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The past and prognosis of mining cessation impact on river sediment pollution

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Abstract

Purpose Remediation of mine sites is often aimed at reduction of river pollution after cessation of mining activity. However, the effects of these works overlap natural attenuation processes and their efficiency cannot be recognized well without detailed studies of the fluvial environment. The aim of the study is to predict changes of channel sediment pollution in rivers affected by mine sites after their rehabilitation, planned after mining cessation.

Materials and methods This study utilizes data on sediment pollution in three rivers polluted by lead-zinc mining, sampled over a time span of over 20 years in southern Poland. The former and the present pollution levels have been compared between the Biała Przemsza and Sztoła Rivers continuously influenced by mine waters and the Chechło River where mining has ceased. The observed changes in sediment pollution in the three catchments have been interpreted in terms of the impact of mining cessation on sediment pollution and the observed rate of the pollution changes in the past, reconstructed also from reservoir sediments. Results and discussion In the drainage basins of the Chechło River, a mine has been closed and the rapid drop in metal concentrations in channel, and reservoir sediments was observed within a few years. In the other two drainage basins where lead-zinc mines still operate, the metal concentrations remained at the same level. Based on the similar catchment and pollution characteristics, a drop in metal concentration in these two rivers is expected soon after ore exhaustion. Nonetheless, considering the large contamination of soils and overbank sediments in the three drainage basins due to a long mining history, the drop will be followed by a very long period when their concentrations are raised above a permissible level.

Conclusions The study has shown that the natural fluvial attenuation processes can decrease heavy metal concentrations in channels cut in fine-grained alluvia within a few years after mine closure by one order of magnitude but the rate of the decrease depends on the frequency and magnitude of floods which will take place after mining cessation. Efforts made to mitigate pollution at mine sites may not be as efficient as fluvial processes to rapidly decrease channel sediment pollution below permissible levels.

Keywords Anthropogenic impacts · Metals · Natural attenuation · Remediation of contaminated sediment · River

1 Introduction

Mining and reworking of metal ores can be a source of river pollution with heavy metals over distances exceeding hundreds of kilometers. Metals can be discharged directly into a river by waters pumped out from a mine or through artificial drainage of ore beds by a system of adits. Heavy metals are

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AGH-University of Sciences and Technology, Mickiewicza St. 30, 30-059 Krakow, Poland also mobilized from wastes and tailings exposed to atmospheric conditions. They reach a river system due to weathering and erosion of wastes or tailing dam failures (Byrne et al. 2012). Metals are mobilized from mine sites both during mine operation and also long after mining cessation, affecting aquatic biota and sediments (Resongles et al. 2014). This is the reason why remedial actions are undertaken in order to mitigate the mining impact on downstream river systems (von der Heyden and New 2004). These actions are very complex, expensive, and can last for many years (Mebane et al. 2015). Unfortunately, despite the efforts in mine site remediation, numerous river systems remain contaminated to levels far exceeding permissible values (Bird 2016). Effects of remediation works in these river systems are hampered by

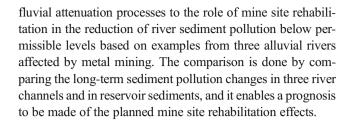


pollution of floodplain and channel sediments, which are secondary pollution sources, mobilized in high flow conditions (Žák et al. 2009).

Whereas most historical metal mines have been abandoned leaving them without post-closure reclamation, at present, large mines are closed down according to schemes prepared to minimize environmental risk. The Olkusz-Trzebinia Zn-Pb ore exploitation area is an example of a region in southern Poland where ores have come to an end, and a scheme of the closure is planned. The ore body has been extracted here at least since the twelfth century, but at the beginning of the twenty-first century only four mines had been active. One of these mines, located in Trzebinia in the southern part of the region, was already closed down in 2009. The mine discharged up to 0.5 m³ s⁻¹ of waters containing sediments polluted mainly with zinc, lead, and cadmium into the Chechło River and the river sediments were characterized in the 1990s by their very high pollution level (Ciszewski 1997). A similar pollution degree with these metals at that time was also observed in the other two rivers in the northern part of the region: Biała Przemsza and Sztoła (Fig. 1). These rivers have been affected by mine waters directed from the other three mines of the Olkusz-Trzebinia region (Ciszewski 1998). The aim of the study is to compare the efficiency of natural



Fig. 1 Location of research area and sampling sites: 1. Olkusz-Trzebinia lead-zinc ore mining area, 2. ore processing plants, 3. closed mines, 4. active mines, and 5. sampling sites of bottom and overbank sediment profiles



2 Materials and methods

Exploited ore bodies in the region occur in karstified carbonates abundant in water, and mining activity is hindered by extremely high inflows of waters (Motyka and Czop 2010). The enormous amount of mine water, comprising about 60% of mean low water discharges of the Biała Przemsza River and up to 100% of the Sztoła River discharge, is pumped out of the mines in the north of the region (Ciszewski 1998; Morman and Czop 2012). Besides pollution with metals, river waters are characterized by their high turbidity, sulphates, and carbonates as well as high Mg and Ca content (Aleksander-Kwaterczak and Ciszewski 2016). The level of pollution in the Chechło River waters, in the southern part of the region, has never been regularly tested. Besides being exposed to mine waters of a similar characteristic to that in the northern part, the river was also affected by a meat producing plant, a rubber plant and some smaller plants, and municipal effluents from Chrzanów and Trzebinia (Ciszewski 1997).

The bottom sediments of the three river channels were sampled in the years 1992-1993 in cross-sections situated along the whole river's length downstream from metal mines; in each cross-section, sediments were sampled from the following sites: pools, channel bars, near-bank locations, and banks above water table (Ciszewski 1998). The Biała Przemsza was sampled again after a large flood in 1998 (Ciszewski 2001). The current text presents data for Cd and Zn concentrations in fraction < 1 mm of samples in the two types of points of the previous channel cross-section. One type of the points represents in-channel sediments in locations near the river banks where mainly fine, silty sediments, transported as suspended matter, accumulate. The other type of the points represents channel bars where mainly sandy sediments, transported as bed load, accumulate. In the year 2016, channel sediments of the three rivers were again sampled from the same two types of points at locations situated within dozens of meters from the original ones and compared to the unpublished data from 1992 to 1993 for Chechło and Sztoła and from 1998 for Biała Przemsza. Moreover, bottom sediment samples were taken in two 60–78-cm-long profiles from two water reservoirs in subsidence basins (called further upper and lower) in the lower reach of the Chechło River valley. The sediment profiles were sampled with Eijkelkamp multisampler and sectioned in 3-6-cm intervals (the lower



one) and every 7–14 cm (the upper one). The third profile, 90cm long, was sampled from outcrop of overbank sediments of the Chechło River in 10-cm intervals. The sediment profiles of subsidence basins were located about 1 km apart, whereas overbank sediments were sampled about 500 m downstream from the lower profile, outside the lower subsidence basin. The samples were homogenized and representative subsamples were taken for metal analyses. All the channel sediment samples were wet sieved through a 1-mm sieve and extracted in Teflon bombs in 1:1 HNO3 and then filtered through a 0.45-µm filter according to the procedure described earlier (Ciszewski 1998, 2001). Samples of sediment profiles were wet sieved through a 0.063-mm sieve and digested with HNO₃ according to EPA 3051. All Cd and Zn concentrations in channel and profile sediments were determined using atomic absorption spectrometry. A standard quality control procedure was applied, including blind duplicates and reference materials with an error less than 10%. The ¹³⁷Cs activity in the sediment profile was determined via the emission peak at

661 keV by high-resolution gamma spectrometry (HPGe, Canberra Gx4020).

3 Results and discussion

Investigations of the Chechło River sediments show their very high pollution levels both in 1992 and at present (Fig. 2). The highest concentrations of Cd and Zn occur within a roughly 2-km-long reach downstream from the mine with the extreme values for Cd reaching 700 mg kg⁻¹ and 10% for Zn. Downstream, the concentrations decrease by one order of magnitude in near-bank sediments, whereas in active sediments, they drop by three orders of magnitude on average. The minimum Cd values reach about 1 mg kg⁻¹ and Zn concentrations are somewhat higher than 300 mg kg⁻¹. There are important differences between changes observed over the river course in near-bank and active channel sediments. Generally, in near-bank sediments sampled on both occasions,

Chechło River

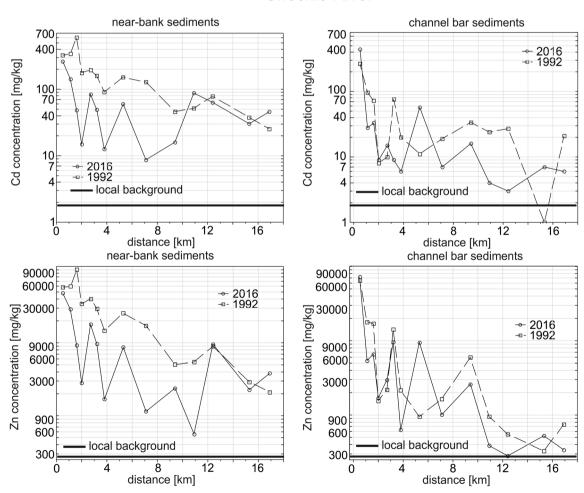


Fig. 2 Zn and Cd concentrations in-channel sediments of the Chechło River in 1992 and in 2016, local background values after Ciszewski (1997)



the downstream decrease of metal content is more proportional than in active channel sediments where concentrations are more variable.

The difference between metal concentrations in 1992 and in 2016 is well seen in Fig. 2. Over that period, the concentrations in samples taken at river banks changed over the entire river reach from a more uniform in 1992 toward a more variable in 2016. These concentrations decreased the most in the upper 10-km-long river reach, whereas in the lowest river course, about 4 km in length, the metal content changed much less. This difference is related to the channelization of the 10km-long reach where the uniform width and bank lining favor more effective winnowing of fine sediments than the lowest reach where the river is meandering. Active lateral erosion of overbank sandy alluvia of this meandering river reach also contribute to the pollution of channel sediments because overbank sediments which outcrop in river banks are characterized by the high content of Zn and Cd (Fig. 3). The highest sediment pollution, exceeding several hundred times the local background values, evidently is related to over 50 years of intensive ore mining, and considering a stratigraphy dominating along this reach (as visually inspected during the course of investigations) characterized by black strata intercalated with bright sandy laminae, these polluted sediments are an important secondary pollution source. The process of metal remobilization is common in areas of former metal mining, where large amounts of pollutants may enter rivers through erosion of finegrained overbank sediment sequences (Žák et al. 2009).

The respective changes are not observed for channel bars of the Chechło River where concentrations changed irregularly in both sampling periods. The large variability of metal concentrations in channel bars is a characteristic feature of channel bed sediments where processes of sediment reworking and mixing of mine-related polluted sediments with Holocene substrate took place (Ciszewski 1997). This local variability, induced by variable intensity of fluvial processes and the bed material size, makes these sediments less suitable for monitoring (Ciszewski 1998).

However, the decrease of sediment metal pollution in the investigated period is evident; it is not clear when the change took place. Because of a lack of sediment quality monitoring on the Chechło River, the estimation could rely solely on

reconstruction of pollution levels by an analysis of sediment sequences. The profile for the analysis was retrieved from the lower reservoir in a reach of the river valley which underwent the subsidence as a result of underground hard coal mining. The subsidence had been a slow process which resulted in a sinking of the whole valley floor over about a 500-m-long reach. It took place at the break of the 1980s and 1990s. The average lowering of the ground was about 1 m; maximum values did not exceed 1.6-1.7 m. The surface of the former floodplain was found at a depth of 50 cm in the profile of bottom sediments (Fig. 4). It comprised more compacted sandy-silty sediments than overlying strata, composed mostly of black muds, rich in organics. The former surface was dated by the high activity of ¹³⁷Cs from the Chernobyl accident in 1986. The floodplain sinking initiated the accumulation of water-borne, strongly polluted sediments at the peak of metal mining activity in the 1990s and at the beginning of the twenty-first century. High and uniform Zn concentrations in the prevailing part of the profile suggest that the river pollution levels were generally stable over that time. The large drop in Zn content observed at a depth of 12 cm can be correlated to the decrease of river pollution levels due to cessation of mine water discharge in 2010. It seems that the drop in sediment pollution levels of the Chechło River, shown in Fig. 2, can be assigned to the same date.

A distribution of Cd content in the profile does not resemble that for Zn, what could be expected considering presence of Cd almost exclusively in excavated sphalerite, and does not fully agree with the local mining history (Fig. 3). Particularly, there is the high Cd concentration peak just over the less permeable floodplain surface and decline of Cd content followed by a distinct peak and decrease toward the profile top. The Cd distribution suggests some post-depositional changes because Cd competes with Zn for adsorption sites and at higher Zn concentrations in solutions, as expected in the pore waters of the profile, Cd adsorption decreases (Wilkins et al. 1995). Lower binding Cd than Zn intensity favors its mobility (Han et al. 2003). Mobilized Cd in backswamp sediment is preferentially adsorbed at profile depths enriched with clay minerals and Fe-hydroxides (Cappuyns and Swennen 2004). In the absence of these minerals, the less permeable sediment strata can constitute a barrier

Fig. 3 Zn and Cd concentrations in overbank sediment profile of the Chechło River

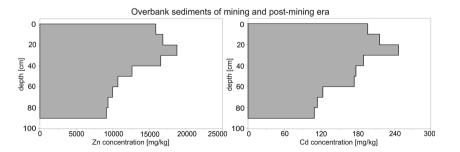
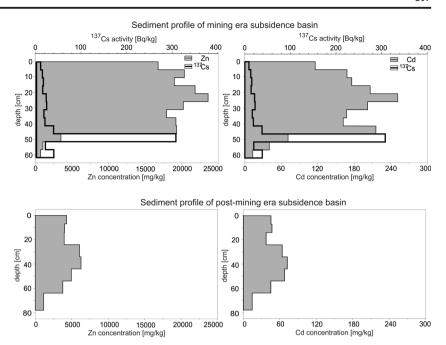




Fig. 4 Zn and Cd concentrations and ¹³⁷Cs activity in profiles of bottom sediments from mining era (lower) and post-mining era (upper) subsidence basins in the Chechło River valley



for waters percolating through the profile as Cd is an element easily translocated down-profile in wetland soils even at neutral or weakly acidic conditions (Kalbitz and Wenrich 1998).

The other profile representing bottom sediments was retrieved from the deltaic section of the upper reservoir in a basin which started to subside around 2007. At present, the inundated surface of this basin is equal to about 5 ha and the average depth is ca. 1.5–2 m. The river flows through the central part of the reservoir. Delta sediments are composed of fine-grained sands in the main channel zone and of mud with high organic matter content just outside this zone. The sediment accretion is rapid in the whole, proximal ca. 150–200 m long zone of the reservoir which is filled with about 1-m-thick sediments. The upper reservoir profile (Fig. 4) comprises these muddy sediments, resembling sediments from the lower basin but of much lower pollution level with Zn and Cd. The differences of maximal concentrations in both profiles are about fourfold and they decline toward the profiles' top. The lowest strata probably represent the former floodplain surface, much less polluted than overlying sediments. The age of the upper reservoir indicates that, contrary to lower profile mainly accumulated during peak river pollution, these sediments mainly represent the post-exploitation period but also the period when production strongly declined after initializing the mine closure. However, we are not able to date back to the beginning of bottom sediment accumulation in the upper profile, but the drop of aquatic sediment pollution within not more than a few years after the mine was closed is evident. The change in sediment pollution after the mine was closed is well reflected because in the upper reservoir a significant part of the sediments conveyed down the stream is trapped even at low and moderate discharges, whereas in the profile of lower reservoir sediment accumulation took place only during less common flood discharges. The much less pollution of the younger than of the older profile sediments agree with the rapid change of the channel Chechło sediments pollution expressed in median values (Table 1). The drop in pollution at channel banks is about threefold and is larger in case of the 75 than 25 percentile. It is related to the drop of the highest values particularly upstream of the sinking area and to the much smaller decrease of both metals over the lower reach of the river. The drop of analogous values in channel bars is less evident in the case of Zn medians and well seen for Cd.

The changes of sediment pollution with Zn and Cd in the Chechło River were particularly rapid in comparison to that of the Sztoła River (Fig. 5). Both rivers are comparable in terms of their average discharge and length; however, the Sztoła River is characterized by the much smaller variability of annual water stages which do not exceed several cm per year (Ciszewski 1998). The regime of this river is controlled by generally constant discharge of mine waters which predominates over waters drained naturally from the catchment (Morman and Czop 2012). The artificial regime favors fine sediment accumulation at river banks. The near-bank sediments are trapped under uniform flow conditions by plant roots growing on unlined banks. The entrapment of fine sediments transported in suspension results in insignificant changes of Zn and Cd concentrations over a distance of 10 km. For this reason, the near-bank sediments represent the ideal location for pollution monitoring. The changes in metal concentrations observed in near-bank sediments of the Sztoła River between both sampling periods are not affected by channel morphology and discharge variability. Because there are virtually no changes for over the last two decades in metal pollution (Fig. 5), we may conclude that the river has been continuously polluted by the mine. This regularity is



Table 1 Median, 25 and 75 percentile of Zn and Cd concentrations (mg/kg) in the Chechło, Sztoła, and Biała Przemsza River sediments and average rank values for Kruskall–Wallis test at *p* < 0,01 (explanations in the text)

River	Location	Year	Zn				Cd			
			Median	25%	75%	Average rank	Me	25%	75%	Average rank
Chechło	Banks	1992	21,450	5450	40,300	40.9	141	51	196	42.9
		2016	6238	2283	9644	29.7	48	16	84	30.7
	Bars	1992	1900	950	14,300	22.7	22	11	72	24.3
		2016	2158	524	6601	20.6	9	6	28	15.9
Sztoła	Banks	1993	19,475	16,862	20,862	23.2	134	124	147	26.1
		2016	20,365	19,562	21,054	25.7	124	120	131	22.8
	Bars	1993	2448	1443	6737	9.5	7	2	44	8.9
		2016	1270	651	7416	7.5	12	1	44	8.1
Biała Przemsza	Banks	1998	4125	3325	13,975	21.9	28	22	54	20.8
		2016	13,056	9164	15,244	27.8	105	62	123	28.8
	Bars	1998	1106	595	4633	11.2	5	3	33	11.9
		2016	1148	392	1968	13.1	9	3	15	12.5

confirmed by very similar medians, 25 and 75 percentile of concentration values at the channel banks for both investigation periods (Table 1). The analogous values for channel bars change less regularly and are higher for Cd and lower for Zn in 2016 than in 1993.

Similarly to the Sztoła River, the Biała Przemsza River has also been permanently polluted with mine waters since the second half of the twentieth century. Maximum Zn and Cd concentrations downstream the 4-km-long culvert, shown in Fig. 6, are similar to those of the Sztoła River. They decrease both in near-bank and in active channel sediments over the roughly 15-km-long reach. Lower, downstream the mouth of the Sztoła River to the Biała Przemsza River, metal concentrations rise in near-bank sediments by one order of magnitude and stay high over the next 15-km-long natural reach through to the confluence of the other small but polluted rivers. The

Fig. 5 Zn and Cd concentrations in-channel sediments of the Sztoła River in 1993 and in 2016

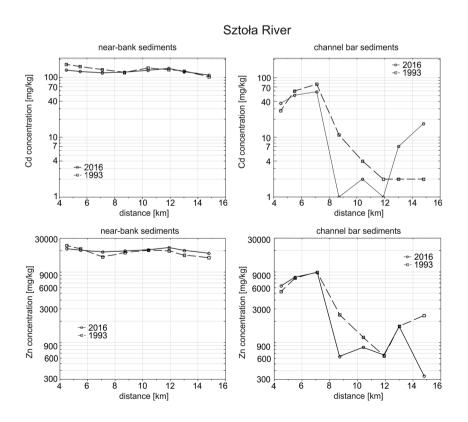
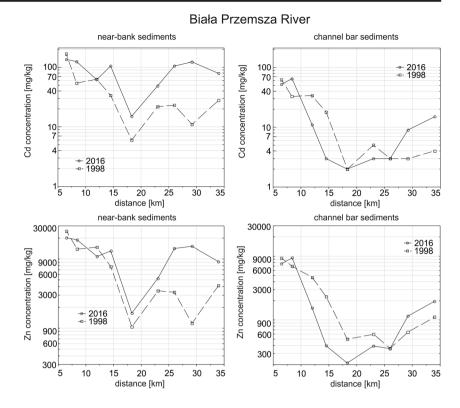




Fig. 6 Zn and Cd concentrations in-channel sediments of the Biała Przemsza River in 1998 and in 2016



concentrations in channel bars over this lower reach also. however, irregularly increase. Considering the conclusion that channel bar sediments may not reflect river pollution tendency well, we refer here to Zn and Cd concentrations in near-bank sediments. Concentrations of these sediments in 2016 rose in almost all locations since the previous sampling in 1998. The increase is the largest in the lower reach and lowest closer to the mine. This regularity is also well seen when comparing median, 25 and 75 percentile values (Table 1). Medians of both metals' content are around three times higher than 18 years earlier, but the higher concentrations are more or less of the same order as suggested by the similarity of the 75 percentile. In contrary to the near-bank sediments, median values and respective percentiles for channel bars are of the same order as before. It suggests persistent transport of fine and polluted sediments which are trapped mainly by plant roots at the channel banks. The rise of metal content in the period following 1998, shown in Fig. 6, continued the tendency observed after the flood in 1997. This flood, which was the largest on record, resulted in a decrease of metal content by as much as two times in the upper investigated reach and showed an increase in some points downstream in comparison to the values observed in 1993 (Ciszewski 2001). Average rank values obtained by the Kruskall–Wallis test at p < 0.01 (Table 1) exhibited the same regularity: for the Chechło River, the drop of both metal contents over the investigated period is remarkable, particularly at the banks, whereas pollution of the Stoła River is the same and the metal content in

the Biała Przemsza sediments increased relatively moderately.

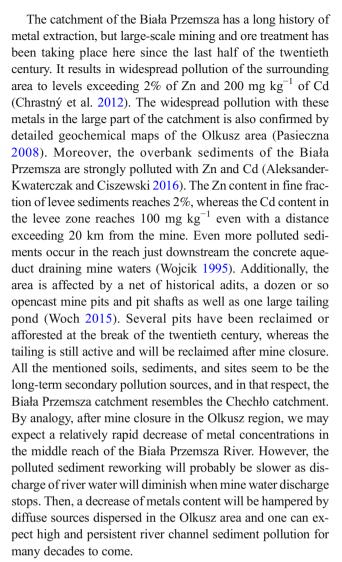
A similar tendency in metal pollution for over the last two decades is also observed for the Przemsza River which receives waters from the Biała Przemsza (Strzeboska et al. 2017). The average pollution levels of that river did not change significantly because the Przemsza is dominated by the influence of waters from the lead-zinc mine discharged via the Biała Przemsza River. The mine is considered to be the largest source of Cd and Zn in Poland, and as long as the mine is active, the original pollution level can be restored within several years over the distance of 10-40 km (Ciszewski 2001). The limited role of floods in reduction of the sediment pollution level was also recognized to be a characteristic for the other perennial rivers with an active pollution source as well as for rivers with strongly polluted overbank deposits which are mobilized during floods and heavy rains (Ciszewski and Grygar 2016). This situation changes as the pollution source becomes inactive, direct discharge of effluents ceases, and natural fluvial processes lead to winnowing of the polluted sediment from the channel. In time, the role of secondary pollution sources is bound to increase.

Soon, after the mine closure in Trzebinia in the Chechło catchment in 2009, the reclamation and of flotation mine tailing pond was undertaken and finished within the following 3 years. Works were not fully successful because this tailing is located on permeable sandy deposits, and waters leaching tailings are easily drained by the two nearby ditches. At present, they remain a source of sulphates, Zn, Pb, and Cd in



amounts exceeding permissible levels in receiving surface waters (Klojzy-Kaczmarczyk and Mazurek 2011). The amount of drained waters polluted with heavy metals, which equals a few dm³ s⁻¹ at average weather conditions, is expected to rise markedly during heavy rains, contributing significantly to river pollution. Moreover, top soil horizons of the Chechło River catchment are strongly polluted with these metals. Based on the data of detailed geochemical maps (1:25000), soils polluted with Zn and Cd in excess of local geochemical background cover the prevailing part of the catchment (Pasieczna 2008). The most polluted soils are at sites that have been mined here for centuries in numerous, but small shafts, dispersed in the middle and northern part of the area. The maximum pollution levels of Zn at these sites exceed 5000 mg kg⁻¹, while Cd is 50 mg kg⁻¹. In addition, pollution of the Chechło overbank sediments, represented by the profile in Fig. 3, is characteristic also for long-term polluted rivers in mining areas (Byrne et al. 2012) and we may expect that sandy floodplain sediments are an additional river pollution source. The polluted soils and sediments constitute a diffuse pollution source activated particularly during longer rainy periods and, indeed, the doubled metal concentrations were observed in river waters after heavy rains in the Chechło River catchment (Michailova et al. (2018) accepted).

It seems that as long as the drop in metal concentrations in the Chechło River in response to mine closure was rapid, future changes will be much slower because metals will be mobilized from the catchment area. Such a scenario confirms the reconstruction of pollution level changes in the other mine-affected catchments where the pulse in waste production was reflected in the peak of dominant metal concentration. In such catchments, where subsequent closure of a mine resulted in a reduced fluvial sediment supply, the drop of the dominant metal content is followed by a decades-long period of the metal content much above a background level (Pirrie et al. 1997). In the drainage area of large-scale mining, the changes observed in response to mining reduction and improvement in ore extraction and treatment technologies are less striking (van Geen et al. 1997). Nevertheless, the drop in metal concentrations in response to mine closure initializes the aquatic system recovery. The rapidity of metals winnowing, i.e., a decrease of metal concentrations particularly in a river reach immediately downstream the mine due to a decrease of pollution plume, dilution with extra sediments, or overbank accumulation, controls the rate of species redistribution and abundance because only a few higher organisms are able to tolerate the extreme metal concentrations.



The virtually artificial discharge of the Sztoła River means that after mining cessation, the river will no longer be supplied with mine water. At present, the river flows over the groundwater depression cone surrounding the mine. The local water table occurs more than 10 m below the river bed and the river is losing about 10% of its discharge over its whole reach. The escape of river waters is limited by colmation of the bed by fine, polluted sediments transported by mine waters (Morman and Czop 2012). In the case of total cessation of mine water discharge, the channel would be almost empty for even 40 years before natural recharge would recover (Motyka and Czop 2010). In this scenario, the Sztoła River channel would remain strongly polluted for much longer than the Biała Przemsza River as the natural discharge of the Sztoła River is by at least one order of magnitude smaller than the artificial one.



In the case of all three rivers, we should expect persistent river sediment pollution despite some efforts undertaken toward land rehabilitation. Mine site rehabilitation is common good practice even if the effects do not occur quickly and harmful elements are episodically dispersed, especially during flood flows (Bird 2016). In many cases, reclaiming of the whole mine-affected area is unreliable and proper measures focused on hot points can significantly diminish pollution load transferred to a river as recorded in some sediment profiles (Schindler and Kamber 2013). It is also known that many efforts and expenses can be lost and, because of the fluvial sediment pollution, works may increase response time to success by only a little (Moore and Langner 2012). It seems that in catchments under consideration, the extent of the polluted area precludes efficient rehabilitation for reasonable costs. Instead, mitigation of river sediment pollution by mine tailing reclamation supported by natural fluvial attenuation processes can be achieved.

4 Conclusions

The study has shown that in the strongly polluted river channels, the drop of heavy metal concentrations after mine closure can be rapid. It can take only a few years to reduce the pollution in some river reaches even by one order of magnitude, but the rate of the decrease depends on the frequency and magnitude of floods which will take place after mining cessation. Natural attenuation processes are particularly effective in rivers reworking fine-grained alluvia in the relatively short, dozen, or so year-long period after mining cessation.

The drop in metals content in channel sediments will be followed by a much longer period with their concentrations raised above permissible values. The length of that period is controlled by the extent of waste, soil, and sediment pollution in a drainage basin. In strongly polluted catchments, efforts made to mitigate pollution at mine sites cannot be efficient enough to rapidly decrease channel sediment pollution below permissible levels.

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