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Research Article

ASSESSMENT OF HEAVY METALS CONTENTS IN BOTTOM SEDIMENTS OF BUG RIVER

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ABSTRACT
The development of industry, agriculture, and transport contributes to an increased environmental pollution by heavy metals. The aim of the study was preliminary assessment of the contents of selected metals (lead, cobalt, copper, chromium, cadmium and nickel) in the sediments of Bug river. The study comprised part of the river flowing through Poland. It was found that the Bug river sediments are not contaminated in respect to the content of tested metals. Based on the analysis of the study results, these metals can be lined up in the following order: Cr > Pb > Cu > Ni > Co > Cd. Statistical analysis showed that copper and chromium occur in Bug river sediments in forms bindings with organic matter in majority of cases. The granulometric analysis of sediments from Bug river revealed the largest percentage of two fractions: 1.0–0.2 mm with average of 47.7 ± 19.77% and 0.2–0.1 mm with average of 20.6 ± 7.7%. These are the dominant fractions with the accumulation of metals in river sediments, which has been confirmed by statistical analysis.

Keywords: Bug river, bottom sediment, heavy metals.

INTRODUCTION

The development of industry, agriculture, and communication contributes to an increased environmental pollution by heavy metals. Heavy metals are stable elements that last in the environment as they are not degraded or destroyed. Therefore, they tend to accumulate in the soil, sediments, and organisms [Yu et al. 2000, Demirezen, Aksoy 2004, Dmochowski et al. 2005, Wiśniowska-Kielian, Niemiec 2005, Sasmaza et al. 2008]. Contamination of rivers due to metals can originate from natural and anthropogenic sources. Heavy metals in the waters are subject to chemical transformations, the adsorption and complexation, then the processes of sedimentation and accumulation in bottom sediments. At low concentrations, most of heavy metals behave as microelements, while in higher concentrations, they are toxic [Nelson et al. 2000]. Analysis of the contents of metals accumulated in bottom sediments of a water reservoir is a valuable material for the description of sources, rate, and ways of metal distribution in the reservoir. Bottom sediments are good indicators of the contamination degree of the river environment [Kucuksezgin et al. 2008, Essien et al. 2009, Skorbiłowicz 2012]. Concentration of metals in sediments strongly exceeds their contents in waters, therefore the chemical analysis of sediments can detect them even when their quantities are low in waters [Bojakowska et al. 2006]. The accumulation of metals in sediments is controlled by a number of environmental factors such as pH, Eh, salinity, type and concentration of organic and inorganic ligands, hydraulic processes in rivers, and sediment grain size (Helios-Rybicka 1991).

The aim of the study was preliminary assessment of the content of selected metals (lead, cobalt, copper, chromium, cadmium and nickel) in bottom sediments of Bug river. The study comprised part of the river flowing through Poland.

MATERIAL AND METHODS

Study area

The Bug river is one of the longest in Poland and also belongs to the largest right tributaries of the Vistula river and left Narew river. The catchment area of the river is situated in the central-eastern Poland, the north-western Ukraine, and south-western Belarus. The Bug river catchment area is 39 420 km^2: 19 284 km^2 in Poland, which represents 49% of the Bug River catchment and 20 136 km^2, i.e. 51% beyond its borders. The average height of the Bug river catchment is 183 meters above sea level and the average river inclination is 0.3‰.

The length of the Bug river is approximately 755 km. The lower section of the river with a length of 207 km (28%) is located on the territory of our country, while 185-kilometer upper section represents 24% of the total catchment area and is situated outside Polish borders. The border zone between Poland, Belarus, and Ukraine stretches through 363 km (48%).

In its lower section, Bug river runs through the lowlands. They consist of sand, here and there covered with illuvial deposits. In the north of Terespol, the river flows through 3–4 meters wide valley located in the surrounding of hills of up to about 80 meters high. The greatest narrowing is found in Melnik surroundings. In a further section, the Bug river valley again expands to about 5 km. In the area of the middle and lower Bug river, arable lands and grasslands dominate. There are also mixed forests, although the lower part is dominated by coniferous afforestation. The main tectonic elements of the ground in Bug river valley is a marginal part cut by numerous faults in a direction perpendicular to its borders. They make this zone is composed of continuous depressions and elevations.

The river cuts across the faults structures on almost the entire length of the valley. Loess is the dominant surface formation on plateau areas. The dammed sediments prevail in the valley coastal zones. The plains are covered with boulder clay and fluvio-glacial formations. Lower terraces are built of sands and gravels covered by high-water flow sediments. Organic sediments, peats, and silts are accumulated in the depressions of terrace surface. In the bottom of the valley, there are also common Aeolian forms – sand dunes and wind-blown sand fields [Dojlido 2003].

The catchment area of Bug river is dominated by arable lands (50.3%) as well as meadows and pastures (17.7%). Forests cover 23.5% of the catchment area. Forests are predominated by pine, fir, spruce, ash, birch, oak, and alder.

There are 25 cities, that differ with economic status, population, and building character, within Bug river catchment. Their economic and industrial growth is a result of localization at the intersection of major rail and road routes, and in the case of Terespol – of the border crossing. Cities with a small number of inhabitants dominate in the river catchment area. Only the three largest cities can be distinguished: Siedlce (72 858 inhabitants), Biała Podlaska (55 424 inhabitants), and Chełm (69 016 inhabitants).

Nine industrial activities can be found in the catchment area. These include tanning, glass, ceramic, machine-metal, light, cement, electrical, food, and tourist industries. The tanning industry is localized in Włodawa, although it is very harmful to the environment. In the vicinity of Wyszków and Chełm, the glass industry facilities operate. In the area from Chełm to Belżec, pottery plants function. Machine-metal industry can be found in larger cities such as Łuków, Włodawa, Biała Podlaska, Chełm, Międzyrzecz, and Siedlce. One of the biggest Polish cement industry facility is localized in Chełm. Branches of high technology, i.e. electrical industry may be encountered in the north-western part of the catchment, more precisely in Węgrów, Woźniki, and Biała Podlaska.

Top-developed industry is the food industry, which can be located in any locality within discussed catchment. The largest sectors of this industry include such branches as baking, dairy, sugar, meat, fruit and vegetable. The second most developed industry is tourism, which is mainly located in areas with rich and beautiful landscape. The main sectors of the industry are farmhouses, tourist attractions (hiking, cycling, and water tracks), accommodations and dining sites.

Bug river, together with its tributaries, is also the receiver of wastewaters from municipal and industrial sources. The main point sources include sewage inflow from the most populous cities, entering the sewage system and that from urban and industrial areas. The contamination factors are also effluents coming from the Ukraine from mines and industrial plants.

Field and laboratory works

The fieldwork was conducted in 2012 in 11 measurement points located on the Bug river (Drohiczyn, Siemiatycze, Mielnik, Niemirów, Krzyczew, Terespol, Sławatycze, Włodawa, Dorohusk, Horodło, and Kryłów). The object for analyses was composed of bottom sediments, in which contents of lead, cobalt, copper, chromium, cadmium, and nickel, were determined. Bottom sediments were collected in the coastal zone, where suspended material is deposited (Bojakowska and Sokołowska 1992, Bojakowska 2001). Several individual surface samples of bottom sediments (from a depth of 5 cm) from beneath the water, from every selected measurement point. After mixing the test material, representative sample (weighing about 1000 g) was achieved. The samples were then dried in air to the "air-dried" status and stored until laboratory tests [Lis and Pasieczna 1995]. Prior to chemical analyzes, the bottom sediment sample was dried at 40 °C and sieved through a nylon mesh of 0.2 mm. Sediments were mineralized with nitric acid [Frančišković-Bilinski et al, 2005, Kunwar P. Singh et al. 2005, Salminen et al. 2005] in a closed microwave system CEM Mar-5. The concentrations of Pb, Co, Cu, Cr, Cd, and Ni in sediments were determined by means of ASA. The measurements were performed using Varian Spectra AA100 equipment. The correctness of the methodology was verified on a base of the reference material NCSDC 733 analysis. Achieved results for tested metals contents were presented in relation to air-dry sediments and compared with the literature data as well as arithmetic mean of these metals in bottom sediments for Poland (fraction < 0.2 mm) [Lis and Pasieczna 1995] and Europe (fraction < 0.15 mm) (Salminen et al. 2005) and soil of Polish Lowlands area [Pasieczna 2003]. To assess the extent of sediment contamination by heavy metals, the proposed classification of aquatic sediments in Poland on the basis of geochemical criteria [Bojakowska and Sokołowska 1998] and on the basis of threshold values taking into account the harmful effects of pollutants accumulated in sediments towards aquatic organisms [Bojakowska 2001], was applied. For sediments, the granulometric composition analysis was carried out, which consisted in separating the raw air-dry sediment samples to size fractions using an appropriate set of sieves (mesh: 2 mm, 1 mm, 0.2 mm, 0.1 mm, 0.063 mm) mounted to a mechanical shaker. After the screening complete, the individual sieve fractions were weighed, then the weight percentage of particular grain size samples in reference to the total sample weight, was calculated. In addition, the sediments were subject to analysis of organic compounds content by means of annealing (550 °C). The pH of sediments and river water was performed applying potentiometry.

The licensed version of Statistica 10 software was used for statistical processing of results. Arithmetic mean, median, standard deviation, and Spearman correlation coefficient values were calculated. The results were also subject to cluster analysis based on the concept of distance of objects or variables in a multidimensional space.

RESULTS AND DISCUSSION

Granulometric analysis of sediments collected from Bug river showed mainly varied character of size distribution and resembled in majority the features of loose and weak loamy sand. It has been reported the largest percentage of two fractions: 1.0–0.2 mm (average 47.7 ± 19.77%) and 0.2–0.1 mm (average 20.6 ± 7.7%). The smallest percentage characterized fraction 0.1–0.063 of sediments (average 7 mm ± 5.09%). The coarsest grain size (fraction > 2 mm) occurred at the measurement points Niemirow (21.43%) and Wlodawa (13.17%). This was due to the fact that the least favorable conditions for deposition of the smallest of loam and dust parts were present at those points (Table 1).

The organic matter content in analyzed samples of bottom sediments ranged from 0.8% to 19.3%. Samples collected in the localities Mielnik (19.3%) and Drohiczyn (11.2%) were characterized by the highest contents of organic matter. The lowest content, not exceeding 3%, was recorded at the measuring points: Sławatycze, Horodło, and Kryłów (Table 2).

The Bug river water was also subject to pH determination, which ranged from 6.2 to 7.5 pH, while the pH of the bottom sediment in most cases was neutral ranging from 6.3 to 7.3 pH.

Table 2 presents the results of heavy metals contents determinations: lead, cobalt, copper, chromium, cadmium, and nickel, in bottom sediments of Bug river flowing through Poland.

Bojakowska et al. [2006] reported that the lead content in uncontaminated sediments gen-

Table 1. Contents of particle fractions in bottom sediments from Bug river

River	Measurement point	Content of particle fraction [% by weight]					
		>2 mm	2.0–1.0 mm	1.0–0.2 mm	0.2–0.1 mm	0.1–0.063 mm	<0.063 mm
Bug	Drohiczyn	8.23	10.37	37.60	20.80	12.60	10.40
	Siemiatycze	12.97	12.87	22.10	24.07	10.32	17.67
	Mielnik	9.47	21.07	30.20	20.20	12.23	6.83
	Niemirów	21.43	11.5	47.80	14.27	2.17	2.83
	Krzyczew	3.93	3.80	37.17	33.60	9.63	11.87
	Terespol	1.35	4.39	85.03	7.30	1.15	0.78
	Sławatycze	2.30	3.13	52.17	28.13	6.77	7.50
	Włodawa	13.17	11.87	57.43	12.77	2.50	2.26
	Dorohusk	3.23	7.47	60.47	23.63	3.06	2.14
	Horodło	3.40	8.50	24.43	26.87	15.00	21.80
	Kryłów	3.30	7.03	70.60	15.23	1.97	1.87
General statistical data							
Minimum		*1.35*	*3.13*	*22.10*	*7.30*	*1.15*	*0.78*
Maximum		*21.43*	*21.07*	*85.03*	*33.60*	*15.00*	*21.80*
Arithmetic mean		*7.5*	*9.2*	*47.7*	*20.6*	*7.0*	*7.8*
Median		*3.9*	*8.5*	*47.8*	*20.8*	*6.8*	*6.8*
Standard deviation		*6.25*	*5.15*	*19.77*	*7.7*	*5.09*	*6.99*

Table 2. Measured indicators for Bug river

River	Measurement point	River water pH	Sediment pH (pH_{H2O})	Organic matter [%]	Pb	Co	Cu	Cr	Cd	Ni
					Heavy metals contents in bottom sediments [mg·kg^{-1}]					
Bug	Drohiczyn	7.1	6.8	11.2	4.8	3.8	3.6	8.3	0.2	2.9
	Siemiatycze	6.8	6.7	6.1	9.1	6.2	12.1	27.2	0.4	7.8
	Mielnik	6.5	6.6	19.3	4.9	3.2	5.3	10.2	0.3	6.1
	Niemirów	6.2	6.7	5.8	4.6	5.9	6.5	9.8	0.4	2.9
	Krzyczew	6.7	6.8	3.1	2.8	3.6	3.4	7.3	0.5	4.2
	Terespol	7.5	7.3	0.8	6.3	4.5	1.9	5.1	1.1	5.3
	Sławatycze	7.4	6.9	1.8	4.2	5.3	4.6	10.2	0.4	5.1
	Włodawa	7.2	6.3	5.9	5.8	3.7	3.6	13.1	0.3	6.1
	Dorohusk	7.7	6.8	3.1	9.3	4.3	4.5	14.1	0.6	1.9
	Horodło	7.5	6.9	2.2	1.1	3.1	2.3	4.1	0.7	3.1
	Kryłów	6.7	6.8	2.8	6.8	4.3	4.1	6.1	0.5	4.2
General statistical data										
Minimum		*6.2*	*6.3*	*0.8*	*1.1*	*3.1*	*1.9*	*4.1*	*0.2*	*1.9*
Maximum		*7.5*	*7.3*	*19.3*	*9.3*	*6.2*	*12.1*	*27.2*	*1.1*	*7.8*
Arithmetic mean				*5.6*	*5.4*	*4.4*	*4.7*	*10.5*	*0.5*	*4.5*
Median				*3.1*	*4.9*	*4.3*	*4.1*	*9.8*	*0.4*	*4.2*
Standard deviation				*5.37*	*2.44*	*1.04*	*2.76*	*6.35*	*0.24*	*1.76*

erally does not exceed 30 mg·kg^{-1}. The pattern of lead concentration variability in bottom sediments coincides substantially with the geochemical pattern of this element in soils. Geochemical Atlas of Poland indicates that the arithmetic mean for lead is 68 mg·kg^{-1} [Lis and Pasieczna 1995] and for Europe it is 38.6 mg·kg^{-1} [Salmine et al. 2005]. Much less contents were recorded in present studies. The lead content in tested sediments ranged from 1.1 to 9.3 mg·dm^{-1} (average of 5.4 ± 2.44 mg·kg^{-1}). The highest lead concentrations above 9 mg·kg^{-1} were recorded at the measuring points Siemiatycze and Dorokhusk. On this basis, it can be concluded that the Bug river bottom sediments are not contaminated with lead. Sediments from the Odra river collected near Nowa Sól – Kostrzyń contained 24–84 mg·kg^{-1} of lead [Jędrzejczak and Czyrski 1990], whereas sediments from the upper Narew river from 9.2 ± 4.65 to 22.9 ± 15.14 mg·kg^{-1} [Skorbiłowicz 2012].

Cobalt in bottom sediments from non-industrialized areas is usually present at levels up to several mg·kg^{-1}. In general, Co content higher than 10 mg·kg^{-1} is associated with anthropogenic activity [Bojakowska and Sokołowska, 1998]. The study showed variations in the content of this element from 3.1 mg·kg^{-1} to 6.2 mg·kg^{-1}. The average cobalt concentration in bottom sediments of Bug river was 4.4 ± 1.04 mg·kg^{-1}, which was less than the average value for European sediments 11.2 mg·kg^{-1}.

The copper content in bottom sediments is related to the type of macierzysta rocks and its average content reaches only a few mg·kg^{-1}. Fox and Pasieczna [1995] reported that differences in the copper content in aquatic sediments is much higher than in the soils. The average copper concentration in soils of Polish Lowlands is 3 mg·kg^{-1} [Pasieczna 2003], while average content of Cu for European sediments is 22.1 mg·kg^{-1} and for Polish sediments 21 mg·kg^{-1}. Bojakowska and Sokołowska [1998] consider value of 6 mg·kg^{-1} the background of copper in Polish sediments. Copper in bottom sediments collected from Bug river occurred in relatively small contents (1.9–4.6 mg·kg^{-1}) and in majority of studied samples it appeared below geochemical background. The exception were measurement points localized in Siemiatycze (12.1 mg·kg^{-1}), Niemirów (6.5 mg·kg^{-1}) and Mielnik (5.3 mg·kg^{-1}).

The highest contents among the tested metals were recorded in the case of chromium. The amount of chromium in studied bottom sediments ranged from 4.1 to 27.2 mg·kg^{-1} (average 10.5 ± 6.35 mg·kg^{-1}). Variable chromium contents were observed in the sediments along with the river flow. The largest concentration of the element occurred at the measurement point in Siemiatycze (27.2 mg·kg^{-1}), then chromium quantities decreased to the point in Włodawa (13.1 mg·kg^{-1}) and Dorohusk (14.1 mg·kg^{-1}); at subsequent points, chromium concentrations were even lower. The tanning industry using chromium compounds in technological processes, is localized in the Bug river catchment. In part, this may be the cause of increased contents of this element in studied sediments in the case of discharging the wastewaters from tanneries into Bug river water. Considering achieved results in a view of natural geochemical chromium contents provided by Bojakowska and Sokołowska [1998] at the level of 5 mg·kg^{-1}, it must be noted that the amount of chromium has not been exceeded only in 2 examined sediment samples. In contrast, Lis and Pasieczna [1995] argued that natural concentration of chromium in bottom sediments of not contaminated rivers reaches 10 mg·kg^{-1}. In relation to this value, chromium content has been exceeded in five samples of analyzed sediments. It is also worth mentioning that results of Cr content achieved for sediments from Bug river were much lower than the average value for bottom sediments in Europe (92.8 mg Cr·kg^{-1}).

Cadmium contents in the range from 0.2 to 1.1 mg Cd·kg^{-1} (average 0.5 ± 0.24 mg· kg^{-1}) was observed in collected samples of bottom sediments. The geochemical criteria allowed to classify 8 sediment samples to geochemical background, while 2 samples (Dorohusk – 0.6 mg·kg^{-1} and Horodło – 0.7 mg·kg^{-1}) to the first geochemical class [Bojakowska Sokołowska 1998]. Surprisingly high concentrations of cadmium were observed in bottom sediments from Bug river in Terespol (1.1 mg·kg^{-1}) (II geochemical class). The border crossing is localized in Terespol, which is associated with an intense traffic flows. Wang et al. [2006] claimed that among variety of human activities contributing to the natural environment degradation, broadly understood transport is one of the most important. On areas of high development density, products of liquid fuels combustion cannot be quickly and freely moved, which in turn is the cause of very high concentrations of heavy metals in urban atmospheric air [Sutherland 2000]. Increase in heavy metal contamination of aquatic ecosystems localized on areas with intense traffic, is very disturbing trend. These results of cadmium concentrations in bottom sediments from Bug river in Terespol seem to confirm the thesis about the impact of road transport on the content of cadmium in aquatic environment.

Nickel levels in Polish aquatic sediments are similar to those in soils. On Polish Lowland, nickel content does not exceed 10 mg·kg^{-1} [Piaseczna 2003]. The arithmetic mean nickel content in the case of sediments from surface waters for Polish amounts to 11 mg·kg^{-1} and to 35.2 mg·kg^{-1} for Europe. The study upon bottom sediments in Bug river showed that nickel concentrations ranged between 1.9–7.8 mg·kg^{-1} with average of 4.5 ± 1.76 mg·kg^{-1}. The maximum contents were recorded in Siemiatycze (7.8 mg·kg^{-1}) as well as Mielnik and Włodawa (6.1 mg·kg^{-1}).

Statistical analysis of achieved results indicated that copper and chromium contents were significantly positively correlated with the content of organic matter (r_{Cu} = 0.64, r_{Cr} = 0.65). There was also a positive correlation between copper and chromium (r = 0.69 at $p < 0.05$), which may indicate that both elements have similar pollution sources. The cluster analysis (CA) was also performed in relation to the variability of metals concentrations and sediment grain size, water and sediments pH value, and organic matter. Figure 1, being the result of cluster analysis, illustrates two separate main groups and two subgroups. Two fractions are present in main groups. Analysis of particle size distribution of bottom sediments collected from Bug river showed the largest percentage of two fractions: 1.0–0.2 mm (average 47.7%) and 0.2–0.1 mm (average 20.6%). These are the dominant fractions in sediments of Bug river. The accumulation of metals in bottom sediments is affected by sediment grain size. However, the first subgroup consisted of the following components: fraction > 2 mm, 2.0–1.0 mm fraction, organic matter, chromium, fraction 0.1–0.063 mm, and fraction < 0.063 mm, whereas the second subgroup consisted of: sediment and water pH, and metals: lead, cobalt, nickel, copper, and cadmium.

When comparing the results of present studies with those carried out in other rivers [Wardas 2001, Wiśniowska-Kielian Niemiec 2005, Samecka-Cymerman Kempers 2007, Skorbiłowicz 2012], it is evident that heavy metals contents in bottom sediments from Bug river were lower. It has been found that lead, copper, cadmium, and nickel were at a geochemical background level, while cobalt and chromium were within the range of I geochemical class (Table 3).

CONCLUSIONS

1. Granulometric analysis of bottom sediments from Bug river revealed the largest percentage of two fractions: 1.0–0.2 mm (average 47.7 ± 19.77%) and 0.2–0.1 mm (average 20.6 ± 7.7%). These are predominant fractions in Bug river sediments having the largest impact

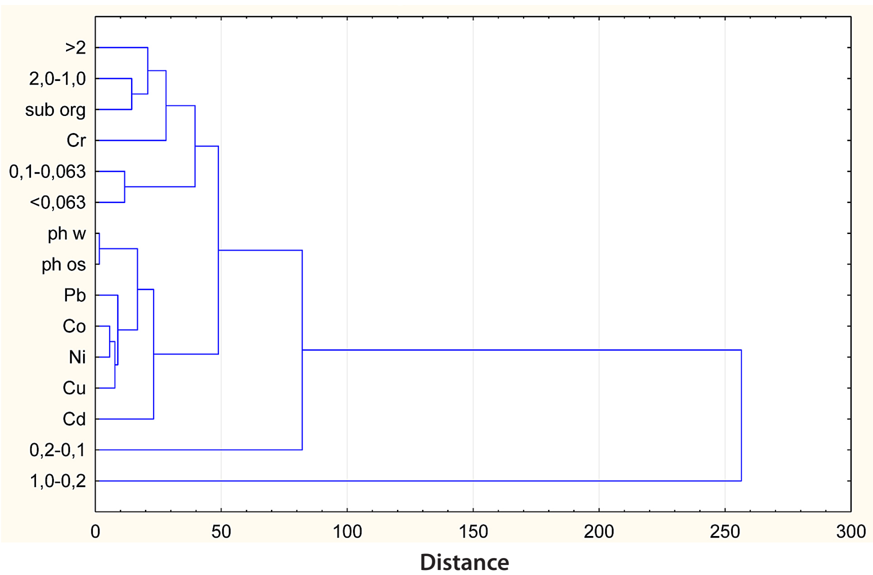

Figure 1. Cluster analysis (CA) by Ward for Bug river

Table 3. Geochemical classification of bottom sediments from Bug river on a base of arithmetic mean value [Bojakowska Sokołowska 1998, Bojakowska 2001]

Bug river	Pb	Co	Cu	Cr	Cd	Ni
	[mg·kg^{-1}]					
	background	I	background	I	background	background

on metals accumulation in bottom sediments, which was confirmed by statistical analysis. Fraction 0.1–0.063 mm (average 7 ± 5.09%) was characterized by the lowest proportion.

2. It was found that bottom sediments from Bug river, due to the contents of lead, cobalt, copper, chromium, cadmium and nickel, were not contaminated. Results of studies upon metals contents allow to line them up in the following sequence: Cr > Pb > Cu > Ni > Co > Cd.
3. Statistical analysis showed that copper and chromium in bottom sediments of Bug river occurred in majority as bound to organic matter.

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