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**Akumulacja metali ciężkich w osadach dennych rzeki
Białej Przemszy jako wskaźnik ich rozprzestrzenienia drogą wodną
z górniczo-hutniczego ośrodka przemysłu cynku i ołowiu ***

**The accumulation of heavy metals in the bottom sediments
of the River Biała Przemsza as an indicator of their spreading
by water courses from the centre of the zinc and lead mining
and smelting industries**

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Abstract — The level of concentration of 12 microelements in the bottom sediments of the River Biała Przemsza and its tributaries (the watershed of the Upper Vistula) was investigated for the evaluation of its usefulness as an indicator of the pollution of the environment of head waters. It was found that the total content of microelements in the sediments chiefly depends on the degree of pollution of the water and does not show any distinct connection with the amount of organic and silt substances in the sediments. The content of microelements in the sediments is a good indicator of the spreading of mineral pollution by water courses. The degree of its usefulness partly depends on the properties of the aquatic environment. Zinc shows the greatest migration with the waste waters from the mining and smelting centre of the zinc and lead industries.

The pollution of rivers with heavy metals, especially with lead, zinc, and mercury is nowadays of wide interest. This arises from the ever more frequent occurrence of their high concentrations in the aquatic environment of the industrial complexes and from the possibility of their negative influences. The degree of contamination of aquatic environments with heavy metals cannot always be precisely evaluated on the basis of their content in the head waters, as a part of these elements is often distributed with the suspended matter and accumulated in the bottom sediments or on the flood terraces of river valleys (Hellmann, 1972, Pasternak et al. 1973). Thus in many cases the concentration of such metals in the sediments must be known, while the occurrence of microelements in the sediments of various water bodies is as yet considerably less known than that in the waters.

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The principal aim of the present investigations was to examine the possibility of using the concentration of microelements in the sediments as an indicator of the spread of mineral pollution from the mining and smelting works of the zinc and lead industries. A further purpose of these studies was to obtain data on the level of the content of microelements in the river sediments under the influence of this type of pollution, as it is known that in the regions where the ore deposits are exploited and various metals processed the quantitative level of individual microelements in the soils (Skawina 1967, Golub 1967, Paluch, Karweta 1970, Davies 1971, Kowaliński et al. 1972, Turski, Baran 1972, Burkit et al. 1972) and in the waters (Prokofiev, Sachbasova 1969, Złata, Koncz 1971, Pasternak 1973) may be found to be different.

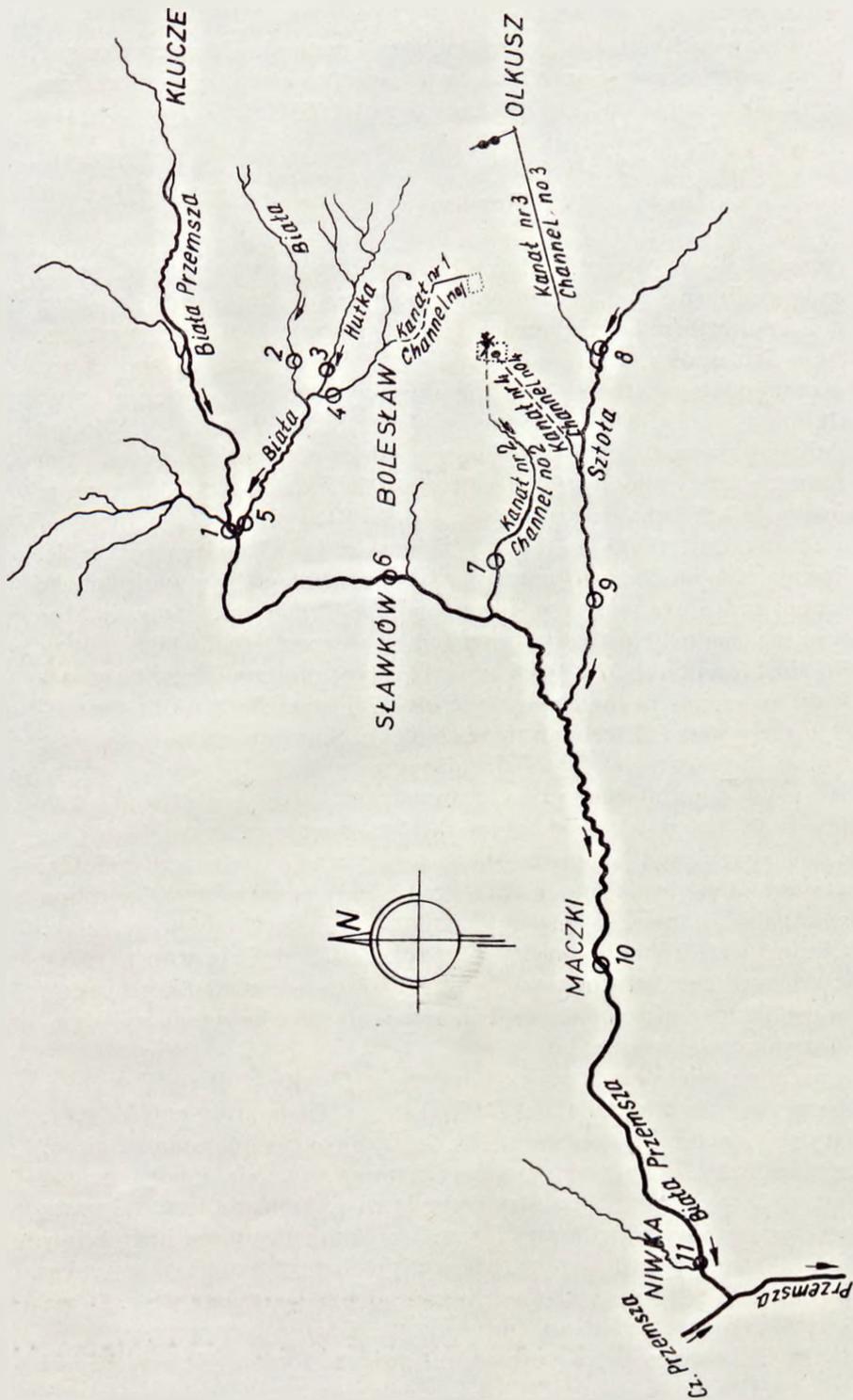
The territory and method of investigations

The River Biała Przemsza together with the Czarna Przemsza form the left tributary of the River Vistula — the River Przemsza. In the substratum of the upper part of the catchment basin of the Biała Przemsza the lime stones, marls, and orebearing dolomites dominate, and in the lower part the Carbon schist and sandstones. All these rocks are covered with glacial sands, on which soils of the granulometric composition of loose and weakly loamy sands have formed, hence the river has a sandy bottom in its whole course.

In 1972 the sediments for the investigation were sampled from the Biała Przemsza during normal and high water levels. They were collected at each station along a 50-metre sector. A sketch of the water network of the Biała Przemsza and the distribution of the sampling stations are presented in fig. 1.

Besides the waste water from the lead and zinc mining and metallurgic works, the investigated river receives a large load of organic pollution from the cellulose and paper plant in Klucze. The polluted waters from the mines are fed into the river through its two left tributaries, i. e. the streams Biała and Sztola. At present the flotation effluents, very rich in suspension, from the ore dressing plant are drained through channel No 1 (fig. 1) to the Biała, while the Sztola receives considerable amounts of water from the depth of the mine through two channels (Nos 3 and 4), these waters containing some amounts of very fine suspended matter originating from the pulverized rocky background of the exploited ore. The polluted waters from the smelting works are neutralized and fed to the Biała Przemsza directly through channel No 2, some domestic wastes being mixed with them. The general chemical composition of these waters and the content of microelements in them were presented in an earlier work (Pasternak 1973).

The granulometric composition of sediments was determined aeometrically using Casagrande-Prószyński's method, the reaction electrometrically, and



Ryc. 1. Schemat rzeki Białej Przemszy i rozmieszczenie punktów poboru prób osadów

Fig. 1. The scheme of the River Biała Przemsza and the distribution of the sampling stations of sediments

the content of organic matter according to Alten's method. The microelements in the air dry sediments were examined by the spectral emission method using the variant of one addition (Gliński, Grajpel 1965).

Results

The results of the determination of more important physico-chemical properties of sediments and of the total content of microelements are given in Table I in the form of the means from two terms of investigations.

As these data indicate, the sediments of the Biała Przemsza and of the natural sectors of its tributaries have the granulometric composition of loose or weakly loamy sands. The sediments of the artificial channels Nos 1 and 2 have different granulation. The dissimilarity does not result so much from the difference in the content of the colloidal silt fraction (< 0.002 mm) in the sediments of the channels (given in Table I) as from the number of granules of the dust fraction (0.11–0.02 mm) not included in the table. The sediments of both channels contain a large percentage of granules of the dust fraction and are among the group of dust formations. Moreover the sediment of the channel No 1 is characterized by an exceptionally high content of the silty dust fraction (0.02–0.006 mm). This type of granulometric composition of the sediments in the channels results from the character of the suspensions in the waters fed through these channels. Some quantitative increase in the size of the dust and also of the silty dust granules in relation to the sediments in the natural sectors of the investigated streams is also noted in the sediments of the polluted sector of the Sztola stream.

Thus the investigated sediments show a very small content of colloidal silty fractions and are hence not characterized by any greater mineral sorption complex, capable of more intensive assimilation of zinc and other metals. As may be inferred from the results of Nachsina and Feldmann (1971), in the investigated channels the capacity of the silt to assimilate the sediments of heavy metals may even be lowered in a measure by the presence of considerable amounts of calcium.

In connection with the organic pollution of the River Biała Przemsza, its sediments have an acid reaction (Table I) in the whole investigated sector. A similar acid reaction is also shown by the sediments of the upper, clean, sector of the Biała stream, whose water contains relatively small amounts of calcium and magnesium, the valley being in its greater part wet and peaty and almost wholly covered with pine forest. The sediments of the Biała below the outflow of channel No 1 and the sediments in channel No 2 have a weakly acid reaction. The sediments of the Hutka and Sztola streams (Fig. 1) are characterized by a neutral or alkaline reaction. This is connected with the fact that the first stream is fed with some amounts of water flowing out of the adits of old zinc ore mines, drilled in calcium-dolomite rocks, while the

Table I. Średnie ogólne zawartości mikroelementów, ilastych frakcji, organicznej materii i odczyn osadów dennych rzeki Białej Przemszy i jej dopływów

Table I. Mean total contents of trace elements, clay fraction, organic matter, and reaction of the bottom sediments of the River Biała Przemsza and its tributaries

Rzeka (P) River Potok Stream	Nr punktu No of point (Miejscowość) Locality	PPM										Ilaste frakcje Clay fraction		Organic matter %	pH		
		Cu	Zn	Pb	Wn	Ni	Co	Cr	Mo	V	Ba	Sr	D			<0.02 mm	<0.002 mm
Biała Przemsza (P)	1 (Kuzniańska Nowa)	6.6	10	7	44	2.0	1.0	25.0	5.6	1.0	31	6.2	36	3	3	2.40	5.1
Biała (P)	2 (Laski)	4.8	12	6	58	1.0	1.0	1.3	4.6	1.0	14	3.0	32	4	4	1.30	5.2
Hutka (P)	3 (Laski)	9.6	54	124	76	1.0	1.0	2.4	36.0	1.0	96	14.8	36	5	3	0.70	7.0
Kanał nr 1 Channel no 1	4 (Laski)	128.0	10000	420	750	26.0	24.0	58.0	20.0	58.0	300	58.0	86	47	8	7.40	7.3
Biała (P) ujście - mouth	5 (Kuzniańska Nowa)	15.6	800	124	140	17.2	21.0	8.0	20.0	20.0	38	12.4	50	5	4	3.40	6.6
Biała Przemsza (P)	6 (Starków)	7.0	100	50	89	6.0	4.8	20.0	9.8	7.4	67	21.4	40	4	2	2.50	5.3
Kanał nr 2 Channel no 2	7 (Bukorne-St. Wiel.)	56.0	20000	500	720	29.0	17.2	21.0	14.0	120.0	450	36.0	26	8	1	17.80	6.7
Stola (P) powyżej ujścia kanałów 3 i 4 above the mouth of channels no 3 and 4	8 (Bukorne)	4.0	200	100	80	20.0	9.0	5.0	38.4	16.2	26	13.8	30	2	2	0.33	7.3
Stola (P) poniżej ujścia kanałów 3 i 4 below the mouth of channels no 3 and 4	9 (Bukorne-Las)	5.8	2500	1200	850	50.0	12.8	50.0	46.8	57.8	126	51.8	54	11	2	1.17	7.4
Biała Przemsza (P)	10 (Wierzb)	26.5	1500	150	120	30.0	5.6	100.0	23.2	13.1	48	14.3	42	3	2	2.03	6.4
Biała Przemsza (P)	11 (Bisza)	12.0	880	29	205	3.5	-	80.0	7.6	3.0	108.0	11.0	40	4	2	1.70	5.6

second flows in its whole length through the territory of carbonate rocks and receives a load of waters from the depth of working mines.

In the sandy sediments of the River Biała Przemsza fairly small amounts of organic matter are contained (1.7—2.5 per cent, Table I). This results on the one hand from the fact that a great part of the organic pollution in its water occurs in soluble form, and on the other that the fine suspension of cellulose fibres sedimentates with difficulty from the rapidly flowing water. In the sediments of the clean sectors of the tributaries of the Biała Przemsza the content of organic matter is very low. Similarly as in the clean streams, small amounts of organic matter occur in the sediments of the polluted (with mineral substances) sector of the Sztoła stream, while the sediments of channels Nos 1 and 2 contain considerable amounts of organic matter. In the first channel this is probably connected with the residues of organic reagents used in the process of ore flotation (xanthogenate, resin oils) on the surface of the suspension granules sedimentating on the bottom, and in the second with the admixture of domestic wastes to its waters.

The quantitative level of microelements in the sediments of the Biała Przemsza above the point of inflow (through the lower sector of the Biała stream) of the polluted waters from the centre of zinc and lead ore dressing is, with the exception of chromium, only slightly higher than in the sandy sediments of clean rivers recently investigated by the author (unpublished data). It seems to indicate, similarly as the results of water analyses (Pasternak 1973), that the influence of wastes from a cellulose plant on the concentration of heavy metals in the environment of their receivers is relatively small.

In the clean upper sector of the Biała stream the concentration of microelements in the sediments is in the range regarded as the natural level. Not until the inflow of the strongly polluted waters of channel No 1 does it considerably increase. Moreover, it must be mentioned here that in the sediments of the relatively clean Hutka stream the content of some microelements (Pb, Zn, Mo, Ba, Sr) was also higher than the average (Table I). It seems that this phenomenon is brought about by feeding the stream with certain amounts of waters flowing from the above-mentioned old adits and by the occurrence of ore-bearing rocks in the substratum of the whole catchment basin.

In the sediments of the final sector of channel No 1, i. e. at a distance of about 5 kilometres from the source of pollution, the concentration of the majority of the investigated trace elements is very high. Particularly high values of the content of zinc, lead, copper, manganese, and barium were found (Table 1). The concentration of lead and zinc in the sediments of this sector of the discussed channel is higher than in the sediments of that sector of the Czarna Przemsza and Vistula which is most polluted with metals (Pasternak et al. 1973). This would suggest that the concentrated flotation effluents are rather slowly purified from these metals in the first kilometres

of their course. The content of large amounts of copper in the sediments of channel No 1 supports the earlier observation of the author (Pasternak 1973) that the small content of this element in the water of the channel, in spite of the fact that it is added in the flotation processes, results from its intense physico-chemical precipitation into the sediments. This precipitation is chiefly brought about by the higher oxygen saturation of the water and by its alkaline reaction. Thus any precise data on the amount of copper compounds carried out with the flotation effluents may only be obtained on the basis of the determinations of heavy metals in the bottom sediments. The specific granulometric composition of sediments in the channels (many granules of dusty fraction) seems to suggest that a certain percentage of lead and zinc may pass from the water into the sediment in the form of fine dusty granules of the ore remaining in the flotation effluents.

In consequence of the inflow of the polluted water of channel No 1 a considerably increased level of the majority of the investigated micro-constituents is still noted in the sediments of the Biała stream in its final sector (i. e. some kilometres below the mouth of the channel). Yet the increased content of zinc and lead in the sediments of the outflow sector of the Biała is not so high as might be inferred from the degree of pollution of the channel and the voluminal relation of the polluted to clean waters. This phenomenon probably results from the high capacity of self-purification of the Biała, which flows in a bed with wide marshy overflow-arms thickly overgrown with various flowering plants favourably influencing according to many authors (Rodziller, Zotov 1971) the processes of water purification, especially from suspended solids. It may be concluded from what has been said above that the influence of pollution from this stream on the concentration of microelements in the River Biała Przemsza is as yet insignificant. This is supported by the results of the determination of microelements in the sediments of the Biała Przemsza a little below the mouth of this stream (Sławków), where the amount of these elements in the sediments is only insignificantly higher than in the upper sector of the river not influenced by the zinc wastes. In general, in this part of the Biała Przemsza only the content of zinc and lead in the sediments increases in a slightly higher degree, the content of chromium even decreasing. This is chiefly connected with the great dilution of the water of the Biała stream, polluted with heavy metals, with the water of the Biała Przemsza.

Channel No 2, flowing into the Biała Przemsza in its further course, is lined with the sediments of still higher content of some microelements than that found in channel No 1. Above all, in the sediments of channel No 2 very large amounts of zinc are found. The concentration of zinc in the sediments of this channel is higher than that in the sludge, usually rich in heavy metals, of the settling tanks of industrial towns (Hellmann, Grifatong 1972). It almost reaches the values of its average concentration in the silty sediments of a fiord in western Norway, strongly polluted by the

metallurgic industry, and investigated by Skei et al (1972). In the sediments of channel No 2, besides zinc, considerable amounts of lead, vanadium, and barium are also recorded. The content of copper is lower by half than that in the sediments of channel No 1. Such a high accumulation of zinc and lead in the sediments of channel No 2 indicates that a large load of these metals gets into the River Biała Przemsza both with its neutralized waters and with the transported suspension.

In the clean sector of the next tributary of the Biała Przemsza, i. e. the Sztola stream, the majority of microelements occur in the sediments in amounts similar to those usually found in loose sands. Exceptions are the contents of Zn, Pb, Mo, and V which are many times higher than in the average clean aquatic environment. The increased level of zinc and lead in the sediments of this stream undoubtedly results from the natural influence of ore-bearing carbonate rocks occurring in the substratum of the catchment basin. It is worthy of note here that the natural influence of zinc and lead deposits is more distinctly observable in the composition of microelements in the sediments than in the waters (Pasternak 1973). The increase in zinc and lead concentration in the surface waters in the territories where ore deposits of these metals occur (Wales) was also observed by Abdullah, Royle (1972). The investigations of Görlich F., Görlich Z. (1956) suggest that the spread of zinc and lead through porous carbonate rocks does not occur in consequence of diffusion but by way of their salts being washed by the migrating solutions. According to Zlot a and Koncz (1971), the degree of dissolving zinc and lead which are contained in the natural rocks or in the mining and smelting wastes collected in the vicinity of industrial centres of non-ferrous metals by the rain water is influenced by the concentration of carbon and sulphur dioxides in their environment.

In the sediments of the Sztola stream below the inflow of waters from the depth of the mine the concentration of trace elements, with the exception of copper, rapidly increases, reaching a fairly high level, the greatest increase being noted in the concentration of zinc, lead, and manganese. However, in the composition of microelements in the sediments of this sector of the Sztola the high content of lead is most striking, as it is more than twice as high as in the sediments of channels Nos 1 and 2. In the sediments of the polluted sector of the Sztola the exceptionally high concentration of lead and at the same time lower content of zinc (in relation to channels Nos 1 and 2) results in the decreased relation Zn/Pb. Skei et al. (1972) found that in the sediments of the investigated polluted fiord the relation Zn/Pb resembles that in flowing waters. It seems that in the discussed stream this relation may be regarded as an indicator of the kind of water pollution as far as the initial sector of its tributary only is concerned, as in the waters from the depth of the mine a considerable part of these metals spreads in the form of fine suspension, which may undergo certain qualitative differentiation down the stream in consequence of, for example, poorer solubility of lead

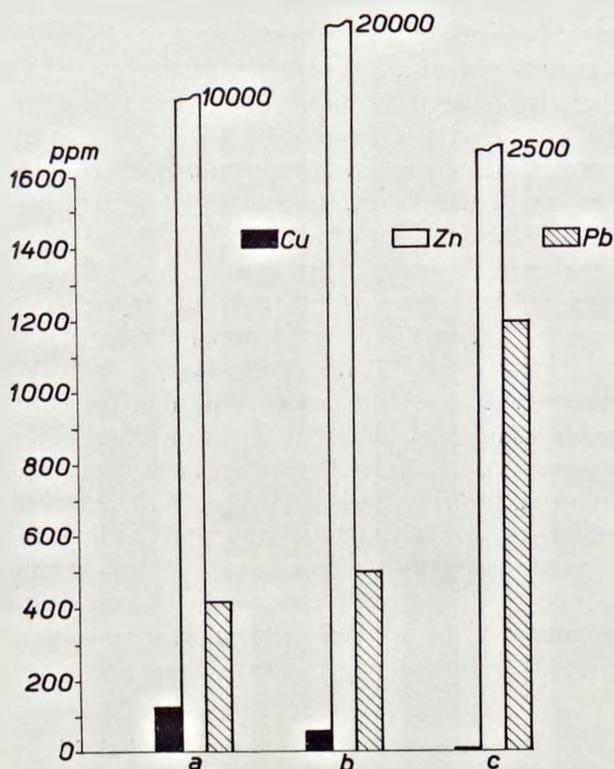
in the water and of the differences in specific weight of the granules of its minerals. This supposition is supported by a faster decrease in lead than in zinc down the stream (Pasternak 1973). In the Sztola stream a certain amount of heavy metals also passes from the water to the sediments, together with the remnants of organisms living in it. Butterworth et al. (quoted according to Burkitt et al. 1972) found that some organisms living in this kind of polluted water are characterized by a high content of heavy metals.

A considerably increased content of the majority of the investigated microelements still occurs in the sediments several kilometres below the mouth of channel No 2 and the Sztola stream (Maczki). Above all, in the sediments of this part of the river the quantitative level of zinc considerably increased. The concentration of lead increased in a lesser degree as compared with the sediments of the less metal polluted upper sector of the river. In the sediments of this part of the Biała Przemsza the fairly high content of chromium and copper is also remarkable, indicating the inflow of pollution from sources other than those discussed in this sector of the river.

In the final sector of the Biała Przemsza the content of microelements, with the exception of copper, undergoes a marked decrease, though it is still on a level many times higher than in the sandy sediments of other clean rivers investigated by the author. The amount of zinc in the sediments of this sector of the Biała Przemsza even exceeds the concentration recorded in the loamy sediments of the not very clean southern part of Lake Michigan (Shimp et al. 1971) and of fairly clean dam reservoirs (Pasternak 1972).

The occurrence of increased amounts of zinc and lead in the sediments of the lower course of the river indicates that the spread of these metals by waterways is fairly high in this type of industrial complex. Especially zinc migrates in the aquatic environment over great distances. The amount and quality of microelements in the sediments of channels and streams, particularly in the immediate vicinity of the sources of pollution, depends above all on the load and kind of pollution of the flowing water. The comparison of the concentrations of zinc and lead in the sediments of channels Nos 1 and 2 and of the lower sector of the Sztola stream (which carry various kinds of polluted waters) indicates (Fig. 2) that the largest load of zinc and lead is carried to the neighbouring surface waters with the wastes from the smelting works, a slightly smaller one with the flotation effluents from the ore dressing plants, and the smallest with the waters from the depth of the mine. In general, in the sediments of the initial sector of the channels and polluted streams the content of zinc and lead is much greater than in the land soils occurring in the immediate vicinity of the source of emission of dusts containing these metals (Skawina 1967, Paluch, Karweta 1970, Burkitt et al. 1972, Turski, Baran 1972). However, no dependence between the concentration of microelements and the content of organic and also of silt substances was found in the sediments of the investigated environment, though it usually

occurs in the sediments of relatively clean lakes (Shimp et al. 1972) and dam reservoirs (Pasternak 1972).



Ryc. 2. Porównanie średnich zawartości cynku, ołowiu i miedzi w osadach: a — kanału nr 1 odprowadzającego wody poflotacyjne; b — kanału nr 2 odprowadzającego wody hutnicze; c — potoku Sztola poniżej zrzutu wód dołowych kopalni

Fig. 2. The comparison of the content of zinc, lead, and copper in the sediments of: a — channel No 1 carrying the flotation effluents; b — the channel No 2 carrying the smelting wastes; c — the Sztola stream below the inflow of waters from the depth of the mine

Conclusions

1. The total content of microelements in the sediments under flowing waters may be regarded as an indicator of their mineral pollution, as between the quality and quantity of microelements in the sediments and in the water a distinct relation exists.

2. Nevertheless, the degree of usefulness of this indicator partly depends on the properties of the aquatic environment. A more distinct characterization of the influences of mineral pollutions or of natural orebearing rocks in the catchment basin on the environment may be obtained on the basis of the content of microelements in the sediments than on that of their content in

the waters, in the case of highly oxidated and alkaline environments which favour rapid physico-chemical precipitation of certain metals from the water. However, in the streams and rivers which carry waters of almost neutral reaction and rich in dissolved organic substance, in which the majority of metals remain longer (because of better conditions of solubility and formation of chelate compounds), it is at the same time necessary to know the concentration of microelements in the water.

3. The level of zinc and lead content in the sediments of the receivers of polluted waters from the discussed branch of industry is very high.

4. The spread of certain metals in the longitudinal profile of streams and rivers is fairly considerable. Among the investigated microelements zinc is observed to migrate by water the most. The concentration of zinc and lead in the sediments is approximately the function of the distance from the source of pollution.

5. The highest percentage of microelements is transported by the waste waters from mining plants in the form of fine suspension.

STRESZCZENIE

Badano poziom stężenia 12 mikroelementów w osadach dennych rzeki Białej Przemszy i jej dopływów w aspekcie możliwości jego wykorzystania jako wskaźnika mineralnego zanieczyszczenia wód płynących oraz rozprzestrzeniania się metali ciężkich drogą wodną z górniczo-hutniczego ośrodka przemysłu cynku i ołowiu. Określono także niektóre podstawowe fizykochemiczne właściwości tych osadów. Stężenie mikroelementów w wodzie, na tle jej ogólnego składu chemicznego, przedstawiono w innej publikacji. Zawartość mikroskładników w osadach oznaczono metodą spektralnej analizy emisyjnej.

Między innymi stwierdzono, że całkowita ilość mikroelementów w osadach (tabela I) zależy głównie od stopnia zanieczyszczenia wody, nie wykazuje natomiast wyraźnego związku z ilością substancji organicznych i ilastych. Poziom zawartości niektórych metali ciężkich w osadach bezpośrednich odbiorników zanieczyszczonych wód rozpatrywanej gałęzi przemysłu jest bardzo wysoki. W dalszych odbiornikach, na skutek oczyszczania się wody, jest funkcją odległości od źródła zanieczyszczenia. Stężenie mikroelementów w osadach stanowi dobry wskaźnik rozprzestrzeniania się drogą wodną mineralnych zanieczyszczeń przemysłowych lub wpływu naturalnych skał kruszczo-nośnych występujących w zlewni rzeki. Stopień jego wskaźnikowej przydatności zależy w części od właściwości rozpatrywanego środowiska wodnego. W mocno natlenionych i alkalicznych środowiskach lepszy obraz zanieczyszczenia czy wpływu wspomnianych skał uzyskuje się na podstawie danych o mikroelementach w osadach niż w wodach. Natomiast w ciekach posiadających wodę o niższym pH i sporej ilości organicznych substancji, mogących zwiększać utrzymywanie się w niej metali ciężkich (tworzenie się chelatowych połączeń), konieczna jest znajomość mikroelementów w wodzie. Z rozpatrywanych obiektów przemysłowych rozprzestrzenianie się niektórych mikroelementów jest dość znaczne. Największy ładunek cynku i ołowiu jest wynoszony do okolicznych cieków z wodami odpadowymi huty, nieco mniejszy ze ściekami pollotacyjnym zakładu wzbogacania rud, a najmniejszy z wodami dołowymi kopalni (ryc. 2). Spośród badanych mikroelementów najbardziej migruje drogą wodną cynk.

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