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Contamination and Environmental Risk Assessment of Heavy Metals in Sediments of Dobczyce Reservoir and Its Tributaries – a Literature Review**

Abstract: Most transported particles released from a catchment area are deposited and accumulated in the sediment layers of water reservoirs. Along with mineral particles, contaminants originating from human activity and natural processes are added to such aquatic systems (e.g., heavy metals). This is an especially important issue when a reservoir is being used as a source of drinking water. The main aim of this study was the environmental risk assessment of the sediment in the Dobczyce Reservoir and two of its tributaries (the Raba River and Wolnica Stream) during the years of 2004–2007 and 2016, 2017. Substantial variations in heavy metals were found due to the land use and catchment management. The potential ecological risk showed an uneven distribution, and the overall ecological risk level ranged from low to moderate.

Keywords: sediments, heavy metals, Dobczyce Reservoir catchment, environmental risk assessment

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1. Introduction

The sediments of water reservoirs are where most transported particles released from catchment areas are deposited and accumulated. Therefore, they can play a pivotal role in determining the quality of water reservoirs since they act as carriers and sinks for contaminants, reflect pollution history, and provide a record of catchment inputs into an ecosystem [1]. Contaminants (e.g., heavy metals) get into the aquatic ecosystem both as a result of human activity (e.g., municipal waste, agricultural cultivation) as well as natural processes (e.g., atmospheric and fluvial transport). The composition of sediments depends primarily on weather conditions, human activity, and the geochemical formation of the catchment [2].

The main aim of this study was an the environmental risk assessment of the sediments of the Dobczyce Reservoir (which is typical of the sub-mountain part of the Polish Carpathians) and two of its tributaries (the Raba River and Wolnica Stream). The data from different years was derived from previous investigations conducted by Reczynski et al. [2], Szarek-Gwiazda [3], and our own research (No. 2016/23/N/ST10/01292).

2. Study Area

The Dobczyce Reservoir (49°52N 20°02E) was constructed on the Raba River (60 km from the river's source) and put into operation in 1986. It is situated 30 km to the south of the city of Cracow (around 800,000 inhabitants) in the Raba River valley between the towns of Myślenice and Dobczyce; it supplies more than 50% of the drinking water for the Cracow agglomeration. The reservoir consists of three different hydrological sectors: the Myślenice Basin, Dobczyce Basin, and Wolnica Gulf. The total length of the reservoir is about 10 km, the maximum depth is 26.9 m, the average depth is 11.9 m, and the width varies from 0.8 to 1.3 km. The direct reservoir catchment has a surface of 78.2 km^2. Besides the Raba River, several other tributaries also flow into Dobczyce Reservoir (Fig. 1), with a total catchment area of 64.4 km^2. The right tributaries include the Trzemieśnianka, Bulinka, Ratanica, and Brzezówka Streams, and the left tributaries include the San, Dębnik, and Wolnica Streams [1, 3, 4–7].

The Raba River is one of the main tributaries of the right bank of the Vistula River (in southern Poland). The Raba River catchment area is 1537 km^2, 137 km long, and has an average slope of 4.4‰. The river's source area is between the towns of Rabka-Zdrój and Nowy Targ in the Gorce Mountains (the Sieniawska Pass region) at an altitude of 785 m above sea level. It flows to the north and then to the northeast. Its course can be divided into three main parts: (i) the upper course within the Beskidy range (with a length of 60 km and average slope of 8.5‰); (ii) the middle course within the Carpathian foothills (with a length of 34 km and average slope of 2.3‰); and (iii) the lower course within the Sandomierz Basin (with a length of 43 km and average slope of 0.6‰). Moreover, 86% of the Raba River catchment is within the Carpathian area; this determines its typical mountain character, with a narrow valley, rock-covered bed, fast currents, and big drops. The Dobczyce Reservoir is also situated in this area [6].

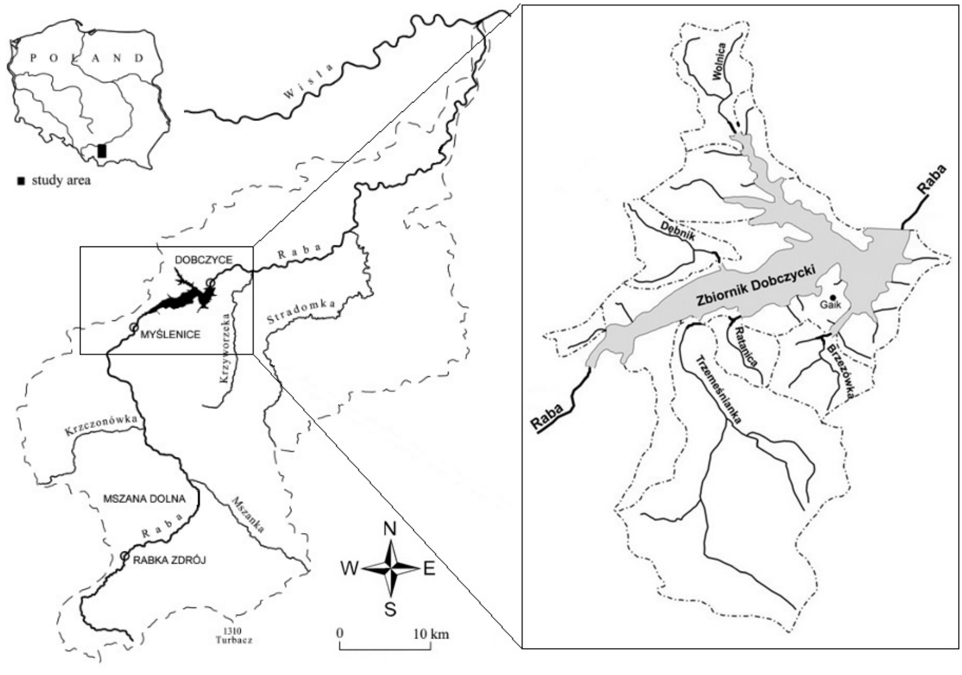

Fig. 1. Raba River catchment with main network of tributaries and location of Dobczyce Reservoir

Source: [5, 8]

The Raba River catchment is located in a moderately warm region; only the parts that are higher than 750 m above sea level feature a cool temperate region. The temperature in the catchment area shows great diversity, which is related to the various landforms. The average annual temperature ranges from 2.7°C (the Gorce Mountains) to 7.9°C (Myślenice). However, the prevailing part of the catchment has an average annual temperature of 6–8°C. The average annual rainfall ranges from 600 mm to 1200 mm; the minimum is in February (4.7% of annual rainfall), and the maximum is in July (17.1% of annual rainfall) [6, 9].

The Raba River and Wolnica Stream differ in terms of land use due to their locations in the catchment. Figure 2 presents the types of land use according to a uniform legend of land cover for Europe (Corine Land Cover – Coordination of Information on the Environment).

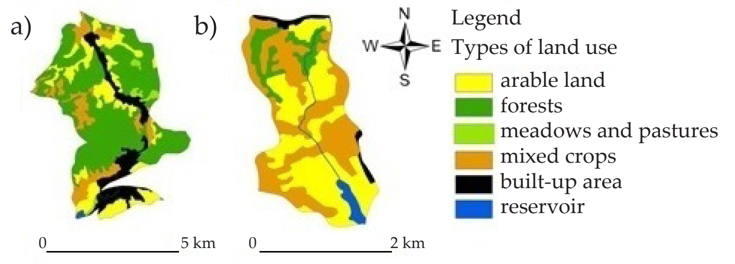

Fig. 2. Land use of Raba River (a) and Wolnica Stream (b) according to Corine Land Cover

Source: [10]

The vegetation cover in the Raba River is diverse. The land-use structure of the Raba River shows that it is dominated by forests, which constitute 54.6% of the area. Arable land (18.4%), meadows and pastures (1.9%), and mixed crops (12.1%) make up a smaller share of the coverage area. Built-up areas cover 13% of the total area. The situation is slightly different in the Wolnica Stream catchment, where arable land (42.6%) and meadows/pastures (42.1%) cover most of the land, while forests take up only 9.4% of the area. Built-up areas (3.1%) have the smallest proportion of the land-use structure [10].

3. Methods

The sediment samples (10- to 15-cm-thick layers) were collected from the Dobczyce Reservoir (the Myślenice Basin, Wolnica Gulf, and Dobczyce Basin) in June of 2004, 2005, and 2006. They were taken by an Ekman device and placed in polyethylene containers. The laboratory work consisted of drying and homogenizing the samples. For the quantitative analysis, the samples were wet digested with HNO_3 and $HClO_4$ acids in a microwave system. The use of atomic absorption spectrometry, flame technique (FAAS), and flame photometry revealed the elemental composition of the samples (Fe, Mn, Cu, and Zn); The AAS electrothermal technique (ETAAS) was used for the determination of Cd, Cr, and Pb content [2].

Also, sediment samples (0- to 5-cm-thick layers) from the Dobczyce Reservoir (the Myślenice Basin, Wolnica Gulf, and Dobczyce Basin) were taken in May 2007 with a polyethylene bucket tube with a diameter of 4 cm. Before the analysis, the samples were dried at 105°C for 48 hours. The samples were mineralized in concentrated HNO_3 in a microwave system with automatic temperature control. They were then analyzed for selected heavy metals (Cd, Pb, Cu, Zn, Mn, and Fe) using atomic absorption spectrophotometry (AAS) [3].

In August 2016 and October 2017, sediment samples were collected by hand from the surface layer of the Raba River and Wolnica Stream (both in the estuaries). The laboratory work consisted of separating the sediment from the water by filtration and vacuum pump, centrifuging samples, and air drying. The dry samples were mineralized in 65% HNO_3 Suprapur in a closed microwave system at 175°C for 10 min (EPA 3051 method). Metal measurements (Cd, Cu, Pb, Ni, Zn, Mn, and Fe) were performed with the use of atomic absorption spectrophotometry.

For the assessment of the degree of pollution of heavy metals in the sediments, four parameters were used: the geo-accumulation index (I_{geo}), contamination factor (CF), pollution load index (PLI), and EF (enrichment factor). Furthermore, the sediment quality guidelines (SQGs) for metals in freshwater ecosystems that reflect TECs and PECs were evaluated to determine whether the contaminants in the sediment might harm aquatic organisms.

4. Results and Discussion

The concentrations of heavy metals in the sediment in the investigated areas are presented in Tables 1 and 2.

Considering the concentration changes in the sediments of the Dobczyce Reservoir in during the years of 2004–2007, it may be concluded that increases in the Cr and Cd content as well as a decrease in Pb were observed. The concentration of Mn and Fe did not change substantially. These heavy metals came from the Raba River due to natural weathering processes and washing out from the soil; they occur as dissolved compounds and suspended matter [11]. Also, substantial differences were found depending on the prevailing land use in the catchment. This was related to the inflow of agricultural contaminants from the catchment as well as from municipal pollution (e.g., an outlet of the Dębnik Stream in the Wolnica Gulf). These may also be caused by other factors such as the presence of biological or geochemical barriers that are conducive to the accumulation of heavy metals in sediment; e.g., gulfs overgrown by macrophytes or groundwater supplies rich in heavy metals [3]. In the catchment where agriculture was the dominant activity, concentrations of Mn and Zn were much higher, while all of the remaining metals were detected at higher levels in the catchment with more diverse land use.

The geo-accumulation index (I_{geo}) [12] determined the metal contamination in the sediments and was calculated using the following formula:

$$I_{\text{geo}} = \log_2 \frac{C_n}{1.5B_n} \tag{1}$$

where:

C_n – metal concentration in sediment sample,
B_n – geochemical background value,
1.5 – allows for analysis of natural fluctuations in content of given element in environment and very small anthropogenic influences.

Table 1. Average concentration [mg/kg] of heavy metals in sediment of Dobczyce Reservoir with SQGs

Heavy metals	TEC/PEC	Dobczyce			
		2004*	2005*	2006*	2007**
Cd	0.99/4.98	0.55	0.54	0.79	0.92
Pb	35.8/128	73.0	34.5	34.0	24.7
Cu	31.6/149	23.5	30.5	24.5	25.45
Zn	121/459	108.5	106.5	192.5	104.4
Mn	–	1000	950	900	945
Fe	–	22,000	23,450	19,650	24,650
Cr	43.4/111	40.5	29.5	43	–
Ni	22.7/48.6	–	–	–	–
"–" – no data available					

Source: [2*, 3**]

Table 2. Concentration [mg/kg] of heavy metals in sediment of Raba River and Wolnica Stream with SQGs

Heavy metal	TEC/PEC	Raba		Wolnica	
		2016	2017	2016	2017
Cd	0.99/4.98	0.30	0.70	0.45	0.42
Pb	35.8/128	12.9	19.1	11.8	9.5
Cu	31.6/149	27.8	27.8	11.4	11.0
Zn	121/459	93.2	85.7	77.8	56.9
Mn	N.D.	811	635	536	619
Fe	N.D.	25,721	29,107	16,344	17,387
Cr	43.4/111	N.D.	N.D.	N.D.	N.D.
Ni	22.7/48.6	63.745	60.233	15.244	14.273

N.D. – no data available

The data was calculated using the geochemical background values given by Turekian and Wedepohl (Cd – 0.3 μg/g; Pb – 20 μg/g; Cu – 45 μg/g; Zn – 95 μg/g; Mn – 850 μg/g; Fe – 4.6%) [13] and the local geochemical background values (Cd – 2 μg/g; Pb – 45 μg/g; Cu – 40 μg/g; Zn – 110 μg/g; Mn – 140 μg/g; Fe – 1.3%) for the sediments of the upper Vistula River given by Helios-Rybicka [14]. The scale consisted of seven grades (0–6) ranging from uncontaminated to highly contaminated. According to the geochemical background values given by Turekian and Wedepohl, the majority of the sampling sites characterized the sediments as uncontaminated ($I_{geo} < 0$) to moderately contaminated ($1 < I_{geo} < 2$). Sediments from the Dobczyce Reservoir (2004–2007) were classified as uncontaminated by Cu, Mn, and Fe, and variously contaminated by Zn, Cd, and Pb (Tab. 1). Igeo's values also indicated that the sediments of the Raba River and Wolnica Stream (2016, 2017) were classified as uncontaminated. However, according to the local geochemical background values, sediments from the Dobczyce Reservoir, Raba River, and Wolnica Stream were only Mn-contaminated (mostly $1 < I_{geo} < 2$). The remaining concentration did not exceed these values.

The contamination factor (CF) [15] was used to determine the overall contamination of the sediment at each monitoring site. It was calculated by the following equation:

$$\mathrm{CF} = \frac{C_{\mathrm{metal}}}{B_n} \tag{2}$$

where:

C_{metal} – concentration of each heavy metal in sediment,
B_n – background concentration.

According to the CF values, the sediment contamination levels consisted of four grades (ranging from low to very high contamination) [16]. The CF values were computed by comparing the metal levels in the sediments and the two different background concentrations. The data calculated with the Turekian and Wedepohl background showed that, in the Dobczyce Reservoir, the highest CF was found for Cd (especially in 2007) and Pb (2004), which indicated considerable contamination ($3 \le \mathrm{CF} < 6$). The lowest CF factor ($\mathrm{CF} < 1$) was for Cu and Fe (2004–2007), while Mn and Zn (2006) indicated moderate contamination ($1 \le \mathrm{CF} < 3$). Also, the highest CF value was found for Cd in the Raba River (especially in 2017) and Wolnica Stream, which indicated moderate contamination ($1 \le \mathrm{CF} < 3$). The situation was slightly different when the comparison was performed with the use of the local background [14]. In all of the data sets, the level of contamination was low ($\mathrm{CF} < 1$) except for Fe (which indicated moderate contamination [$1 \le \mathrm{CF} < 3$]) and Mn (which indicated very high contamination [$\mathrm{CF} > 6$]) in the Dobczyce Reservoir as well as considerable contamination ($3 \le \mathrm{CF} < 6$) in the Raba River and Wolnica Stream.

The extent of the pollution by the metals was assessed by employing the pollution load index (PLI) method for a cumulative indication of the pollution of the site quality [17]. It was calculated by the following equation:

$$\mathrm{PLI} = (\mathrm{CF1} \times \mathrm{CF2} \times \mathrm{CF3} \times \mathrm{CF}n)^{\frac{1}{n}} \tag{3}$$

where:

n – number of metals,
CF – contamination factor of each metal (as mentioned above).

In each case, the PLI was calculated using the geochemical background and local background. The results did not differ substantially among the investigated water bodies. The lowest PLI index was found in Wolnica Stream (2016, 2017) and the Raba River (2016, 2017), which suggested that the sediments were uncontaminated (<1 not polluted), while the highest PLI index (>1 polluted) was found in Dobczyce Reservoir (2004–2007).

The anthropogenic effect on the sediments was determined by the EF (enrichment factor), which required a normalizing metal to mitigate the disparities caused by the heterogeneous nature of the sediments. In this case, Fe was used as a reference heavy metal since it is capable of absorbing the metals and has been linked to fine-grained fractions [18]. The enrichment factor was calculated using the formula based on the equation suggested by Buat–Menard [19]:

$$\mathrm{EF} = \frac{\frac{C_n}{C_{\mathrm{ref}}}}{\frac{B_n}{B_{\mathrm{ref}}}} \tag{3}$$

where:

C_n – content of examined heavy metal in investigated area,
C_{ref} – content of reference heavy metal in same area,
B_n – content of examined heavy metal in background,
B_{ref} – content of reference heavy metal in background.

Seven contamination categories were established on the basis of the EF, ranging from no enrichment to extremely severe enrichment [20]. The EF results calculated using the Turekian and Wedepohl background in the Dobczyce Reservoir showed minor to moderately severe enrichment for Cd and Pb. In other cases, heavy metals presented moderate enrichment (EF 3–5). On the other hand, the results from the Raba River and Wolnica Stream showed minor enrichment (EF < 3) for almost all heavy metals except for Cu (which indicated no enrichment [EF > 1]) and Cd (which showed moderate enrichment [EF 3–5]). The situation was slightly different when the comparison was performed with the use of the local background (Helios-Rybicka) [14]. In all of the data sets, the EF indicated no enrichment (EF < 1) except for Mn (which ranged from minor to moderate enrichment).

The concentration of heavy metals in the sediments was also compared to the corresponding SQGs for each metal. When the heavy metal content is below the TEC (below which harmful effects are unlikely to be observed), no toxic effects are expected, and the sediment contamination can be tolerated by aquatic organisms [21]. Above the PEC (above which harmful effects are likely to be observed), the contaminants probably have a noxious impact on the aquatic organisms. Between the TEC and PEC, the sediments were predicted to be neither toxic nor nontoxic. The PEC levels were exceeded for Ni in the Raba River (2016 and 2017), suggesting possible detrimental effects on the aquatic organisms. Cd, Pb, Cu, and Zn were below the TEC levels in the Raba River and Wolnica Stream (2016 and 2017), as was Ni in the Wolnica Stream. In other cases, the concentration of heavy metals was between the TEC and PEC levels, which suggests the moderate contamination of the sediments and a moderate impact on the biota.

Heavy metals behave differently in a sedimentary and diagenetic environment and, thus, have different potentials for remobilization and uptake by the biota [22]. The accumulated heavy metals may be remobilized from sediments by natural processes and human changes in external parameters, such as pH changes due to acid rain or complexing agents [23]. The accumulation of heavy metals in fish tissues depends on many factors, especially the concentration of heavy metals in the environment, time exposure, fish species, intake path of heavy metals, tissue affinity, age, fish size, or environmental conditions. The Dobczyce Reservoir is slightly contaminated with heavy metals; therefore, the long-term impact on reservoir biocenosis is expected in this area [6]. In terms of the relationship between the concentration of heavy metals in the sediments and fish tissues (*Abramis brama* (L.), *Rutilus rutilus* (L.), *Sander lucioperca* (L.)), risk was found by Szarek-Gwiazda et al. [24]. Concentrations of heavy metals in fish tissues showed significant variation. The lowest concentrations were in the muscle tissues, while the the highest were found in the organs responsible for metabolism (kidneys, gills, livers). Generally, higher concentrations of heavy metals were found in *Abramis brama* (L.) *and Rutilus rutilus* (L.), both of which feed on food found in the silt. Therefore, they were more exposed to heavy metals deposited in the sediments [6, 24]. Also, concentrations of accumulated heavy metals in individual parts of the macrophytes (roots, stem, leaves) varied depending on the species, environmental conditions (such as metal concentration in the water and sediment, exposure time, pH, redox potential), metal intake paths, transport mechanisms, or interactions between metals [25]. Among the studied macrophytes (*Myriophyllum spicatum* (L.), *Najas marina* (L.), *Polygonum amphibium* (L.), *Phragmites australis* (Cav.) Trin. ex Steud.), the ability to accumulate heavy metals was found. Higher concentrations were in the submerged parts (*Myriophyllum spicatum* (L.), *Najas marina* (L.)). In addition, there was an increased accumulation of Cd and Mn in the shoots of *Myriophyllum spicatum* (L.) and *Najas marina* (L.) due to the fact that these metals have the greatest potential mobility in sediment layers [6, 24]. Also, sediment-dwelling organisms might be exposed to concentrations of heavy metals (Cr, Pb, Zn, Cd) that especially cause genetic changes. The most common sediment invertebrates in the Dobczyce Reservoir are Oligochaeta and Chironomidae; the occurrence of Ephemeroptera, Coleoptera, Chaoboridae, and Spheridae is rare [6, 26].

The contamination risk of drinking water uptake was taken into consideration based on published six-grade sequential chemical extraction studies of heavy metals and the ambient physicochemical conditions in the Dobczyce Reservoir [6, 27]. A greater variability of the chemical forms of heavy metals was observed in the coastal part of the Dobczyce Reservoir. Within this site, Cd, Mn, Pb, Zn, and Cu revealed considerable potential mobility in the sediments, while Mn and Cd were moderately mobile. The potential mobility and bioavailability of the heavy metals in the sediments of the Dobczyce Reservoir were affected by the dynamics of the hydrological and catchment factors, eutrophication processes in the reservoir, and heavy metal content in exchangeable, carbonate, and easily reducible forms that are sensitive to changes in the environmental conditions (pH, redox potential) [6]. Moreover, it should be mentioned that water is taken from the tower intake (61.4 km from the source of the Raba River on the left bank of the Dobczyce Reservoir), which has the ability to take up water from three different levels. Based on the laboratory tests, the water is taken from the best quality level [6].

5. Conclusions

The investigated water bodies present substantial variations in heavy metals due to land use and catchment management. During the investigated years, increases in Cr and Cd concentrations have been noticed in the Dobczyce Reservoir. The concentrations of such elements depend largely on human activity. In the agricultural catchment, Mn and Zn were at much higher concentrations than in the catchment with more diverse land use. Heavy metal concentration in sediments displayed deviations from the geochemical background and local background values. Various contaminations were found using the geochemical background for Zn, Cd, and Pb in the Dobczyce Reservoir, while the sediments in the Raba River and Wolnica Stream were uncontaminated. However, the local geochemical background values in all of the sediments indicated only Mn contamination. According to the contamination factor that incorporated the geochemical background values, a considerable contamination of Cd (especially in 2007) and Pb (2004) was found in the Dobczyce Reservoir. On the other hand, moderate Cd contamination was established in the Raba River and Wolnica Stream. In comparison to the local background values, the sediments were only contaminated with Mn. The highest PLI index was found in the Dobczyce Reservoir, while the sediments in the tributaries were uncontaminated. PEC levels were exceeded for Ni in the Raba River, suggesting possible detrimental effects on the aquatic organisms. The enrichment factor using the Turekian and Wedepohl background showed mostly moderately severe enrichment in the Dobczyce Reservoir as well as minor enrichment for almost all of the heavy metals in the Raba River and Wolnica Stream. In contrast, when using the local background, EF indicated no enrichment except for Mn (which ranged from minor to moderate enrichment). For the majority of the water bodies, the heavy metal levels were between the TEC and PEC levels, which suggests moderate contamination. To sum up, the heavy metal concentrations studied in the Dobczyce Reservoir over the years were typical of slightly polluted water reservoirs. Therefore, further research and monitoring of the sediment quality should be maintained in future seasons due to the use of the Dobczyce Reservoir for drinking purposes throughout the year.

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Metale ciężkie w osadach dennych Zbiornika Dobczyckiego i jego dopływów oraz ocena zagrożenia środowiskowego – przegląd literaturowy

Streszczenie: Zbiornik wodny to obszar, na którym większość transportowanych cząstek uwalnianych ze zlewni osadza się i akumuluje w osadach dennych. Razem z cząstkami mineralnymi do ekosystemów wodnych przedostają się również inne zanieczyszczenia (np. metale ciężkie), które są efektem działalności antropogenicznej, a także procesów naturalnych. Jest to szczególnie niebezpieczne, gdy zbiornik używany jest jako źródło wody pitnej. Głównym celem tego badania była ocena zagrożenia środowiskowego w osadach dennych Zbiornika Dobczyckiego oraz jego dopływów: Raby i potoku Wolnica w różnych latach (odpowiednio 2004–2007, 2016, 2017). Stwierdzono, że różny sposób użytkowania i zagospodarowania zlewni miał wpływ na znaczne wahania stężeń metali ciężkich, a ocena zagrożenia środowiskowego wskazała zanieczyszczenie od niskiego do umiarkowanego.

Słowa kluczowe: osady denne, metale ciężkie, zlewnia Zbiornika Dobczyckiego, ocena zagrożenia środowiskowego