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ORIGINAL PAPER

EFFECT OF THE KARLINO OIL FIELD OPERATIONS (ZACHODNIOPOMORSKIE PROVINCE) ON METAL BIOACCUMULATION IN AQUATIC PLANTS IN RIVERS WITHIN THE BALTIC DRAINAGE AREA*

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Abstract

The research was conducted on two rivers in Western Pomerania: the Parsęta and the Radew. Both rivers flow in close vicinity of the Oil and Natural Gas Field in the village of Krzywopłoty, on the outskirts of Karlino. Rivers in the area near the Baltic Sea in Poland typically carry products of agricultural and household origin. Only occasionally, there are also point-like pollution sources of industrial origin. Pollutants from these sources, among which heavy metals are noted, accumulate in aquatic plants which grow in the river beds. The objective of the research was to evaluate the effect of a petroleum mine on the quality of the aquatic environment and aquatic plants in the Radew and Parsęta rivers based on the determination of metal bioaccumulation in plants. The analytical material (water and plants) was collected in the summer of 2012 and 2013, from 9 sites on each river. The content of Cu, Ni, Cd, Pb and Zn was determined with atomic absorption spectrophotometry. Analysis of the metal content in aquatic plants growing along the course of a river gives good representation of the metal distribution and can also be used to reflect the actual effect of point-like pollution sources on the aquatic environment. Results of an investigation into the effect of natural gas and oil field operations on metal bioaccumulation in aquatic plants of rivers in Western Pomerania are presented in this paper. The content of metals in river water was low and can be ordered as follows: Cd<Cu<Zn<Ni<Pb. The concentrations of metals in water were: Ni (0.003-0.053 mg Ni dm^{-3}), Cd (0.001-0.013 mg Cd dm^{-3}), Pb (0.075-0.113 mg Pb dm^{-3}), Zn (0.010-0.066 mg Zn dm^{-3}) and Cu (0.003-0.040 mg Cu dm^{-3}). Heavy metal levels in aquatic plants were relatively low. The content of metals in aquatic plants was the following: Ni (1.61-27.48 mg Ni kg^{-1}), Cd (0.52-6.96 mg Cd kg^{-1}),

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Pb (7.45-60.15 mg Pb kg^{-1}), Zn (13.09-125.63 mg Zn kg^{-1}) and Cu (1.54-36.97 mg Cu kg^{-1}). The levels of these metals in water and plants in both rivers can be ordered as follows: Cd<Cu<Zn<Ni<Pb. The bioaccumulation factor values compared to water levels can be arranged in the following order: Pb<Cd<Ni<Cu<Zn. Point-like effects of pollutants on the aquatic environment were found. In addition, self-purification of rivers was seen, as conditions before the contamination were restored over relatively short sections.

Keywords: water, rivers, aquatic plants, bioaccumulation, metals.

INTRODUCTION

All compounds received by reservoirs in a drainage area affect their intrinsic components, which include bottom sediments, aquatic plants and animals as well as water. Aquatic plants are good indicators of pollutants which accumulate in reservoirs (Samecka-Cymerman, Kempers 2001*a*, Skorbiłowicz 2009). The accumulation of chemical compounds has adverse effects on aquatic ecosystems, even if their levels are low. Together with natural precipitation, economic, agricultural and household activities are sources of pollution. Irrespective of their geographical location, flowing waters frequently collect pollutants from very large drainage areas, hence it is highly likely that the accumulation of captured compounds will be significant. The types of activities in a drainage area are often varied and thus anthropogenic effects differ (Peng et al. 2008, Jastrzębska et al. 2010, Pokorny et al. 2015, Senze et al. 2015).

Rivers in the area near the Baltic Sea in Poland typically carry products of agricultural and household origin (Maciejewska, Wybieralski 2007, Maciejewska et al. 2009). Only occasionally, there are also point-like pollution sources of industrial origin. Pollutants from these sources, among which heavy metals are noted, accumulate in the aquatic plants. The analysis of metal concentrations in plants along the river course gives good representation of spatial distribution of metals and can also be used to reflect the actual effect of point-like pollution sources on the river environment.

The presence of particular types of pollution, except typical and most common urban, agricultural and industrial contaminations, has been noted in the catchments of some rivers. The petroleum mine in Karlino is an example of a source of industrial pollution. Its impact is mainly caused by the emission of products derived from the extraction petroleum and processing of petroleum products. The composition of oil is dominated by hydrocarbons, the share of which is nearly 75%, while other components include sulfur, oxygen, nitrogen as well as heavy metals (Wolicka E. 2010). Two Pomeranian rivers, the Radew and the Parsęta, flow in the vicinity of the mine. Their relatively close location has a potential impact on the quality of the aquatic environment.

The objective of the research was to evaluate the effect of the petroleum mine on the quality of the aquatic environment and aquatic plants in the Radew and Parsęta rivers based on the determination of metal bioaccumulation in plants.

Study area

Two rivers in Western Pomerania, the Parsęta and the Radew, were selected for the study. Both flow in the close vicinity of the Oil and Natural Gas Field in Krzywopłoty, operated by the Polish Oil and Gas Mining S.A. in Warsaw, Zielona Góra Branch, Crude Oil and Natural Gas Company in Karlino, and located on the outskirts of Karlino. Field operations include natural gas and crude oil mining. According to the company's policy, all field activities involve environmentally friendly management of local deposits and high-energy waste gases. Owing to these activities, solid fuel can be replaced by gas fuel, and emission of pollutants can be reduced by almost 80%. One of the priorities for the Karlino field is to secure a continuous supply of natural gas to the municipality (Report on the state of the environment WIOŚ 2008).

The analytical material included river water and aquatic plants collected from each river at 9 sites in the summer of 2012 and 2013 (Table 1, Figure 1).

The springs of the Parsęta are near the town Parsęcko near Szczecinek, situated in the Drawskie Lake District. The river enters the Koszalin Coastal Region near the town Pustary and flows into the Baltic Sea in Kołobrzeg. The Parsęta is 139 km long and drains an area of 3151 km^2. The Radew River, which joins the Parsęta in Karlino, is its main tributary. Białogard, Karlino and Kołobrzeg are the major towns along the Parsęta River. Small hydroelectric power plants operate on the river: near Rościno (53.0 km), in Storkowo (116.3 km) and in Pustkowo (121.2 km). There is a fishing district which comprises the river's waters from the springs to the boundary between inland waters and internal marine waters. The Parsęta River Valley is a special habitat protection area (the Parsęta Drainage Area). There is a paddling trail on the Parsęta (Report on the state of the environment WIOŚ 2008).

The Radew River is a right-side tributary of the Parsęta. It is 83 km and has a drainage area of 1091.5 km^2. The river's springs are near the village Żydowo in the Bytów Lake District. The river flows through the following towns: Żabiniec, Bobrowo, Mostowo, Rosnowo, Niedalino, Wronie Gniazdo, Białogórzyno, Nosowo, Parsowo and Karlino. Its mouth is located in Karlino. Small hydroelectric power plants operate on the river in Rosnowo (44.7 km), Niedalino (36.0 km) and Karlino (0.53 km). The river valley has been designated the status of a protected land area known as the Radew River Valley and incorporated into the Natura 2000 network: Radew, Chociela and Chotla River Valleys. There is a paddling trail on the Radew River between Żydowo and Karlino (Report on the state of the environment WIOŚ 2008).

Table 1

Characteristics of the sampling sites on the Parsęta and Radew rivers

No.	Radew		Parsęta	
	site, depth (m), width (m), geographical coordinates			
1.	Cybulino 1.50 3.00	N:54° 2′ 19.87″; E:16° 37′ 8.31″	Storkowo 0.50 1.05	N:53° 45′ 58.37″; E:16° 30′ 55.05″
2.	Kurowo 1.00 3.00	N:54° 3′ 3.95″; E:16° 34′ 55.44″	Stare Dębno 1.00 4.00	N:53° 52′ 22.23″; E:16° 10′ 36.51″
3.	Mostowo 7.00 189 ha Rosnowskie lake	N:54° 4′ 27.01″; E:16° 23′ 35.05″	Osówko 0.50 20.00	N:53° 53′ 49.92″; E:16° 5′ 11.14″
4.	Rosnowo 8.00 189 ha Rosnowskie lake	N:54° 4′ 28.01″; E:16° 17′ 43.76″	Byszyno 2.00 19.00	N:53° 56′ 56.88″; E:16° 3′ 4.45″
5.	Niedalino 0.70 8.00	N:54° 3′ 40.36″; E:16° 10′ 1.65″	Białogard 2.00 10.00	N:54° 0′ 8.69″; E:15° 58′ 48.61″
6.	Bardzlino 2.00 7.00	N:54° 3′ 47.71″; E:16° 6′ 56.87″	Karlino 2.00 30.00	N:54° 1′ 42.01″; E:15° 52′ 25.31″
7.	Nosowo 2.50 8.00	N:54° 6′ 23.34″; E:16° 0′ 14.78″	Wrzosowo 4.00 40.00	N:54° 6′ 3.86″; E:15° 48′ 32.02″
8.	Karlinko 3.00 6.00	N:54° 2′ 42.21″; E:15° 54′ 2.68″	Ząbrowo 5.00 25.00	N:54° 5′ 30.34″; E:15° 38′ 26.55″
9.	Karlino 5.00 10.00	N:54° 1′ 57.58″; E:15° 52′ 46.51″	Kołobrzeg 9.00 60.00	N:54° 10′ 47.49″; E:15° 33′ 45.99″

MATERIAL AND METHODS

Water from each site was collected into polyethylene bottles with a 1 dm^3 bucket, and filtered through 0.45 Whatman 1 filters in a laboratory. Next, water samples for determination of metals were digested with concentrated HNO_3. Subsequently, digested samples were cooled down to room temperature and diluted with distilled water to 25 cm^3. The metals in water (Cu, Ni, Cd, Pb, Zn) were assaysed with atomic absorption spectrophotometry on a Varian Spectra AA-110/220 apparatus (PB-10/I – 1998).

Plants collected at each site (whole plants, N = 3) were dried in a labora-

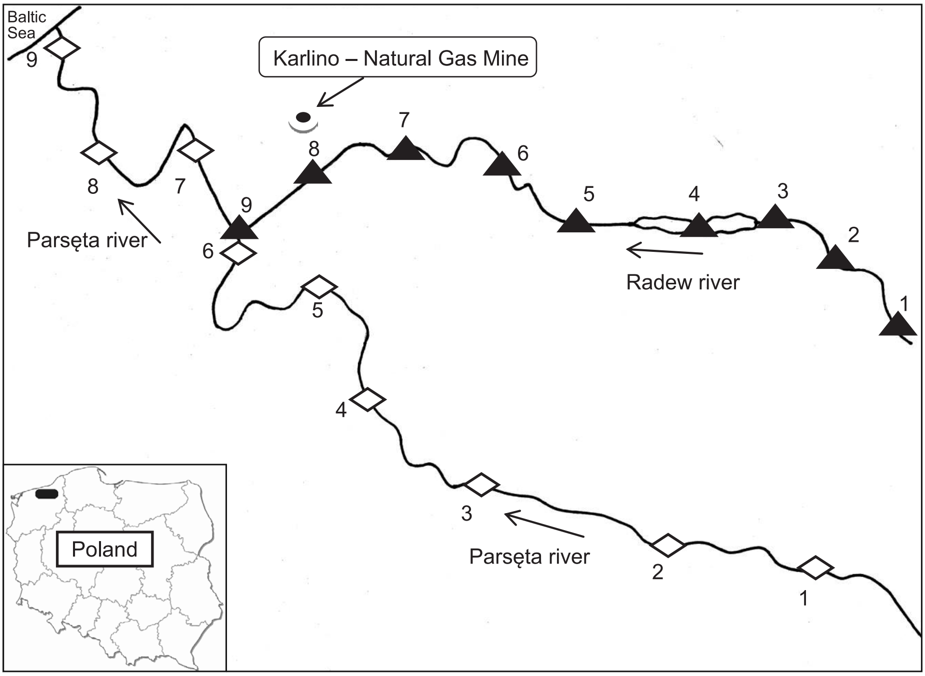

Fig. 1. Study area and location of the sampling sites

tory at room temperature to obtain dry mass and then cut and pulverised. Homogenised samples of 0.5 g of air dry were weighed in an HP-500 Teflon vessel. An amount of 10 cm^3 of concentrated HNO_3 (min. 69% puriss. P. a, Reag. ACS, Reag. ISO; Sigma-Aldrich) was added to each sample, which was then left to stand at room temperature for 24 hours. Subsequently, the samples were placed in a CEM Mars 5 microwave oven and digested in a 3-step digestion process. When cooled down to room temperature, digestion products were transferred into test tubes and diluted with distilled water to 25 cm^3. Metal assays in plants (Cu, Ni, Cd, Pb, Zn) were performed with atomic absorption spectrophotometry using a Varian Spectra AA-110/220 apparatus (PB-10/I – 1998).

The metal bioaccumulation factor (BCF) as the ratio of their content in an aquatic plant (C_{MR}) to the concentration in water (C_{MW}) was calculated according to formula (Jezierska, Witeska 2001).

$$BCF = \frac{C_{MR}}{C_{MW}} \cdot$$

The normality of the distribution of the analyzed metals was verified using the Shapiro-Wilk's *W* test (Zar 1999). Plotting the principal component and classification analysis (PCCA) ordination of water and plant samples and projecting concentrations of the metals in water and plants on the factor plane provide information about similarities between the samples and illustrate the correlation between the original variables and the first two factors (Legendre, Legendre 1998).

The differences between the sites close to and further from the Oil and Natural Gas Field (Karlino) were evaluated by the Kruskal-Wallis non-parametric ANOVA followed by a post-hoc test. Statistical confidence was set at $p = 0.05$. All statistical calculations were conducted using R version 3.3.2 or Statistica 13.1.

RESULTS AND DISCUSSION

Twenty-two species of aquatic plants were collected, and the reed canary grass (*Phalaris arundinacea* L.) was most widespread species in both rivers (Table 2).

Metal concentrations in waters of the rivers are collated in Table 3. Cadmium, nickel and copper levels were higher in the first year of the study. The lead concentrations were very similar in both years. The lowest values were seen for cadmium and the highest ones – for lead.

Heavy metal levels in aquatic plants were relatively low. The results are shown in Table 4. The content of the metals was: Cu (1.54-36.97 mg Cu kg^{-1}), Ni (1.61-27.48 mg Ni kg^{-1}), Cd (0.52-6.96 mg Cd kg^{-1}), Pb (7.45-60.15 mg Pb kg^{-1}) and Zn (13.09-125.63 mg Zn kg^{-1}). The levels of the metals in both rivers can be ordered as follows: Cd<Cu<Zn<Ni<Pb.

The bioaccumulation factor for metals in aquatic plants was calculated with reference to metal levels in water. The highest values in plants from both rivers were seen for zinc in the first year (on average, 1983 for the Radew and 3218 for the Parsęta) and for copper in the second year (1814 and 1537, respectively) – Table 5. The lowest mean values were noted for lead. The bioaccumulation factors for the analyzed metals in both rivers can be ordered as follows: Pb<Ni<Cd<Cu<Zn.

Significant correlations were found between nickel, cadmium and copper levels in plants and between nickel, cadmium and copper levels in water ($r = 0.5015$, $r = 0.3200$ and $r = 0.1300$) at $p < 0.05$ (Table 6). Other correlations were found between metals in aquatic plants: Ni-Cd, Ni-Pb, Ni-Zn, Ni-Cu, Cu-Zn, Cd-Pb, Cd-Cu, Pb-Zn and Pb-Cu. The correlations found in water were between the following pairs of metals: Ni-Cd, Ni-Pb, Ni-Cu, Cd-Pb and Cd-Cu.

Similar metal levels (nickel, cadmium, lead) as those found in the Radew and Parsęta appeared in plants collected from different rivers in Poland. Lower values were found for copper and zinc (Skorbiłowicz, Wiater 2003, Jastrzębska et al. 2010, Rajfur et al. 2010, Gwoździński et al. 2014, Klink et al.

Table 2

The occurrence of macrophyte species in the sampling sites on the Parsęta and Radew rivers (numbers in the table indicate numbers of the sites)

Species	Sites			
	Parsęta River		Radew River	
	2012	2013	2012	2013
Berula erecta (Huds.) Coville	4, 9	2, 3, 8	1, 3, 4	1, 2, 4, 8
Callitriche cophocarpa Sendtn.	7, 9	3		
Carex acuta L.			1	
Carex riparia Curt.		1		2
Elodea canadensis L.		5	2	
Epilobium hirsutum L.	2, 8	6, 7, 9	5	
Epilobium palustre L.	4	2, 7		
Equisetum fluviatile L.	1			
Galium palustre L.		7		
Glyceria fluitans (L.) R.Br.			9	4
Glyceria maxima (Hartman) Holmb.			2, 3, 5, 8	2, 3, 5, 8, 9
Mentha aquatica L.	1	3, 4	2	2, 3
Myosotis palustris Roth	1			2, 7
Phalaris arundinacea L.	2, 3, 4, 5, 6, 8, 9	4, 6, 7, 8	4, 7, 9	4, 5, 9
Phragmites australis (Cav.) Trin. ex Steud	1			8
Potamogeton pectinatus L.			5	
Roripa amphibia L.	6			
Solanum dulcamara L.	2, 6	7		4
Sparganium emersum Rehmann	2, 4, 7	2, 3, 4, 5	6, 7, 8	7
Scrophularia umbrosa Dumort		2, 6		
Veronica anagallis-aquatica L.	2, 3	3	9	

2013, Pokorny et al. 2015), but higher ones were in the mountains. When these results are compared with literature data, it is concluded that the metal levels in water and aquatic plants in the Radew and Parsęta were higher than, for example, in rivers flowing from the Karkonosze Mountains (southern Poland), although they were affected by the local industrial operations (Samecka-Cymerman, Kempers 2001*a*, Samecka-Cymerman, Kempers 2001*b*, Samecka-Cymerman et al. 2005).

A comparable lead content, higher zinc content and lower copper content as found in the Parsęta and Radew River in our study were observed in the Parsęta river and its tributaries in 2003-2007 by Maciejewska, Wybieralski (2007) and Maciejewska et. al. (2009). Among the investigated surface water bodies in western Poland also affected by industrial activity (metal, food, pharmaceutical, and chemical industries), only nickel levels were lower than

Table 3

Heavy metal concentrations (mg dm^{-3}) in the water of the Parsęta and Radew rivers

River	Year	Ni	Cd	Pb	Zn	Cu
		min-max ($\bar{x} \pm$ SD)				
Parsęta	2012	0.033 - 0.053 0.044 ± 0.003	0.009 - 0.013 0.011 ± 0.001	0.080 - 0.105 0.093 ± 0.008	0.010 - 0.019 0.014 ± 0.003	0.016 - 0.021 0.018 ± 0.001
	2013	0.003 - 0.009 0.005 ± 0.002	0.001 - 0.002 0.001 ± 0.001	0.075 - 0.113 0.092 ± 0.010	0.013 - 0.026 0.020 ± 0.004	0.003 - 0.040 0.008 ± 0.011
Radew	2012	0.039 - 0.053 0.044 ± 0.003	0.009 - 0.012 0.010 ± 0.001	0.086 - 0.107 0.092 ± 0.006	0.012 - 0.066 0.022 ± 0.014	0.016 - 0.023 0.019 ± 0.001
	2013	0.004 - 0.008 0.006 ± 0.001	0.001 - 0.002 0.002 ± 0.001	0.075 - 0.099 0.087 ± 0.007	0.013 - 0.029 0.021 ± 0.004	0.003 - z0.031 0.005 ± 0.005

Table 4

Heavy metal concentrations (mg kg^{-1}) in aquatic plants from the Parsęta and Radew rivers

River	Year	Ni	Cd	Pb	Zn	Cu
		min-max ($\bar{x} \pm$ SD)				
Parsęta	2012	5.10 - 27.48 13.77 ± 7.72	0.03 - 6.98 3.30 ± 2.10	9.79 - 60.16 26.54 ± 16.66	23.26 - 125.65 43.89 ± 24.37	2.35 - 36.98 11.71 ± 8.14
	2013	3.08 - 13.35 6.17 ± 2.73	0.52 - 5.05 1.79 ± 0.62	8.04 - 27.06 17.18 ± 4.69	17.21 - 72.19 34.22 ± 14.73	1.72 - 22.43 7.88 ± 4.68
Radew	2012	5.46 - 13.40 8.19 ± 1.97	1.17 - 2.53 1.74 ± 0.37	7.47 - 18.19 13.34 ± 2.58	26.58 - 73.73 37.26 ± 10.61	2.85 - 14.37 6.35 ± 2.57
	2013	1.61 - 8.34 4.44 ± 2.03	0.56 - 2.32 1.75 ± 0.42	7.89 - 23.29 17.02 ± 3.52	13.08 - 42.57 28.94 ± 8.49	1.53 - 20.01 5.43 ± 4.87

Table 5

Bioconcentration factor (BCF) of heavy metals for the Parsęta and Radew rivers

River	Year	Ni	Cd	Pb	Zn	Cu
		min-max ($\bar{x} \pm$ SD)				
Parsęta	2012	118 - 639 314 ± 178	93 - 687 308 ± 195	103 - 1523 395 ± 365	1463 - 10132 3218 ± 1796	133 - 1933 650 ± 446
	2013	538 - 2989 1318 ± 578	434 - 2620 1494 ± 530	95 - 301 191 ± 51	873 - 5269 1808 ± 919	114 - 5902 1814 ± 1433
Radew	2012	104 - 320 187 ± 51	102 - 269 173 ± 47	80 - 211 147 ± 32	507 - 4915 1983 ± 909	155 - 764 338 ± 139
	2013	252 - 1463 753 ± 370	473 - 1704 1121 ± 318	90 - 296 200 ± 47	566 - 3101 1331 ± 573	274 - 5261 1537 ± 1242

Table 6

Correlation coefficient matrix – content of metals in water and aquatic plants

Metals	Ni - P	Ni - W	Cd - P	Cd - W	Pb - P	Pb - W	Zn - P	Zn - W	Cu - P	Cu - W
Ni - P	1.00									
Ni - W	0.50*	1.00								
Cd - P	0.86*	0.30*	1.00							
Cd -W	0.51*	0.99*	0.32*	1.00						
Pb - P	0.82*	0.16*	0.85*	0.17*	1.00					
Pb - W	0.14*	0.27*	0.17*	0.31*	0.03	1.00				
Zn - P	0.25*	0.26*	0.05	0.23*	0.14*	0.15*	1.00			
Zn - W	-0.17*	-0.16*	-0.21*	-0.15*	-0.19*	-0.13*	-0.11	1.00		
Cu - P	0.58*	0.20*	0.50*	0.19*	0.60*	0.26*	0.52*	-0.23*	1.00	
Cu - W	0.33*	0.64*	0.23*	0.64*	0.14*	0.11	0.12	-0.03	0.13*	1.00

W – water, P – plant;
Significant correlations ($P < 0.05$) shown*.

in the Parsęta and Radew (Samecka-Cymerman, Kempers 2003, Skorbiłowicz 2009). This was probably due to nickel contained in the Karlino petroleum airborne deposits. Our analysis of the plant material shows similar levels of the metals except cadmium, whose concentration was lower.

The water of the Wieprz River investigated by Bojakowska et al. (2010) in an area of natural gas, crude oil and hard coal extraction had higher cadmium and lead levels than in the rivers of the coastal region (the Radew and the Parsęta), but it contained less copper, nickel and zinc. Similar metal concentrations were found in lake waters affected by a copper smelter (Lubin, Poland). Copper, lead and zinc levels in macrophytes growing in water bodies located in that area were higher than in the coastal rivers, although similar to the latter with respect to nickel and cadmium (Samecka-Cymerman, Kempers 2004). Investigations into the levels of heavy metals in lowland dam reservoirs show higher levels of Ni and Zn, a lower level of lead and similar levels of cadmium and copper (Czamara, Czamara 2008, Senze et al. 2009, Wojtkowska 2014). Significantly higher content of metals was found in reservoirs connected with mines (Aleksander-Kwaterczyk et al. 2010). In our study, significantly higher values of Pb in water (p = 1.377e-07) but lower values of Zn (p = 0.015) were found in water downstream in both rivers (Figure 2). Also, significantly higher values of Cu in plants (p = 0.001) were found in plants growing more downstream in both rivers. While the lead content is directly related to the mine's influence, the copper concentration in plants may also depend on soil conditions. The lower zinc content (p = 0.015) in the water of both rivers in their lower course would have been affected by the flow of pollutants which either do not contain this element, or its level is considerably lower.

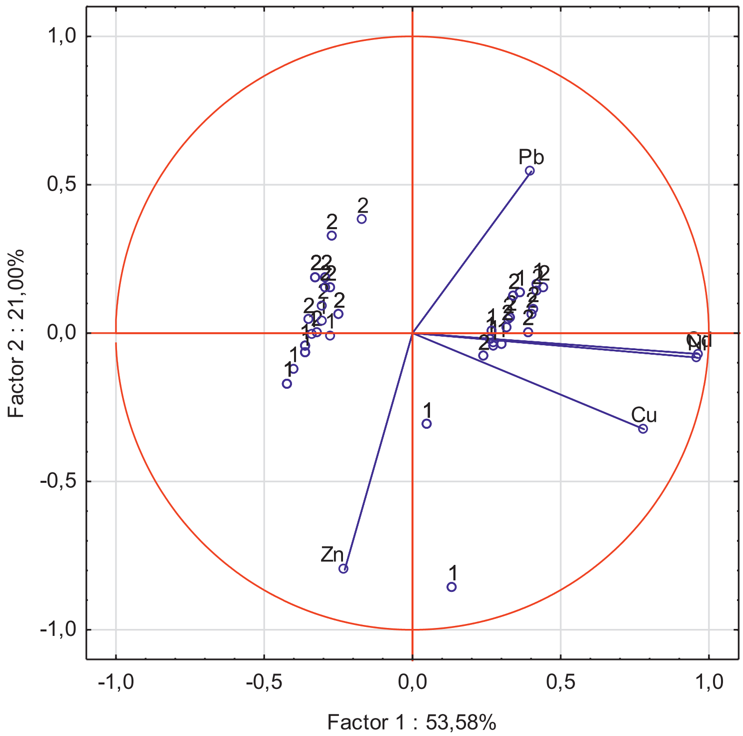

Fig.2. Ordination plot for the study sites by PCCA based on concentrations of metals in water and projection of f metals on the factor plane

Concentrations of the analyzed metals in the coastal rivers changed along their flow. The sites located on the Radew and Parsęta rivers in the closest vicinity of the Crude Oil and Natural Gas Field in Karlino had higher (but not statistically significantly) levels of all metals compared to samples taken upstream and downstream from these sites.

Summary

The evaluated waters of the Parsęta and Radew rivers may be considered as having low pollution levels in terms of chemical components. Zinc and copper levels were consistent with water purity class I. However, cadmium, lead and nickel levels exceeded the environmental quality standards for priority substances (Journal of Laws 2014).

Metal levels in aquatic plants were low, and the bioaccumulation factor values compared to water arranged the metals in the following order: Pb<Cd<Ni<Cu<Zn.

Significantly higher values of Pb (p = 1.377e-07) as well as lower values of Zn (p = 0.015) were found in water downstream in both rivers. Also, significantly higher values of Cu ($p = 0.001$) in plants were found downstream in both rivers. To our best knowledge, the content of Pb is directly connected to the impact of the Karlino mine field, but the Cu concentration in plants could depend on soil condition. The lower zinc content ($p = 0.015$) in river water downstream could have been a consequence of the influx of pollution without any zinc or else with small amounts of this metal.

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