Environmental Impact of Differently Remediated Hard Coal Overburden and Tailings Dumps a Few Decades after Remediation

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Abstract The historical hard coal mining area of the districts of Zwickau and Lugau/Oelsnitz in Saxony (Germany) is a source of heavy metals and arsenic polluting the adjacent ground- and surface waters. Heavy metals and metalloids like Zn, Cd, Ni, Co, As and Mn are transported, partially as fine precipitates, to the adjacent river Zwickauer Mulde and with that to the river Elbe. The dumps are partially older than 150 years, and some of them were remediated more than 50 years ago. Today we still can learn from the more or less successful remediation measures after some decades of application. In this paper, three different dumps are presented and differences between their remediation measures and the success of it are shown. As a result, a successful long-term stability of a covered dump and a covered dump part, respectively, could be shown.

Key Words Hard coal mining dumps, acid mine drainage, prevention of AMD formation, sulfate reduction

Introduction

The environmental impact of mining tailings, overburden and waste rock materials containing sulfidic minerals like pyrite and/ or marcasite FeS_2 is well known at least since decades (Evangelou 1995). This environmental impact is due to biogeochemical weathering processes, in which iron and sulfur mineral oxidizing bacteria play an important role. Since many years interdisciplinary approaches have been developed for the treatment and mitigation of these problems. Most discussions and experiences about the mitigation of AMD occurred only since around the last 20 years, when such environmental issues became more the subject of public interest. It is especially important to find out very effective and low-cost remediation methods, because some of the main problems of AMD treatment at closed or abandoned mining sites are the huge, often remote areas with AMD generation and contamination, and on the other side the only limited available financial sources for remediation of these large areas.

In this study, a few dumps in a historical hard coal mining district of Zwickau/Oelsnitz in Saxony in Germany were investigated to evaluate the success of former revegetation and remediation measures 30 to > 60 years ago. The coal mining at this place was very typical, as it also was and is performed at many other places of the world. More than 80 dumps of the former hard coal mining are located in the area of Zwickau/Oelsnitz; some of these dumps were now investigated in a research project to study their environmental impact and to learn more about the long-term behaviour of such sites, especially about success and problems of the remediation measures. The investigation of such sites represents a rare opportunity because most remediation measures reported internationally are less than 20 years ago (Willscher 2001). Thus, such rare informations about the action of remediation measures after many decades are very valuable.

The hard coal mining sites of Zwickau and Lugau/ Oelsnitz

The hard coal mining sites of this area are situated in the South of East Germany, in the mountainous area of Saxony. Hard coal mining in the area of Zwickau in Germany started already more than 660 years ago. Coal mining activities were growing in the time of industrialization (since 1830); today, the hard coal mining is closed since several decades, but the generation of acid mine drainage (AMD) represents a continuing environmental problem. Ground and surface waters around the dumps are contaminated with Mn, Co, Ni, Zn, As and Cd, and an enrichment of the metall(oid)s is observed in the sediments of the surface waters in the district of Zwickau. The metall(oid)s are transported in their dissolved and particle associated forms to the adjacent creek (the Zwickauer Mulde) which results in downstream contamination of the river Elbe. Investigations of the seepage waters from the dumps show a discharge of heavy metals and sulfate into the ground and surface waters, partially in mass flows of kilograms up to several tons/year (Hertwig 2007). This environmental impact is observed even though a partial recultivation of the dumps occurred up to 60 years ago.

Three dumps (of more than 80) with different environmental conditions were chosen and investigated in the former coal mining area of Zwickau and Oelsnitz:

- Oe10: Closed since >50 years, no topsoil addition, partially natural topsoil generation, planted trees (birch), AMD generation
- Zw10: Closed since > 30 years, partially no topsoil covering and spare tree vegetation (microsomia), heat generation (90°C), parts of the dump covered by a domestic landfill site (covered, with gas collection and -utilization)
- Zw45: Closed since > 64 years, compacted loam and topsoil covers, planted trees and gardens, nearly no acid generation

The dumps consist of coarse to fine grained material due to former processing, and have enhanced water permeability with a porosity of around 40%. The dumps contain pyrite in locally changing concentrations, also depending on the depth of the layers, and typical coal accompanying, heavy metals containing minerals, e.g. sphalerite, galena and arsenopyrite (Frenzel 1874).

Samples from different depths, drainage-, surface- and groundwater samples were taken from the sites. They were investigated for their mechanical, geological, geochemical, biogeochemical, physico-chemical and geohydrological characteristics.

Remediation and environmental impact of the dumps Dumps only with revegetation, no topsoil addition

Two dump sites with a revegetation with trees were investigated, but there occurred no additional topsoil addition (Oe10, parts of Zw10). Pioneer trees (e.g. birch) were planted there to reduce wind and rain erosion processes, but without a previous addition of a topsoil layer. In that time, a natural topsoil development was expected during the growth of pioneer vegetation. Neither the problems of AMD generation were known in that time, nor the relationships between mass transport of oxygen and water into the dumps and biogeochemical weathering processes.

The revegetation of dump Oe10 occurred around 50 years ago, the revegetation of dump Zw10 "only" 20—30 years ago. Dump Oe10 obtains the higher pyrite and total sulfur content (up to 4.35% and up to 4.07%, respectively); hence this dump site also has the highest potential of all three compared sites for the generation of AMD.

Both dumps are characterized by steep slopes, so that a natural topsoil formation and growth of natural vegetation are difficult. As a result, steep dump material can slide down, and oxygen and rain water can penetrate better into core zones of the dump, with the result of a higher weathering activity. Hence, the seepage water of Oe10 has a constant temperature of around 16°C even in winter time which demonstrates increased biogeochemical activities in the dump. The seepage water is acidified (pH 3.61, see Fig. 1, pH_{min}) and the redox potential is with up to 732 mV quite high (Fig. 1, Eh max), and increased amounts of metall(oid)s are dissolved in the seepage- (up to 337 mg/L Zn) and groundwaters. The concentrations of many other contaminants were also the highest of all three investigated dump sites. The oxidation grade of the sulfur (ratio of oxidized S species to total sulfur) in the coal containing layers is 12.8-57.7% (Fig. 1), depending on the depth of the layers. The surface of the dump is not full covered with naturally formed topsoil; hence rainwater and air can trickle and diffuse into the dump, respectively. Without any further remediation measures, AMD generation and contamination of ground and surface waters with metall(oid)s will continue here over further centuries.

Uncovered parts of dump Zw10 have up to today, even after 30 years, partially no natural topsoil formation and only sparse tree vegetation (microsomia). Some slopes of the dump are partially too steep, hence the natural soil formation and natural growth of vegetation are difficult, and slides can occur. The so-called "canyon-formation" at the slopes due to the former dumping process and later by erosion results in a better air diffusion into highly exposed parts of the dump Figure 1 Comparison of characteristic data of dump material and seepage waters from uncovered (Oe10, part of Zw10) and covered dumps (Zw45, part of Zw10) in the former hard coal mining area Zwickau/ Oelsnitz (E_h – redox potential, Ec – electrolytical conductivity, Oxgrad – oxidation grade of sulfur)



with local thermal conversion of coal and pyritic material (up to 90 °C), which can result in a volume contraction, fissure formation, new slides and better access of oxygen to the unconverted coal remainders (high surface area, high porosity). The uncovered parts of the dump show an extended weathering (31–58% oxidation grade of the sulfur in the coal containing layers, Fig 1), an increased acidification (up to pH 4.4, Fig 1 pH_{min}), higher redox potential (up to 533mV, Fig 1 E_{h max}) and sulfate generation (up to 5.5 g/L).

In summary, a revegetation only with trees and no topsoil addition seems to be not sufficient to minimize water and oxygen penetration into the dumps. There is no complete and tight topsoil cover on top and at the slopes of the dumps; thus the porous dump material partially lays on the surface without any soil or plant covering. Hence, water and oxygen have still a good access to these parts of the dumps.

Dumps with a covering and revegetation

After the closedown of the dump Zw10, from 1985—1997 a domestic landfill was sited on parts of it, with 57% of the area on top of former coal sludge ponds (ponds of milling and flotation remainders on an area of 12.2 hectare, where an inflow of around 10.22 Mio m³ of flotation sludges was operated), and 33% of the area on the former mining dump. After the end of operation time, the landfill was sealed and covered until 2001 with soil material, and since then the landfill gas is collected and utilized as energy source.

Whereas uncovered parts of the dump are exposed to an extended weathering (see section before), the covered parts seem to be stabilized and immobilized. These parts show a neutral pH (up to 7.42, Fig 1 pH $_{max}$) and partially a very low redox potential (down to -254 mV). These reducing conditions resulted in a long-term stabilization of the coal sludges underneath; organic substance containing seepage waters from the landfill stimulate sulfate reduction processes in the coal sludges, which could be confirmed in microbiological investigations (Willscher 2010).

The dump Zw45 has a similar total age as dump Oe10, but the end of operation time occurred already 77 years ago. Already > 66 years ago, a first loam and topsoil covering was carried out, and a first use with gardens on top for food supply followed since this time. After WWII trees were cut for heating; in the 50-ies and 60-ies a repeated remediation of the full surface area was carried out with loam and topsoil covers, with planting of new trees and utilization of some area on top for gardens. Since the 80-ies nearly all the top area is used for gardens. The seepage waters of the dump are nearly neutral (pH 6.4-7.15, Fig. 1), and the redox potential is moderately low (104-335 mV, Fig 1). Because of the complete sealing, the oxidation grade of the sulfur in the coal containing layers is relatively low (3.1-11.72%, Fig 1). Concentrations of heavy metals in the ground-and seepage waters are only low, although the highest heavy metal concentrations in the solid phase were measured in this dump. Because of the low oxidation grade and the low acidification of the site, the metals mostly remain in the fraction of undissolved minerals.

These topics, together with the low concentrations of contaminants in the ground- and seepage waters show the effective action of sealing covers on such a dump. Inflowing waters containing organic substance (from gardens and groundwaters) give an additional stabilizing and immobilizing effect; they stimulate the sulfate reduction, as it was shown in the geomicrobiological investigations (Willscher 2010). One problem today is only the low thickness of the topsoil covers of \approx 30 cm, which does not prevent the rise of contaminants with particles (by garden works and digging animals) or in dissolved form by capillary forces. Hence, a further growth of root vegetables is not recommended anymore to people in the gardens. As a result of these investigations, the covering and revegetation of the dumps is minimizing the penetration of rain water and of air into the dump cores, which results in a lower oxidation of the sulfidic minerals in the coal material and subsequently in a minimization or prevention of AMD generation.

Comparison of the seepage formation rates and seepage contaminant loads at the different dump sites

The different remediation measures at the dumps resulted in different seepage water rates for groundwater recharge; so 237 mm/a were calculated (by the program BOWAHALD) for the dump Oe10 with some natural topsoil formation and tree vegetation; 100 mm/a were estimated at the sealed area of Zw10, whereas 350 mm/a are generated at the unsealed area. 125 mm/a of seepage water are generated at the covered dump Zw45, plus additionally 25 mm/a of irrigation water in the gardens. Thus, comparing the seepage loads of the whole dumps (sum of drainage and seepage water of the total base contact area of the dumps), it is evident that the seepage load of sulfate in dump Zw45 (covered and revegetated) is 85% lower than in the dump Oe10 without a covering and revegetation only (data referred to a unit area). The seepage loads of Zn, Ni and Cd in dump Zw45 are 98.6%, 98.5% and 99.98% lower, respectively, compared to the uncovered dump Oeio with only revegetation. As a conclusion, the covering or sealing of the dumps, combined with final vegetation helps significantly to diminish the amount of trickling waters and oxygen diffusion, and therefore also reduces mass transport processes in the dump core. Finally, by these measures, combined with a slight input of organic substance, the biogeochemical weathering of sulfidic minerals can be reduced in a very large extent and over a long period of time, and a long-term stabilization and immobilization of the sulfidic minerals can be achieved.

Conclusions

As results of the substantial investigations of this work, a successful long-term stability of a covered dump and a covered dump part, respectively, could be shown. The sealing and covering is resulting in a high reduction of seepage waters, in nearly no acidification of the dump material, and a good immobilization of the contained metals. Sufficient counts of sulfate reducers were found in all samples with a relatively neutral pH and an adequate content of organic carbon. A simple revegetation of a dump surface may help against erosion processes, but it can not prevent the AMD generation due to the further penetration of water and oxygen into the dump core. A natural soil formation to cover the dumps does not fully occur at such sites; therefore a covering with additional material (partially compacted) is required. The remediation measures of the investigated hard coal mining dumps were carried out decades ago, this means "yesterday"; using our modern, advanced knowledge, even better results should be obtained in future remediation works.

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