

Open cast mines as river sediment and pollutant sinks. The example Mulde Reservoir (East Germany)

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Abstract

Since 1975, the Mulde River has flown through the abandoned open-cast lignite mining pits at Muldenstein near Bitterfeld (Germany) forming a lake named Mulde Reservoir. Ever since, the Mulde Reservoir has built up a sediment and pollutant deposit protecting the downstream water bodies and sediment quality in the lower Elbe stretch extending to the port of Hamburg area and the North Sea. Therefore, the Mulde Reservoir is an example for beneficial effects of riverine flow-through of pit lakes.

Core drillings have revealed that the sediments currently deposited in the Mulde Reservoir still have to be regarded as highly polluted as to their contents of As, Cd, Cr, Hg, Ni, Pb, Zn, according to the classification of the Pollutant Sediment Management Concept (PSMC) of Elbe River. Arsenic and the mentioned metals originate from former metal mining and processing in the upper part of the catchment of the Mulde River. However, a decrease in pollutant inflow since 1990 has been documented, correlating with the long-term changes observed in the upper course of the Mulde River. As far as organic pollutants are concerned, the lake sediments do not show any excessive concentrations, apart from a few exceptions.

From a geochemical and hydraulic point of view, there is presently no need to remove the contaminated sediments from the Mulde Reservoir. Main activities in sediment management should focus on further improving stabilization and efficiency of the Mulde Reservoir sedimentation zone and its transregionally vital ecosystem services. Given the present retention capacity of the Mulde Reservoir, it will continue to serve as an important sediment trap of supra-regional significance for the downstream water courses for at least another 770 years.

Key words: pit lake, Mulde Reservoir, sediment deposit, sediment management, sediment quality

Introduction

Open cast lignite mining released a lot of pit lakes in Germany (Schultze et al. 2013). Further lakes will form in future when currently operated mines are abandoned and remediated. Like natural lakes and also reservoirs (Wetzel 2001; Knoll et al. 2013; Frings et al. 2014), pit lakes are basically sinks for many kinds of matter in the landscape and in river systems. They act as big sediment traps. Biogeochemical processes inside the lake may produce additional sediment, e.g. via sedimentation of dead plankton or precipitation of calcite due to high photosynthetic activity in the lake (Wetzel 2001). In the Island Copper Mine pit lake (Vancouver Island, Canada), this is used to remove and deposit metals from acid rock drainage as technology to protect the environment (Wen et al. 2015).

Regions with long lasting metal mining activities often experienced contamination of river water, river sediments, floodplain soils and even estuaries with metals (Macklin et al. 1997; Krüger et al. 2005; Olias et al. 2006; Foulds et al. 2014), in extreme dimensions in case of breaches of tailings ponds (e.g. Hudson-Edwards 2016). The legacy of mining and related smelting and industrial activity often lasts long in aquatic sediments and floodplain soils (e.g. Coulthard et al. 2003; Förstner 2004; Resongles et al. 2014). Managing such sediments is complicated due to resuspension by floods and since dredging (e.g. in harbors) causes resuspension and high effort for treatment and safe deposition of the dredged material (Detzel et al. 1998; Förstner 2004).

The so called Mulde Reservoir (Germany) is basically a pit lake which is used as a reservoir. The Mulde River is flowing through it. Much of the inflowing suspended matter load is deposited in the Mulde Reservoir. We present the results of sediment investigations and the cleaning of the river water caused by sedimentation of the suspended matter. The focus is on arsenic and metals.

Study site

The Mulde Reservoir (fig. 1) is one of the pit lakes resulting from lignite mining in the northern part of the Central German lignite mining district (Schultze et al. 2010; Eissmann and Junge 2015). As indicated by the pit lakes shown in panel A of fig. 1, Mulde Reservoir is embedded in a region which was heavily impacted by lignite mining. Mulde Reservoir originated from the mine Muldenstein which was operated from 1955 to 1975. During this time, 126×10^6 t of lignite were produced and 439×10^6 t of overburden were excavated (Liehmann et al. 1998). When mining ceased, the remaining mining void was filled with water from Mulde River (initially $3\text{--}10$ m³/s, beginning at April 30, 1975; Liehmann et al. 1998; Böhme et al. 1994). Eventually the complete Mulde River was diverted into the new lake at March 4, 1976. In this way, the no longer used natural stretch of Mulde River and the accompanying floodplains were included into mine Goitsche which was operated a few kilometers to the west of mine Muldenstein since 1949 and which ended up in the two pit lakes immediately west of Mulde Reservoir in fig. 1 (Liehmann et al. 1998; Eissmann and Junge 2015).

In order to use the new lake as a reservoir, a dam was constructed at the outflow. It allows for a maximum water level of 82 m above sea level (a.s.l.) in case of high flow in Mulde River. The water level can be controlled between the minimum water level of 76.1 m a.s.l. and the normal goal of water level management at 79.25 m a.s.l. (Böhme et al. 1994). At the latter level, the Mulde Reservoir has a surface area of about 6.1 km² and a maximal depth of 32 m (Junge 2015). The maximum storage volume is 135.5×10^6 m³ containing an exceptional storage volume for flood protection of 17.5×10^6 m³ and a normally managed storage volume of 18.0×10^6 m³ (Böhme et al. 1994). In 2009, a fish pass was added to the outflow dam to allow for re-colonization of the upper stretches of Mulde River and its tributaries by salmon (Wouters 2010).

Panel C of fig. 1 shows a bathymetric map of Mulde Reservoir. The lake basin consist of two sub-basins, Main Basin and Friedersdorf Basin which are connected by a channel-like part of the former mine void. There is a ridge-like structure crossing the Main Basin from south west to north east (fig. 1, panel C). It is the remnant of a paleo-beach-dune-system from the time when the lignite forming peat was deposited about 20 million years ago (Eissmann 2002).

In the upper part of the catchment, Mulde River has two branches, Freiburger Mulde and Zwickauer Mulde. Downstream the confluence of both branches, the full name is Vereinigte Mulde (fig.1A). However, we use only Mulde River for simplification.

The catchment size of Mulde Reservoir is 6,709 km² and the long term average annual inflow is $2,097 \times 10^6$ m³/a (Böhme et al. 1994). The highest elevation of the catchment is 1,243 m a.s.l. (Peak Klinovec, Czech Republic). The nearest gauge upstream the Mulde Reservoir with a long term hydrological record is located in Bad Dübén, about 15 km upstream (fig. 1, panel A). For the period 1961 to 2012, the average low flow was 15.8 m³/s, the average flow was 64.7 m³/s and the average high flow was 487 m³/s (all data for hydrological years; LHW 2015). The highest daily average flow of the period 1961-2012 was 2,200 m³ at August 14, 2002 (LHW 2015). A further exceptional flood occurred in June 2013 with a maximum daily average of 1,720 m³/s (Junge 2015).

Active mining in the upper part of the catchment area of Mulde River began in 1168 and basically ending in the 1990s (Hösel et al. 1997; Greif 2015). It was mainly metal mining with some hard coal mining around Zwickau. Panel A of fig. 1 shows the major centers of the mining in the Ore Mountains where the headwaters of Mulde River are located. However, there were also smaller mining activities between the marked centers. Mining in the Ore Mountains was almost exclusively underground. It started with silver and went on with tin and a second boom of silver mining (beginning in the 13th century). In the 18th century, also bismuth, cobalt and nickel became relevant while in the 19th century lead, zinc, fluorite and barite were in the focus of the mining activities. Hard coal and in particular uranium were the main targets of mining in the 20th century (Hösel et al. 1997). All the time, mining

was accompanied by smelting and in the last two centuries also by diverse metal using industries (Greif 2015).

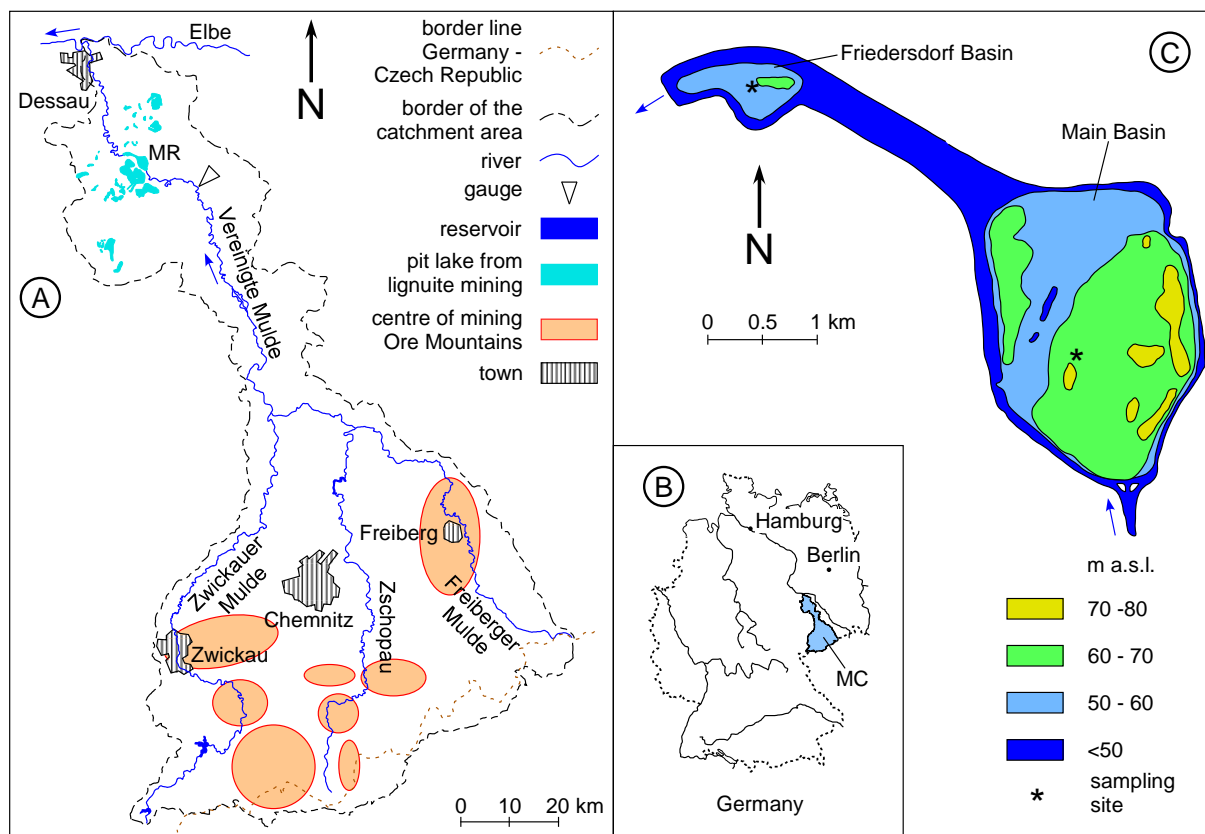


Figure 1 Location of the study site. Panel A: Catchment area of Mulde River including Mulde Reservoir (MR). Panel B: Location of the catchment area of Mulde River (MC) in Germany. Panel C: Bathymetric map of Mulde Reservoir

Methods

Sediment cores were extracted by gravity coring (UWITEC, Mondsee, Austria) from the Main Basin and from the Friedersdorf Basin (fig. 1, panel C) in April 2002, September 2002, July 2012 and July 2013. Samples were taken from the cores according to the visible layers.

Samples were prepared according to Junge et al. (2004) and Klemm et al. (2005), i.e. only the fraction <20 µm was analyzed. Analyses were performed by the laboratories of the group Pollutant Dynamics in Catchments of the Saxon Academy of Science (Leipzig, Germany) and of UFZ Department Analytics (Leipzig, Germany) for samples from 2002 and by Labor Eurofin (Freiberg, Germany) for samples from 2012 and 2013 according to standard methods (DIN EN ISO 17294-2, DIN EN 1483, DIN ISO 10382, DIN EN 11308, DIN 38414 S24). Organic pollutants were analyzed only from cores taken in 2012 and 2013, i.e. only in the uppermost about 30 cm of sediment. For more details see Junge et al. (2004), Klemm et al. (2005) and Junge (2013).

Results and discussion

Concentrations of contaminants and their evaluation

Collected sediments showed clear layering (fig. 2). Light colored layers were separated by dark colored layers rich in organic material. The light colored layers were identified as originating from floods and the dark colored layer as representing periods between floods. Together with the detection of the Tschernobyl peak by ¹³⁷Cs activity, reliable dating of the layers was possible (Junge et al. 2004). Based on the found concentrations and knowledge of changes in the catchment area regarding treatment of waste water, handling of wastes in mining and industry and the general development of industrial activities in eastern Germany after German reunification in 1990, four different periods of

sediment deposition can be distinguished in Mulde Reservoir: the period of highest contaminations from the time before 1990, a first period of decreasing contamination from 1990 to 2002, the deposits of the exceptional flood in August 2002 and a second period of decreasing contamination after 2002. Arsenic and metals concentrations are presented in tab. 1 according to these periods. Fig. 2 shows the results for a core taken in the Friedersdorf Basin in July 2013, including the fresh deposits of the exceptional flood in June 2013.

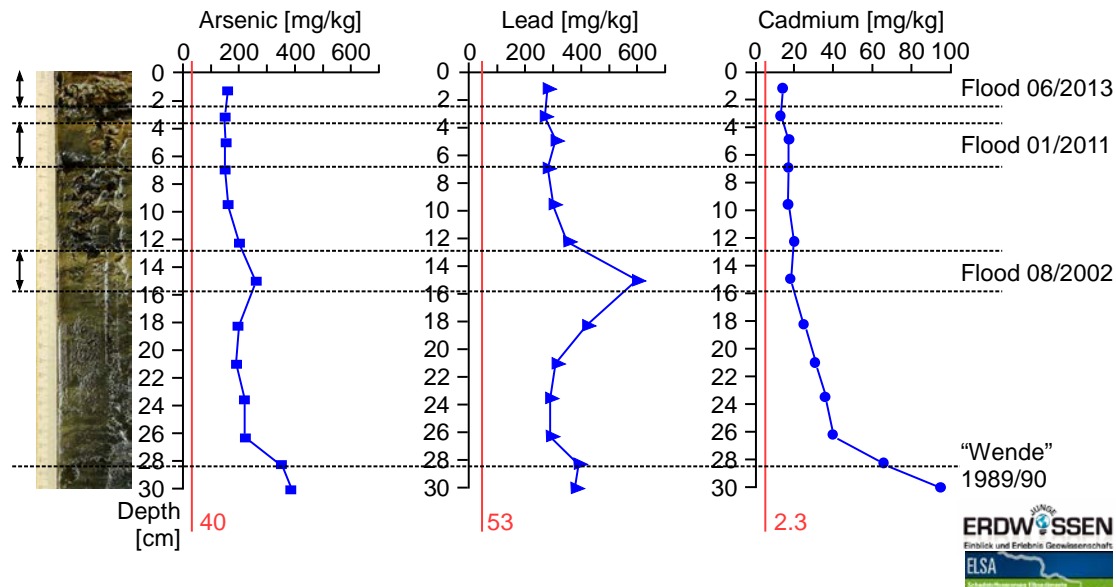


Figure 2 Sediment characteristics of a core taken in Friedersdorf Basin in July 2013. From left to right: Photograph of the sediment core, concentrations of arsenic, lead and cadmium in mg/kg versus depth in the sediment (in cm). Vertical red lines and red numbers indicate the upper thresholds of the PSMC in mg/kg (ICPE 2014). “Wende” is the common German term for the crash of the former German Democratic Republic and the German reunification 1989/90.

Three relatively thick light colored layers are visible in the photograph (fig. 2). They result from the three flood events in August 2002 (peak flow at gauge Bad Dübener 2200 m³/s), in January 2011 (peak flow at gauge Bad Dübener 730 m³/s) and in June 2013 (peak flow at gauge Bad Dübener 1720 m³/s). They represent the ranks 1, 2 and 6 in the ranking of daily average peak flows of floods in the data series 1961 to 2013 (LHW 2012). Concentrations of arsenic, lead and cadmium show a general decreasing trend with the highest decrease rate at the beginning of the 1990s, linked to the stop of mining and many industrial activities in the catchment. Later on, remediation of former mining and industrial sites and implementation of adequate waste water treatment became the most relevant reasons for improvement of the sediment quality (e.g. Greif 2015). The flood event of 2002 caused a considerable temporal increase in the concentration of lead which was much less pronounced for arsenic and cadmium and for the other two flood events. The background is the considerable erosion of mining and smelting wastes by the flood in 2002 which were deposited immediately at the banks of the Mulde River near Freiberg (Klemm et al. 2005).

In order to assess sediment quality and the extent of sediment contamination as well as to develop measures for improvement of sediment quality and prioritize such measures in the framework of a polluted sediment management concept (PSMC) according to the EU Water Framework Directive (EU 2000), the International Commission for the Protection of the Elbe River (ICPE) developed thresholds for a set of 29 contaminants which were identified to be most relevant for sediments in the catchment of Elbe River (ICPE 2014). These thresholds are included into tab. 1 for evaluation of the measured concentrations. While concentrations below the lower threshold indicate that good sediment quality can be reached without problems, exceedance of the upper threshold indicates the need for a source-related assessment of the risk (ICPE). All analyzed metals except chromium exceeded the upper threshold (tab. 1). Sources of metals and of arsenic are known and diverse measures were already undertaken or underway to reduce the contamination of the suspended matter in Mulde River and, thus, the concentrations of contaminants in the sediments deposited in Mulde Reservoir (Paul et al.

2013; Greif 2015). However, the work is not finished yet and widespread diffuse contaminations of soil exist around historical smelters or in aquatic sediments in the tributaries of Mulde River, which may be mobilized during floods and can hardly be fully controlled in future (Förstner 2004).

Table 1 Concentrations of metals and arsenic in the sediment of Mulde Reservoir (averages of all analyses available for the different deposition periods) and thresholds of PSMC. Depth ranges mean depth below sediment surface. MB – Main Basin, FB – Friedersdorf Basin, data in parentheses - range from minimum to maximum; n – number of available analyses.

	As mg/kg	Cd mg/kg	Cr mg/kg	Cu mg/kg	Hg mg/kg	Ni mg/kg	Pb mg/kg	Zn mg/kg
Lower threshold of PSMC	7.9	0.22	26	14	0.15	3	25	200
Upper threshold of PSMC	40	2.3	640	160	0.47	3	53	800
Period 2002-2012	188	21	77	140	0.8	78	292	1593
Depth ranges	(130-300)	(16-27)	(62-87)	(110-170)	(0.46-1.1)	(67-100)	(210-370)	(1200-2060)
MB: 0-50 cm	n=32	n=32	n=32	n=32	n=32	n=32	n=32	n=32
FB: 0-12 cm								
Flood 2002	230	24	88	175	1.0	82	549	1685
Depth ranges	(220-240)	(15-32)	(82-93)	(160-187)	(1.0-1.0)	(78-90)	(313-866)	(1240-2103)
MB: 50-55 cm	n=3	n=8	n=3	n=8	n=3	n=8	n=8	n=8
FB: 12-15 cm								
Period 1990-2002	286	45	122	198	0.82	99	309	2044
Depth ranges	(170-440)	(17-136)	(74-220)	(73-400)	(0.57-1.4)	(59-160)	(88-477)	(527-3450)
MB: 55-110 cm	n=14	n=59	n=14	n=59	n=14	n=59	n=59	n=59
FB: 15-30 cm								
Period 1975-1990	471	104	237	372	1.5	136	386	2847
Depth ranges	(260-620)	(38-230)	(150-380)	(159-600)	(0.9-2.6)	(89-220)	(243-590)	(1595-5000)
MB: 110-200 cm	n=13	n=47	n=13	n=47	n=13	n=47	n=47	n=47
FB: 30-50 cm								

Organic pollutants were investigated in the sediment of Mulde Reservoir first time in 2012 and 2013. Concentrations were highest in the old sediments originating from the period before 1990. The majority of concentrations were well below the upper threshold of the PSMC (ICPE 2014), in particular in surface layers of the sediment. Only DDT (1,1,1-trichloro-2,2-bis(p-chlorophenyl)ethane), some of its metabolites and fluoranthene considerably exceeded the PSMC thresholds(ICPE 2014), slightly even in the surface sediment. DDT and its metabolites certainly have two sources: widespread remnants of DDT use in the past and known contaminations resulting from a production site of DDT and other chlorinated organics in Delitzsch. The Lober-Leine-Kanal, a creek draining the area around Delitzsch, enters the Mulde River only about two kilometers upstream the inflow of the Mulde Reservoir.

Retention of contaminants in Mulde Reservoir

Zerling et al. (2001) summarized the results of investigations of the metal and arsenic content of suspended matter entering and leaving the Mulde Reservoir in the period 1992 to 1997 (tab. 2). The most important finding was that the deposition of suspended matter in the Mulde Reservoir considerably contributes to the limitation of the contamination of Elbe River, its sediments and flood plains downstream the mouth of Mulde River and also the North Sea (tab. 2). This is even more important as the Mulde River is one of the main contributors of metals and arsenic for Elbe River (e.g. Pepelnik et al. 1997). Only for mercury, the Mulde Reservoir is not such a relevant sink. The mercury introduced into Elbe River by the Mulde River almost completely originates from the former chlorine production in Bitterfeld and related contaminations, located downstream the Mulde Reservoir (Hintelmann and Wilken 1994; Brandt 2003). Our sediment investigations (fig. 2, tab. 1) confirm the results of Zerling et al. 2001.

Table 2 Load of arsenic and metals trapped in Mulde Reservoir (averages for the period 1992-1997) related to total input as suspended matter by Mulde River and to total load of Elbe River at Schnackenburg(Germany) (Zerling et al. 2001).

	Cu	Pb	Zn	Cd	Cr	Ni	Co	As	U
Trapped load [t/a]	22	35	192	4.1	12	12	3.2	19	5.0
Trapped load [% of input]	71	84	50	72	71	32	56	52	39
Trapped load [% of load of Elbe River]	23	51	16	90	21			27	

An important aspect regarding sediment quality evaluation and trapping of contaminants is the long term stability of the binding of arsenic and metals in Mulde Reservoir sediments. Conditions in the sediment are reductive and neutral. Accordingly, the formation of sulfides of very low solubility can be considered. There is no evidence of mobilization of contaminants like the decrease of their concentration in deeper layers over time when comparing results from 2002 with that of 2012 and 2013. In addition, the length of diffusion paths to the sediment surface is permanently increasing due to the deposition of new sediment, in particular for the most contaminated sediments in deeper layers. Close to the inflow, older sediments are gradually covered by sand and gravel which are deposited there and already filled up a small part of Mulde Reservoir and formed the two small islands in the inflow part (fi. 1, panel C). Since no changes in redox state or acidification have to be expected and sediment resuspension can be excluded at the lake bottom (the main potential causes for re-mobilization of contaminants from sediments according to Förstner (2004)), there is no considerable risk that the sediments turn from being a sink to being a source of contaminants within timescales relevant for planning in river basin management. Gradual covering of contaminated sediments by sand and gravel is even comparable with artificial capping, one of the options for in situ treatment of contaminated sediments (Förstner 2004).

The deposition of sediments, sand and gravel will fill up the Mulde Reservoir gradually. This raises the question “How long will Mulde Reservoir remain a considerable trap for contaminants from the catchment of Mulde River?” Estimates of sediment accumulation based on the sediment investigations and considering the dynamics of sedimentation in the Main Basin and the Friedersdorf Basin result in a period of about 650 years for continued sedimentation in the Main Basin and additional about 120 years of continued sedimentation in the Friedersdorf Basin, i.e. in summary about 770 years (for details of the applied methodology see Junge 2013).

Conclusions

Mulde Reservoir as an example demonstrates the potential beneficial effect of riverine flow-through of pit lakes. In this case, the effect is even relevant for part of the North Sea. Valente et al. (2015) support our findings and their evaluation: Considerable retardation of metals and arsenic in reservoir sediments was identified in the Rio Tinto basin which is also highly contaminated by long lasting mining and source of contaminations of global relevance (Olias et al. 2006). This is in line with evaluation of river flow-through of pit lakes by McCullough and Schultze (2015). Basically, reservoirs or natural lakes can have comparable effects. However, this finding and the resulting management option to mitigate environmental adverse impacts of mining via riverine flow-through of pit lakes, reservoirs or natural lakes does not allow for being less strict in preventing contaminations already during mining and as close as possible to the mining sites. Not only the river stretches downstream of pit lakes like Mulde Reservoir need to be protected, but also the ones upstream of them.

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