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EVALUATION OF THE ALUMINIUM LOAD IN THE AQUATIC ENVIRONMENT OF TWO SMALL RIVERS IN THE BALTIC SEA CATCHMENT AREA*

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Abstract

The study covered the aquatic environment of two small rivers in Western Pomerania, Poland, such as the Czerwona and the Grabowa. Its purpose was to determine aluminium bioaccumulation in the aquatic environment by testing water, bottom sediments and aquatic plants. Samples were taken in the summers of 2008-2011. pH, electrolytic conductivity and aluminium concentration were determined in water samples, while the sediment and plant samples were submitted to analyses of the aluminium content. The water pH oscillated between 6.04 and 8.95, while the electrolytic conductivity ranged from 440 to 1598 μS cm^{-1}. The aluminium concentration in the river water was up to 0.138 mg Al dm^{-3} in the Czerwona and up to 0.425 mg Al dm^{-3} in the Grabowa. The maximum aluminium content in the bottom sediments was 47.01 mg Al kg^{-1} in the Czerwona River and 26.15 mg Al kg^{-1} in the Grabowa River. The maximum aluminium content in the aquatic plants sampled from the Czerwona was 91.63 mg Al kg^{-1}, and from the Grabowa – 1,077 mg Al kg^{-1}. The bioconcentration factor (BCF) of aluminium for the Czerwona River ranged from 5.06 to 24,052, and for the Grabowa River – from 1.10 to 70,132. The concentration factor (CF) of aluminium in the bottom sediments oscillated between 66.72 and 23,492 in the Czerwona River and between 14.81 and 2,763 in the Grabowa. The aluminium content in the two rivers was relatively low in the water, sediments and aquatic plants, which is typical of environments without strong anthropopressure. The values fell within the limits set by environmental water quality standards. The low aluminium accumulation degrees in the biotic and abiotic components indicate that the environments of the two rivers have a low load of aluminium compounds.

Keywords: aluminium, water, aquatic plants, bottom sediments, rivers, accumulation.

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INTRODUCTION

Aluminium is the third (after oxygen and silicon) most common component of the Earth's crust. Aluminium concentrations in soil largely depend on local human activities, hence in industrial areas and in their vicinity, Al soil levels can be several-fold higher than elsewhere. Studies conducted near a large urban center in southern Poland, in areas adjacent to an industrial complex, which includes a cement plant, have shown an aluminium concentration in arable land of up to 14.80 g kg^{-1} (CIESIELCZUK et al. 2011). In another industrialized area (coal mines), the aluminium concentration in farmland was 32.42 g kg^{-1} (KUCHARCZAK-MORYL et al. 2014).

Aluminium is common in plants, and its actual concentration depends on the species and soil conditions. Plants growing in acid soil accumulate large amounts of aluminium. Naturally, excess aluminium is harmful to plants, as it interferes with the uptake and transport of nutrients, damages cellular membranes (KOCHIAN et al. 2005), and affects adversely cell division and DNA synthesis (SILVA et al. 2000).

Aluminium solubility strongly depends on the pH of water, namely it increases in solutions with a pH < 6.0 and pH > 8.0 (GRZEBISZ et al. 2005). The concentration of aluminium in Poland's rivers is relatively low, usually less than 0.05 mg Al dm^{-3}, as most of the rivers have water with alkaline and close to neutral reaction (SZCZEPAŃSKI et al. 2010). Aluminium is easily absorbed by bottom sediments in the form of metastable compounds and can be activated when water acidity raises (WIDŁAK, WIDŁAK 2013). Aluminium accumulates in the food chain in all elements of an aquatic environments (OBERHOLSTER et al. 2012), and is poorly absorbed by organisms from water containing increased amounts of calcium, magnesium and fluorine ions. In fish, the precipitation of aluminium on gills is particularly harmful (ALSTAD et al. 2005).

The study focused on the accumulation of aluminium in surface water, as this compartment of the environment gathers pollutants both from atmospheric precipitation and surface runoff. The study was designed to determine the presence and accumulation of aluminium in two lowland rivers in Western Pomerania, exposed to agricultural and urban management.

MATERIAL AND METHODS

The study included two rivers in the Province of West Pomerania (*województwo zachodniopomorskie*): the Grabowa and the Czerwona (Figure 1). The Grabowa River is 74 km long and has a catchment area of 536 km^2. Its upstream section flows through an upland and woods. Afterwards, it cuts through a lowland falls into the Wieprza River about a kilometre before the

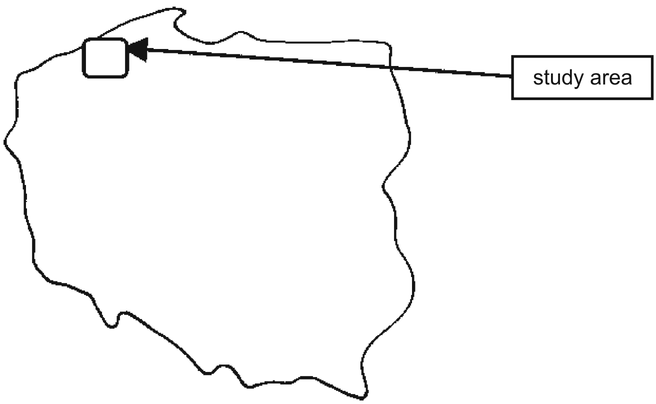


Fig. 1. The study area

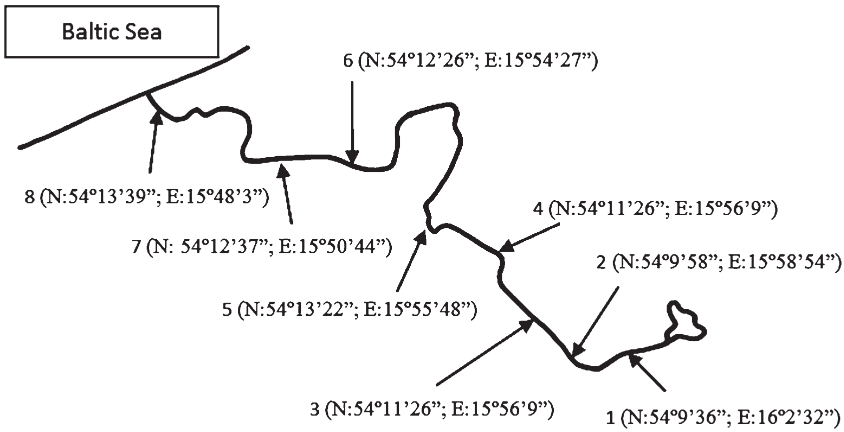
latter feeds its water to the Baltic Sea. The geological structure of the catchment is mostly made up of sand deposits. The river valley bottom is predominantly covered with peat and mostly drained. The river is used for recreational purposes, both as part of a kayaking trail and an angling site (Report WIOŚ 2008).

The Czerwona River is 28.6 km long and has a catchment area of about 143 km^2. It flows from Parnowskie Lake. It is a typical lowland river, with a small channel gradient and a slow current. In its immediate vicinity, there are numerous farm buildings. The geological structure of the catchment is predominated by tills, and small depressions are filled in with peat (Report WIOŚ 2008).

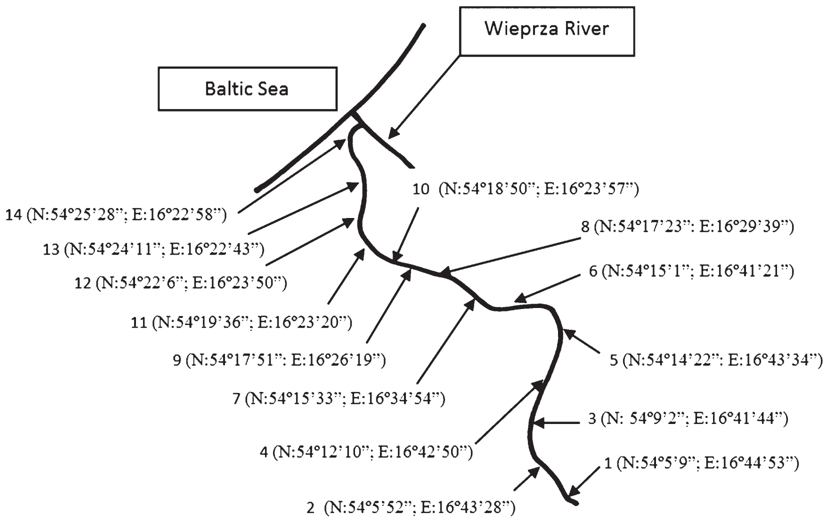
The study material comprised water, bottom sediments and aquatic plants. The research covered a four-year period, from 2008 to 2011. Samples were obtained from locations along the entire lengths of the two rivers. They were collected once a year, at the end of July/the beginning of August (Figures 2, 3). Water was sampled with a sampler and filtered through 0.45 Whatman 1 filters. Then, the pH of water was measured by the potentiometric method (PN-90/C-0454/01) and its electrolytic conductivity was tested by the conductometric method (PN-EN 27888:1999).

Because the research objective was to assess the loading of the rivers' aquatic environment with aluminium, plant samples comprised only perennial plants so as to be able to obtain a possibly comprehensive pattern, pertaining to the four-year period of the study as well as some earlier time. Both submergent and emergent plants were sampled, depending on their presence at a given site (Table 1). At some sites it was impossible to collect both types of plants. In total, 21 plant species were collected: 15 emergent and 6 submergent.

The plants were sampled whole, together with the root, stem and leaves. Afterwards, they were dried at a temperature of 22°C until dry mass was



Rys. 2. Location of the sampling sites on the Czerwona River



Rys. 3. Location of the sampling sites on the Grabowa River

obtained. Air-dried and homogenized samples were digested in a microwave oven (MARS 5, CEM, USA), in concentrated HNO_3, using a three-step mineralization process. Bottom sediments were sampled directly into canvas bags, dried at room temperature, pounded in a mortar and passed through a two -millimetre mesh sieve. Air-dried and homogenized samples were subjected to three-step mineralization with HNO_3:$HClO_4$ (3:1) in a microwave oven (MARS 5, CEM, USA).

The aluminium content in all the tested materials was determined using atomic absorption spectrophotometry (Varian Spectra AA110/220 unit, Au-

Table 1

Macrophytes recorded at different sampling sites

Species	Sampling sites	
	Czerwona River	Grabowa River
Berula erecta (Huds.) Coville		8, 9
Callitriche cophocarpa Sendtn.		12, 14
Carex riparia Curt.	1, 2, 5, 6	4, 7, 9, 10
Cladium mariscus (L.) Pohl.		3, 4, 14
Elodea canadensis L.		5, 6, 12, 13, 14
Epilobium hirsutum L.	1, 2, 3, 5	3, 6, 8, 11
Glyceria maxima (Hartman) Holmb.	1, 3, 5	1, 2, 4, 6, 8, 9, 11, 12, 14
Hydrocharis morsus-ranae L.	8	
Lythrum salicaria L.	6	9
Mentha aquatica L.	5	4, 6, 11, 13
Myosotis palustris Roth	3	3, 6, 13
Phalaris arundinacea L.	1, 2, 3, 5, 6, 7, 8	2, 5, 7, 8, 9, 10, 11, 12, 13, 14
Phragmites australis (Cav.) Trin. ex Steud	7, 8	1, 7, 8, 9, 10, 11
Potamogeton pectinatus L.		4, 5, 6, 8, 12
Potamogeton perfoliatus L.		8, 9, 14
Solanum dulcamara L.		6, 8, 9, 14
Sparganium emersum Rehmann	2, 5, 6, 7	13, 14
Spirodela polyrrhiza (L.) Schleiden	7	
Sium latofolium L.	2	12
Rumex hydrolapathum Huds.	8	8
Veronica anagallis-aquatica L.		5, 6, 9

stralia). The correctness of the analytical procedures was verified using the Certified Reference Material BCR No. 60, Lagarosiphon major (The Commission of the European Communities, The Community Bureau of Reference – BCR). The reference concentration in BCR No. 60 was 4027±84 mg kg^{1}.

The aluminium concentration factor (CF) was calculated as a ratio of the aluminium content in sediment to its content in water, whereas the aluminium bioconcentration factor (BCF) was a ratio of the aluminium content in plants to its content in water. The statistical analysis of the results was carried out using Statistica 10.0. Significant Pearson's (r) correlation coefficients were evaluated using Anova at a 5% significance level.

RESULTS AND DISCUSSION

Water

Absorption of metals by aquatic plants depends on the pH of water and its mineralization level. Acidic environments are conducive to the release of metals from sediments and consequently boost their concentration in water. The waters of the two rivers usually had neutral pH (Table 2).

Water mineralization, expressed as electrolytic conductivity, reflects the loading of water with mineral compounds, principally calcium and magne-

Table 2

Physical and chemical properties of water, bottom sediments and aquatic plants sampled from the Grabowa and the Czerwona Rivers in 2008-2011

Sampling site	Physical and chemical properties	Year of study			
		2008	2009	2010	2011
		min.-max. average			
The Czerwona	pH	6.54-7.28 6.92	6.31-7.39 6.93	6.71-6.79 6.75	7.10-8.95 8.01
The Grabowa		7.05-7.43 7.24	6.04-7.54 6.98	6.59-6.96 6.75	6.74-7.05 6.86
The Czerwona	electrolytic conductivity ($\mu S\ cm^{-1}$)	651-796 713.2	549-846 664.8	671-927 773.7	675-1,598 857.7
The Grabowa		446-502 463.4	437-486 453.7	485-613 508.4	496-572 512.7
The Czerwona	aluminium in water ($mg\ Al\ dm^{-3}$)	0.057-0.095 0.074	0.056-0.116 0.098	0.069-0.138 0.110	0.001-0.003 0.002
The Grabowa		0.048-0.147 0.116	0.087-0.425 0.251	0.164-0.273 0.193	0.008-0.009 0.010
The Czerwona	aluminium in bottom sediments ($mg\ Al\ kg^{-1}$)	12.02-42.96 20.72	7.549-35.96 18.01	6.739-35.63 12.87	7.15-47.06 20.19
The Grabowa		3.965-25.32 11.99	4.942-26.15 12.02	4.255-25.51 12.03	5.081-25.96 12.46
The Czerwona	aluminium in aquatic plants ($mgAl\ kg^{-1}$)	1.851-25.00 7.782	0.583-91.63 14.07	0.612-47.12 6.014	0.941-45.70 6.875
The Grabowa		2.012-1077 156.6	0.231-45.31 7.481	0.351-90.71 12.22	2.031-860.5 71.50
The Czerwona	concentration factor (CF)	166.6-464.4 278.4	78.39-381.7 189.9	66.72-269.2 116.2	3215-23492 10076
The Grabowa		34.11-282.5 130.9	14.81-183.4 63.59	19.89-155.7 61.65	529.3-2763 1367
The Czerwona	bioconcentration factor (BCF)	26.1-362.8 75.79	5.065-796.8 103.8	5.621-689.5 56.18	376.5-24052 416.6
The Grabowa		15.96-10478 1506	1.102-312.9 39.05	2.09-550.8 65.71	206.7-70132 7955

sium carbonates, chlorides, sulphates and other minerals such as aluminium. The electrolytic conductivity of the two rivers fell within the range from 437 to 1,598 μS cm^{-1}

In the middle section of the rivers, the water had lower mineralization than upstream and downstream. Particularly high values were recorded at site No. 1, where water flows through wooded and upland areas. A higher electrolytic conductivity at the outflow into the Baltic Sea indicates that the value was affected by seawater. The most probable cause was an earlier forcing of seawater into the river. Throughout the four-year study, conductivity values for the Grabowa were higher than those for the Czerwona.

The aluminium concentration in the water of both rivers oscillated between 0.001 and 0.425 mg Al dm^{-3} (Table 2). It grew downstream, towards the outflow into the Baltic Sea.

A number of surface water bodies in Poland and abroad present a range of aluminium concentrations very similar to the ones found in the two coastal riversexamined herein. Examples include small rivers in Lower Silesia, flowing near the city of Wrocław (0.009-0.035 mg Al dm^{-3}) and streams in the Karkonosze Mountains (0.011-0.018 mg Al dm^{-3}), which were studied by SAMECKA-CYMERMAN and KEMPERS (2003). Another example is the Thames, which collects urban and industrial pollution from its catchment (NEAL et al. 2000). Also, rivers in industrial regions in Poland (near the town of Legnica and in Upper Silesia) as well as in Russia have aluminium concentrations similar to those in the coastal rivers we examined (RODUSHKIN et al. 1995, SAMECKA-CYMERMAN, KEMPERS 2003, 2004).

A significant impact on the aluminium concentration in water is exerted by the water pH and the more acid it is, the higher aluminium concentration in water (KLUCZKA et al. 2012). However, aluminium concentration in the water of acidic lakes in the US near New Jersey did not exceed 0.4 mg Al dm^{-3} (SPRENGER, MCINTOSH 1989), while in acidified streams in the Rocky Mountains it was higher (0.69-2.00 mg Al dm^{-3}) than in coastal rivers (MCKNIGHT, BENCALA 1990). Also, higher values were recorded in European highlands (in Poland and the Czech Republic) subject to acidification – from 0.145 to 1.17 mg Al dm^{-3} (WRÓBEL 1993, VESELÝ et al. 1998). Aluminium concentrations higher than in the Czerwona and Grabowa have been found all over the world (in waters subject to various forms of anthropogenic influence, mainly connected with industry) and developed municipal management (JORDÃO et al. 2002, GUIBAUD et al. 2003, GUNKEL et al. 2011).

The correlation between the aluminium concentration in water and its pH was significant only in the Grabowa in 2009 (r = -0.507; $P < 0.05$) and the Czerwona in 2011 (r = 0.601; $P < 0.05$).

Bottom sediments

The aluminium concentrations in the bottom sediments of the two rivers oscillated between 3.965 and 47.06 mg Al kg^{-1} (Table 2). The lowest values in the whole four-year research period in both rivers were recorded in samples taken in the middle section of the rivers.

The correlation coefficients between aluminium concentrations in the water and in the sediments of the Czerwona River, except for the last year of the study, were higher than for the Grabowa. For both rivers, the correlations proved significant. Irrespective of being positive or negative, the correlation coefficients calculated for the Grabowa River, with the highest value (r = -0.219; $P < 0.05$) achieved in 2009. On the other hand, the highest correlation coefficient for the Czerwona River occurred in 2008 (average correlation r = 0.521; $P < 0.05$).

The correlations between aluminium concentrations in the sediments and the pH of the water were significant, and higher for the Czerwona than for the Grabowa. The correlations for the two rivers were the highest in 2009, reaching r = -0.341 ($P < 0.05$) for the Grabowa and r = -0.610 ($P < 0.05$) for the Czerwona. The dependence between the aluminium concentration in the sediments and conductivity was weak for samples from the Grabowa. For the Czerwona, the correlation was positive and high in two consecutive years (2009 and 2010), amounting to r = 0.632 and r = 0.610 ($P < 0.05$), respectively.

The concentraions of Al found by analyzing the sediments were low and similar to those recorded for streams in the Karkonosze Mountains, which also have a typically wooded catchment area (Samecka-Cymerman, Kempers 1999). Likewise, bottom sediments of rivers outside Poland are characterized by similar value ranges (Baruah et al. 1996, Carpentier et al. 2002, Yalcin et al. 2007). Higher aluminium concentrations in river sediments have been found in lowland areas of Lower Silesia (Samecka-Cymerman, Kempers 2003, Szalińska et al. 2010).

Ałuminium concentrations in bottom sediments in acidified lakes tend to be higher than in the Czerwona and Grabowa rivers (Sprenger, Mcintosh 1989) and in those receiving industrial wastewater or located in the immediate vicinity of such areas (Samecka-Cymerman, Kempers 2001, 2003, 2004). In areas with potentially low pollution in northern and eastern Poland, aluminium concentrations are similar (Trojanowski, Antonowicz 2005). Also, stagnant water bodies around the world, including lakes and marshes, have been found to have similar aluminium concentrations as the Czerwona and Grabowa rivers (Rossi et al. 1993, Baudo, Beltrami 2001, Jordão et al. 2002, Mwamburi 2003, Léopold et al. 2008).

Vegetation

During the four years of the study, the aluminium content in plants of the Czerwona River oscillated between 0.583 mg Al kg^{-1} in common reed and 91.63 mg Al kg^{-1} purple loosestrife (Table 2). This is also the range of values for emergent plants, which predominate in the Czerwona.

The lowest aluminium concentration in plants sampled from the Grabowa was 0.231 mg Al kg^{-1}, and the highest reached 1,077 mg Al kg^{-1} (Table 2).

The less common submergent macrophytes had more aluminium (on average, 240.2 mg Al kg^{-1}) than emergent plants (on average, 14.47 mg Al kg^{-1}). The minimum value was also the lowest one for all emergent plants. The highest value was also the maximum for submergent plants.

The submergent plants growing in the Grabowa River had more aluminium than those from the Czerwona. This can be explained by the the Grabowa's much larger catchment area and length. The Grabowa catchment also has more sources of pollution, which can affect the aluminium content in the water and consequently in plants (Report WIOŚ 2008). Furthermore, more plant samples were taken from the Grabowa than from the Czerwona. Additionally, the makeup of submergent species in the two watercourses was different, which in combination with the unique tendency of individual species to accumulate metals (Senze et al. 2009) might have affected the results.

In the Czerwona, emergent vegetation was found to have more aluminium than submergent plants. Although the plant samples were rinsed after being collected, it cannot be ruled out that atmospheric precipitation before the study had been intensive and affected the settlement of aluminium compounds on plants. Fewer submergent plant samples were taken from the Czerwona than from the Grabowa.

The correlation coefficient for aluminium concentrations in water and plants, similarly to the water-sediment correlation, was higher for the Czerwona than for the Grabowa, except in 2010. The correlation between the aluminium content in plants and the pH of the river water was complete ($r = 1.000$, $P < 0.05$) for the Grabowa in 2008. For both rivers, the correlation between the aluminium content in plants and electrolytic conductivity was significant. Analysis of the correlation between the aluminium content in sediments and in plants indicated a relationship similar to that between water and plants. Likewise, for these values, the correlations were weak, with the highest value reaching barely $r = -0.296$ ($P < 0.05$) in the Czerwona River (2008).

The aluminium concentration in aquatic plants growing in stagnant or flowing water varies widely and depends on a number of environmental factors as well as on plant species. Of much importance is also the composition of the substrate and water from which plants take nutrients. Studies of submergent and emergent plants growing in heavily anthropogenized areas show that such vegetation contains more aluminium than the emergent aquatic plants sampled from the Czerwona or the Grabowa Rivers (Samecka-Cymerman, Kempers 2001, 2003, 2004, Senze, Kowalska-Góralska 2013).

Aluminium concentration factors (sediments-water)

Aluminium accumulation expressed as a concentration factor (CF) in the bottom sediments of the rivers ranged from 14.81 to 23,492 (Table 2). With the exception of 2009, the highest aluminium CFs were recorded for site No. 13. The aluminium accumulation in the bottom sediments of the rivers covered by the study, expressed as the CF, was typical of water with a moderate level of pollution (Kozubek, Marek 2002).

Aluminium bioconcentration factors (vegetation-water)

The biocencentration factor for aluminium in the Czerwona River oscillated between BCF = 5.065 in common reed and BCF = 24052 in greater pond sedge (Table 2). The high aluminium BCF in 2011, as compared to notably lower values in the previous years, resulted from the very low aluminium concentration in the Czerwona River water.

The aluminium bioaccumulation in the acquatic plants of the Grabowa oscillated between 1.102 (reed canarygrass) and 70,132 (Canadian pondweed) – Table 2. BCFs were significantly higher in the first and last years of the study than in the other two years. In all the syears, the aluminium BCFs for submergent plants were substantially higher than for emergent ones.

The BCFs calculated in the study proved similar to those typical of submontane areas in the Czech Republic that are exposed to relatively little industrial pollution (Senze, Kowalska-Góralska 2013). Macrophytes in rivers flowing in submontane and wooded regions have limited opportunities to absorb aluminium from sediments, because aluminium compounds in such sediments are scarce. Acquatic plants in the Legnickie Copper Mining Region also accumulate aluminium to a similar degree (Samecka-Cymerman, Kempers 2003, 2004), but the aluminium accumulation in Lower Silesian rivers that collect pollution from the area of Wrocław agglomeration, compared to the above results, was very high, with the values ranging from BCF = 89,300 to BCF = 125,000 (Samecka-Cymerman, Kempers 2003). The most probable explanation is the large concentration of aluminium in bottom sediments, from which – under appropriate circumstances – plants are able to absorb the metal.

CONCLUSIONS

1. The pHs of the waters in the two rivers were close to neutral and did not stimulate the growth in aluminium concentrations.

2. The aluminium concentration in aquatic plants was relatively low and typical of environments subject to weak anthropopressure.

3. The aluminium concentrations in the two rivers covered by the study do not pose any danger to the environment of Western Pomerania or to the areas they flow through before feeding into the Baltic Sea.

4. There was no effect of the aluminium content in the water and sediments on the level of aluminium in aquatic plants.

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