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ASSESSMENT OF MICROSCALE VARIATION OF HEAVY METAL POLLUTION OF THE BYSTRZYCA RIVER ALLUVIA DOWNSTREAM FROM LUBLIN

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Abstract. Fluvial sediments accumulated in the bottoms of river valleys downstream from large cities are characterised by higher levels of heavy metal content, which poses a threat to the environment and humans. This study presents a comprehensive assessment of the degree of pollution of four alluvial sediment profiles (80 samples), collected from the bottom of the Bystrzyca river valley downstream from Lublin, conducted with the use of five geochemical indices. Channel deposits and sediments (alluvial soils) sampled from the floodplain were analysed. The content levels of the six heavy metals under study were as follows: Cd: 10.6–291.2 mg/kg, Cr: 53.1–292.4 mg/kg, Cu: 20.4–223.1 mg/kg, Ni: 2.9–19.3, Pb: 39.3–280.3, Zn: 108.9–991.4 mg/kg. The horizontal and vertical variation of the pollution level was linked with the history of anthropogenic pressure on the one hand, and the geomorphological location of a given profile on the other. Heavy metal content in the samples did not show any correlation with grain size composition, organic matter content, and Fe and Mn content. Cadmium was the element whose concentration levels were comparable with those in alluvial sediments of rivers in industrialised areas while the indices for the other metals showed varied levels of pollution: from low to high. However, the ecological risk is high for all samples as indicated by the synthetic potential ecological risk index, which takes into account the toxicity of all the metals under study.

Keywords: alluvia, alluvial soils, Bystrzyca river, heavy metals

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INTRODUCTION

Human activity causes increased levels of various harmful substances in the environment. Intensive accumulation of heavy metals takes place in the bottoms of river valleys in industrial and urban areas. This is why sediments occurring there contain high levels of these elements harmful to people and nature (Bojakowska 1994; Lis, Pasieczna 1995; Thonon 2006), which poses a threat of pollution to alluvial soils and possible pollution of water if the polluted sediments are mobilised. The condition of the geochemical environment of fluvial sediments is discussed primarily with regard to industrial areas and valleys of large rivers (Ciszewski 2002; Szwarczewski 1998). The impact of the character of fluvial processes on the variation of pollution level in sediments occurring in river valleys is also stressed (Ciszewski 1997; Martin 2000).

Studies conducted so far indicate increased levels of heavy metals in the alluvia in the Bystrzyca river downstream from Lublin (Bojakowska, Sokołowska 1996; Zgłobicki 2008). They provide information on the pollution of the surface layers of alluvial sediments and soils in the context of the Bystrzyca river bottom. The objective of this study was to determine the spatial, microscale variation of heavy metal pollution in alluvial sediments depending on their geomorphological position. Therefore, the vertical variation of the content of six heavy metals in four alluvial sediment profiles was analysed in detail.

MATERIALS AND METHODS

Lublin is a provincial capital with about 350 thousand inhabitants. The city is a source of sanitary, industrial and rainwater wastes; the latter is not treated. In the past, the amount of pollutants reaching the Bystrzyca river was considerably influenced by the large industrial plants (currently closed) operating in the city and the lack of a modern wastewater treatment plant until the early 1990s. The maximum pollutant discharge to the Bystrzyca river occurred in that period (Michalczyk *et al*. 1997). The situation began to improve starting form 1991 when the biological treatment of wastewater started at the treatment plant in Hajdów.

The studies were conducted in the Bystrzyca valley at Turka, downstream from the municipal treatment plant in Hajdów (Fig. 1). The sample collection points were thus located downstream from all sources of pollutant discharge related to the city of Lublin. The mean flow rate of the Bystrzyca is about 2.8 m^3/s in Lublin and 4.9 m^3/s at the point where it flows into the Wieprz river (Michalczyk *et al*. 1997).

Samples were collected at four points located within a straight stretch of the river channel (T4), a convex part of the meander channel (T2) and a floodplain

4 metres (T1) and 2 metres (T3) from the channel (alluvial soils). At each point, a continuous core down to the depth of 40 cm was collected and then divided into 2 cm fragments. It is estimated that a layer of such thickness could have accumulated over a period of a few years (Zgłobicki 2008).

The samples were dried and sieved through a 0.2 mm sieve. Heavy metal content in fluvial sediments is commonly examined in such a fraction (Bojakowska, Sokołowska 1998). Heavy metal content (Cd, Cr, Cu, Ni, Pb and Zn) was determined with an Epsilon 5 X-ray spectrometer (ED-XRF). Based on the data obtained, the indices (commonly used in literature) showing the degree of environmental pollution with heavy metals and the related threats were calculated: 1) geoaccumulation index (I_{geo}), enrichment factor (*EF*), pollution index (*Pi*), index of ecological risk (Er_i), potential ecological risk index (*RI*) (Müller 1979; Håkanson 1980; Ergin *et al*. 1991).

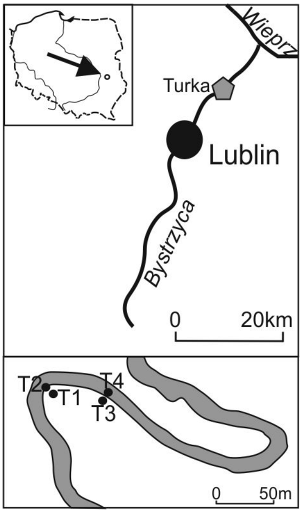

Fig. 1. Location of the sample collection points

The geoaccumulation index is defined with the following formula:

$$I_{geo} = \log_2 \left({C_n}/{1.5\, C_{ref}} \right) \tag{1}$$

where: C_n denotes the concentration of the metal; C_{ref} denotes the value of the background for the metal.

Figures provided in the publication by Bojakowska and Sokołowska (1998) were used as the background values. It should be mentioned that analytical methods applied by Bojakowska and Sokołowska (1998) and in our study were different (AAS and ICP MS versus ED-XRF). However, background values obtained by the ED-XRF method do not exist for Poland while mentioned values are in common use.

Six categories of pollution are distinguished (Table 1).

TABLE 1. POLLUTION LEVELS AS PER THE GEOACUMULATION INDEX AFTER MÜLLER (1979)

I_{geo}	Pollution level
<0	Unpolluted
0–1	Unpolluted to moderately polluted
1–2	Moderately polluted
2–3	Moderately to strongly polluted
3–4	Highly polluted
4–5	Highly to extremely polluted
>5	Extremely polluted

Enrichment factor is defined with the following formula:

$$EF = \frac{C_n / C_{ref}}{B_n / B_{ref}} \tag{2}$$

where: C_n is the metal concentration in a sample; C_{ref} is the background value for the metal under study; B_n is the reference metal concentration in the sample.

Fe, Al, Ti, Mn, Sc, Zr or Li can be a reference element (Fe was used as a reference element in this study). B_{ref} is the background value for the reference metal.

Five categories of pollution are distinguished based on the value of the enrichment factor (Table 2).

TABLE 2. POLLUTION CATEGORIES AS PER THE ENRICHMENT FACTOR AFTER ERGIN *ET AL.* (1991)

EF	Pollution level
<2	Minimal
2–5	Moderate
5–20	Significant
20–40	Very high
>40	Extreme

Pollution index (*Pi*) can be defined with the following formula:

$$Pi = \frac{C_n}{C_{ref}} \tag{3}$$

See the explanation of the indices above; pollution categories are shown in Table 3 below:

TABLE 3. POLLUTION LEVELS AS PER POLLUTION INDEX AFTER SHI AND WANG (2013)

Pi	Pollution level
≤1	Unpolluted
1–2	Low pollution
2–3	Moderate pollution
>3	High pollution

The index of ecological risk factor is defined with the following formula:

$$Er_i = T_i \times \frac{C_n}{C_{ref}} \tag{4}$$

T_i is the toxic-response factor for the metal. Håkanson (1980) provides the following values for this index: Cd – 30, Cr – 2, Cu – 5, Ni – 5, Pb – 5, Zn – 1. Five categories of pollution are distinguished (Table 4).

TABLE 4. CATEGORIES OF INDEX OF ECOLOGICAL RISK AFTER HÅKANSON (1980)

Er_i	Potential ecological risk
<40	Low
40–80	Moderate
80–160	Considerable
160–320	High
>320	Very high

Potential ecological risk (*RI*) is defined as the sum of the index of ecological risk factors for metals in a given sample (Soliman *et al*. 2015). Four categories of the index are distinguished (Table 5).

$$RI = \sum_{i=1}^{n} Er_i \tag{5}$$

TABLE 5. CATEGORIES OF POTENTIAL ECOLOGICAL RISK AFTER HÅKANSON (1980)

Er_i	Potential ecological risk
<150	Low
150–300	Moderate
300–600	Considerable
>600	High

RESULTS AND DISCUSSION

Silt (2-63 µm) is the dominant fraction in the samples under study, and its content ranges from 74 to 84%. Total organic carbon content varies from 2.8 to 6.1%. High heavy metal levels were found in the sediments under study. They were, on average, 6 to 200 times higher than the geochemical background determined for fluvial sediments in Poland (Bojakowska, Sokołowska 1998). The background was exceeded most in the case of Cd (40–200 times) and Cu (6–17 times) and least in the case of Zn (4–12 times) (Table 6). Based on the data obtained, the studied sediments (mean content) can be assigned to the following geochemical classes, according to the typology of fluvial sediments by Bojakowska and Sokołowska (1998):

a) Cd – third class (<20 mg/kg), T1, T3 and T4; non-class (>20 mg/kg), T2
b) Cr – second class (<100 mg/kg), all profiles
c) Cu – second class (<100 mg/kg), all profiles
d) Ni – first class (<30 mg/kg), all profiles
e) Pb – second class (<200 mg/kg), all profiles
f) Zn – second class (<1,000 mg/kg), all profiles

TABLE 6. MEAN, MINIMUM AND MAXIMUM HEAVY METAL CONTENT IN STUDIED PROFILES

Profile	Cd	Cr	Cu	Ni	Pb	Zn
			Mean content [mg/kg]			
T1	20.8	94.0	67.7	7.5	83.3	385.5
T2	106.8	165.0	104.7	19.3	143.1	564.4
T3	18.9	92.1	49.3	6.3	89.2	230.9
T4	16.3	78.7	42.6	5.3	57.7	252.2
			Minimum content [mg/kg]			
T1	12.0	62.8	37.0	4.8	51.0	198.4
T2	14.4	71.5	40.7	5.4	55.0	260.5
T3	12.0	74.0	31.6	4.5	67.1	108.9
T4	10.6	53.1	20.4	2.9	39.3	132.3
			Maximum content [mg/kg]			
T1	29.8	123.9	77.9	9.3	94.7	473.9
T2	291.2	292.4	223.1	46.7	280.3	991.4
T3	22.7	110.8	64.0	7.9	101.6	333.9
T4	19.3	98.1	66.4	7.5	74.8	372.8
Geochemical background[1]	0.5	6.0	6.0	5.0	10.0	48.0

[1] – after Sokołowska and Bojakowska (1998)

The following pattern occurred for mean, minimum and maximum concentrations in all profiles: Zn>Cr>Pb>Cu>Cd>Ni. Only in the case of maximum content levels in profile T2, the Cd concentration was slightly higher than the Pb concentration.

The highest mean metal concentrations occurred in the profile located within the meander bar; the concentrations were similar in the case of the other profiles, they were only slightly higher in profile T1. The greatest difference occurred for Cd whose mean content in profile T2 was five times higher than in the other profiles. In the case of the other metals, their mean content was usually about twice as high.

The minimum content levels of the particular metals were similar in the case of all profiles analysed. For cadmium, however, there still were from 20 to 28 times higher than the geochemical background. In the case of the other metals, these levels were from two to seven times higher. The maximum concentrations were the highest in profile T2. For cadmium, they were 10 times higher than in the other profiles. A similar pattern occurred in the case of Ni. The smallest differences between the profiles were found for Zn and Cr. The maximum-to-minimum ratios were as follows: a) Cd – 30, b) Cr – 5.5, c) Cu – 11, d) Ni – 16, e) Pb – 7, f) Zn – 9.2.

Standard deviations of metal content in profiles T1, T3 and T4 are small or medium (they reach maximum 30% of the mean content in profile T1 and T4). In profile T2, the deviations for all metals are medium (20–50% of the mean metal content) or high (over 50% of the mean metal content). Cr and Pb content in the particular profiles shows small variation except for profile T2 where its deviation from the mean is almost 50% for Cr and over 55% for Pb. Ni shows medium deviations in all profiles except T2 where the standard deviation is over 70%. Cd and Cu shows the biggest variation of content and deviation from the mean in a given profile. The greatest deviations for both metals occur in profile T2. In three profiles, Zn shows medium standard deviations (from 30 to over 46%), while in profile T1 the variation is small (16%).

The occurrence of significant correlations between heavy metal content in the samples was found (Table 7). These correlations were the strongest in profile T2, slightly weaker in profile T3, and the weakest in profiles T1 and T4. In profile T2, the correlation coefficient for nearly all metals was higher than 0.8; it was slightly lower (0.54–0.74) only in the case of the Ni–Cr and Cu–Cd correlation. Similarly high coefficients were found in profile T3, except for the correlation between Cr and the other elements (0.4–0.5). In profile T3, the strongest correlations (>0.9) occurred between the concentration of Ni and Cd, Ni and Pb, Cu and Zn, Pb and Cd. In the case of other pairs of metals, the correlation coefficients ranged from 0.5 to 0.6. Similar relationships occurred in profile T1: the highest correlation coefficients (>0.9) were found for Ni and Cu, Ni and Zn, Cu and Zn, Cu and Pb, Zn and Pb.

The correlations between the content of the metals studied and the concentrations of Mn and Fe were distinctly weaker. In the case of Mn, these correlations were negative. There was a moderate correlation between TOC and Cr, Cu and Cd, no correlation was found between heavy metal content and clay fraction content (from 15 to 25%), i.e. the factor that influences the binding of heavy metals in sediments (Zgłobicki *et al*. 2015).

TABLE 7. COEFFICIENTS OF CORRELATION BETWEEN CONCENTRATIONS OF SELECTED ELEMENTS (DATA FOR ALL SAMPLES)

	Cd	Cr	Cu	Ni	Pb	Zn
Cd	1	0.95	0.92	0.73	0.87	0.86
Cr		1	0.93	0.84	0.94	0.90
Cu			1	0.76	0.91	0.96
Ni				1	0.91	0.84
Pb					1	0.90
Zn						1

Distinct patterns occur in the vertical variation of metal content in the profiles under study (Fig. 2). In the case of profile T1, no distinct changes were found with increasing depth. In profile T2, concentrations increase considerably with increasing depth. A slight or moderate decrease of concentrations was found in profile T3, and a slight or moderate decrease in profile T4. In samples up to the depth of 20 cm, the highest concentrations of metals occur in profiles T1 and T2.

The indices used in the study can be divided into two groups. The first one consists of indices based on the comparison of the content levels with the geochemical background and reference elements. They are calculated separately for each element. The geoaccumulation index, enrichment factor and pollution index belong to this group. In the case of the second group of indices, the toxicity of metals is taken into account. Indices in this group can be determined for the specific elements (index of ecological risk factor) and can have a synthetic character taking into account the toxicity of sediment resulting from the elements it contains (potential ecological risk index). Cd is regarded as a particularly toxic element (T_{ri}=30) while Zn and Cr are the least toxic (T_{ri} 1 and 2 respectively) (Håkanson 1980). Due to the different structure of the indices, a certain variation occurred in the degree of profile pollution (Table 8):

a) Geoaccumulation index: a medium level of heavy metal pollution of the studied profiles was found in most cases. It was low for Ni, high for Cr, and high and extremely high for Cd (profile T2).

b) Enrichment factor: a medium or considerable degree of pollution usually occurs; it is very high for Cd, and minimal for Ni.

c) Pollution index: high pollution for most metals and profiles; Ni content indicates the lack of pollution with Ni.

d) Index of ecological risk factor: low for metals and profiles except for Cd where a very high risk was found as well as for Cu and Pb in profiles T1 and T2.

e) Potential ecological risk index: high for all profiles, very distinct increase of the index value in profile T2 approximately at the depth greater than 20 cm.

The results obtained are consistent with the data on heavy metal content in surface layers of channel sediments in the stretch of the Bystrzyca valley under study, published by Bojakowska and Sokołowska (1996). In the case of Cd and Cr, however, the maximum concentrations found are distinctly higher (3–4 times) while for Cu and Pb they are an order of magnitude higher than the content levels determined by Plak *et al*. (2006) for alluvial soils (Table 9). It should be emphasised that these results cannot be compared in a straightforward manner due to the different analytical methods used.

The geoaccumulation indexes obtained for the studied profiles remain at a similar level as the ones obtained for large rivers in Poland while they are lower than indices for rivers of heavily industrialised areas (Table 10). Only in the case of Cd, they are comparable with the data obtained for the Stoła river sediments (vicinity of Tarnowskie Góry). Thus, it can be concluded that a strong anthropogenic pollution with Cd occurs in the alluvia under study.

TABLE 8. AVERAGED INDEXES OF POLLUTION LEVEL IN STUDIED PROFILES

Index		T1	T2	T3	T4
Geoacumulation index (pollution level)	Cd	4.8	6.4	4.5	4.4
	Cr	3.4	4.0	3.3	3.1
	Cu	2.9	3.3	2.3	2.2
	Ni	0.0	1.0	-0.3	-0.5
	Pb	2.5	3.0	2.5	1.9
	Zn	2.4	2.8	1.4	1.8
Enrichment factor (pollution level)	Cd	28.0	127.9	21.0	29.1
	Cr	10.9	17.5	8.9	11.7
	Cu	7.2	10.9	4.6	6.0
	Ni	1.0	2.4	0.7	0.9
	Pb	5.5	8.9	5.1	5.2
	Zn	5.1	7.4	2.6	4.5
Pollution index (pollution level)	Cd	44.2	213.5	35.6	32.6
	Cr	15.8	27.5	15.0	13.2
	Cu	11.5	17.4	7.8	7.3
	Ni	1.6	3.8	1.2	1.1
	Pb	8.7	14.3	8.6	5.8
	Zn	8.2	11.7	4.5	5.4
Index of ecological risk	Cd	1325.4	6405.1	1069.5	979.2
	Cr	31.7	55.0	30.0	26.3
	Cu	57.7	87.2	38.3	36.4
	Ni	7.9	19.2	6.1	5.4
	Pb	43.7	71.5	43.0	29.3
	Zn	8.2	11.1	4.5	5.4
Potential ecological risk		1474	6649	1192	1082

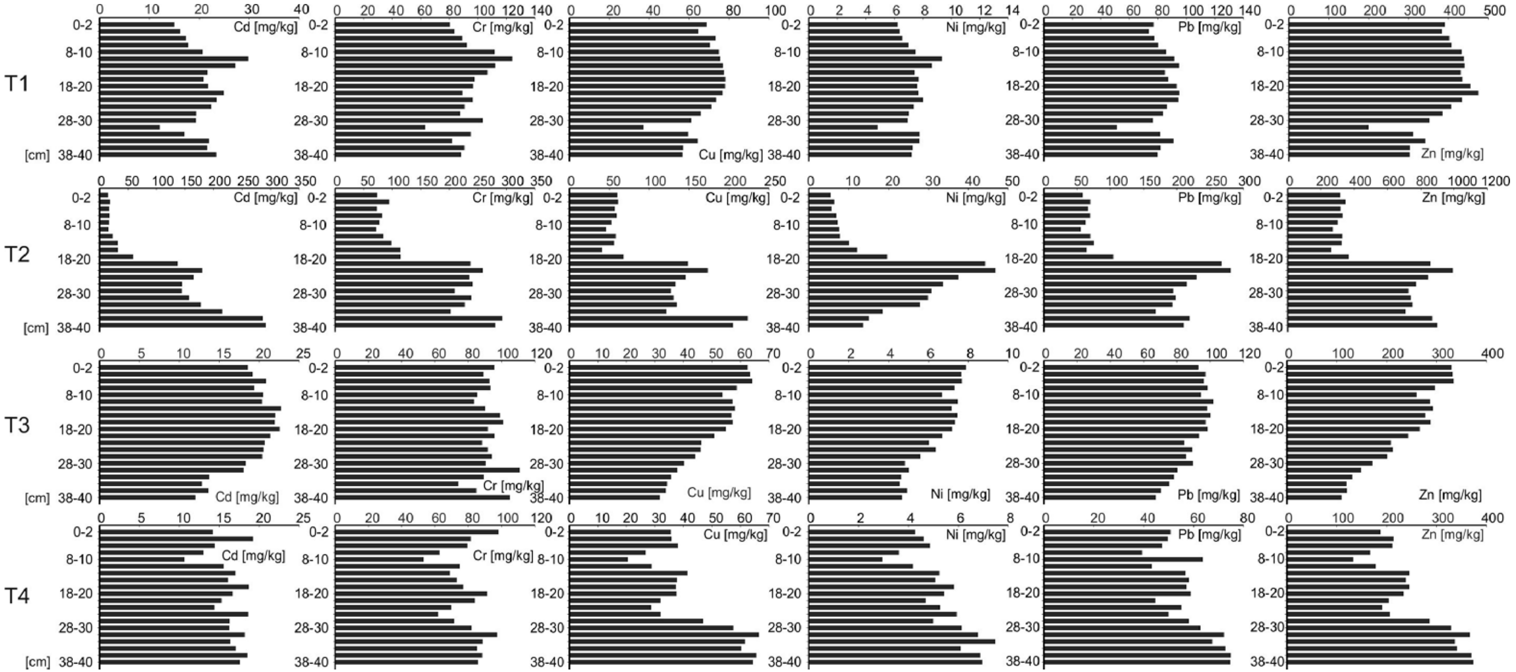

Fig. 2. Vertical variation of heavy metal concentration in the studied profiles

TABLE 9. HEAVY MENTAL CONTENT IN CHANNEL SEDIMENTS AND ALLUVIAL SOILS IN THE BYSTRZYCA RIVER VALLEY AT TURKA AND DOWNSTREAM FROM HAJDÓW

	Cd [mg/kg]	Cr [mg/kg]	Cu [mg/kg]	Ni [mg/kg]	Pb [mg/kg]	Zn [mg/kg]
Channel sediments[1]	20–34	29–39	59–79	41–71	62–79	357–488
Alluvial soils Bystrzyca[2]	–	–	0.44–5.73	0.03–3.66	0.04–14.04	–
Channel sediments, alluvia[3]	15–24	75–99	33–73	4.6–7.8	52–97	204–420

[1] – Bojakowska and Sokołowska (1996), [2] – Plak *et al*. (2006), [3] – this study (mean data for the 0–20 cm range)

TABLE 10. GEOACCUMULATION INDICES FOR FLUVIAL SEDIMENTS FOR THE SELECTED RIVERS IN POLAND

River	Cd	Cr	Cu	Ni	Pb	Zn
Wisła[1]	2	–	2	–	2	2
Odra[1]	2	–	3	–	3	3
Warta[1]	1	–	1	–	1	1
Przemsza[1]	3	–	2	–	4	3
Kaczawa[1]	1	–	3	–	3	2
Stoła[2]	5.2÷7.7	-1.3÷0.4	–	-1.6÷0.4	3.4÷7.3	3.6÷5.3
Bystrzyca[3]	3.4÷5.4	-1.5÷0.4	1.4÷2.6	-1.5÷0.4	0.6÷1.7	0.4÷1.7

[1] – Bojakowska and Sokołowska (1998), [2] – Rubin *et al*. (2011), [3] – this study

The vertical variation of heavy metal content in the profiles under study results from changes in the anthropogenic discharge into the Bystrzyca river and varied geomorphological location. The highest metal content levels occur in the area of present intensive accumulation of alluvial sediments (profile T2). Distinctly higher concentrations in the lower part of the profile (deeper than 20 cm) are related to the accumulation of polluted sediments in the period before the Hajdów wastewater treatment plant was put into operation, and to the larger discharge of pollutants from industrial sources (automotive factory, chemical and food processing plants). The fact that natural Cd concentrations are distinctly exceeded can be linked primarily with the operation of the Passenger Automobile Factory. The accumulation of this type of sediments ended in the late 1980s and early 1990s. The profiles located on the floodplain (T1 and T2) are characterised by moderate pollution levels resulting from the fact that metal discharge (during the overbank stage) does not occur every year. Slightly higher concentrations in profile T1 result from its location within the neck of the meander where the material carried by the water is accumulated. Profile T3 is located in the zone where the water returns to the river channel. The lowest metal concentrations were found in profile T4 located within a straight stretch of the river channel. On the one hand, it is a place where sediments accumulate; on the other, erosion occurs here during high flow stages, which results in the detachment of sediments containing heavy metals. Alternating layers contain slightly greater and slightly smaller amounts of metals, which is linked with the intensity of high flows of the Bystrzyca. Also in this case, deeper-lying (older) sediments contain slightly greater amounts of metals but their increase with increasing depth is not so abrupt and considerable as in profile T2. The studies conducted so far confirm the role of channel and overbank processes in the spatial variation of heavy metal concentration in alluvia (Ciszewski 1998). It is also emphasised that the maximum accumulation of contaminants of this type occurred in the past, in the 1960s, 1970s or 1980s depending on the location (Sokołowska, Szwarczewski 1998; Szwarczewski 2002).

The vertical variation of heavy metal content can be used for sediment dating and determining the sedimentation rate (Ciszewski 2001). The distinct decreases of pollution levels occurring in profiles T1 and T2 make it possible to assess the present sedimentation rate, assuming that their timing corresponds to the time when the Hajdów wastewater treatment plant was put into operation and the output of many industrial plants declined (late 1980s and early 1990s). This rate is about 0.4 cm/year in the case of the floodplain and 0.9 cm/year in the case of channel sediments.

CONCLUSIONS

1. In small areas in the bottoms of river valleys, the level of sediment pollution with heavy metals can vary significantly. The level of pollution is influenced by the geomorphological location of a given profile.
2. The vertical variation of heavy metal concentration in sediments, primarily in the active channel zone, is linked with the history of anthropogenic discharge of pollutants into the catchment.
3. Natural factors such as the grain size distribution, Mn and Fe content do not have a significant impact on heavy metal content in the alluvia of the Bystrzyca river.
4. Sediments in the Bystrzyca river bottom show varied levels of pollution with heavy metals. The pollution is the highest in the case of Cd (whose content exceeds the geochemical background 200 times in some locations) and occurs in all profiles. The pollution level for the other metals is usually moderate.
5. The specific indices show varied levels of pollution of sediments, from low to very high. However, in the case of the synthetic potential ecological risk index, which describes the total toxicity of a given sample, the pollution level of all profiles is high.

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