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Differentiation of the concentration of heavy metals and persistent organic pollutants in lake sediments depending on the catchment management (Lake Gopło case study)

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Abstract. This paper presents the results of the study on the concentration of heavy metals and persistent organic pollutants (POPs), including PAHs and PCBs, in the bottom sediments of Lake Gopło. This lake is significantly elongated (about 25 km); its longitudinal profile is diversified, and there are deeps and thresholds impeding the flow of water. The shoreline is varied, which is characteristic of tunnel valley lakes. The catchment has a typical agricultural character with a point arrangement of industrial centres. The analysis of the diversity of the concentration of heavy metals and POPs was based on 37 samples from two representative cores: one collected in the northern part of the lake, the catchment of which shows an industrial character, and the second one in the southern part where the catchment is agricultural in character.
In the sediments, the content of the following heavy metals was analysed: Cu, Pb, Cd, Zn, Ni, Cr, Hg and As, as well as PAHs and PCBs. The sediment age was determined by the ^{210}Pb dating method. In order to assess the contamination level of the bottom sediments with heavy metals, the contamination factor (CF) and degree of contamination (DC) were calculated. Moreover, the impact of the changes in the catchment's land use over the past 100 years was determined. The results showed that the sediments from the industrial part of the lake significantly exceed the geochemical background for both the heavy metals from the group identified as industrial pollution and from the group of agricultural pollutants. The southern core shows only a slight increase in the amount of pollution from the agricultural group, lack of industrial pollution and a low degree of contamination. A slight increase in persistent organic pollutants is also recorded, without any apparent effect on the state of the deposited sediment. The ^{210}PB dating enabled the main stages of human impact to be determined: the pre-industrial revolution, from the beginning of industrialisation to the 1950s, intensive human impact from the 1960s to the 1980s, and a gradual decrease in the human impact starting from the 1990s. In addition, attention was paid to the changing sedimentation rate.

Key words
lake sediments,
heavy metals,
PAHs,
PCBs,
Lake Goplo,
Poland

Introduction

Striving for continuous development, people have become a major threat to the surrounding environment. Discharging pollution from industrial, agricultural and municipal activities to the surface water bodies causes their accumulation in water reservoirs, particularly in their bottom sediments. The composition and quantity of contaminants depend on many factors, including the management of the lake catchment in which the sewage receiver is located.

Pollutants entering lakes has changed their natural chemical composition; it has also increased the potential adverse effects on the biotic elements and, increasingly, on human health. Lakes are natural receivers of any contaminants, and studies of accumulated sediments are being increasingly used to assess the environmental pollution with heavy metals and harmful organic substances (Lindell et al. 2001).

The study included Lake Gopło, whose catchment management in the last 100 years transformed from agricultural into industrial-agricultural in the northern part, while the southern part of the catchment is still considered to be a typical farming area. Lake Gopło shows a large north-south extent and is fragmented by thresholds into multiple pools hindering the migration of contaminants. This contributed to the development of two zones: the northern one with a strong predominance of industrial pollution, and the southern one with the predominance of agricultural pollution.

The aim of the study was to analyse the diversity in the concentrations of heavy metals and persistent organic pollutants in the lacustrine sediments depending on the management of the two delimited parts of the catchment (northern and southern) and thus to determine the impact of industrial and agricultural development of the individual parts of the catchment on the pollution of Lake Gopło. In addition, the two separate parts of the lake were compared and the changes that took place during and after the period of economic change were indicated.

Research Area

The research area was the Lake Gopło catchment of an area of 1342 km^2 together with the lake, which covers an area of 21.5 km^2 and the capacity of which is 78.5 M m^3. The large surface area of the catchment in relation to the lake determines the high values of Schindler's coefficient, which amounts to 17. This corresponds to the third class of susceptibility of the lake to degradation (Kudelska et al. 1994). This means under natural conditions Lake Gopło is not resistant to the negative impact exerted by the catchment.

Lake Gopło is located in the catchment of the Noteć, which in turn is part of the river basin of the Odra. The water system in this area has a trellis character, i.e. both meridional and latitudinal. Lake Gopło, along with the Noteć flowing through it, is a major component of the meridional axis. The other watercourses are the tributaries of the Noteć and drainage ditches emptying into Lake Gopło. Only a few watercourses are rich in water throughout the year. Most of them run water during the spring thaw and are dry in the summer (Skawińska et al. 1996). The Noteć catchment area, and thus the catchment area of Lake Gopło, records low precipitation, high evaporation, adverse geological conditions and intensive farming activities (Przytuła et al. 2013).

For centuries the entire basin of Lake Gopło was agricultural. However, in the second part of the 20th century industry began to dominate in its northern part. In this way, the northern part of the Lake Gopło basin has changed its character to industrial while the southern part has remained agricultural. In the northern part urban centres (Kruszwica, Strzelno, Radziejów) and smaller towns (e.g. Dobre) are located. Specifically, the development of Kruszwica, initiated at the end of the nineteenth century, led to the perception of that part of the lake as industrial. In addition, during this period the Noteć was engineered and became navigational, and a sugar factory was built, among other things. In the post-war period new industries were opened, including a winery, a fruit and vegetable processing plant as well as an oil and fat processing plant. At the same time, the southern part was mainly under the influence of agricultural activity with the increasing participation of recreation and tourism, especially around the lake. The southern part of the basin is one of the most densely populated agricultural areas in Poland and has a dense and evenly distributed network of rural settlement. One of the main sources of linear pollution in the catchment of Lake Gopło is the national road DK62 connecting Strzelno and Radziejów. It runs from west to east across Kruszwica, surrounding Lake Gopło from the north. It can be a source of various pollutants, including PAHs and heavy metals (Fig. 1).

Starting from the 1970s, at the peak of industrialisation, there was a rapid deterioration in water quality in Lake Gopło. This was due to the discharge of untreated household refuse as well as industrial and farming wastewater into the lake. The bad ecological status of the lake persisted until the beginning of the twenty-first century. Today, all wastewater is treated. This allows the results of the lake studies conducted in the years 2008–2013 to show a slow improvement in the water quality. The vast majority of the studied parameters, especially the physico-chemical ones, mean the lake water can be classified as Class I, and only the values of a few parameters determine a lower water purity grade. Currently, the physico-chemical state is rated as good, while the ecological potential as bad. Such a situation is mainly due to agricultural pollution, including the presence of nitrogen and phosphorus in the waters supplying Lake Gopło (Table 1).

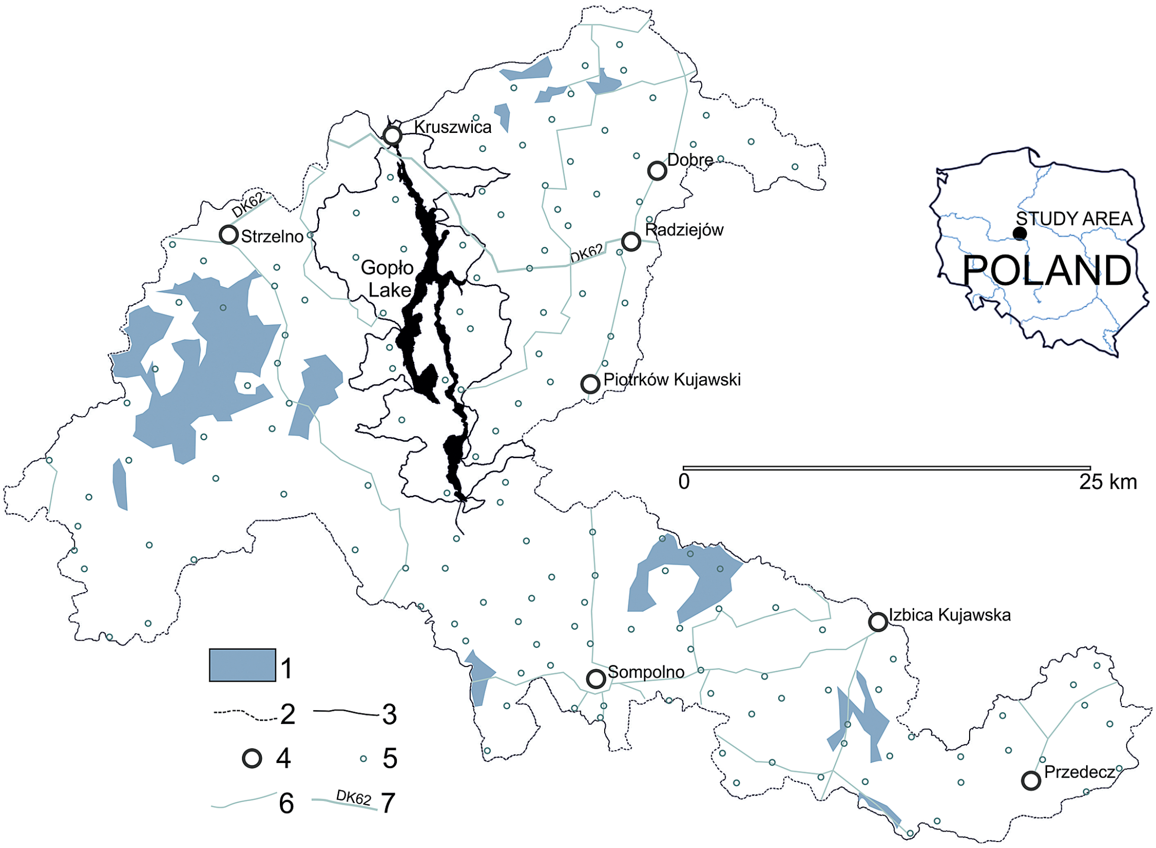

Fig. 1. Study area against the settlement network and the total catchment area: 1 – forests and woodlands, 2 – total catchment, 3 – direct catchment, 4 – towns, industrial centres, 5 – villages, 6 – roads, 7 – national roads (developed on the basis of hydrographical data of the Map of the Hydrographical Division of Poland made by the Department of Hydrography and Riverbed Morphology, Institute of Meteorology and Water Management, at the request of the Minister of the Environment and funded by the National Fund for Environmental Protection and Water Management, MPHP 2007, updated in 2010, as well as OpenStreetMap and the data of the Corine Land Cover project of 2006, updated in April 2012)

Table 1. Changes in the water purity classes of the tributaries of Lake Gopło: 1971–2013

The three-stage classification until 2004, according to the Regulation of the Minister of the Environment, Natural Resources and Forestry of 5 November 1991 (Journal of Laws No. 116, item. 503), distinguished three classes of water quality (Roman numerals I, II, III) and unclassified water (NON). This regulation was repealed on 01 January 2005 with the amendment of the Water Law. The five-stage classification, in force since 2008, according to the Regulation of the Minister of the Environment of 11 February 2004 on the classification for presenting the status of surface water and groundwater, the method of monitoring as well as interpreting the results and presenting the status of these waters (Journal of Laws No. 32, item 284), includes five water purity classes I, II, III, IV and V. The five-stage classification does not include the term "unclassified".

Tributary (Fig. 2)	1971	1976	1985	1989	1995	2002	2007	2008–2013
W1	II	II/III	III	III	III/NON	NON	NON	–
W2	II	II	III	–	III/NON	–	NON	–
W3	I	II	III/NON	–	II	NON	NON	V
W4	I	II/III	III/NON	–	II/III	–	NON	–
W5	I	III	NON	–	NON	–	NON	–
W6	I	NON	NON	–	NON	NON	NON	–
W7	I	NON	NON	–	NON	–	NON	III/V
W8	I	II	NON	NON	III	–	NON	III

Source: own elaboration based on the findings of the National Environmental Monitoring in 1971–2013 (Konarska et al. 1985; Goszczyński, Jutrowska 1996; Makarewicz 2003; Szatten 2007; http://www.wios.bydgoszcz.pl/webmapa/)

As already mentioned, the studied catchment is dominated by agricultural areas, which take up 82.6% of the total catchment area of the lake. The participation of forests is small (11.4%). Forests do not form extensive complexes, but are found in a few small enclaves. Urbanised and industrial areas represent only 1.5% of the total catchment area, while the surface waters take up 4.5%. The detailed data on the total and direct catchment management of Lake Gopło is presented in Table 2.

Table 2. Management of the Lake Gopło catchment

Lake catchment management	Code	Catchment total %	Catchment direct %
Discontinuous urban fabric	112	1.1	1.3
Industrial or commercial units	121	0.1	0.1
Dump sites	132	0.3	0.0
Non-irrigated arable land	211	71.6	66.4
Fruit trees and berry plantations	222	0.0	0.1
Pastures	231	3.3	2.1
Complex cultivation patterns	242	4.0	4.2
Land principally occupied by agriculture, with significant areas of natural vegetation	243	3.6	4.3
Broad-leaved forest	311	3.6	0.8
Coniferous forest	312	5.6	2.0
Mixed forest	313	2.0	2.4
Transitional woodland-shrub	324	0.2	0.0
Inland marshes	411	1.8	3.1
Water bodies	512	2.7	13.0

Source: developed on the basis of the project Corine Land Cover 2006 (updated in April 2012)

Methods

In order to document the existing diversity of heavy metals and POPs in two areas of the lake, i.e. industrial and agricultural, two cores were compared in the present study. The first core was from before the increased development of collected in the northern part (52.66691°N, 18.32697°E), while the other in the southern part of the lake (52.51774°N, 18.37985°E). The undisturbed cores were collected from the deepest parts of the lake with the core of the author's own design. It was assumed that the basal parts of the cores contain sediments the industry. In this way it was possible to document any changes in the bottom sediments which occurred during the period of the strongest human pressure, i.e. over the past 120 years (Fig. 2).

In both cores a total of 39 samples of bottom sediments were obtained with a resolution of 2.5 cm. Their basic physical and chemical characteristics were determined, including the content of organic matter and carbonates by loss on ignition (Heiri et al. 2001) and the particle size by laser diffraction (Krawczykowski et al. 2012). The content of copper (Cu), lead (Pb), cadmium (Cd), zinc (Zn), nickel (Ni), chromium (Cr), mercury (H) and arsenic (As) were determined. Depending on the concentration of the metal, the samples were analysed by atomic absorption spectrometry: in a graphite tube or by flame, hydride or cold-vapour atomisation (Buchalska, Krata 2006). In the samples the content of persistent organic pollutants was also determined, including PAHs by HPLC with the FLD detector (ISO 13877:2004), and PCBs using GC with the ECD detector (PN-EN ISO 6468:2002).

The northern core (N) was dated using ^{210}Pb. The analyses were performed with the alpha spectrometry method using the procedures tested previously on lake sediments (Pempkowiak et al. 2006; Tylmann et al. 2007; Tylmann et al. 2013). In the second, southern core (S) the age was estimated on the basis of metal content and correlation with the neighbouring cores (Ciszewski 2010).

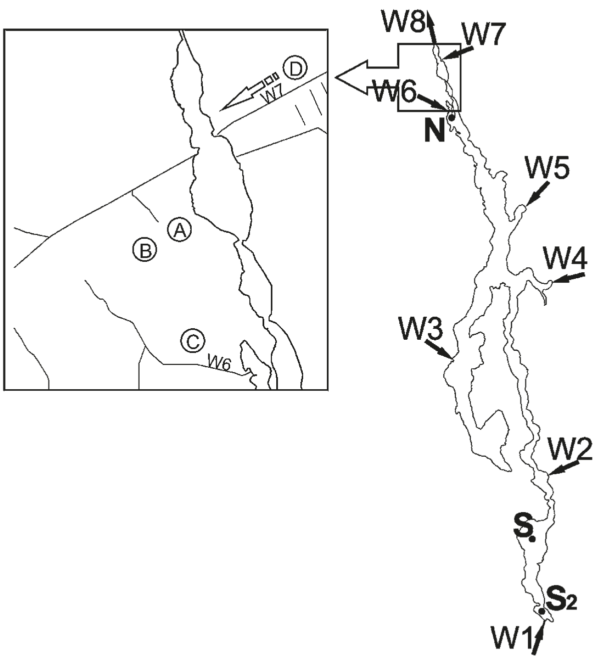

Fig. 2. Monitored tributaries of the lake, location of the sediment cores and sources of pollution: W1 – Ślesin Canal, W2 – Canal Gopło-Świesz, W3 – Canal Ostrowo-Gopło, W4 – supply from Radziejów, W5 – supply from Gocanów, W6 – Łagiewniki Ditch, W7 – Bachorze Canal, W8 – Noteć; N – northern core, S – southern core, S_2 – secondary core ^{210}Pb dated; A - sugar factory (Kruszwica), B - oleochemical plant, C - factory of wines and processed fruit and vegetables, D - sugar factory (Dobre)

Assessment of the degree of contamination of the bottom sediments of Lake Gopło with heavy metals was based on the values of the contamination factor (CF) and the degree of contamination (DC). This method is not based solely on a single metal threat, but it gives a picture of overall risks (Yisa et al. 2012). For these reasons it was chosen for detailed analyses and assessment.

In order to determine the state of contamination of bottom sediments, in addition to the contamination factor CF, the categories adopted by Håkanson (1980) were also applied (Table 3):

$$CF = C_{0\text{-}1} / C_n$$

where: $C_{0\text{-}1}$ expresses the mean concentration of metal in the sediment, C_n is the geochemical background – the assumed values were as proposed by Bojakowska and Sokołowska (1998) – Table 4.

Table 3. Classification of the contamination factor CF

Contamination Factor	Classification
CF < 1	Low
1 ≤ CF < 3	Moderate
3 ≤ CF < 6	Considerable
6 ≤ CF	Very high

Table 4. Geochemical background

Heavy metals	Background
Copper (Cu)	7
Lead (Pb)	15
Cadmium (Cd)	<0.5
Zinc (Zn)	73
Nickel (Ni)	5
Chromium (Cr)	6
Mercury (Hg)	<0.05
Arsenic (As)	<5

In order to determine the degree of contamination (DC), the partial coefficients CF were added up and the classification by Håkanson (1980) was carried out (Table 5):

$$DC = \Sigma\ (CF^{\text{Cu, Zn, Pb, Cd, As, Hg, Ni, Cr}})$$

where: CF is the contamination factor for individual heavy metals.

Table 5. Classification of the degree of contamination DC

Degree of contamination	Classification
DC < 8	Low
8 ≤ DC < 16	Moderate
16 ≤ DC < 32	Considerable
32 ≤ DC	Very high

Results and Discussion

In general, the sediment in the northern part of Lake Gopło is primarily coarse silt of the grain size increasing towards the top. Fine-grained sediments allow the accumulation of heavy metals due to the larger surface of the grains with respect to their volume (Solomons, Förstner 1984; Martincic et al. 1990). In the southern part of the lake, according to the classification by Markowski (1980), there is calcareous gyttja, while in the northern loamy-calcareous gyttja with interbeddings of loamy gyttja (Citkowska 2013). In the samples the content of organic matter was observed in the sediment, varying from 11.3 to 19.4%. The average value for the northern core was 15.3%, while for the southern 12.9%. The organic matter content is correlated with the increased levels of heavy metals in the sediments (Jain, Sharma, 2003) – Fig. 3.

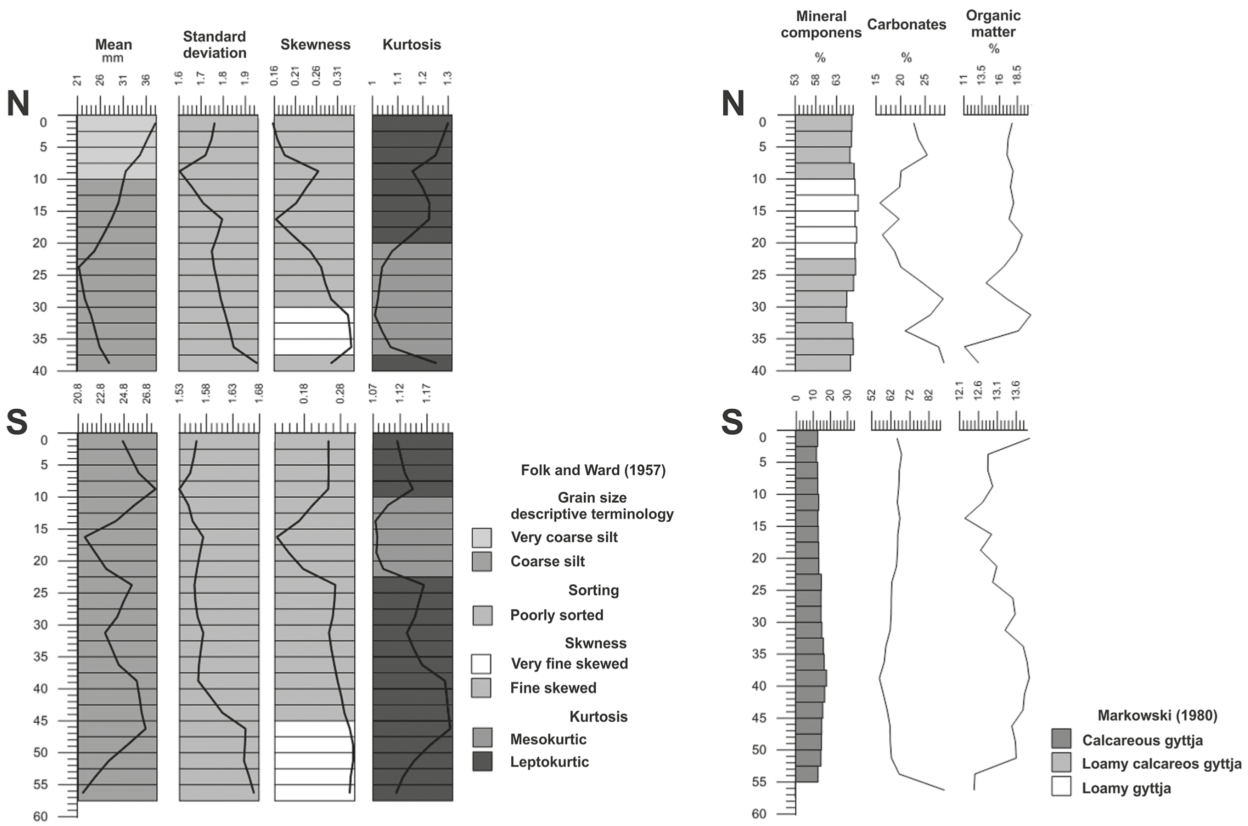

Fig. 3. Cores from the northern (N) and southern (S) parts of Lake Gopło; granulometric features by Folk and Ward (1957) and sediment classification by Markowski (1980)

The content of heavy metals and POPs are distinctly differentiated (Table 6). In the case of the sediment from the northern (industrial) part, all average values exceed the geochemical background, which is not the case in the core from the southern (agricultural) part. The observed difference was mainly due to the catchment management (Jancewicz et al. 2012).

Table 6. Contents of heavy metals, PAHs and PCBs in the bottom sediments of Lake Gopło for 39 samples divided into the northern and southern parts

mg/kg	N			S		
	Min	Max	Mean	Min	Max	Mean
Cu	14.0	126.0	72.4	2.2	12.0	6.8
Pb	23.0	292.0	148.9	8.2	20.0	13.6
Cd	0.07	0.98	0.7	0.03	0.3	0.18
Zn	67.0	901.0	554.2	21.0	42.0	29.6
Ni	0.0	16.0	8.2	0.0	11.0	3.9
Cr	2.0	14.0	9.2	0.0	2.1	0.9
Hg	0.16	9.0	3.3	0.013	0.08	0.04
As	2.4	6.0	4.4	1.7	4.6	2.9
PAHs	0.4	13.4	4.8	0.06	0.6	0.3
PCBs	0.0008	0.08	0.02	0.0004	0.006	0.0014

The contamination factor CF allowed a detailed assessment of various types of heavy metal pollution and sediment classification with respect to the geochemical background. The overall assessment of the pollution level of Lake Gopło was based on the degree of contamination DC (Fiori et al. 2013).

Figure 4 presents the summary for both indicators. About 50% of the samples in the northern part showed a very high level of the contamination factor; over 40% of the samples show moderate values of this indicator. In the southern core only about 15% of the samples have moderate values of the indicator. This affects the degree of contamination, which gives the deposit from the northern part of the lake the top – very high – level of this indicator. The higher rates for the deposits of the southern core do not affect the overall state of this part of the lake.

Anthropogenic pollution, such as PAHs and PCBs which do not occur naturally in the environment, were found in small amounts in the sediments of Lake Gopło. They are more clearly pronounced in the northern part, which is related to the pollution runoff from urban areas surrounding the lake. Accumulation of heavy metals such as zinc, cadmium, nickel and lead may also be due to the industrial development of that part of the catchment. In contrast, arsenic, copper and mercury are treated as pollution from agricultural sources and clearly indicate the spatial dominance of this type of human activity in the catchment of Lake Gopło (Jancewicz 2009).

The northern profile allowed at least four stages related to human activity to be designated: pre-industrial period, from the beginning of industrialisation until the 1950s, intensive human impact from the 1960s to 1980s, and a gradual decrease in human impact starting in the 1990s.

It is also possible to observe a downward trend in the rate of sedimentation in the northern part of the lake after 1990, when the rate and intensity of human impact decreased. Such a change in the southern, agricultural part is not observed. On the contrary, in that part of the lake a slower but steady increase is observed, reflecting the dominance of agriculture on the catchment of Lake Gopło. The impact of industry was more intense, but once a significant reduction in pollution took place, its role quickly diminished (Fig. 5).

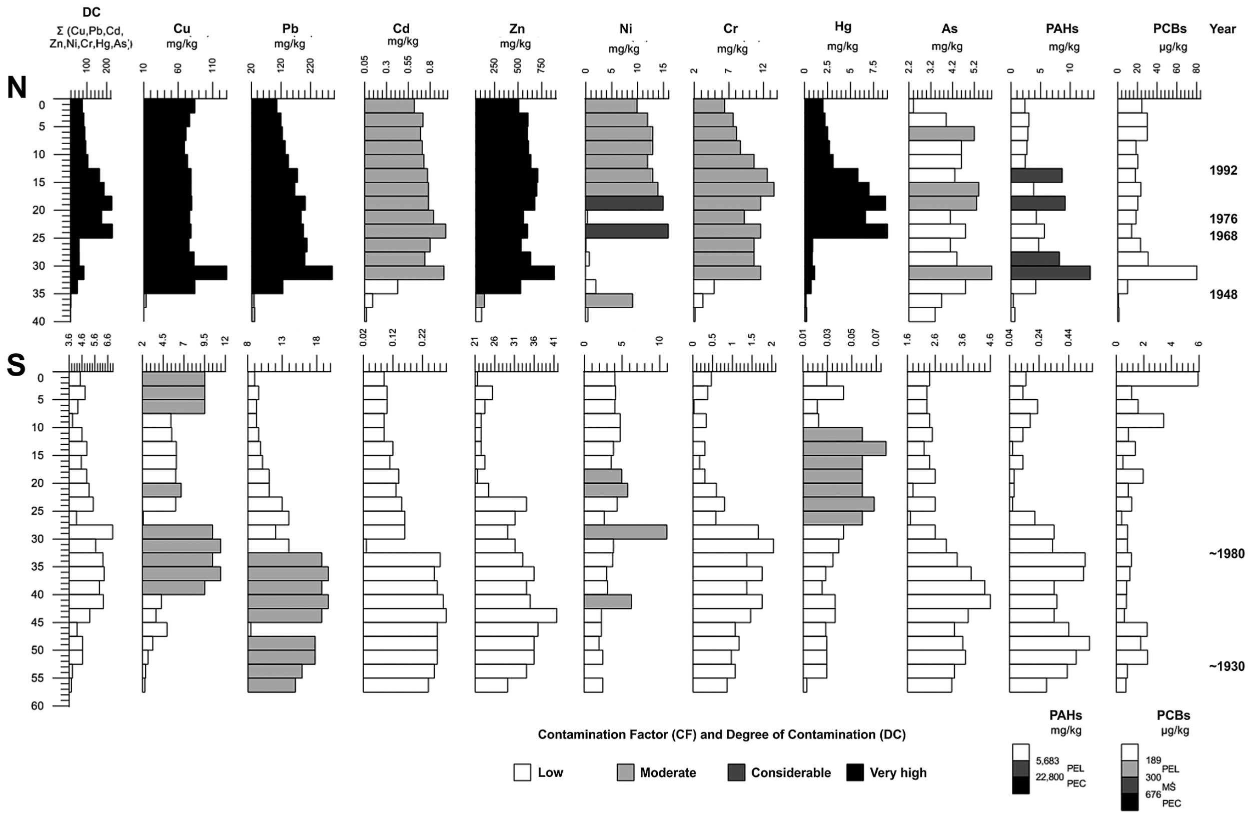

Fig. 4. Concentration of heavy metals with the contamination factor and the degree of contamination by Håkanson (1980), as well as concentration of persistent organic pollutants – PAH and PCB: MŚ – regulation of the Ministry of the Environment, PEC – probable effect concentration, PEL – probable effects level (Decree of the Ministry of the Environment of 16 April 2002, MacDonald et al. 2000) with the results of the deposit ^{210}Pb dating and dating based on the metal content and the correlation between adjacent cores

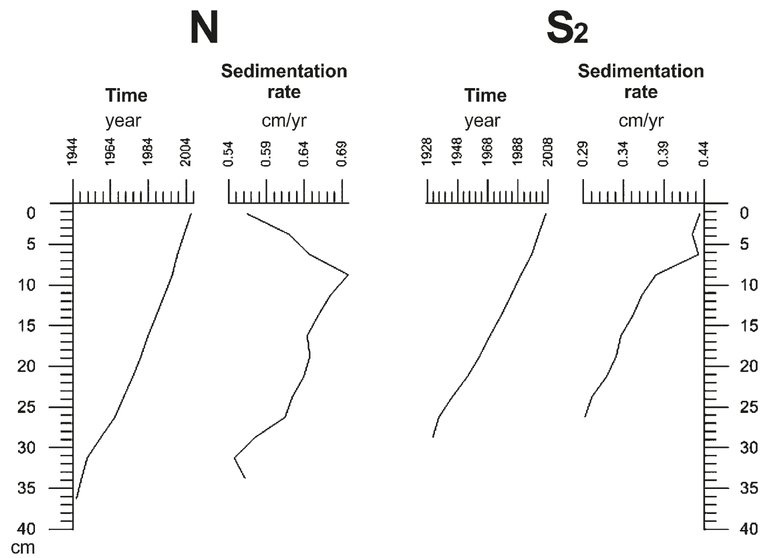

Fig. 5. Changes in sedimentation rate in the northern and southern parts of Lake Gopło

Conclusions

Assessment of the contamination state of the Lake Gopło sediments using the contamination factor and degree of contamination showed significant differences in the pollution of both parts of the lake. Modern dualism has been revealed: the northern part is distinctly polluted by industrial and urban centres located in close proximity to Lake Gopło. While the southern part, despite higher contamination factors associated with agriculture, in the final balance has a low degree of contamination.

What is clearly indicated in the amount of contaminants in the sediments is the main stages of anthropopressure:

a) from before the Industrial Revolution, which forms the local geochemical background,
b) from the beginning of the Industrial Revolution until the 1950s, which is the stage of a growing amount of pollution,
c) intensive anthropopressure between the 1960s and 1980s, when very significant deterioration in the quality of sediments is observed,
d) a gradual decrease in the anthropopressure in the 1990s, when the protective measures lead to rapid improvement.

The tests clearly indicate the change in the causes of the current pollution of the lake. The hitherto dominant negative role of industrial pollution has been reduced to a minimum. At the same time, the role of agricultural pollution has remained at a similar level since the 1960s, when fertilisers were widely introduced in agriculture. It seems that in the coming years, the biggest problem in the field of water protection will be connected with the negative impact of farming on lakes and rivers.

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