heavy metal concentrations in lacustrine zone were lower than those in riverine and transitional zones. Significantly higher concentrations of heavy metals created clusters in the riverine and transitional zones, which were characterized by a predominance of thick organic deposits (C_{org}). The highest Cu, Ni and Zn concentrations were observed in the riverine zone, which suggests that those elements were intensively absorbed by rush plants growing in this part of the reservoir. The highest levels of Cd, Cr, Pb and As were observed in the transitional zone, colonized by submerged vegetation (transects II and III). The lacustrine zone (transect IV) created a cluster consisted of the sites 9 and 10, characterized by the predominance of mineral deposits and significantly lower levels of organic compounds and heavy metals in bottom sediments. The above results suggest that plants growing and organic debris accumulated in the upper part of the reservoir significantly contributed to the elimination of toxic metals supplied to the reservoir from the catchment area. The observed spatial distribution of heavy metals can be attributed mainly to differences in the thickness and type of bottom sediments (organic vs. mineral) and the degree of deposit hydration.

The Łoje reservoir is surrounded by an agricultural and forest catchment, hence the contamination of its sediments with heavy metals, seems relatively small. Concentrations of the analyzed heavy metals typically fell within the lowest to average amounts detected in other reservoirs located in Poland (Table 4), and beyond: in Hungary (Nguyen et al. 2005), the USA (Gewurtz et al. 2007), or China (Jiang et al. 2012).

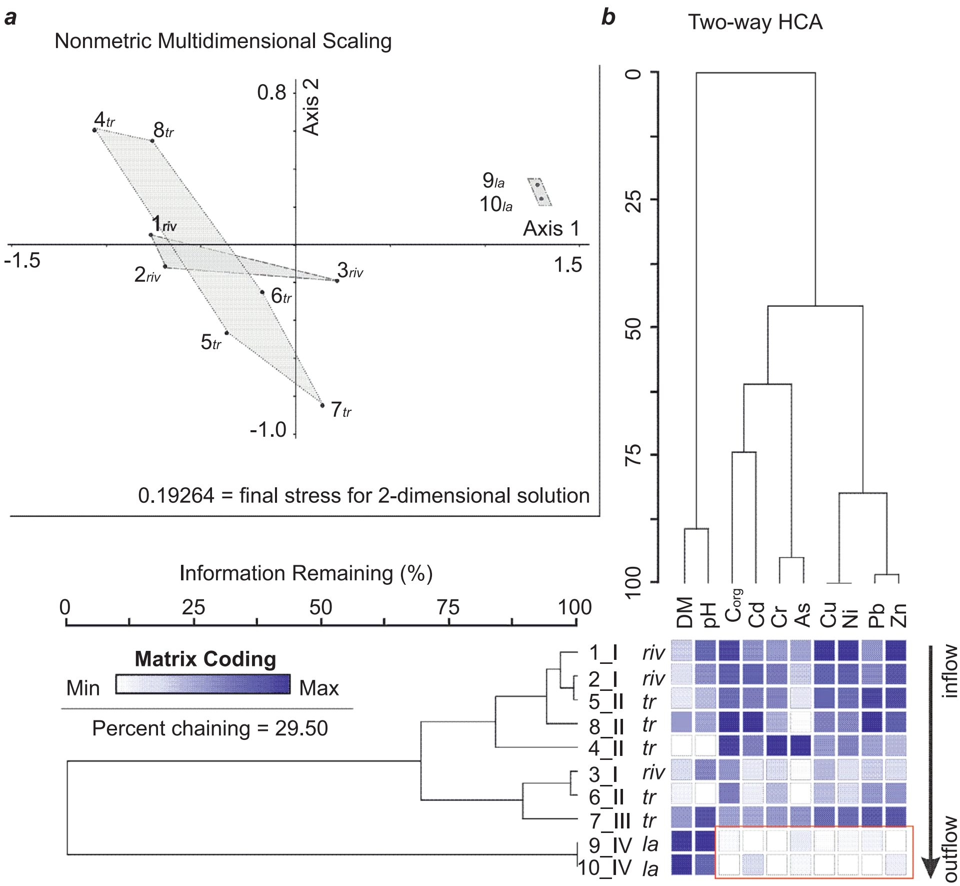

Fig. 2. (*a*) Non-Metric Multidimensional Scaling of sites based on metal contents in bottom sediments. (*b*) Two-Way Hierarchical Cluster Analysis. A distinct drop in heavy metals and C_{org} contents in the lacustrine zone cluster is marked with the red box. Denotations: I-IV – cross-sectional transects (I – riverine zone (*riv*), II and III – transitional zone (*tr*), IV – lacustrine zone (*la*)); 1-10 – sediment sampling sites

Both Threshold Effects Level (TEL) and Probable Effects Level (PEL) showed the potential effect of heavy metals on the lake ecosystems (Table 5). The ecotoxicological assessment of bottom sediment determines heavy metals' content at which:

a) there is no adverse effect on aquatic organisms because the content of the analyzed element is lower than the TEL value,
b) there is a sporadic adverse effect on aquatic organisms because the content of the element is higher than TEL but lower than PEL,
c) there is a frequent adverse effect on aquatic organisms because the content of the element is higher than PEL.

According to the above criteria, the bottom sediments in the Łoje reservoir did not contain heavy metals in amounts exceeding the PEL value (c),

Table 4

The content of heavy metals in sediments of selected dam reservoirs (mg kg^{-1})

Reservoir	Cd	Cr	Cu	Ni	Pb	Zn
Brzózki (Wiatkowski et al. 2008)	0.1	-	12.9	17.2	18.0	122.7
Brody Iłżeckie (Smal et al. 2015)	2.4	41.4	16.6	14.3	69.2	345.0
Dzierżno Małe (Kostecki et al. 1998)	-	-	27.1	22.9	106.6	560.0
Krępna (Jasiewicz, Baran 2006)	0.7	-	30.0	42.7	15.3	79.8
Łoje	0.4	11.0	7.2	9.1	20.5	39.9
Młyny (Gałka, Wiatkowski 2010*a*)	-	-	67.0	48.0	73.0	176.0
Mściwojów (Wiatkowski et al. 2008)	3.3	-	21.0	19.9	21.9	49.0
Psurów (Gałka, Wiatkowski 2010*b*)	-	-	32.1	7.0	18.2	90.6
Włocławek (Dojlido, Taboryska 1991)	-	750.0	190.0	-	100.0	620.0
Zasławice (Jasiewicz, Baran 2006)	0.5	-	12.4	10.7	15.0	93.8
Zalew Zegrzyński (Wojtkowska 2014)	12.4	-	43.6	-	35.2	72.9
Zalew Zemborzycki (Smal et al. 2015)	0.5	5.9	7.1	6.1	54.2	43.1

Table 5

Ecotoxicological bottom sediment contamination criteria (acc. to Smith et al. 1996) (mg kg^{-1})

Index	Cd	Cr	Cu	Ni	Pb	Zn	As
TEL	0.6	37	36	16	35	123	6
PEL	3.5	90	197	42	91	315	17

which would signify an adverse effect on aquatic organisms. At the sampling points located in the neighborhood of the river flow through the riverine and transitional zones, we have found the contents of Cd, Ni and Pb in the sediments higher than TEL level and lower than PEL (b), what can be the evidence of sporadic effect of the river on the water ecosystem (Smith et al. 1996). Based on the potential ecological risk assessment (PER) we concluded that the ecological risk resulted from the contamination of bottom sediment of the Łoje reservoir with heavy metals is low (Table 6).

Despite the differences heavy metals concentrations and the time of exposure, as well as their toxic effects on aquatic environments they are reliable indicators of ecosystem health (Singh et al. 2005). The determination of toxic pollutants in sediments is fundamental to the solution of many environmental problems and can be of great importance for local people's health (Graney, Eriksen 2004) and ecosystem functions (Li et al. 2013). The example of the Łoje reservoir showed that low values of ecotoxicological indices are

Table 6

Potential ecological risk assessment (PER) of heavy metals in surface sediments from the Łoje Reservoir

Index	E_r									
	riverine zone			transitional zone					lacustrine zone	
	transect I			transect II			transect III		transect IV	
	1	2	3	4	5	6	7	8	9	10
Cd	27.0	36.6	10.8	30.0	30.6	9.6	22.8	47.4	6.6	15.6
Cr	3.9	5.3	2.2	11.0	4.9	2.8	3.9	3.2	1.1	1.1
Cu	13.6	7.9	2.9	4.3	7.8	2.9	7.3	5.6	0.9	0.9
Pb	5.9	6.5	3.9	5.3	12.2	5.8	10.2	13.7	2.4	2.1
Zn	1.2	0.9	0.2	0.2	1.0	0.4	0.9	0.7	0.1	0.1
As	3.4	2.7	1.7	6.1	2.0	1.6	3.6	1.7	1.9	1.8
RI	55.9	61.9	24.7	60.9	63.5	29.1	55.6	80.2	22.1	31.6

particularly satisfying when the reservoir's water is intended to consumption or is of ecological importance. Similar outcomes of the studies are reported for Baihua Lake in China as a source of drinking water (HUANG et al. 2009). Ecological risk posed by heavy metals in bottom sediments was also reported for a hyperthrophic Taihu Lake (YIN et al. 2011, JIANG et al. 2012), or Dongting Lake, the second largest lake in China (LI et al. 2013).

CONCLUSIONS

Bottom sediments accumulated in the Łoje reservoir are characterized by the spatial variation that is typical of flow-through water bodies. The highest amounts of organic sediments accumulated in the transitional and riverine zone of the reservoir. These sediments were found to contain the highest levels of heavy metals. The lacustrine zone was dominated by mineral fractions, and consequently the accumulation of heavy metals there was much lower. The criteria applied to assess the contamination of bottom sediments in the Łoje reservoir led to the conclusion that water drained from the agricultural and forest catchment flowing to the reservoir did not cause its excessive contamination. The concentrations of heavy metals determined in the samples ranged from the natural content of soils (the geochemical background) to sediments weakly or poorly contaminated (mostly due to the presence of Cd, Cr, Cu, Ni and Pb in the riverine and transitional zones of the reservoir, along the water flow). Among the heavy metals potentially responsible for the deterioration of the bottom sediment contamination class, only cadmium, nickel and lead occurred were on a level that causes sporadic adverse effects on aquatic organisms.

The ecotoxicological criteria used in this paper do not consider the potential for bioaccumulation in aquatic organisms nor the associated hazards to the species that consume aquatic organisms (i.e., wildlife and humans). Nevertheless, it is important to combine the results with other tools, such as bioaccumulation tests and tissue residue guidelines, to evaluate more fully the potential effects of sediment-associated contaminants in the environment.

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