Archives of Environmental Protection
Vol. 47 no. 4 pp. 40–54

PL ISSN 2083-4772
DOI 10.24425/aep.2021.139501

How we can disrupt ecosystem of urban lakes – pollutants of bottom sediment in two shallow water bodies

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Keywords: Nutrients, pollution, heavy metals, PAH, bottom sediments, municipal sewage

Abstract: The research covered two lakes: Karczemne and Domowe Małe, which served as receivers for rainwater and municipal or industrial sewage. The sediment cores were obtained using a Kajak tube sampler. Analyses of HM, PAH and PCB were done by the AAS, ICP-AES and GC MS methods. OM, SiO_2, TH, Ca, Mg, CO_2, Fe, Al, Mn, TN and TP were measured. The research showed that the sediments of Lake Karczemne, into which the untreated municipal sewage was discharged, are characterized by a high content of P. It was found that the sediments accumulate toxins, OM and pollutants characteristic for various industries. Karczemne Lake which collected municipal and industrial wastewater, contained a high content of Pb, Cu and PAH in the sediments, and Domowe Małe Lake, receiving stormwater, contained high concentrations of PAH. Research has shown that one of the most important tools for selecting an appropriate method of lake restoration is the analysis of the spatial distribution of pollutants in the bottom sediments. Thanks to such an analysis of the composition of the bottom sediments and the correlation between the components of the sediments and their sorption properties, the restoration of the Karczemne Lake using the Ripl method was planned and the possibility of restoration of the Domowe Małe Lake in this way was eliminated.

Introduction

Good quality of water in all European societies is a special value that is reflected in legal acts concerning ecological policies of the states of the community. Among the legal conditions governing the preservation of natural heritage in the field of aquatic ecosystems, the most important document is the Water Framework Directive (DIRECTIVE 2000/60/EC of 23 October 2000). All Member States of the European Union adjust their legal acts to the guidelines developed in this document. One of the key problems is achieving or maintaining the so-called good water status. Poland, as a country with limited water resources, must intensively strive to achieve environmental goals defined for individual types of surface waters. Lakes play an important role in the circulation of water and matter in lakeland areas in Poland. They are particularly vulnerable to pollution, because a variety of human activities in their catchment area (agriculture, use of fertilizers and plant protection products, animal husbandry and breeding, burning garbage, cutting down forests, industry, urbanization) have changed their natural chemical settings. As a result of the transformation of the natural environment excessive loads of various categories of substances: nutrients, organic matter, pesticides and herbicides are introduced to water bodies (Algül and Beyhan 2020, Juśkiewicz et al. 2015). The result is a deterioration of water quality, decrease in biodiversity, the disappearance of habitats and ecological corridors, the vanishing of precious fish species and the influx of invasive alien species, including aquatic plants (Nasr et al. 2006). The most dangerous aspect of the civilization development is the transformation of lakes into receivers for rainwater, municipal and industrial sewage (Grochowska et al. 2014, 2019, Joniak et al. 2013). Huge loads of nutrients and organic substances (nitrogen and phosphorus), toxic substances and heavy metals (HM) are introduced to water with sewage (Wang et al. 2018, Wilson 2018, Sojka et al. 2018).

All substances introduced to surface water participate in various chemical and biological processes that lead to their sedimentation and accumulation in bottom sediment (Håkanson 1980). Most chemical pollutants, including nutrients, heavy metals and toxic organic compounds, accumulate in bottom sediments as a result of adsorption and co-precipitation and may be a secondary source of pollution (Håkanson 2014, Kowalczewska-Madura et al. 2011 2018). Binding of chemical compounds by the bottom sediment is not permanent and in favorable conditions comprising aerobic conditions, redox potential, pH, temperature, sediment's chemical composition, and the concentration gradient between the overlying and interstitial waters, the elements can be released to water again (Bing et al. 2011).

Heavy metals (e.g. Hg, Pb, Cd, Cr, Ni, Cu, Zn, Bi) occurring in lake ecosystem are very dangerous due to their toxicity, persistence and bioaccumulation (Waisberg et al. 2003, Bocca et al. 2004). Polychlorinated biphenyls are used, for example, in the production of transformers, plastics, protective paints and lubricants. They are ionic compounds with very low solubility in water, which are easily bioaccumulated in aquatic organisms (especially in mollusks), and they have a negative effect on plant growth, inhibiting the photosynthesis process and reducing the content of chlorophyll (Lidell et al. 2001, Jeremiason et al. 2011). Polycyclic aromatic hydrocarbons are produced during the processing of fuels, mainly coal and crude oil. These are the compounds that are harmful to aquatic organisms. They can be endogenously synthesized by some bacteria and plants (Jeremiason et al. 1999, Sanders et al. 1996).

In Poland and in the world, many different restoration techniques are used to restore the good condition of lakes. The bottom sediment removal (Ripl method) is a radical, but highly effective, restoration method for shallow, heavily degraded reservoirs (Ripl 1976a, 1976b, Zamparas et al. 2018).

During the design of lake restoration using Ripl method the spatial distribution of nutrients and organic pollutants, toxic compounds and heavy metals should be carefully examined. Knowing the sediment composition allows to accurately determine the sediment layer to be removed to obtain a clear improvement of the environmental conditions in the lake. Moreover, it is extremely important to know the content of toxic substances, because the sediment excessively loaded with such substances must be disposed of after being extracted from the bottom of the water body. The processing, management and utilization of deposits is often the biggest logistical and economic challenge.

The goal of the research was to analyze the content of mineral, organic and toxic pollutants in the bottom sediments of two urban lakes, which act as receivers for rainwater and municipal sewage.

Material and methods

Description of the research objects

Karczemne Lake is a flow-through, hypertrophic water body located in Kartuzy town (Kashubian Lake District), and Domowe Małe Lake is located in Western Masuria, in the Pasym group, in the physical and geographic area: Mrągowo Lakeland – Masurian Lakeland Voivodeship (Kondracki 2011) (Fig. 1). The lakes are situated in the Vistula basin.

An area of Karczemne Lake is 40.4 ha, and its maximum depth is 2.3 m (Table 1).

In the mid-1950s Karczemne Lake was transformed into municipal, storm and industrial (dairy and slaughterhouse wastewater, brewery, furniture and paint factories and petrol stations) sewage receiver. For over 30 years of the raw municipal sewage went to Karczemne Lake through six sanitary sewers (Fig. 2). In 1982, a mechanical-biological sewage treatment plant was launched. In the 1990s almost the entire city was connected to sanitary sewerage system, and it was only in 2010 that the management of storm water began to be organized by building settling tanks and storm water separators. Until 2018, the storm water drainage network in Kartuzy covered only part of the city – 17%. This situation meant that during heavy rainfall, local flooding occurred and the excess of rainwater and snowmelt from streets that did not have a rainwater drainage system penetrated the sanitary sewerage system, overloading it. The connection of the existing storm water drainage system to the sanitary sewerage system caused the raw sewage loading into the lake. From 2018 the rainwater has been discharged into the lake by six collectors. Actually, the total catchment area of Karczemne Lake covers 5.15 km^2 (57% forests and 43% urbanized

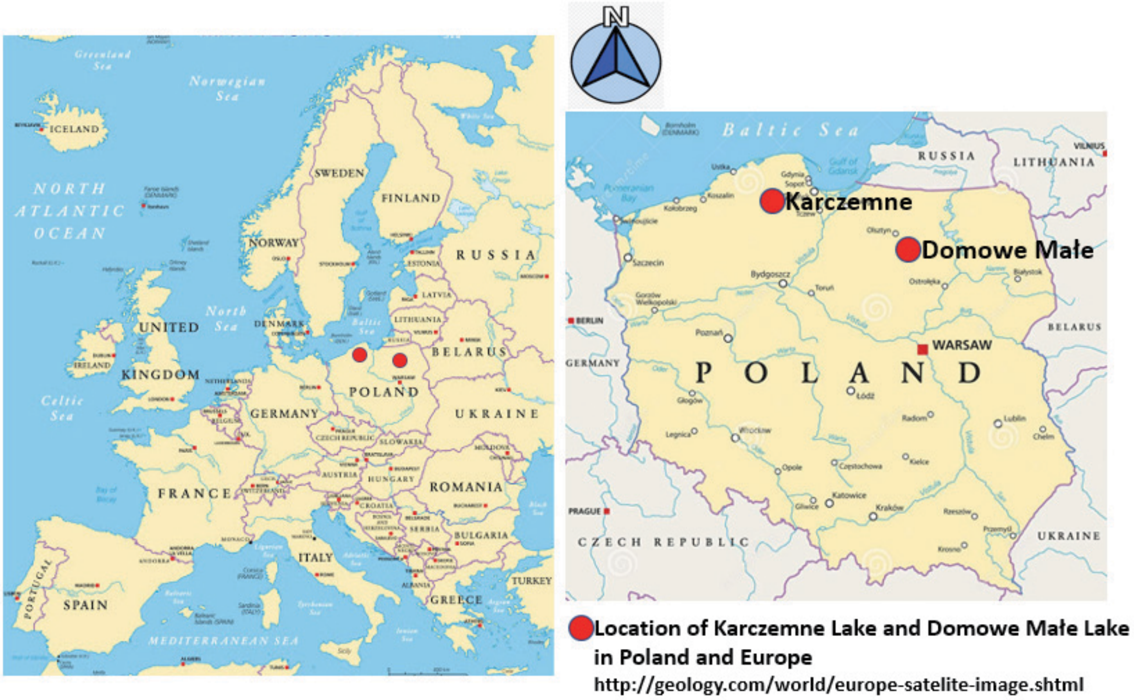

Fig. 1. Location of studied lakes in Poland and Europe

areas), and the direct catchment of the lake, excluding the area covered by the drainage system collecting rainwater, covers 0.45 km^2 (20.7% barren lands, 79.3% forests). The Kashubian Lake District, where Karczemne Lake is located, is covered with Quaternary sediments of glacial origin (Piasecki 1960). The geological structure includes: Jurassic and Cretaceous formations from the Mesozoic group as well as Tertiary and Quaternary formations from the Cenozoic era. The lower layer consists of sediment made of gray siltstones with dark inserts. There are marls, sands and sandstones, as well as loams above.

Domowe Małe Lake is a small, polymictic, flow-through urban lake. According to the latest measurements and morphometric studies (Wróbel 2012), the lake has a water table area of 11.4 ha and reaches a maximum depth of 4.9 m, with an average depth of 3.05 m (Table 1). The total catchment area of Domowe Małe Lake covers an area of approximately 5.90 km^2 (Planter et al. 2005) with majority of this area being the catchment of Domowe Duże Lake located above. Nearly 85% of this area is urban, and the remaining 15% is grassland. The catchment surface of Domowe Małe Lake covers about 1.2 km^2 and has been delineated taking into account the areas of the city from which rainwaters are drained. The main pollution factor in Domowe Małe Lake was rainwater discharged before the modernization of the rainwater drainage system by 15 collectors into the lake (Fig. 3).

Table 1. Morphometric coefficients of the examined lakes

Parameter	Karczemne	Domowe Małe
Geographical coordinates	54°19’42”N 18°11’27”E	53°34’02”N 20°59’41”E
Elevation of normal water table (m AMSL)	203.7	140.6
Lake area (ha)	40.4	11.8
Maximum depth (m)	3.2	4.9
Average depth (m)	1.98	3.05
Relative depth	0.0050	0.0142
Depth indicator	0.62	0.62
Volume (thousand m^3)	798.3	360.6
Maximum length (km)	1.28	0.49
Maximum width (km)	0.44	0.33
Elongation	2.9	1.5
Average width (km)	0.32	0.24
Lenght of shoreline (km)	3.2	1.4
Indicator of shoreline development	1.4	1.2

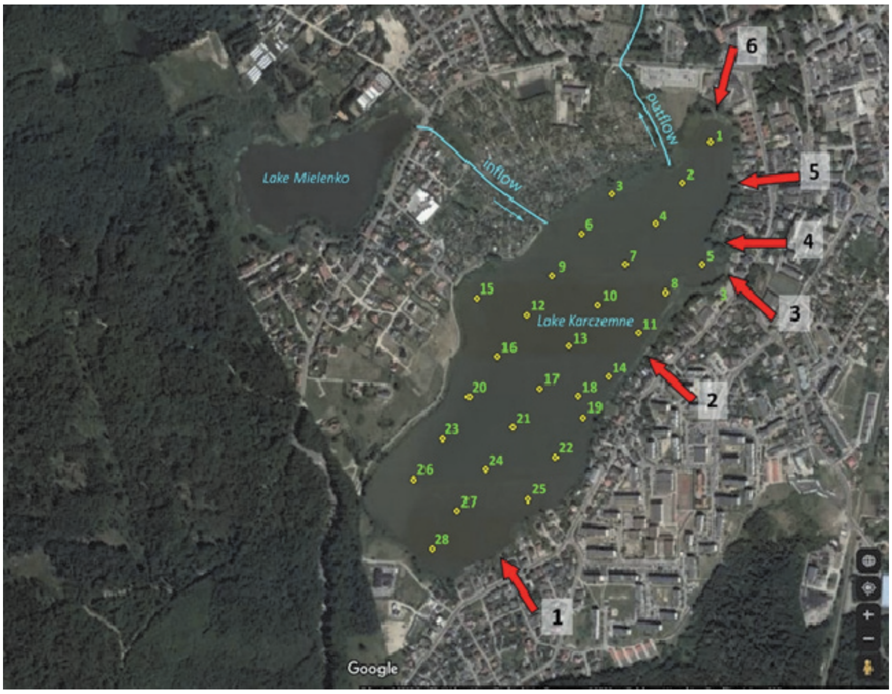

Fig. 2. Location of sampling points on Karczemne Lake (no. 1–28) and distribution of historical and modern rainwater outlets (red arrows)

Some of them, especially on the north side, captured rainwater from polluted areas, including a transport and machine base, a detention center and road junctions. Currently, there are 4 collectors equipped with pre-treatment units. The remaining inlets were removed, and the rainwater from about 50 ha of urban areas was redirected through the band sewage system to pre-treatment units and further introduced to the Domowy Canal located on the south-eastern edge of the lake, constituting the outflow from the lake (Łopata 2010). The Domowy Canal discharges water to the Sawica River.

In the area of the Masurian Lake District, where Domowe Małe Lake is located, glacial deposits lie on the Tertiary formations. In the ground there are Miocene loams and sands (Piaścik 1996).

Bottom sediment collection

The bottom sediment sampling was carried out on 7, 8, 9 and 11 March 2018 (Karczemne Lake) and 12, 13, 14 June 2018 (Domowe Małe Lake). The samples were collected at the research stations, the location of which is shown in Figs. 2, 3. The sediment cores were obtained using a Kajak tube sampler (KC Company, Denmark) and individual cores were divided into layers with a thickness of 30 cm (a – 0–30 cm, b – 30–60 cm, c – 60–90 cm, d – 90–120 cm, e – 120–150 cm, f – 150–180 cm) and confectioned as separate samples. It was assumed that if the lakes were restored by the method of bottom sediments removing, the thinnest layer to be removed would be 30 cm thick.

Studies of spatial variability of chemical properties of bottom sediment of studied lakes

The spatial distribution of bottom sediment sampling points of research lakes were planned according to EPA (2001). In accordance with the guidelines included in the Ordinance of Ministry of the Environment (2015) on waste recovery outside installations and equipment six aggregate samples were prepared for analyzing the content of POPs, PAHs and HM in the bottom sediments. Karczemne Lake: No. 1 – homogenized sample 1–15, layer 0–60 cm, No. 2 – homogenized sample 1–15, layer 60–120 cm, No. 3 – homogenized sample 16–28, layer 0–60 cm, No. 4 – homogenized sample 16–28, layer 60–120 cm, No. 5 – homogenized sample 5, layer 0–60 cm, No. 6 – homogenized sample 5, layer 60–120 cm. Domowe Małe Lake: No. 1 – homogenized sample 1–20, layer 0–60 cm, No. 2 – homogenized sample 1–20, layer 60–120 cm, No. 3 – homogenized sample 1–20, layer 120–180 cm.

Analyses of HM, POPs and PAHs were done by the AAS, ICP-AES and GC MS methods. The coefficient of variation (CV) for the repeated analysis was 2% (Kaca 2003). The usefulness of deposits for development was verified on the basis of the Ordinance of the Ministry of the Environment of 1 September 2016 on the method of conducting an assessment of the soil surface pollution (Journal of Laws of 2016, item 1395) and concentrations of toxic substances caused by pollution.

The sediment, after being transported to the laboratory, was weighed to determine the volume and density of the fresh sample. Then the samples were dried to constant weight, and the volume and density were redetermined (Brzozowska and Gawrońska 2009). The following parameters were measured in bottom sediments: organic matter – OM (as a loss on ignition at 550°C after regeneration CO_2 saturated water – weight analysis), silica – SiO_2 (weight analysis after preliminary digestion of a sediment sample in a mixture of a strong mineral acids: H_2SO_4, $HClO_4$, HNO_3 1:2:3), TH (titration methods with $C_{10}H_{14}N_2O_8Na_2$ and $C_{20}H_{12}N_3O_7SNa$, alkaline environment)

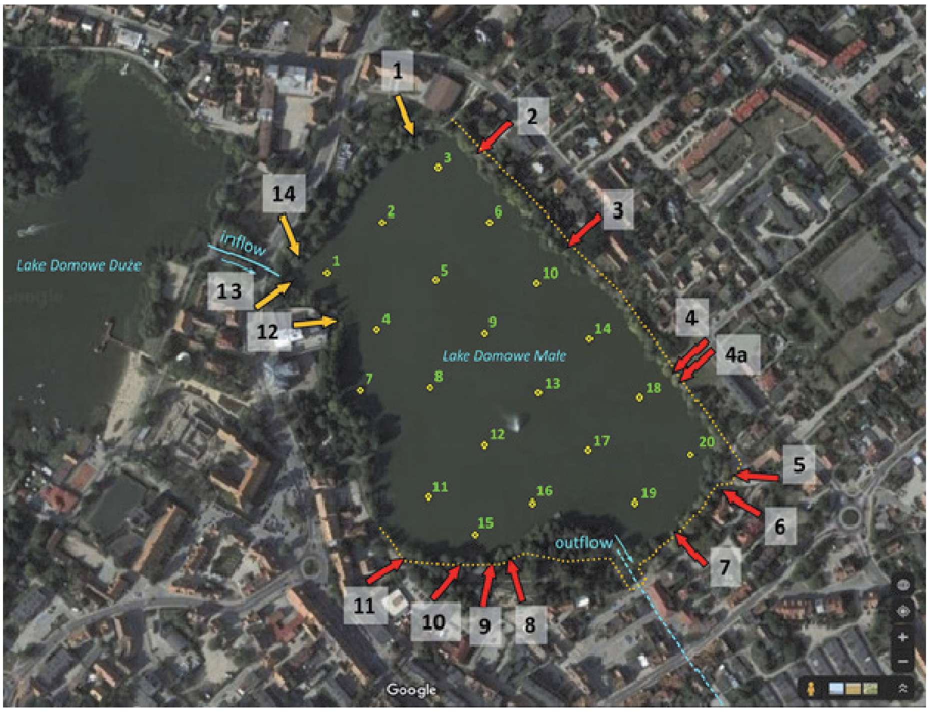

Fig. 3. Location of sampling points on Lake Domowe Małe (no. 1–20) and distribution of historical (red arrows) and modern (orange arrows) rainwater outlets. A dotted line marks the newly built collector that intercepts most of the storm sewage

Ca – as CaO (titration methods with $C_{10}H_{14}N_2O_8Na_2$ and $C_8H_8N_6O_6$, alkaline environment), Mg – as MgO (difference in total hardness and calcium concentration), CO_2 (as a loss on ignition at 1000°C), Fe – as Fe_2O_3, Al – as Al_2O_3, Mn – as MnO (spectrophotometric method, Merck's tests), TN – N (by distillation method in a Büchi B-324 apparatus of a previously mineralized sediment by Kjeldahl method), TP – as P_2O_5 (spectrophotometric method, Merck's tests). Chemical analyses of bottom sediments were performed in accordance with Hermanowicz et al (1999).

Some of the results of the research on bottom sediments from Karczemne Lake (i.e. the content of TP and POPs, PAHs and HM) were taken from the publication of Grochowska et al 2020.

The results were statistically analyzed (mean values, standard deviation, standard error, one-way ANOVA, p=0.05, Tukey's HSD) using a Statistica 13.3 software package (2018). The alternative tested hypothesis was the presence of significant differences of mean values of chemical components of first 0–30 cm layer of bottom sediments and mean values of these components in layers: 30–60 cm, 60–90 cm, 90–120 cm, 120–150 cm, 150–180 cm. The relationships between particular sediment compounds were analyzed using correlation coefficient (n=9). An assessment was made using Statistica 13.3 software package (Tibco Inc., Palo Alto, CA, USA).

Results and discussion

Organic and inorganic suspensions settling to the bottom of water bodies may be of both autochthonous and allochthonous origin – a substance flowing into the lake from the surrounding catchment with surface runoff or formed as a result of the destruction of the banks (silica, clays, loams, detritus, silt, gravel, sand) (Augustyniak et al. 2017).

The mean contents of chemical composition in bottom sediments of Karczemne Lake was quite variable. The sediments that accumulated in the central part of the lake basin, up to a depth of 60 cm, can be considered mixed according to the Stangenberg nomenclature (Augustyniak et al. 2017) because none of the components was present in amounts exceeding 50% of the dry mass (Figs. 4, 5).

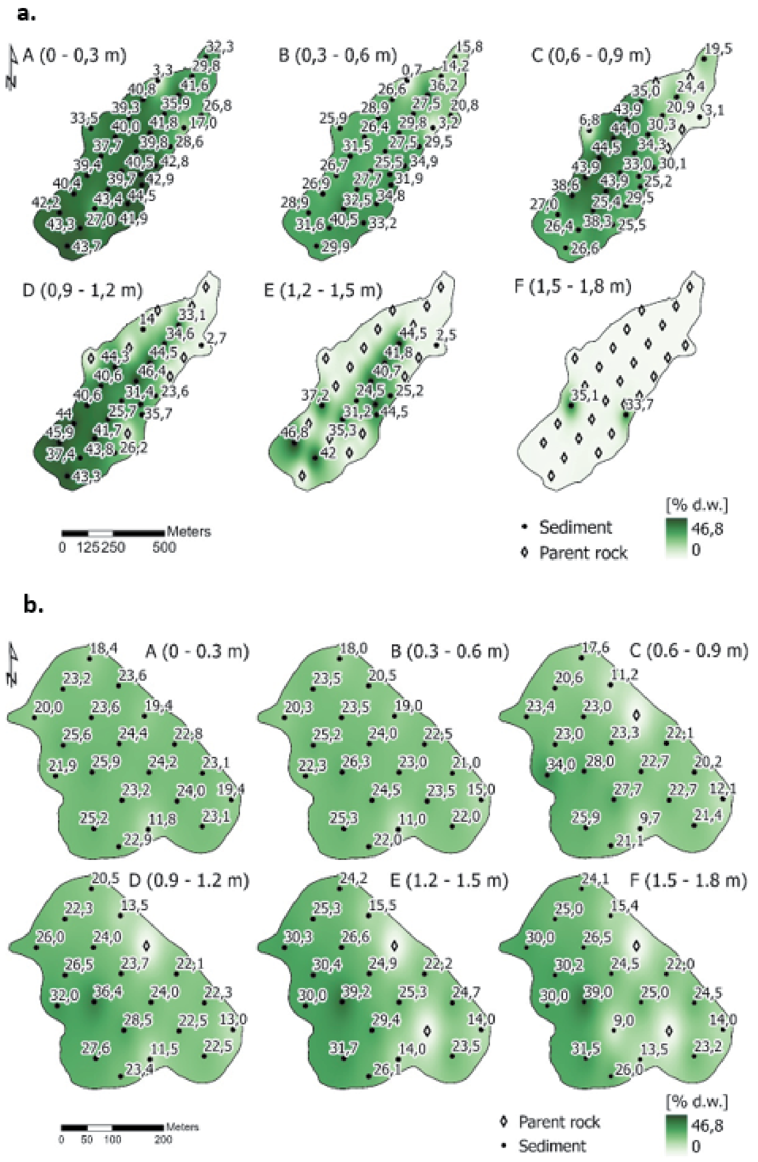

Fig. 4. The spatial distribution of organic matter (% of OM in d.w.) in bottom sediments of studied lakes (a – Karczemne Lake, b – Domowe Małe Lake)

In the coastal regions of the lake and in deeper levels of sediment from the central part of the lake bowl, the dominant component was silica, accounting for up to 92% of the dry mass. These bottom deposits can be described as silicate. The organic matter in these sediments ranged from 15.8 to 43.5% of the dry mass (Figs. 4, 5). In the case of Domowe Małe Lake, average chemical composition of bottom sediments was not significantly differentiated. The sediments accumulated in the entire body of water are mainly of silicate nature (Prosowicz 2008), because the proportion of silicates usually exceeded 50% of the dry mass composition of the sediment. Only at deeper sites (in layer a), the content of silicates was slightly lower than 50% (Fig. 5). The source of silica in the bottom sediment of lakes are primarily dead diatom cells (Liu et al. 2014, Liu et al. 2019), and the increased silica content in deposits of Domowe Małe Lake is probably related to the inflow of rainwater, containing significant amounts of sand washed from the surface roads and streets. The organic matter in these sediments comprised 9.74 to 39.18% of dry mass (Fig. 4). Contamination of the reservoir with organic matter flowing in from rain collectors in the past, before the modernization of the sewage system, was visible in the form of a higher share of organic matter in the deepest collected layer of sediment (c).

In the process of increasing the fertility of lakes, the content of nutrients – nitrogen and phosphorus – plays a particularly important role. It is estimated that about 90% of the total amount of these two elements in the lake's ecosystem is stored in upper layer of bottom sediments. Nitrogen is transferred from the water to the sediment mainly in the form of sedimenting allo- and autochthonous matter (Augustyniak et al. 2017). The amounts of N in the sediments of Karczemne Lake varied from 0.20% to 2.50% N in dry weight (Fig. 6).

Lower concentrations of nitrogen were found in the northern part of the lake. In the rest of this water body, the bottom deposits were richer in nitrogen, especially in the areas of the maximum depth of the basin. The amounts of nitrogen in the bottom sediments of the Domowe Małe Lake varied from 0.47% to 2.0% N in d.w. Higher nitrogen contents were found where more organic matter was deposited. Statistical analysis

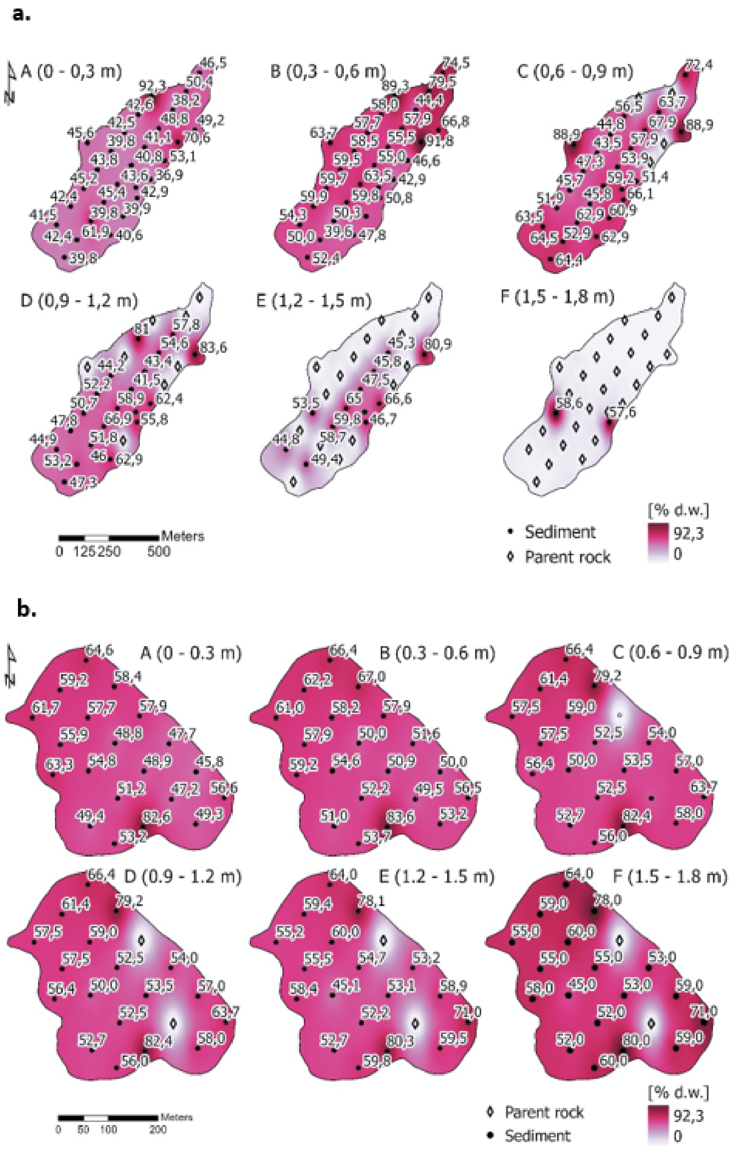

Fig. 5. The spatial distribution of silica (% of SiO_2 in d.w.) in bottom sediments of studied lakes (a – Karczemne Lake, b – Domowe Małe Lake)

did not show any significant differences in nitrogen content between particular layers of bottom sediments of the studied lakes (Table 2).

The process of phosphorus deposition in bottom sediments is more complicated. From the water column it goes to the sediments mainly as a result of coagulation and sorption by active iron, manganese and aluminum hydroxides, as well as a result of sedimentation with organic matter and mineral suspensions, which serve as the basis for the crystallization of apatite (Augustyniak et al. 2019).

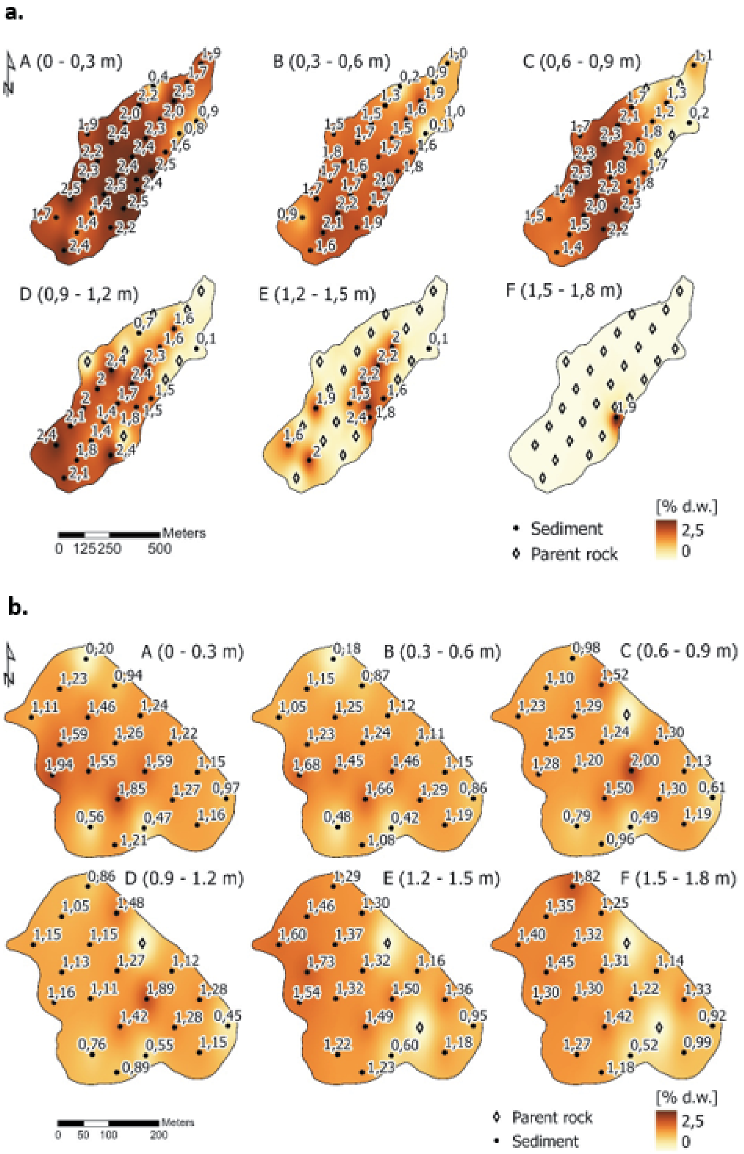

Fig. 6. The spatial distribution of nitrogen (% of N in d.w.) in bottom sediments of studied lakes (a – Karczemne Lake, b – Domowe Małe Lake)

Table 2. Results of one – way ANOVA analyses for investigated variables in bottom sediments of Karczemne and Domowe Małe lakes

VARIABLE	Karczemne		Domowe Małe	
	F values	p values	F values	p values
CaO	8.724885	0.000001	1.652789	0.155546
MgO	0.825046	0.534576	2.725275	0.025095
CO_2	11.42967	0.000000	12.12495	0.000000
Fe_2CO_3	5.190431	0.000267	4.057341	0.002452
Al_2CO_3	0.826815	0.533351	0.879362	0.498830
N	2.255090	0.054130	2.316618	0.050924
P	28.69913	0.000000	1.555680	0.182037

Bold – statistically significant differences in the content of the chemical component in bottom sediments between the 0–30 cm layer and the following layers: 30–60 cm, 60–90 cm, 90–120 cm, 120–150 cm, 150–180 cm

It is assumed that the amount of phosphorus in sediment is determined mainly by the sorption capacity of the sediments, which depends on the content of iron, manganese and aluminum, i.e., the elements that make up the so-called sorption complex, clay minerals and organic matter. On the other hand, silicates show an antagonistic effect in relation to phosphorus sorption. Taking into account the silicate nature of most of the sediments of the Domowe Małe Lake, the amount of deposited phosphorus is average, despite the high pollution of the lake. The content of total phosphorus in the sediments ranged from 0.16% P_2O_5 in d.w. up to 0.89% P_2O_5 in d.w. (Fig. 7). The bottom sediment of Karczemne Lake contained high amounts of P, ranging from 0.1 to 7.3% of the dry weight, and the maximum amount was found at research point number 5: 31.856 mg P g^{-1} d.w. in the surface sediments of layer a (0–30 cm) (Fig. 7). Such extremely high concentrations of phosphorus in bottom sediments have not been recorded in any other lake in the world. Statistical analysis showed significant differences in the phosphorus content between the layers of the collected sediments (Table 2). This situation is related to the municipal sewage inflow. The highest amounts were recorded in the surface layer – 30 cm, which was formed during the period of the greatest pollution, associated with the expansion of the city, and thus – with increased sewage inflow. High concentrations of phosphorus were found in bottom sediments formed near the mouths of sewage collectors. According to Grochowska et al. (2020) the analysis of particular phosphorus percentages of the total phosphorus showed that the dominant types were phosphorus associated with organic matter (up to 80%) and calcium-related phosphorus (up to 45%). Residual phosphorus made up 25% of TP at some research points. Phosphorus bound mainly with aluminum at some sites occurred in higher amounts, exceeding 20% of TP. The most mobile fractions of phosphorus – phosphorus loosely bound to sediment and phosphorus sensitive to redox potential changes made up a small percentage of the total phosphorus (typically only a few percent).

The content of calcium and magnesium in the sediments of Karczemne Lake was low: from 1.0 to 7.4% CaO in the dry weight of the sediment and from 0.2 to 3.5% MgO in

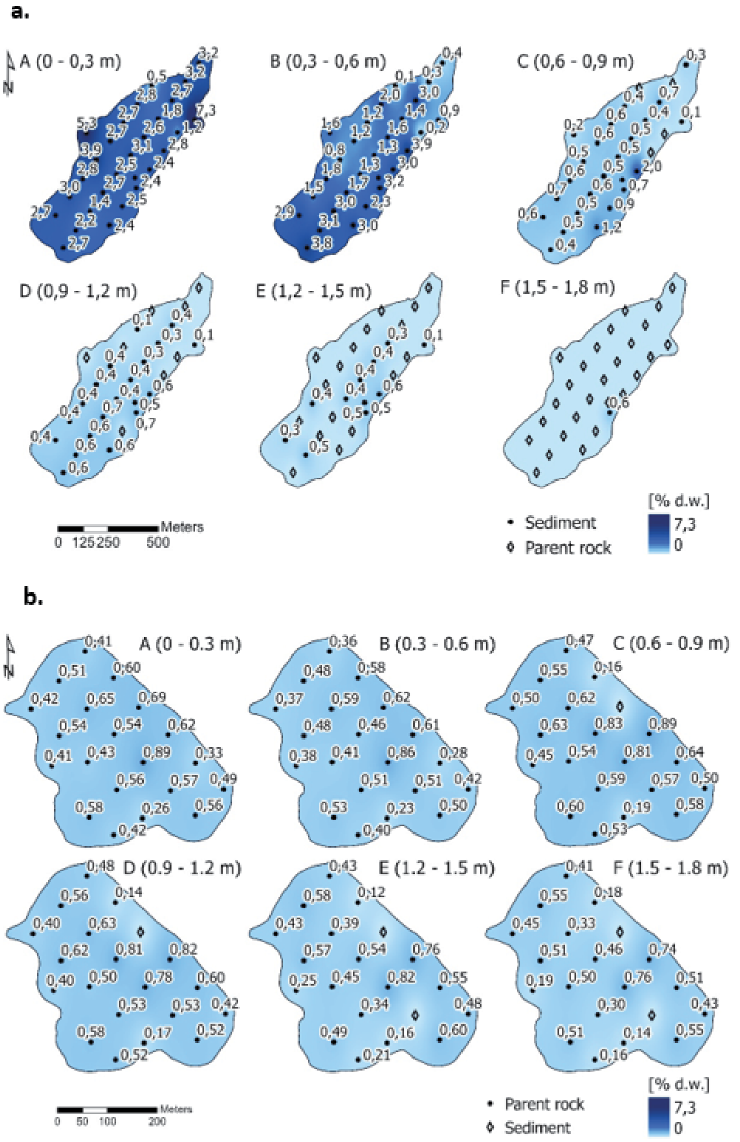

Fig. 7. The spatial distribution of phosphorus (% of P_2O_5 in d.w.) in bottom sediments of studied lakes (a – Karczemne Lake, b – Domowe Małe Lake)

the dry weight of the sediment, and they were mainly in the form of carbonates. The content of calcium in the sediments of Domowe Małe Lake ranged from the values considered low – 1.11% CaO in d. w. through the average, even above 10% CaO in d.w., to a maximum of 12.49% CaO in d.w. Magnesium contents in Domowe Małe Lake were small and ranged between 0.12% MgO in d.w. and 3.42% on d.w. Calcium and magnesium were mainly in the form of carbonates. This is evidenced by the close relationship between calcium, magnesium and carbon dioxide, which constituted from 0.48 to 9.34% d. w. (Fig. 8).

The main components of the sorption complex of the bottom sediments of Karczemne Lake were iron (approximately 3.5% d.w.) and aluminum (approximately 3.0% d.w.). The phosphorus compounds with iron are unstable due to the variable valence of iron (Jansson 1987, Roden and Emonds 1997). Under anaerobic conditions, iron turns into a reduced form, releasing phosphates into the water. In the dry weight of Domowe Małe Lake bottom sediments, the elements building the sorption complex were present in the following amounts: from 0.01 to 0.15% MnO, from 0.53 to 3.26% Fe_2O_3 and from 0.54 to 3.19% Al_2O_3 (Fig. 8). Taking into account the results of the studies by Augustyniak et al (2019), bottom sediments of Karczemne Lake are characterized by high sorption capacity, while Domowe Małe Lake has average sorption capacity.

The chemical composition of the sediments accumulating in a lake is the result of many factors, including lithological structure of the catchment, the type of soil cover and topography, climatic conditions and internal factors in the reservoir. In urbanized areas, the chemical composition of lake sediments depends on various forms of anthropopressure, these are: industrial and municipal sewage discharges, leaks from landfills, dust and gas pollution of the atmosphere (Birch and Taylor 1999). Determination of the degree of bottom sediments contamination with heavy metals based on world or national standards (Migaszewski and Gałuszka 2003, LAWA 1998) is not always sufficient because of local geochemical anomalies. Lithology can be different even within the same research area. It should be taken into account the type of sediment, environment of sedimentation and granulometric fraction of the sediment which was proven in the sediments of lake Wigry (Prosowicz 2008).

With regard to the geochemical background for the lakes of the Kashubian Lake District (Bojakowska and Sokołowska 1996), there were found increased levels of some elements (Cd, Cu, Cr, Hg, Pb and Zn) in the sediments of the nearby Klasztorne Lake (the same group of lakes in Kartuzy). Our research confirmed this observation in relation to Karczemne Lake. The spatial distribution of pollutants in the bottom sediments proves the impact of anthropopressure, especially in the discharge of sewage from industrial plants.

In bottom sediments of Karczemne Lake in vertical and horizontal distribution of As, Cr, Zn, Ni and Hg content, exceedances beyond the limit values were not found. An acceptable standards were exceeded in the case of Cu and Pb. In samples No 1 – No 4, Cu content varied in the range: 25.9–123.0 mg/kg d.w., and Pb content varied from 31.6 to 126.0 mg/kg d.w. In sample No 5, Cu content was 247 mg/kg d.w., compared to the standard of 150 mg/kg d.w.; and Pb – 317 mg/kg d.w., compared to the standard of 200 mg/kg d.w. (Table 3).

At site 5 (sample No. 5), where industrial plants (paint factory, brewery, slaughterhouse) and a fuel and equipment station for buses were located in the adjacent area, the highest concentrations of the following metals were usually found: Cr, Zn, Cd, Cu, Pb and exceeding the permissible standards (according to OME 2015, 2016) for Cu and Pb. According to LAWA (1998), this classifies the surface layer of bottom sediments as heavily polluted (class III). The deeper layer is not contaminated (class I). It should be noted that the selective treatment of the site 5 does not include the physical and chemical differences in bottom sediments collected from different areas of sedimentation. However, it clearly shows the influence of anthropogenic pollution, with pairs of co-precipitating metals Cd / Cr, Cu / Cr and Cd / Cu characteristic for detritus gyttja (Prosowicz 2008). In an industrialized area, the content of lead, copper and mercury decreases with distance from the coastline (Kishe and Machiwa 2003).

The highest concentrations of As, Ni, Hg were recorded in sample no. 3 (from the upper, south-western part of the lake). In this area, the content of Cr, Zn, Cd and Cu was clearly higher than in the north-eastern part (samples No. 1 and 2), which

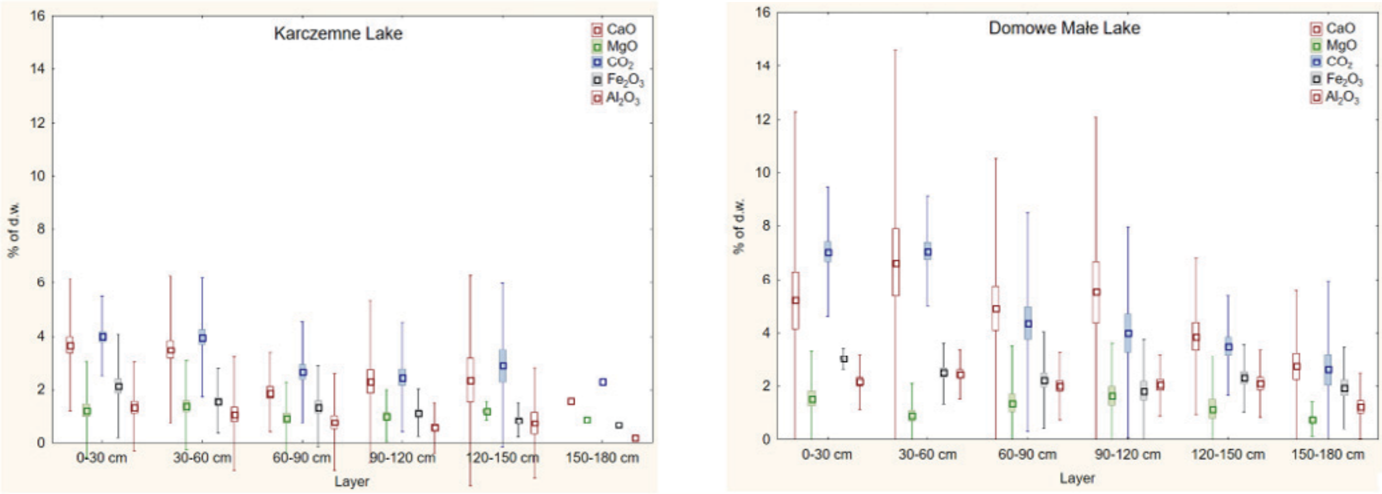

Fig. 8. The content of CaO, MgO, CO_2, Fe_2O_3 and Al_2O_3 (as% in d.w.) in bottom sediments of studied lakes

Table 3. Organic pollutants and heavy metal contents in bottom sediments of studied lakes

Parameter	Norm	Karczemne						Domowe Małe		
		Sample No. 1 mean/SD	Sample No. 2 mean/SD	Sample No. 3 mean/SD	Sample No. 4 mean/SD	Sample No. 5 mean/SD	Sample No.6 mean/SD	Sample No. 1 mean/SD	Sample No. 2 mean/SD	Sample No. 3 mean/SD
As	<30 mg/kg d.w.	9.8/0.98	5.0/–	13.3/1.30	6.0/0.61	12.7/1.30	5.5/0.55	20.8/2.10	14.3/1.40	9.4/0.95
Cr	<200 mg/kg d.w.	29.1/5.5	18.8/2.3	40.1/4.8	33.2/4.0	44.1/5.3	16.7/2.0	38.7/4.6	28.8/3.5	19.8/2.4
Zn	<1000 mg/kg d.w.	406/65	104/17	503/80	152/24	843/130	34.9/5.6	271/43	202/32	131/21
Cd	<7.5 mg/kg d.w.	0.543/0.08	0.050/–	0.794/0.12	0.050/–	0.846/0.13	<0.05/–	<0.05/–	<0.05/–	<0.05/–
Cu	<150 mg/kg d.w.	93.5/9.4	25.9/2.6	123.0/12.0	38.4/3.8	247.0/25.0	10.5/1.1	56.2/5.6	62.2/6.2	50.2/5.0
Ni	<75 mg/kg d.w.	19.9/2.0	13.8/1.4	25.8/2.6	21.9/2.2	22.5/2.3	10.8/1.1	24.8/2.5	16.1/1.6	15.1/1.5
Pb	<200 mg/kg d.w.	97.2/9.7	31.6/3.7	126.0/13.0	49.4/4.9	317.0/32.0	9.9/0.9	119.0/12.0	76.5/7.7	46.4/4.6
Hg	<1.0 mg/kg d.w.	0.60/0.09	0.22/0.03	0.68/0.10	0.26/0.04	0.30/0.04	<0.05/–	0.56/0.08	0.46/0.07	<0.05/–
Benzo(a)pyrene	<1.0 mg/kg d.w.	1.21/0.30	0.41/0.03	2.10/0.53	0.39/0.10	9.75/2.40	<0.01/–	7.52/1.90	1.16/0.29	0.62/0.16
Benzo(a)anthracene	<1.5 mg/kg d.w.	1.13/0.28	0.36/0.09	1.69/0.42	0.24/0.06	13.00/3.30	0.09/0.02	6.00/1.50	1.370.34	0.67/0.17
Benzo(b)fluoranthene	<1.5 mg/kg d.w.	1.88/0.47	0.69/0.17	3.57/0.89	0.55/0.14	14.40/3.60	<0.02/–	12.10/3.00	2.62/0.66	1.04/0.26
Benzo(k)fluoranthene	<1.5 mg/kg d.w.	0.80/0.20	0.27/0.07	1.38/0.34	0.25/0.06	5.77/1.44	<0.01/–	4.90/1.23	1.12/0.28	0.47/0.12
Benzo(g,h,i)perylene	<1.0 mg/kg d.w.	1.44/0.36	0.55/0.14	2.65/0.66	0.72/0.18	7.34/1.80	0.24/0.06	7.99/2.00	2.28/0.57	0.53/0.13
Dibenzo(a,h)anthracene	<1.0 mg/kg d.w.	0.30/0.07	0.02/–	0.48/0.12	0.02/–	3.24/0.81	<0.02/–	1.69/0.42	0.49/0.12	<0.02/–
Indeno(1,2,3-cd)pyrene	<1.0 mg/kg d.w.	1.75/0.44	0.98/0.24	1.86/0.47	0.92/0.23	8.03/2.00	0.13/0.03	12.75/–	3.19/0.80	1.43/0.36

indicates the industrial character of the drained area. Nickel and lead concentration could be linked to particulate matter transported in storm water runoff from urban and industrial areas (Wakida et al. 2008).

The research by Fu et al. (2014) showed that there is a close relationship between ammonium nitrogen, total nitrogen, orthophosphates, total phosphorus, COD-Mn, and metal sedimentation. The positive correlations between trace metals, organic matter and sediment granulometry were confirmed by Bartoli et al. (2012) in the river sediments. The role of organic matter and sediment grain size in relation to the accumulation of heavy metals in sediments has been emphasized (Wakida et al. 2008). According to Grochowska et al. (2020), site 5 was characterized by an extreme amount of total phosphorus, that has never been recorded anywhere else – 31.856 mg P/g in d.w. Our research showed the high concentrations of these components in bottom sediments, lake water and inflows. A significant ($p < 0.05$) correlation between phosphorus and all determined metals was confirmed, but no correlation was found between metals and organic matter (Fig. 9). It can be concluded that the presence of heavy metals in bottom sediments favors phosphorus binding, as evidenced by its high levels in the sediment of Karczemne Lake. Research made by Bojakowska and Sokołowska (1996) shows that heavy metals form easily migrating complexes with humic and fulvic acids. Probably the type of organic matter lying at the bottom of the analyzed lakes is not conducive to the binding of heavy metals.

The bottom sediments of Lake Karczemne, according to LAWA (1998), belong to class II – moderately polluted only in terms of nickel content. The content of the remaining metals classifies them as heavily contaminated, and even, in the case of cadmium – heavily/very heavily contaminated. Cadmium is the most mobile among the determined heavy metals. It accumulates both in plant and animal organisms, and is introduced into the food chain (Barbusiński and Nocoń 2011). Many authors emphasize that toxicity tests are a good complement to chemical tests in procedures for assessing the quality of bottom sediments and the environmental risk arising from the presence of chemicals (Mamindy-Pajany et al. 2011). Moreover, the release of heavy metals from bottom sediments is highly dependent on their different chemical forms, which generally exhibit different physical and chemical properties in terms of chemical interactions, potential toxicity, bioavailability and mobility (Gabarrón et al. 2017, Kang et al. 2017). Heavy metals in bottom sediments can migrate up to the sediment--water interface and they can be released to the overlying water when environmental conditions (e.g. DO, redox potentials and temperature) change, leading to potential hazards to the aquatic environment and organisms (Dhanakumar et al 2015, Chen et al. 2017).

In bottom sediments of Domowe Małe Lake in vertical and horizontal distribution of As, Cr, Zn, Ni, Cu, Pb and Hg content, exceedances beyond the limit values were not found. The As content varied in the range: 9.45–20.8 mg/kg d.w., Cr content varied in the range: 19.8–38.7 mg/kg d.w., Zn from 131.0 to 271.0 mg/kg d.w., Cd was below 0.05 mg/kg d.w., Ni content varied in the range 15.1–24.8 mg/kg d.w., and Hg content from 0.05 to 0.56 mg/kg d.w., Cu from 50.2 to 62.2 mg/kg d.w. and Pb between 46.4 and 119.0 mg/kg d.w. (Table 3).

According to OME (2016), the bottom sediments in Domowe Małe Lake do not exceed the limit values for contamination with heavy metals. According to the LAWA (1998) classification, the content of chromium, cadmium and nickel was in the class I (non-contaminated), the content of copper and zinc was in the class II (recommended limit values for contamination), which indicates a low environmental load with these elements. The lead content was higher and it increased with the depth: in layer a – class I, b – class I–II (uncontaminated/moderately polluted). A likely reason for the low loading of heavy metals is the silicate nature of the bottom sediments. They are characterized by a low content of organic matter (in the upper, layer a,

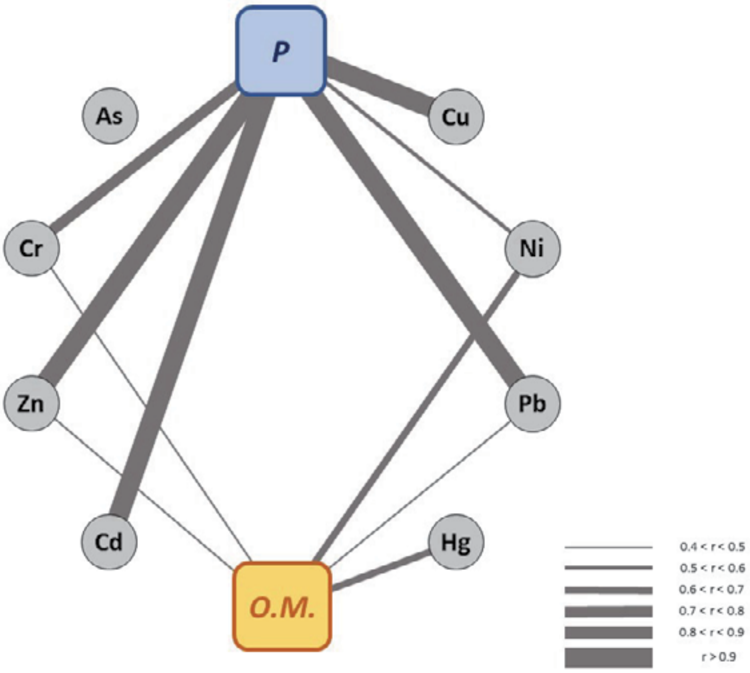

Fig. 9. Correlation strenght between HM content and phosphorus and organic matter concentrations

11.79–25.9% in dry weight) and moderate amounts of nitrogen and phosphorus. That does not support the deposition of heavy metals (Wakida et al. 2008, Fu et al. 2014). There were no significant correlations between organic matter, phosphorus and the heavy metals determined. This allows us to conclude that organic matter promotes metal sedimentation, but does not determine it. The percentage content of organic matter increased in the deeper layers. They contained more total phosphorus, but historically the use of the catchment was so favourable and it did not supply heavy metals. Significant correlations were found between the coexisting metals (As / Cr and As / Pb 0.998, Cr / Zn 0.999, Cr / Pb 0.997 $n = 3$, $p < 0.05$), proving their common origin.

Polycyclic aromatic hydrocarbons (PAHs) occurring in the environment can be products of natural processes, such as pyrolysis of terrestrial organic matter during natural fires, diagenetic changes of organic material in the process of coal and oil formation or during volcanic eruptions. They can also be a product of the activity of living organisms (synthesis by plants, the activity of microorganisms that decompose dead organic matter) (Alves et al. 2010, Bojakowska et al. 2012).

In bottom sediments of Karczemne Lake, in sample No. 1 exceedances were found in the content of benzo(a)pyrene, benzo(b)fluoranthene, benzo(g,h,i)perylene and indeno(1,2,3-cd)pyrene (Table 3). In sample No. 3 exceedances were found in the content of benzo(a)pyrene, benzo(a)anthracene, benzo(b)fluoranthene, benzo(g,h,i)perylene and indeno(1,2,3-c,d) pyrene. In sample No. 5 exceedances were found in the range of benzo(a)pyrene, benzo (a)anthracene, benzo(b) fluoranthene, benzo(g,h,i)perylene, benzo(k)fluoranthene, dibenzo(a,h)anthracene and indeno(1,2,3-c,d)pyrene (Table 3). In bottom sediments of Domowe Małe Lake, in sample No. 1, the content of benzo(a)pyrene was exceeded – 7.52 mg/kg d.w., benzo(b)fluoranthene – 12.1 mg/kg d.w., benzo(g,h,i)perylene – 7.99 mg/kg d.w., indeno(1,2,3-c,d)pyrene – 12.75 mg/kg d.w., benzo(a)anthracene – 6.0 mg/kg d.w., benzo(k)fluoranthene – 4.90 mg/kg d.w., and dibenzo(a,h)anthracene – 1.69 mg/kg d.w. In sample No. 2 the content of benzo(a)pyrene was exceeded – 1.16 mg/kg d.w., benzo(b)fluoranthene – 2.62 mg/kg d.w., benzo(g,h,i)perylene – 2.28 mg/kg d.w., indeno(1,2,3-c,d)pyrene – 3.19 mg/kg d.w. In sample No. 3 only the content of indeno(1,2,3-c,d)pyrene was exceeded – 1.43 mg/kg d.w. (Table 3).

Combustion of fossil fuels, their processing and discharge of municipal or industrial wastewater are the main sources of anthropogenic emissions of these compounds to the environment. In the aquatic environment, they can accumulate in the bottom sediment of water bodies, therefore they are used as indicators of anthropogenic pollution (Cappacioni et al. 1995, Stogannis and Laane 2015).

The sediments of the investigated urban lakes contained high levels of ΣPAHs in the surface layer, exceeding the maximum values given by Bojakowska et al. (2012) in the group of 150 Polish lakes (53.391 µg/kg d.w.). In Karczemne Lake a maximum amount of ΣPAH was 61.500 µg/kg d.w., and in Domowe Małe Lake – 81.500 µg/kg d.w. This proves the serious anthropogenic pollution of the studied lakes over the last several decades. The deeper sediment layers of the examined lakes did not exceed the safe level of the analyzed PAHs (Table 2).

Ratios between selected PAHs can be used to identify the sources of emissions of these pollutants (Stogiannis et al 2015, Pohl et al 2018, Katsoyiannis et al 2007, Yunker et al 2002).

Based on the available results, the following ratios were calculated for the sediments of the studied lakes: IcdP-indeno(1,2,3-c,d)pyrene, BghiP-benzo(g,h,i)perylene, BbF-benzo(b)fluoranthene; BkF-benzo(b)fluoranthene, BaP-benzo(a)pyrene (Table 4). The obtained results indicate traffic pollution (products of combustion of liquid fuels – gasoline, diesel, lubricants and particles of worn tires), as well as emissions from combustion of other fuels (coal, wood) as the main sources of PAHs pollution in the surface layer of bottom sediments of the studied lakes. The analysis of correlation showed that the contents of the four, five- and six-ring hydrocarbons in the bottom sediments of the analyzed lakes are closely correlated ($r \geq 0.8$, $p < 0.05$), which, according to Bojakowska et al. (2012), can confirm their common origin from combustion processes. Moreover, the content of the tested PAHs is significantly correlated mainly to the content of Pb and Cr (with exception of indeno(1,2,3-c,d)pyrene) and benzo(a)anthracene), as well as to As (with exception of benzo(a)pyrene, benzo(a)anthracene and dibenzo(a,h)

Table 4. Selected PAH ratios in the bottom sediment of analyzed lakes comparing to literature references

PAH ratio	Reference value range	Origin	Reference	Karczemne (min-max)	Domowe Małe (min-max)
Indeno(1,2,3-c,d)pyrene / (Indeno(1,2,3-c,d)pyrene + Benzo(g,h,i)perylene)	< 0.2 0.2–0.5 > 0.5	Petrogenic Petroleum combustion Grass, wood and coal combustion	Yunker et al. 2002	0.3–0.6	0.6–0.7
Indeno(1,2,3-c,d)pyrene / Benzo(g,h,i)perylene	<0.25 >0.25 0.25–1.0 >1	Strong petroleum input Pyrogenic sources Mixed sources Combustion sources	Stogiannidis and Laane 2015	0.7–1.8	1.4–2.7
Benzo(b)fluoranthene / Benzo(k)fluoranthene	2.5–2.9 3.7 1.3–1.6	Aluminum smelter emissions Coke oven Gasoline and diesel combustion	Yunker et al.2002; Stogiannidis and Laane 2015	2.0–2.5	2.2–2.5
Benzo(a)pyrene / Benzo(g,h,i)perylene	<0.6 >0.6	Non-traffic emissions Traffic emissions	Katsoyiannis et al. 2007	0.04–1.3	0.5–1.2

anthracene), which confirms the anthropogenic nature of the pollution. However, no significant correlations were found between contents of the analyzed PAHs and the content of organic matter in the bottom sediments of the studied lakes. Also Bojakowska et al. (2012) found no correlations between TOC and most of analyzed PAHs in the sediment of 150 Polish lakes. They reported a significant correlation between TOC and three-ring PAHs (acenaphthylene, fluorene and phenantrene) and only one five-ring PAH (perylene). It is in concordance with Stogiannidis and Laane (2015), who maintain that perylene is a product of organic matter diagenesis in soil and bottom sediment.

The differentiation of the content of various components in the bottom sediments of two shallow city lakes, which served as a sewage receiver, was analyzed. Rainwater, raw domestic sewage, dairy and slaughterhouse wastewater, sewage from paint factories and petrol stations were discharged to Karczemne Lake. Domowe Małe Lake mainly collected rainwater flowing from built-up areas and a large road junction and a road transport base. This situation very clearly differentiated the composition of bottom sediments in both lakes. The sediments of Karczemne Lake contain extremely high concentrations of phosphorus, as well as heavy metals characteristic of such industries from which sewage was produced. Significant amounts of silica, which makes difficult phosphorus binding, as well as PAHs and PCBs, were recorded in the sediments of Domowe Małe Lake.

Conclusion

Research has shown that one of the most important tools for selecting an appropriate method of lake restoration is the analysis of the spatial distribution of pollutants in the bottom sediments. Theoretically, both monitored lakes, as shallow, degraded reservoirs, are predisposed to apply the Ripl method (removal of bottom sediments). This is the most effective method for rejuvenating the lake and deepening its basin. Monitoring of the spatial distribution of pollutants in Karczemne Lake showed that the removal of various sediment thicknesses (30 cm, 60 cm, 75 cm and a maximum of 90 cm) will enable the removal of nutrient loads from the lake, which is essential to stop the degradation of the ecosystem, and that the exposed sediment will contain lower amounts of phosphorus and what is important, in an inactive form (residual phosphorus and calcium-bound phosphorus). In addition, the content of other sediment components (organic matter, silica, Al, Fe, Mn) and the correlations between these components and phosphorus additionally confirm that the application of the Ripl method is the optimal restoration solution for Karczemne Lake. After removing the most loaded sediment layer, the sediment remaining in the lake, thanks to its good sorption properties, will act as a trap for phosphorus.

On the other hand, the study of the spatial distribution of pollutants in the sediments of Domowe Małe Lake eliminated the possibility of restoration of this reservoir using the Ripl method. The amounts of pollutants in the entire analyzed sediment cores are similar, and therefore the entire sediment should be removed to the parent rock – it is environmentally and economically unjustified. Moreover, if only the so-called modern sediments with a thickness of 1 m will be removed, as suggested in the Ripl method, the effect would not be achieved, because the exposed sediment would have a similar composition to the one, that was removed. Another argument eliminating the use of the Ripl method in Domowe Małe Lake is the low sorption capacity of the sediments and the high silica content in the sediments, which limits phosphorus binding. The removal of 1 m layer of sediment would not restore the main role of this element in the lake ecosystem, which is binding and storing pollutants.

Acknowledgements

The author would like to thank the Kartuzy City Office and Maciej Kowarzyk for funding this study. Project financially supported by the Minister of Science and Higher Education within the range of the program entitled “Regional Initiative of Excellence” for the years 2019–2022, project No. 010/RID/2018/19, amount funding 12.000.000 PLN.

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Jak można zniszczyć ekosystem jeziora miejskiego – zanieczyszczenie osadów dennych dwóch płytkich zbiorników wodnych

Streszczenie: Celem badań była analiza zawartości i rozmieszczenie zanieczyszczeń mineralnych, organicznych i toksycznych w osadach dennych dwóch zdegradowanych jezior miejskich. Badaniami objęto dwa płytkie jeziora: Karczemne i Domowe Małe (Polska). Rdzenie osadów pobrano za pomocą czerpacza rurkowego typu Kajak i podzielono na warstwy o grubości 30 cm. Analizy metali ciężkich, WWA i PCB wykonano metodami AAS, ICP-AES i GC MS. W próbkach osadów zmierzono zawartość: MO, SiO_2, TO, Ca, Mg, CO_2, Fe, Al, Mn, TN i TP. Badania wykazały, że osady Jeziora Karczemnego, do którego odprowadzane były nieoczyszczone ścieki komunalne, charakteryzuje bardzo wysoka zawartość P. Ponadto stwierdzono, że w osadach dennych osadzają się toksyny, MO i zanieczyszczenia charakterystyczne dla różnych gałęzi przemysłu. Jezioro Karczemne, które odbierało ścieki z komunalne i przemysłowe, zawierało w osadach dużą zawartość Pb, Cu i WWA, a Domowe Małe, przyjmujące głównie wody opadowe zawierało wysokie stężenia WWA. Badania wykazały, że jednym z najważniejszych narzędzi umożliwiających dobór odpowiedniej metody rekultywacji jezior jest analiza przestrzennego rozkładu zanieczyszczeń w osadach dennych. Dzięki takiej analizie składu osadów i korelacji między komponentami osadów oraz ich właściwości sorpcyjnych zaplanowano rekultywację Jeziora Karczemnego metodą Ripla i wyeliminowano możliwość rekultywacji tym sposobem Jeziora Domowego Małego.