Oceanological and Hydrobiological Studies

International Journal of Oceanography and Hydrobiology

Volume 45, Issue 4, December 2016
pages (493-504)

ISSN 1730-413X
eISSN 1897-3191

# Comparison of selected methods used in the assessment of contamination with heavy metals in littoral sediments of lakes

by

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DOI: **10.1515/ohs-2016-0042**
Category: **Original research paper**
Received: **January 26, 2016**
Accepted: **March 23, 2016**

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## Abstract

This study compared selected methods of assessing the pollution in the littoral sediments by heavy metals (Zn, Cu, Pb, Ni and Cd) in 13 lakes of the Borecka Forest. For this purpose, the ecotoxicological criteria (PEL TEL), the geoaccumulation index ($I_{geo}$), the contamination factor (CF) and the contamination degree ($C_d$) were applied. Characteristic values for both Poland (GBP) and the Earth's crust (GBE) were assumed as the geochemical background. In addition, the organic (TOC) and inorganic (TIC) carbon were determined in the analyzed sediments. Regardless of the method or adopted background value, the highest pollution level was in the littoral sediments of Lake Szwałk Mały and Lake Kacze. This was primarily associated with increased concentrations of Cd (1.25 and 0.64 mg $kg^{-1}$, respectively), which was considered as "likely to have toxic effects on organisms" (exceeding PEL). Several potential sources of this metal (fertilizers, herbicides, leaky septic tanks, bait fishing, drainage ditches, allochthonous organic matter) were identified. The content of heavy metals in the littoral sediments of other lakes was sufficiently low and they could be considered as basically uncontaminated.

**Key words:** heavy metals, littoral sediments, geochemical criteria

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## Introduction

Heavy metals are elements whose density is greater than 4.5 g cm$^{-3}$ (Streit 1994). They are divided into two groups. The first group consists of elements necessary for the proper functioning of the body in trace amounts, while at higher concentrations they usually exhibit toxic effects. These include iron, manganese, zinc, molybdenum, chromium and nickel. The second group includes metals of unknown metabolic functions, which have toxic properties even in small quantities, such as cadmium, lead, mercury, silver and platinum (Zamorska & Papciak 2010).

The presence of heavy metals in aquatic ecosystems is a result of natural processes and anthropogenic activities (Shaikh 2013). Rock weathering is one of the natural sources of metals. However, the content of elements of this origin is usually low and is referred to as "geochemical background", which is specific for a given region. On the other hand, human activity has contributed to a significant increase in the concentration of heavy metals in the aquatic environment (Wei & Wen 2012). Currently, metals pose a serious threat to organisms because of their resistance to degradation and the capacity for accumulation, including bioaccumulation and biomagnification (Tao et al. 2012). This particularly applies to cadmium, copper, nickel, lead, zinc, mercury and chromium. Even small concentrations of heavy metals may adversely affect the proper functioning of living organisms and cause mutagenicity, teratogenicity and carcinogenicity (Ociepa-Kubicka & Ociepa 2012; Sardar et al. 2013).

Heavy metals in natural waters undergo various chemical transformations, adsorption and complexation followed by sedimentation and accumulation in sediments (Allen 1995). Depending on the properties of individual metals, as well as environmental and hydrological factors related to the channel morphology, the speed and character of water flow, they become a more-or-less permanent deposit in sediments (Rabajczyk & Jóźwiak 2008). The littoral zone usually has the highest biological diversity within a lake due to good oxygenation, a large amount of incoming light as well as the presence of shady locations and varied terrain structure. It is also the most sensitive area of environmental and hydrological variability, interacting directly with the land (Kajak 2001). Therefore, an assessment of the contamination level in water bodies with heavy metals should also include analysis of their content in littoral sediments.

The objective of the study was to assess the pollution of littoral sediments with heavy metals (Zn, Cu, Pb, Ni and Cd) in lakes of the Borecka Forest, using different methods based on the geochemical criteria for Poland and the Earth's crust. The Borecka Forest, located in north-eastern Poland, has a high density of lakes, which makes it one of the most important ecological habitats and an attractive place for tourists. Although the lakes differ in the degree of anthropopressure and morphometric and direct catchment parameters, there is no information on the sediments of these lakes, especially in terms of heavy metal content.

## Materials and methods

### Study sites

The study covered thirteen lakes, located in north-eastern Poland (Fig. 1). According to the physico-geographical regionalization of Poland, the studied lakes are located in the mesoregion of Ełk Lakeland, which is part of the Masurian Lake District (Kondracki 2001). The lakes are situated within the Borecka Forest, which is unusually valuable because of its natural qualities including a dense forest complex covering an area of approximately 230 km$^2$.

The largest lake is Lake Łaźno, occupying an area of over 562 ha with a maximum depth of about 18 m. The deepest lake is Lake Krzywa Kuta, with a depth of 26.5 m. The smallest (area 1.8 ha) and shallowest lake (maximum depth 0.8 m) is Lake Kacze (Table 1).

The direct catchment areas of the examined lakes are characterized by different types of land use. The direct catchment of Lake Łękuk is covered mostly by agricultural lands (about 69%). Catchments of Lake Łaźno, Lake Szwałk Mały, Lake Szwałk Wielki, Lake Wolisko, Lake Krzywa Kuta and Lake Litygajno have relatively large surface areas and the contribution of agricultural land above 15%. The highest percentage of wasteland is observed in catchments of Lake Litygajno (12.5%) and Lake Łaźno (12.2%), whereas the highest percentage of building areas – in catchments of Lake Łaźno (5.4%) and Lake Łękuk (4.5%). No wasteland or buildings are present in the catchments of five lakes (Lake Biała Kuta, Lake Kacze, Lake Smolak, Lake Ciche and Lake Dubinek; Table 2).

### Sediment sample collection and analysis

The research on the littoral sediments in lakes of the Borecka Forest was carried out in August 2010. The study sites were located in the central part of the zone with helophytes and if the zone of emergent vegetation was absent – at a depth of 0.5-1.5 m, depending on the slope of the lake bottom.

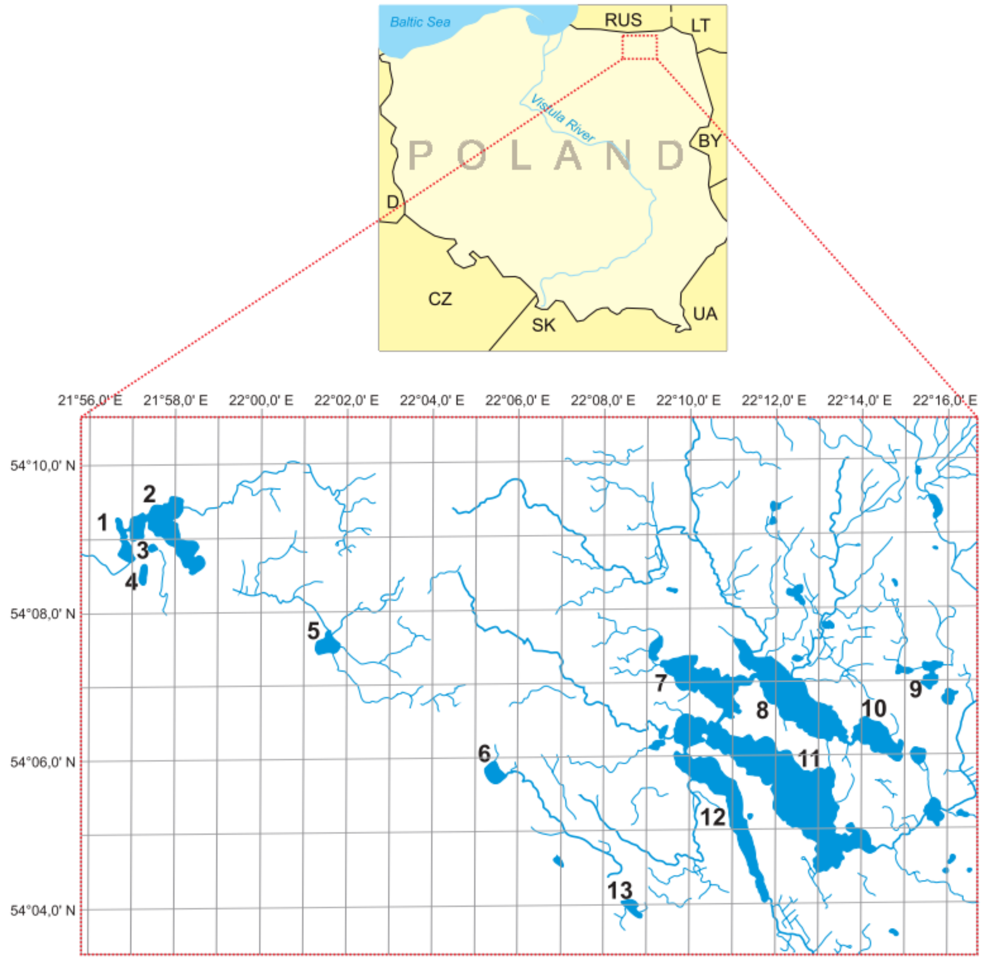


**Figure 1**

Location of the studied lakes: 1. Biała Kuta, 2. Krzywa Kuta, 3. Kacze, 4. Smolak, 5. Łękuk, 6. Wolisko, 7. Pilwąg, 8. Szwałk Wielki, 9. Ciche, 10. Szwałk Mały, 11. Łaźno, 12. Litygajno, 13. Dubinek

**Table 1**

Morphometric parameters of Borecka Forest lakes

| Lake | Catchment | Natura 2000 Code* | Area | Height | Volume | Average depth | Max. depth | Depth factor | Shoreline depth | Shoreline development | Lake length | Average width | Elongation factor |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | - | - | [ha] | [m a.s.l.] | [Mm³] | [m] | [m] | - | [m] | - | [m] | [m] | - |
| Biała Kuta | Pregoła | 3140 | 21.3 | 134.3 | 318.2 | 1.4 | 3.2 | 0.44 | 2700 | 1.65 | 1200 | 177.5 | 6.76 |
| Krzywa Kuta | | 3150 | 131.2 | 134.4 | 7883.1 | 6.0 | 26.5 | 0.23 | 8550 | 2.11 | 2250 | 583.1 | 3.86 |
| Kacze | | 3160 | 1.8 | 134.3 | 5.1 | 0.3 | 0.8 | 0.38 | 490 | 1.04 | 171 | 103.0 | 1.66 |
| Smolak | | 3160 | 7.0 | 135.3 | 125.9 | 1.7 | 5.1 | 0.33 | 1190 | 1.27 | 500 | 140.0 | 3.57 |
| Łękuk | | 3150 | 20.7 | 127.4 | 957.4 | 4.6 | 12.5 | 0.37 | 1900 | 1.18 | 675 | 306.7 | 2.20 |
| Wolisko | Vistula | 3150 | 13.5 | 175.9 | 620.2 | 4.6 | 8.6 | 0.53 | 1780 | 1.37 | 588 | 229.6 | 2.56 |
| Pilwąg | | 3150 | 135.1 | 133.3 | 2025.3 | 1.5 | 3.6 | 0.42 | 7940 | 1.93 | 2320 | 582.3 | 3.98 |
| Szwałk Wielki | | 3150 | 213.4 | 133.4 | 10505.8 | 4.9 | 11 | 0.45 | 9350 | 1.81 | 3800 | 561.6 | 6.77 |
| Ciche | | 3150 | 15.0 | 141.9 | 330.0 | 2.2 | 4.5 | 0.49 | 2560 | 1.86 | 689 | 217.7 | 3.16 |
| Szwałk Mały | | 3150 | 70.4 | 133.6 | 3016.8 | 4.3 | 6.7 | 0.64 | 4900 | 1.65 | 1485 | 474.1 | 3.13 |
| Łaźno | | 3150 | 562.4 | 133.2 | 32017.9 | 5.7 | 18 | 0.32 | 17825 | 2.12 | 6200 | 907.1 | 6.83 |
| Litygajno | | 3150 | 162.1 | 133.1 | 9763.9 | 6.0 | 16.4 | 0.37 | 10675 | 2.37 | 4700 | 344.9 | 13.63 |
| Dubinek | | 3140 | 12.1 | 151.4 | 310.1 | 2.5 | 6.2 | 0.40 | 1660 | 1.35 | 638 | 189.7 | 3.36 |

*shallow lakes (3140), eutrophic lakes (3150), dystrophic lakes (3160)

**Table 2**

The land use in the direct catchments of the analyzed lakes

| Lake | Agricultural land (%) | Wasteland (%) | Building (%) | Forest (%) |
|---|---|---|---|---|
| Biała Kuta | 2.6 | - | - | 97.4 |
| Krzywa Kuta | 17.8 | 1.8 | 2.6 | 77.8 |
| Kacze | - | - | - | 100.0 |
| Smolak | 4.4 | - | - | 95.6 |
| Łękuk | 68.7 | 3.7 | 4.5 | 23.1 |
| Wolisko | 19.6 | 5.5 | 2.7 | 72.3 |
| Pilwąg | 5.2 | 1.1 | 0.5 | 93.2 |
| Szwałk Wielki | 20.6 | 2.8 | 1.1 | 75.4 |
| Ciche | 2.7 | - | - | 97.3 |
| Szwałk Mały | 38.8 | 9.6 | 1.2 | 50.4 |
| Łaźno | 40.7 | 12.2 | 5.4 | 41.7 |
| Litygajno | 15.9 | 12.5 | 2.2 | 69.4 |
| Dubinek | - | - | - | 100.0 |

The number of sediment sampling points for each lake was determined based on its surface area, the shoreline length and the direct catchment land use. It ranged from 3-5 sampling points for the smallest lakes to 5-8 points for the largest lakes. Samples were collected from the 10-cm sediment surface layer using the Kayak sampler, and then mixed to obtain an average sample for further analysis. After drying at room temperature to a constant weight, the samples were grinded in a porcelain mortar and passed through a sieve with 1-mm diameter mesh. The following analyses were performed on the thus prepared sediments:

- total concentration of heavy metals (Zn, Cu, Pb, Ni and Cd) using flame atomic absorption spectrometry (FAAS, spectrometer 280FS AA, Varian) after prior digestion of a sediment sample in a microwave oven (CEM MARSXpress) in the presence of *aqua regia* (a mixture of HCl and $HNO_3$ at 3:1 ratio),

- concentration of total carbon (TC) and total inorganic carbon (TIC) by combustion of dry sediment samples on a TOC analyzer (VCSN, Shimadzu) equipped with a solid sample module SSM-5000A. Total organic carbon (TOC) was calculated as a difference between TC and TIC.

Sediment pollution with heavy metals was assessed on the basis of geochemical criteria. For this purpose, the measured metal concentrations in sediments (in mg/kg of dry mass) were compared to concentrations considered as characteristic of undisturbed conditions, and referred to as 'geochemical background' for Poland (GBP) (Bojakowska 2001).

The potential harmfulness of the analyzed heavy metals on aquatic organisms was determined according to the ecotoxicological criteria using two threshold values of TEL (Threshold Effects Level) and PEL (Probable Effects Level). TEL refers to the concentration of an element above which a toxic effect on the organisms can be observed, while PEL applies to a metal content beyond which toxicity to organisms is often observed (CCME 1999).

The pollution levels in the littoral sediments for a given lake were calculated based on the following indices:

**a) the geoaccumulation index ($I_{geo}$):**

$$I_{geo} = log_2\left(\frac{C_m}{1.5B_n}\right) \quad (1)$$

where: $C_m$ – metal concentration in sediment [mg kg$^{-1}$], $B_n$ – the geochemical background value; two values of $B_n$ were assumed: the value characteristic for Poland (GBP): 73 (Zn), 7 (Cu), 15 (Pb), 5 (Ni), 0.5 (Cd) mg kg$^{-1}$ (Bojakowska 2001) and the concentration corresponding to the natural content in the Earth's crust (GBE): 67 (Zn), 39 (Cu), 17 (Pb), 24 (Ni), 0.1 (Cd) mg kg$^{-1}$ (Taylor & McLennan 1995; Li & Shoonmaker 2003; Choiński et al. 2010). The value of 1.5 is the correction factor arising from the possibility of natural fluctuations in metal concentrations in the environment with low anthropogenic influence. Different classes of sediment quality based on the geoaccumulation index can be distinguished: '0' ($I_{geo}$≤0, unpolluted), '1' (0<$I_{geo}$<1, unpolluted to moderately polluted), '2' (1<$I_{geo}$<2, moderately polluted), '3' (2<$I_{geo}$<3, moderately to strongly polluted), '4' (3<$I_{geo}$<4, strongly polluted), '5' (4<$I_{geo}$<5, strongly to extremely polluted) and '6' ($I_{geo}$>6, extremely polluted).

**b) the contamination factor (CF):**

$$CF = \frac{C_m}{B_n} \quad (2)$$

where: $C_m$ – metal concentration in sediment [mg kg$^{-1}$], $B_n$ – geochemical background for a given metal (GBP or GBE) [mg kg$^{-1}$]. Based on the CF index, the sediment pollution levels can be classified as follows: low (CF < 1), moderate (1 ≤ CF < 3), considerable 3 ≤ CF < 6) and very high (CF ≥ 6) (Håkanson 1980).

**c) the percentage of each metal in the total sediment contamination (% $CF_i$):**

$$\%CF_i = \frac{CF_i}{\sum CF_i} \quad (3)$$

**d) the degree of sediment contamination ($C_d$) (Abraham 2005; Likaku et al. 2013):**

$$C_d = \frac{1}{N}\sum_{i=1}^{N} CF_i \quad (4)$$

where: N – the number of analyzed metals, $CF_i$ – the contamination factor for *i* metal. There are seven categories of sediment contamination levels based on the $C_d$ index: '1' ($C_d$<1, nil to very low), '2' (1.5<$C_d$<2, low), '3' (2<$C_d$<4, moderate), '4' (4<$C_d$<8, high), '5' (8<$C_d$<16, very high), '6' (16<$C_d$<32, extremely high) and '7' ($C_d$≥32, ultra-high) (Likaku et al. 2013).

### Statistical analysis

Means and standard deviations were reported when appropriate. Pearson's correlation analysis was performed to evaluate potential relationships between different variables using Statistica 12.0 (StatSoft Inc.).

## Results

### Total metal concentration in sediments

In most sediments, the concentrations of heavy metals decreased in the following order: Zn>Cu>Pb>Ni>Cd (Table 3). In the case of sediments collected from lakes Łękuk, Ciche and Litygajno, the order was similar to that above, but the average lead concentration was higher than the concentration of copper (Zn>Pb>Cu>Ni>Cd). Only sediments from three lakes were characterized by a specific sequence of heavy metals: Lake Kacze (Zn>Pb>Cu>Ni=Cd), Lake Łaźno (Cu>Zn>Pb>Ni>Cd) and Lake Dubinek (Zn>Cu>Ni>Pb>Cd).

The maximum concentrations of zinc, copper and lead were found in the littoral sediments of Lake Kacze: 42.35 mg Zn $kg^{-1}$, 15.60 mg Cu $kg^{-1}$ and 26.77 mg Pb $kg^{-1}$. The second-highest concentrations of these metals were recorded in sediments of Lake Dubinek, but they were lower by about 36% (Zn), 8% (Cu) and 75% (Pb) compared to metal concentrations

**Table 3**

Heavy metals in sediments arranged in order from the highest to the lowest concentration for a given metal

| Lake | Order of heavy metals |
|---|---|
| Biała Kuta | Zn > Cu > Pb > Ni > Cd |
| Krzywa Kuta | Zn > Cu > Pb > Ni > Cd |
| Kacze | Zn > Pb > Cu > Ni = Cd |
| Smolak | Zn > Cu > Pb > Ni > Cd |
| Łękuk | Zn > Pb > Cu > Ni > Cd |
| Wolisko | Zn > Cu > Pb > Ni > Cd |
| Pilwąg | Zn > Cu > Pb > Ni > Cd |
| Szwałk Wielki | Zn > Cu > Pb > Ni > Cd |
| Ciche | Zn > Pb > Cu > Ni > Cd |
| Szwałk Mały | Zn > Cu > Pb > Ni > Cd |
| Łaźno | Cu > Zn > Pb > Ni > Cd |
| Litygajno | Zn > Pb > Cu > Ni > Cd |
| Dubinek | Zn > Cu > Ni > Pb > Cd |

in sediments from Lake Kacze. On the other hand, the lowest concentrations of zinc, copper and lead were recorded in the sediments of lakes Szwałk Mały and Krzywa Kuta. The littoral sediments of Lake Dubinek was characterized by the highest average content of nickel. The lowest concentration of nickel was observed in sediments of Lake Smolak (Table 4). In the case of cadmium, its maximum content was recorded in littoral sediments of Lake Szwałk Mały. Nearly 50% lower concentration of cadmium was determined in sediments of Lake Kacze. In other lakes, the concentration of this metal did not exceed 0.20 mg Cd $kg^{-1}$. However, the lowest content of cadmium was recorded in sediments from Lake Biała Kuta, Lake Smolak and Lake Dubinek. The average concentrations of heavy metals in the littoral sediments of the examined lakes were at the following levels: 13.28 mg Zn $kg^{-1}$, 6.37 mg Cu $kg^{-1}$, 4.88 mg Pb $kg^{-1}$, 2.18 mg Ni $kg^{-1}$, 0.23 mg Cd $kg^{-1}$ (Table 4).

### Assessment of sediment pollution with heavy metals

#### Geochemical and ecotoxicological criteria

The content of heavy metals in littoral sediments of the examined lakes varied. Based on the geochemical criteria for Poland (GBP) (Bojakowska 2001) (Table 5), it was found that metal concentrations in sediments from nine lakes (Biała Kuta, Krzywa Kuta, Smolak, Łękuk, Wolisko, Szwałk Wielki, Ciche, Łaźno and Litygajno) were below the background value. Whereas, the content of one or more metals in sediments from three lakes slightly exceeded the GBP levels: Lake Pilwąg (Cu), Lake Dubinek (Cu and Ni) and Lake Kacze (Cu, Pb and Cd). Nevertheless, the sediments of

**Table 4**

The average content of heavy metals, total carbon (TC), total organic carbon (TOC), total inorganic carbon (TIC) in littoral sediments in lakes of Borecka Forest

| Lake | Zn | Cu | Pb | Ni | Cd | TOC | TIC | TC |
|---|---|---|---|---|---|---|---|---|
| | (mg kg$^{-1}$) | | | | | g C kg$^{-1}$ | | |
| Biała Kuta | 6.50 | 6.22 | 3.83 | 1.09 | 0.05 | 16.00 | 0.11 | 16.11 |
| Krzywa Kuta | 3.41 | 2.01 | 1.06 | 0.72 | 0.10 | 6.01 | 0.03 | 6.04 |
| Kacze | 42.35 | 15.60 | 26.77 | 0.64 | 0.64 | 494.46 | 0.22 | 494.68 |
| Smolak | 11.31 | 5.62 | 2.89 | 0.05 | 0.05 | 44.58 | 0.12 | 44.71 |
| Łękuk | 12.65 | 5.56 | 6.03 | 2.32 | 0.10 | 7.20 | 3.65 | 10.86 |
| Wolisko | 22.46 | 4.06 | 2.19 | 1.93 | 0.10 | 6.30 | 9.17 | 15.46 |
| Pilwąg | 19.78 | 10.61 | 2.88 | 1.64 | 0.15 | 41.96 | 7.16 | 49.12 |
| Szwałk Wielki | 5.80 | 3.70 | 1.29 | 0.85 | 0.14 | 12.17 | 0.03 | 12.20 |
| Ciche | 8.96 | 2.72 | 2.76 | 1.91 | 0.19 | 8.57 | 0.11 | 8.67 |
| Szwałk Mały | 0.62 | 2.55 | 0.89 | 1.35 | 1.25 | 15.56 | 0.04 | 15.60 |
| Łaźno | 3.46 | 6.31 | 1.90 | 1.43 | 0.14 | 9.79 | 15.11 | 24.90 |
| Litygajno | 8.06 | 3.54 | 4.08 | 1.82 | 0.10 | 22.95 | 0.74 | 23.69 |
| Dubinek | 27.3 | 14.3 | 6.57 | 12.1 | 0.05 | 50.30 | 10.42 | 60.72 |
| Average | 13.28 | 6.37 | 4.88 | 2.18 | 0.23 | 56.60 | 3.61 | 60.21 |
| Min. | 0.62 | 2.01 | 0.89 | 0.50 | 0.05 | 6.01 | 0.03 | 6.04 |
| Max. | 42.35 | 15.60 | 26.77 | 12.10 | 1.25 | 494.46 | 15.11 | 494.68 |
| SD | ±11.81 | ±4.42 | ±6.82 | ±3.03 | ±0.34 | ±132.47 | ±5.14 | ±131.65 |
| V | 112.44 | 144.03 | 71.54 | 71.74 | 68.76 | 42.73 | 70.19 | 45.74 |
| GBP | 73 | 7 | 15 | 5 | 0.5 | - | - | |
| GBE | 67 | 39 | 17 | 68 | 0.1 | - | - | |

SD – standard deviation; V – variability index [%]; GBP – geochemical background for Poland, GBE – geochemical background for the Earth's crust.

**Table 5**

Geochemical criteria of sediment pollution assessment (Bojakowska 2001)

| Metal | Classes of sediment pollution | | | | |
|---|---|---|---|---|---|
| | 0 | I | II | III | IV |
| | geochemical background | unpolluted | moderately polluted | polluted | strongly polluted |
| | (mg kg$^{-1}$) | | | | |
| Zinc (Zn) | 73 | 200 | 500 | 1000 | >1000 |
| Copper (Cu) | 7 | 40 | 100 | 200 | >200 |
| Lead (Pb) | 15 | 30 | 100 | 200 | >200 |
| Nickel (Ni) | 5 | 16 | 40 | 50 | >50 |
| Cadmium (Cd) | <0.5 | 1 | 3.5 | 6 | >6 |

these lakes were considered to be unpolluted (purity class I) (Table 6). Only littoral sediments from Lake Szwałk Mały exceeded the GBP levels for cadmium more than twice and were classified as moderately polluted (purity class II) (Table 6).

According to the ecotoxicological criteria (Table 7), the sediments of lakes Kacze and Szwałk Mały were considered to have a toxic effect on the organisms. These sediments were characterized by an increased content of cadmium (> 0.6 mg kg$^{-1}$). In the case of other lakes, metal concentrations did not exceed the limit value of TEL. Therefore, the sediments from these lakes did not have a toxic effect on the organisms (Tables 4, 7).

### Sediment pollution indices ($I_{geo}$, CF, $C_d$)

The calculated values of the geoaccumulation indices for Zn, Cu, Pb, Ni and Cd in sediments using GBP and GBE as backgrounds are given in Table 8. The $I_{geo}$ indices for Zn in all sediments were below 0. Thus, all sediments showed no contamination with Zn.

**Table 6**

Heavy metal pollution of littoral sediments from lakes in Borecka Forest

| Lake | Zn | Cu | Pb | Ni | Cd | Sediment evaluation |
|---|---|---|---|---|---|---|
| | Sediment purity classes | | | | | |
| BiałaKuta | 0 | 0 | 0 | 0 | 0 | geochemical background |
| KrzywaKuta | 0 | 0 | 0 | 0 | 0 | geochemical background |
| Kacze | 0 | I | I | 0 | I | unpolluted |
| Smolak | 0 | 0 | 0 | 0 | 0 | geochemical background |
| Łękuk | 0 | 0 | 0 | 0 | 0 | geochemical background |
| Wolisko | 0 | 0 | 0 | 0 | 0 | geochemical background |
| Pilwąg | 0 | I | 0 | 0 | 0 | unpolluted |
| SzwałkWielki | 0 | 0 | 0 | 0 | 0 | geochemical background |
| Ciche | 0 | 0 | 0 | 0 | 0 | geochemical background |
| SzwałkMały | 0 | 0 | 0 | 0 | II | moderately polluted |
| Łaźno | 0 | 0 | 0 | 0 | 0 | geochemical background |
| Litygajno | 0 | 0 | 0 | 0 | 0 | geochemical background |
| Dubinek | 0 | I | 0 | I | 0 | unpolluted |

**Table 7**

Ecotoxicological assessment of sediments in rivers and lakes (Macdonald 1994, CCME 1999)

| Metal | TEL | PEL |
|---|---|---|
| | (mg kg$^{-1}$) | |
| Zn | 123 | 315 |
| Cu | 36 | 197 |
| Pb | 35 | 91 |
| Ni | 16 | 42 |
| Cd | 0.6 | 3.5 |

**Table 8**

Values of geoaccumulation indices ($I_{geo}$) of heavy metals in littoral sediments from lakes in Borecka Forest

| Lake | Background | $I_{geo}$-Zn | $I_{geo}$-Cu | $I_{geo}$-Pb | $I_{geo}$-Ni | $I_{geo}$-Cd |
|---|---|---|---|---|---|---|
| BiałaKuta | GBP | -4.07 | -0.76 | -2.55 | -2.78 | -4.00 |
| | GBE | -3.95 | -3.23 | -2.74 | -5.05 | -1.67 |
| KrzywaKuta | GBP | -5.01 | -2.39 | -4.41 | -3.38 | -2.97 |
| | GBE | -4.88 | -4.86 | -4.59 | -7.15 | -0.64 |
| Kacze | GBP | -1.37 | 0.57* | 0.25* | -3.55 | -0.23 |
| | GBE | -1.25 | -1.91 | 0.07* | -7.31 | 2.10*** |
| Smolak | GBP | -3.28 | -0.90 | -2.96 | -3.91 | -3.91 |
| | GBE | -3.15 | -3.38 | -3.14 | -7.67 | -1.58 |
| Łękuk | GBP | -3.11 | -0.92 | -1.84 | -1.69 | -2.95 |
| | GBE | -2.99 | -3.40 | -2.02 | -5.46 | -0.63 |
| Wolisko | GBP | -2.29 | -1.37 | -3.36 | -1.96 | -2.92 |
| | GBE | -2.16 | -3.85 | -3.54 | -5.72 | -0.60 |
| Pilwąg | GBP | -2.47 | 0.02* | -2.97 | -2.19 | -2.33 |
| | GBE | -2.35 | -2.46 | -3.15 | -5.96 | -0.01 |
| SzwałkWielki | GBP | -4.24 | -1.50 | -4.12 | -3.14 | -2.40 |
| | GBE | -4.11 | -3.98 | -4.31 | -6.91 | -0.08 |
| Ciche | GBP | -3.61 | -1.95 | -3.03 | -1.97 | -1.97 |
| | GBE | -3.49 | -4.43 | -3.21 | -5.74 | 0.35* |
| SzwałkMały | GBP | -7.46 | -2.04 | -4.66 | -2.47 | 0.74* |
| | GBE | -7.34 | -4.52 | -4.84 | -6.24 | 3.06**** |
| Łaźno | GBP | -4.98 | -0.73 | -3.57 | -2.39 | -2.39 |
| | GBE | -4.86 | -3.21 | -3.75 | -6.16 | -0.07 |
| Litygajno | GBP | -3.76 | -1.57 | -2.46 | -2.04 | -2.94 |
| | GBE | -3.64 | -4.05 | -2.64 | -5.81 | -0.61 |
| Dubinek | GBP | -2.00 | 0.45* | -1.78 | 0.69* | -3.91 |
| | GBE | -1.88 | -2.03 | -1.96 | -3.08 | -1.58 |

* - unpolluted to moderately polluted sediments (class I); *** - moderately to strongly polluted sediments (class III); **** - strongly polluted sediments (class IV)

The $I_{geo}$ for Cu ranged from -2.39 to 0.57 (GBP) and from -4.86 to -1.91 (GBE). Based on the GBP criterion, the littoral sediments in lakes Kacze, Pilwąg and Dubinek were classified as "unpolluted to moderately polluted" due to positive values of $I_{geo}$, i.e. 0.57, 0.02 and 0.45, respectively. As a result, sediments from these lakes had quality class I. The sediments from the other lakes were unpolluted with Cu (quality class 0). The $I_{geo}$ indices calculated for Pb varied in a wide range from -4.66 to 0.25 (for GBP) and from -4.84 to 0.07 (for GBE). The positive values of $I_{geo}$ were obtained only in the case of sediments from Lake Kacze. Based on $I_{geo}$ values for Pb, these sediments were classified as "unpolluted to moderately polluted". The values of $I_{geo}$ for Ni were negative for all lakes, except for sediments from Lake Dubinek. In that case, the $I_{geo}$ amounted to 0.69, when GBP was used as a background value. This means that Ni content in these sediments was a cause of their deterioration (class I, unpolluted to moderately polluted). On the other hand, $I_{geo}$ for Ni calculated based on GBE gave a negative value (-3.08), which means no sediment contamination with nickel. In the case of Cd, the highest values of $I_{geo}$ were obtained only for sediments collected from lakes Kacze and Szwałk Mały using GBE as a background value (Table 8). Based on the Cd content, sediments from Lake Kacze were classified as moderately to strongly polluted (quality class III), whereas sediments from Lake Szwałk Mały as strongly polluted (quality class IV). Therefore, sediments from these two lakes were characterized by the lowest quality. However, using GBP as a background for Cd, sediments from Szwałk Mały were classified as unpolluted to moderately polluted, and from Lake Kacze – as unpolluted. In addition, sediments from Lake Ciche belonged to quality class I due to a positive value of $I_{geo}$, which, however, was lower than for sediments from lakes Szwałk Mały an Kacze.

Assessment of the sediment pollution by various heavy metals using $I_{geo}$ was reflected in the obtained contamination factors (CF) (Table 9). The CF index also confirmed a very high sediment pollution with Cd using the GBE value for lakes Szwałk Mały and Kacze. In addition, the sediments from other lakes were assessed with the CF index as moderately contaminated with Cd, i.e. sediments from Lake Pilwąg, Lake Ciche

**Table 9**

Contamination factors (CF), the percentage of each metal in the total sediment contamination (%CF) with heavy metals in littoral sediments and the degree of sediment contamination ($C_d$)

| Lake | Background | CF (Zn) | %CF (Zn) | CF (Cu) | %CF (Cu) | CF (Pb) | %CF (Pb) | CF (Ni) | %CF (Ni) | CF (Cd) | %CF (Cd) | ΣCF | $C_d$ |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| Biała Kuta | GBP | 0.09 | 5.8 | 0.89 | 57.5 | 0.26 | 16.5 | 0.22 | 14.1 | 0.09 | 6.1 | 1.54 | 0.31 |
| | GBE | 0.10 | 10.0 | 0.16 | 16.5 | 0.23 | 23.3 | 0.02 | 1.7 | 0.47 | 48.6 | 0.97 | 0.19 |
| Krzywa Kuta | GBP | 0.05 | 6.3 | 0.29 | 38.8 | 0.07 | 9.5 | 0.14 | 19.5 | 0.19 | 25.9 | 0.74 | 0.15 |
| | GBE | 0.05 | 4.5 | 0.05 | 4.5 | 0.06 | 5.5 | 0.01 | 0.9 | 0.96 | 84.6 | 1.14 | 0.23 |
| Kacze | GBP | 0.58 | 9.7 | 2.23* | 37.1 | 1.78* | 29.7 | 0.13 | 2.1 | 1.28* | 21.4 | 6.00 | 1.20 |
| | GBE | 0.63 | 7.0 | 0.40 | 4.4 | 1.57* | 17.4 | 0.01 | 0.1 | 6.41**** | 71.0 | 9.03 | 1.81[2)] |
| Smolak | GBP | 0.15 | 11.5 | 0.80 | 59.5 | 0.19 | 14.3 | 0.10 | 7.4 | 0.10 | 7.4 | 1.35 | 0.27 |
| | GBE | 0.17 | 17.0 | 0.14 | 14.6 | 0.17 | 17.2 | 0.01 | 0.7 | 0.50 | 50.5 | 0.99 | 0.20 |
| Łękuk | GBP | 0.17 | 8.5 | 0.79 | 38.8 | 0.42 | 20.5 | 0.46 | 22.7 | 0.19 | 9.5 | 2.05 | 0.41 |
| | GBE | 0.19 | 11.1 | 0.14 | 8.4 | 0.37 | 21.7 | 0.03 | 2.0 | 0.97 | 56.9 | 1.71 | 0.34 |
| Wolisko | GBP | 0.31 | 19.0 | 0.58 | 35.9 | 0.15 | 9.0 | 0.39 | 23.9 | 0.20 | 12.2 | 1.62 | 0.32 |
| | GBE | 0.34 | 21.1 | 0.10 | 6.6 | 0.13 | 8.1 | 0.03 | 1.8 | 0.99 | 62.4 | 1.59 | 0.32 |
| Pilwąg | GBP | 0.27 | 10.4 | 1.52* | 58.2 | 0.19 | 7.4 | 0.33 | 12.6 | 0.30 | 11.4 | 2.60 | 0.52 |
| | GBE | 0.30 | 13.1 | 0.27 | 12.1 | 0.17 | 7.5 | 0.02 | 1.1 | 1.49* | 66.2 | 2.25 | 0.45 |
| Szwałk Wielki | GBP | 0.08 | 6.9 | 0.53 | 46.0 | 0.09 | 7.5 | 0.17 | 14.8 | 0.28 | 24.7 | 1.15 | 0.23 |
| | GBE | 0.09 | 5.1 | 0.09 | 5.6 | 0.08 | 4.5 | 0.01 | 0.7 | 1.42* | 84.0 | 1.69 | 0.34 |
| Ciche | GBP | 0.12 | 8.4 | 0.39 | 26.6 | 0.18 | 12.6 | 0.38 | 26.2 | 0.38 | 26.2 | 1.46 | 0.29 |
| | GBE | 0.13 | 5.8 | 0.07 | 3.0 | 0.16 | 7.0 | 0.03 | 1.2 | 1.91* | 82.9 | 2.30 | 0.46 |
| Szwałk Mały | GBP | 0.01 | 0.3 | 0.36 | 11.4 | 0.06 | 1.9 | 0.27 | 8.4 | 2.50* | 78.1 | 3.20 | 0.64 |
| | GBE | 0.01 | 0.1 | 0.07 | 0.5 | 0.05 | 0.4 | 0.02 | 0.2 | 12.49**** | 98.8 | 12.64 | 2.53[3)] |
| Łaźno | GBP | 0.05 | 2.9 | 0.90 | 54.7 | 0.13 | 7.7 | 0.29 | 17.4 | 0.29 | 17.4 | 1.65 | 0.33 |
| | GBE | 0.05 | 2.9 | 0.16 | 9.1 | 0.11 | 6.3 | 0.02 | 1.2 | 1.43* | 80.5 | 1.78 | 0.36 |
| Litygajno | GBP | 0.11 | 7.6 | 0.51 | 34.9 | 0.27 | 18.8 | 0.36 | 25.1 | 0.20 | 13.5 | 1.45 | 0.29 |
| | GBE | 0.12 | 8.3 | 0.09 | 6.2 | 0.24 | 16.5 | 0.03 | 1.8 | 0.98 | 67.2 | 1.46 | 0.29 |
| Dubinek | GBP | 0.37 | 7.0 | 2.04* | 38.0 | 0.44 | 8.1 | 2.42* | 45.0 | 0.10 | 1.9 | 5.37 | 1.07 |
| | GBE | 0.41 | 22.2 | 0.37 | 19.9 | 0.39 | 21.0 | 0.18 | 9.7 | 0.50 | 27.2 | 1.84 | 0.37 |

**** – very high contamination level ; * – moderate contamination level; [2)] – low contamination level; [3)] – moderate contamination level and Lake Łaźno using the GPE background and sediments from lakes Szwałk Mały and Kacze using the GBE background.

Sediments from lakes Kacze and Pilwąg showed moderate contamination with Cu when the GBP value was used as a background. In addition, sediments from Lake Kacze were moderately contaminated with Pb, regardless of the background value (GBE or GPE).

Since the degree of sediment contamination ($C_d$) is calculated in relation to all metals present in sediments, its comparison with other indices, i.e. $I_{geo}$ or CF calculated for individual metals can be quite difficult. Nevertheless, the $C_d$ index revealed that although the contamination of sediments in lakes Kacze and Szwałk Mały was mainly caused by Cd (the content of Cd was 71% and 98.8%, respectively), the overall contamination of these sediments was low and moderate, respectively. The values of $C_d$ for the other sediments were below 1. This means that they showed nil to very low contamination.

To sum up, based on the values of pollution indices, Cd, Pb and Cu had the greatest effect on the sediment quality, but it depended on the type of lake. The values of individual indices were affected not only by the total metal concentration in sediments, but also by the background value (GBP or GBE).

### Organic and inorganic carbon in sediments

The content of TOC and TIC in sediments can affect their ability to bind and accumulate heavy metals. In the present study, the sediments are characterized by varying concentrations of organic and inorganic carbon (Table 4). The average content of total carbon (TC) in dry mass of sediments amounted to 60.21 g C kg$^{-1}$. The highest concentrations of TC were observed in lakes Kacze and Dubinek, 494.68 and 60.72 g C kg$^{-1}$ respectively, while the lowest in sediments from lakes Krzywa Kuta and Ciche, 6.04 and 8.67 g C kg$^{-1}$ respectively. The highest content of TOC was recorded in lakes Kacze and Dubinek, while the lowest in lakes Krzywa Kuta and Wolisko. The mean concentration of organic carbon in the analyzed sediments amounted to 56.60 g kg$^{-1}$, which accounted for 94.0% of TC. The maximum concentrations of TIC were recorded in sediments from lakes Łaźno and Dubinek and the minimum concentrations in sediments from lakes Szwałk Mały and Krzywa Kuta. The average content of TIC in the dry mass of sediments from these lakes amounted to 3.61 g kg$^{-1}$ (6.0% of TC) (Table 4). Clearly, the dominant form of carbon in the sediments of most lakes was TOC. The content of TOC in sediments from eight lakes (Kacze, Szwałk Mały, Szwałk Wielki, Smolak, Krzywa Kuta, Biała Kuta, Ciche and Litygajno) ranged from 97 to 100% of TC. On the hand, the TIC dominated only in sediments from lakes Łaźno and Wolisko (about 60% of TC) (Fig. 2).

Statistical analysis revealed strong correlations between the concentrations of individual metals in the examined sediments: zinc and copper (r = 0.85), lead and zinc (r = 0.82) and copper and lead (r = 0.73). A high significant correlation was also found between TOC and Pb content (r = 0.97), TOC and Zn (r = 0.78), and TOC and Cu (r = 0.69) (Table 10).

**Table 10**

Values of Pearson's correlation coefficients between heavy metals, TOC and TIC in sediments

| | Zn | Cu | Pb | Ni | Cd | TOC | TIC |
|---|---|---|---|---|---|---|---|
| Zn | 1 | | | | | | |
| Cu | ***0.85 | 1 | | | | | |
| Pb | ***0.82 | **0.73 | 1 | | | | |
| Ni | 0.34 | 0.49 | 0.03 | 1 | | | |
| Cd | 0.00 | 0.00 | 0.24 | -0.18 | 1 | | |
| TOC | **0.78 | **0.69 | ***0.97 | -0.09 | 0.34 | 1 | |
| TIC | 0.17 | 0.32 | -0.14 | 0.46 | -0.27 | -0.18 | 1 |

(***), (**) significant at $p$<0.01 and 0.05, respectively

## Discussion

In the present study, sediments from different lakes differed in the total concentration of individual metals. Varied metal concentrations in the sediments are determined by catchment factors and the degree of anthropopressure as well as environmental factors, i.e. the nature and structure of sediments, the type and the development stage of the vegetation in the lake (Szafran 2003). Bojakowska and Krasuska (2014) analyzed the content of heavy metals in sediments from 14 lakes located in the Gniezno Lake District (Poland), sediments from 9 lakes outside the power plant cooling system and sediments from 5 lakes within the power plant cooling system. Sediments from lakes within the power plant cooling system are characterized by a considerably higher content of Ba, Zn, Cu, Ni, Hg compared to sediments from the other lakes. Szafran (2003) showed that the lowest concentrations of heavy metals in sediments from lakes Sumin and Rotcze were found in sandy sediments within the area of emergent plants

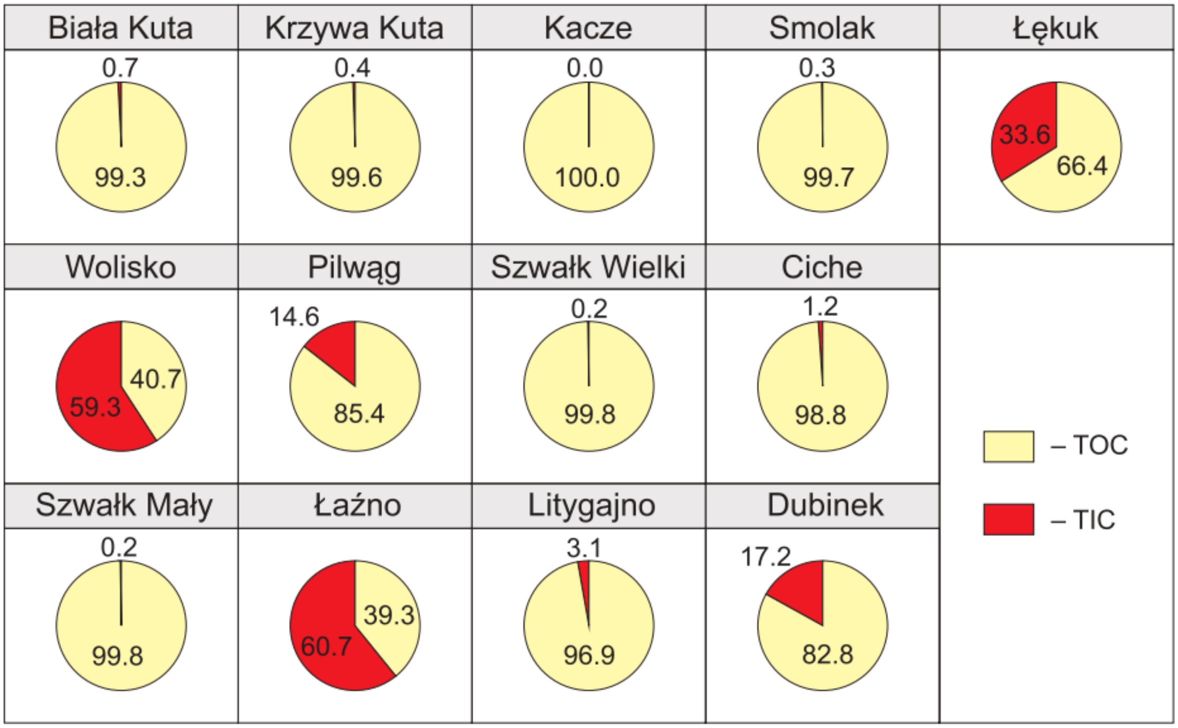


**Figure 2**

The percentage of individual forms of carbon in the littoral sediments in lakes of Borecka Forest

(on average 4.64 mg Zn kg$^{-1}$, 2.77 mg Pb kg$^{-1}$, 1.15 mg Cu kg$^{-1}$, 0.04 mg Cd kg$^{-1}$), and the highest in sediments in the middle part of the lake (on average 147.25 mg Zn kg$^{-1}$, 55.39 mg Pb kg$^{-1}$, 8.08 mg Cu kg$^{-1}$, 1.77 mg Cd kg$^{-1}$). Lower metal concentrations in the littoral sediments compared to the sediments in the middle part of the lake may be due to the bioaccumulation effect. Sediments from the deeper zones of the lake show a higher sorption capacity toward metals. This results from a higher content of organic matter and clay fraction as compared to the littoral sediments. Mielnik et al. (2011) confirmed that heavy metal content in sediments from selected lobelia lakes was spatially differentiated. In most cases, the content of Zn, Cu and Mn was significantly higher in the profundal zone of each of the investigated lakes compared to the littoral zone.

Sediments are generally rich in organic matter. In the present study, the average concentration of TOC in sediments was 56.60 g kg$^{-1}$, which accounted for 94.0% of TC. The content of TOC in sediments can vary within very wide limits and reach a value of even 460 g kg$^{-1}$ (Cieślewicz 2005; Gälman et al. 2008). In the sediments from lakes located on the Wałeckie Lake District, Chodzieskie Lake District, Iławskie Lake District, Drawska Plain and Tucholska Plain (Poland), Cieślewicz & Różański (2010) found the highest concentration of organic carbon in the following lakes: Arkońskie (386 g kg$^{-1}$), Hanki (339 g kg$^{-1}$) and Piaseczno Małe (315 g kg$^{-1}$). The highest concentration of inorganic carbon was determined in the sediments from Lake Wdzydze (65.4 g kg$^{-1}$).

In the present study, the highest $I_{geo}$ and CF values among the analyzed metals in lake sediments were obtained for Cd. It was found that due to the increased content of Cd, the largest contamination occurred in sediments from Lake Szwałk Mały. One possible reason for this may be the surface runoff from agricultural land, which occupies almost 40% of the direct catchment area of the lake. The amount of Cd which is introduced into the soils with the fertilizers and herbicides is undoubtedly much lower than that introduced with the industrial pollutants. The one cause of water pollution with cadmium is using phosphate and calcium-magnesium fertilizers (Bojakowska & Sokołowska 1996). Despite this, their regular and prolonged use may cause accumulation of this metal in the surface soil layers, which then reaches the lake with the runoff waters, melt waters or wind erosion. One of the potential sources of cadmium may also be the effluent from leaking cesspools, located in the area of farms and recreational plots in the vicinity of the lake (Bąk et al. 2013). In addition, Lake Szwałk Mały is very popular in Poland as a carp fishing area.

In this type of lakes, large amounts of groundbait are used. Therefore, it can be assumed that a certain load of cadmium gets into the lake with the bait, mainly to the littoral zone where carp has feeding areas. Additionally, a drainage ditch flows into the lake (about 0.7 m long) from the village of Szwałk, which receives part of the waters from fields and the asphalt road that runs along the shoreline of the lake.

The littoral sediments of Lake Kacze were also characterized by an increased content of cadmium. However, it should be noted that the catchment area of this lake is completely covered by forests and the lake has no surface flow. This small, shallow lake is different from the others by the highest content of organic carbon in the sediment. Therefore, it should be noted that the primary source of cadmium and other heavy metals in the sediments of Lake Kacze could be the weathering processes and bonding of metals, which occurs mainly with a considerable amount of allochthonous organic matter flowing from the catchment (Kabata-Pendias & Pendias 1999; Potarzycki & Maciejewska 2005). Cieślewicz & Różański (2010) found that the cadmium concentration was high (3.40 mg kg$^{-1}$) in the sediments of Slim Lake, which is located in the catchment dominated by deciduous forest.

In the present study, a strong and very strong correlation between TOC and Zn, Cu and Pb may indicate that these metals were bound to organic matter in all analyzed sediments. In addition, these metals were correlated with each other. Although cadmium generally did not correlate with organic carbon (r = 0.34 for all sediments) (Table 10), sediments from Lake Kacze were characterized by a strong correlation between Cd and TOC (r = 0.86 at $p<0.05$) (full data not shown). Cieślewicz & Różański (2010) also found a strong correlation (r = 0.87 at $p<0.05$) between TOC and cadmium in lake sediments. The concentration of organic carbon was high (89-386 g kg$^{-1}$) in these sediments and the total cadmium concentration varied between 0.73 and 3.04 mg kg$^{-1}$. Nickel correlated neither with TOC nor TIC in the sediments.

The correlations between heavy metals and TOC or between metal pairs in sediments were also indicated by other authors. Bojakowska & Krasuska (2014) showed that Pb concentration in sediments coming from the following lake districts: Great Poland, Pomeranian and Masurian was relatively well correlated with the total organic carbon (r = 0.59), aluminum (r = 0.45) and sulfur (r = 0.47) as compared to other analyzed chemical parameters. In the sediments from 13 lakes, Cieślewicz & Różański (2010) showed that the concentration of zinc, copper and lead was also positively correlated ($p<0.05$) with the organic carbon content. Such correlation was not observed for nickel. Similar relationships between the organic carbon content and zinc, copper, lead, and nickel were determined in the present study. If the correlation coefficient between the heavy metal factors is positive, these factors may have a common source and identical behavior during transport (Saeedi and Salmanzadeh, 2012).

## Summary

Due to the lack of any legal regulations regarding the permissible levels of heavy metals in the lake sediments in Poland, the commonly used method of assessing the sediment pollution is to compare the total metal concentrations with the geochemical criteria. A method using the ecotoxicological criteria (PEL TEL) or pollution indices, i.e. the geoaccumulation index ($I_{geo}$), the contamination factor (CF) and the contamination degree ($C_d$) could be more sensitive. A considerable effect on the final quality of sediments has the assumed background value for a given metal (GBP or GBE). As the GBP has been developed specifically for the Polish conditions, it seems to be more appropriate for assessing the purity of lake sediments in Poland. On the other hand, taking into account the very valuable region of Borecka Forest, the GBE can also be appropriate, especially in relation to the most toxic metals such as Cd.

In the present study, the lowest quality was determined for the littoral sediments from two lakes, Szwałk Mały and Kacze, based on the ecotoxicological criteria and pollution indices. The sediments from the other lakes of Borecka Forest were basically unpolluted, regardless of the method used. From the practical point of view, the most useful tool for assessing the quality of lake sediments seems to be the degree of sediment contamination ($C_d$). This index refers to the combined effect of all metals on the sediments. Thus, the overall sediment quality can be estimated and not the quality related to individual metals.

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