

Geochemical and microbial conditions of a lignite coal spoil and overburden area in Middle Germany and environmental impact decades after remediation

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Abstract

During the 20th century, Germany represented one of the world's largest producers of lignite coal. Coal spoil seepage waters from these mine sites can impact billions of cubic meters of ground water for an extended period of time (eternity impacts). Approaches to remediate lignite spoil and overburden material have been ongoing for more than the past 50 years. The relative effectiveness of remediation methods at different lignite coal mining sites can be evaluated based on changes in geochemistry and microbial communities. The results of these long-term measures and the possible improvement of the environmental situation or an ongoing environmental impact give lessons for the advancement of modern remediation methods at current and future coal spoil locations.

In this study, a coal spoil site in Middle Germany with cohesive soil substrates and layers of drift clays was investigated and characterized with geochemical and microbial parameters. The evaluated site partially had a remediated topsoil layer for 40 years, and since this time agricultural cultivation of different plants (grains, legumes, *Poaceae*) on top was performed. The geochemical background data and results of investigation of different microbial populations in the coal spoil substrate are evaluated, and the remaining impact of spoil substrate to the environment is demonstrated. Finally, the results of long-term remediation of such sites, the influence onto microbial life and impact to the environments is compared and discussed.

The investigations resulted in new knowledge and insights into the long-term biogeochemical processes decades after the remediation of coal spoil sites. Measures for the improvement of the actual and future remediation are concluded.

Key words: Lignite coal spoil area, acid mine drainage formation, biodegradation of coal humic matter, ammo-nia accumulation in groundwater, impact on ground and surface water

1. Introduction

In the 20th century, Germany was one of the world's largest producers of lignite coal. Lignite coal represented an important resource for energy and chemical production during a period of important economical growth. Spacious coal spoil areas and the occurrence of large and numerous pit lakes today demonstrate the legacy of the extended open pit lignite coal mining.

Coal spoil seepage waters from these mine sites can impact billions of cubic meters of ground water for an extended period of time (in some cases, perpetuity). Approaches to remediate lignite spoil and overburden material have been ongoing for more than the past 50 years. The relative effectiveness of remediation methods at different lignite sites can be evaluated based on changes in geochemistry and microbial communities (Willscher et al. 2013, 2012a, 2012b). Therefore, lignite coal spoil areas that had been remediated decades ago are investigated today for their changes in geochemical and microbial characteristics. The results of these long-term measures and the possible improvement of the

environmental situation or an ongoing environmental impact, respectively, give lessons for the advancement of modern remediation methods at current and future coal spoil locations.

2. Investigated site and objectives of the study

In the last years, lignite coal spoil sites in the Lusatia (East Germany) with sandy substrates and fast vertical mass transport processes were characterized by geochemical and microbial investigations (WILLSCHER 2013). In this study, another coal spoil site in Middle Germany (the area of Witznitz in the south of Leipzig) with more cohesive soil substrates and layers of calcareous drift clays was investigated (WILLSCHER et al. 2015). The evaluated site partially had a remediated topsoil layer for 40 years, and since this time agricultural cultivation of different plants (grains, legumes, *Pocaceae*) on top was performed.

Objectives of the current investigations were

- to figure out the effects of long-term agricultural utilization and amelioration (since 40 years after recultivation of the spoil area)
- to investigate the influence of cohesive soil substrates and of drift clay (calcareous material) onto the biogeochemical acidification process
- to characterize the effects of groundwater rise onto biogeochemical transformation processes in the coal spoil underground (the final flooding level was accomplished 13 years ago)
- to investigate the effects of different covering layers more than 40 years after remediation, and characterization of the effects of plantation and fertilization on top,
- and to elucidate the fundamental principles of the biogeochemical processes proceeding in the coal spoil substrate

Finally, conclusions should be deduced for the results of the former remediation measures, and recommendations should be given for the further improvement of the environmental situation especially of the ground- and surface waters. As a hypothesis it was assumed, that

- more cohesive soil substrates and drift clays would delay mass transport in the spoil substrate and therefore a retardation of biogeochemical acidification takes place
- agricultural cultivation on the surface for decades could reduce the acidification of the upper spoil layers underneath
- a groundwater rise will shortly establish anaerobic conditions in the saturated zone and therefore maintain the immobilization and elimination of water pollutants like dissolved iron, high sulfate concentrations and acidity

Fundamentals of biogeochemical processes figured out in the Lusatian coal spoil substrates should be confirmed or differences should be figured out.

3. Methods for geochemical and microbial characterization

Main analytical methods for characterization of the chemical- physical and geochemical parameters were the measurement of total content of organic carbon (TOC), total content of inorganic carbon (TIC), total content of sulfur (S_{total}), content of mineral disulfides, content of mineral nitrogen (N_{min}), pH, redox potential E_h , electrolytic conductivity, dissolved sulfate, dissolved Fe (total), Fe(II) and Fe(III), dissolved ammonia, nitrate and nitrite (Ihling et al. 2013).

Microbiological investigation methods of important metabolic groups in the spoil substrate and ground water were the quantification of colony forming units (CFU) of general bacteria and additionally of fungi (aerobic cultivation), microbial sulfate reducers (MPN, anaerobic, for heterotrophic and autotrophic organisms), iron reducers (MPN, anaerobic), heterotrophic nitrate reducers (MPN, anaerobic), autotrophic nitrate reducers (MPN, anaerobic), measurement of the rates of anaerobic ammonification, nitrification and denitrification in flask experiments, and measurement of enzyme activities (urease, amidase and esterase) for an evaluation of the N cycle activity as well as for general microbial activity (Willscher 2012).

A total of 101 samples was taken, including coal spoil samples from different locations and depths, as well as ground- and seepage water samples

4. Main results

4.1 Results of geochemical investigations of the coal spoil substrate and environmental impact

The acidification of the coal spoil substrate essentially depends on the contents of total organic carbon (TOC), total sulfur (S_{total}) and the oxidation grade of the sulfur (ratio of mineral sulfides to oxidized products). The analysis of these parameters in different depths of the coal spoil deposit allows a good investigation of the depth profiles of the coal spoil substrate and its oxidation state (investigations here were accomplished at several sampling points down to a depth of 18 m). All other geochemical parameters basically depend on these main parameters of the coal spoil substrate. The biogeochemical weathering of the tertiary coal spoil substrate finally generates the acidification of the deposit, and dissolved Fe, sulfate, NH_4^+ , dissolved heavy metals and As can be mobilized.

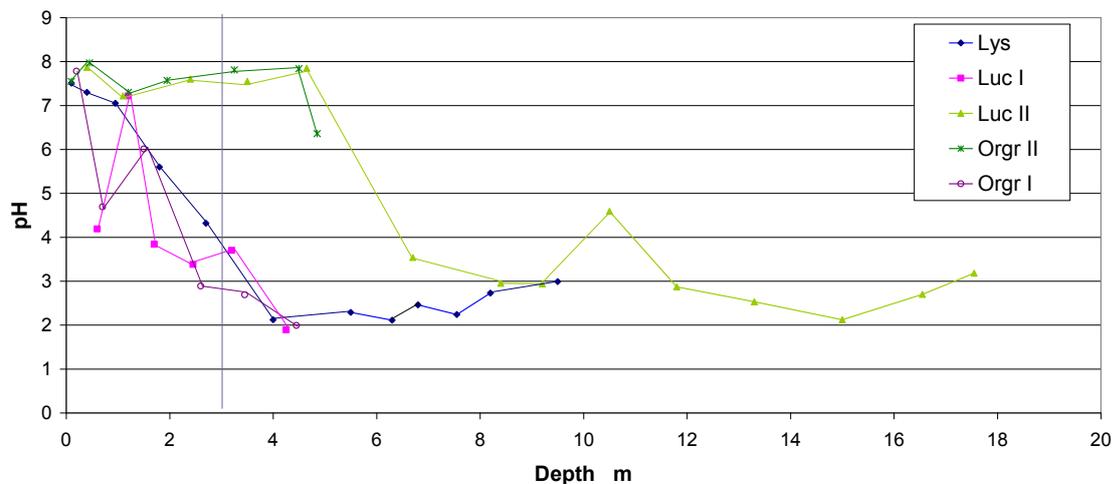


Fig. 1. pH values of the eluates of the samples from several sampling points and different depths (Lys – lysimeter; Luc – Lucerne; Orgr – orchard grass; I – test field I; II – test field II)

In the spoil substrate near the surface (0.6 – 1.0m) partially low pH values were detected (pH 4 – 5), see Fig. 1 (Lucerne I, Orchard Grass I). Due to the technical stacking arrangement, the spoil substrate can be located already that close to the upper top soil layer. Despite the existence of cohesive covering layers at some sampling points, weathering and acidification processes could be detected even in deeper layers down to 18 m (see Fig. 1, Lucerne II). Also in deeper layers down to 18m weathering and acidification processes could be detected. An explanation for the weathering in deeper layers is the former air contact of coal spoil substrate during coal production and during the subsequent stacking process.

At sampling locations with covering layers (clay, cohesive substrates) neutral pH values were detected (see Fig 1 0 – 5m, Lucerne II and Orchard Grass II); at the end of these layers and at the transition to the coal spoil substrate the pH is decreasing within the following 2 – 3m in the depth (see Fig 1).

The biogeochemical weathering process of the coal spoil substrate results in highly increased concentrations of dissolved Fe (up to 1.7 g/l), dissolved sulfate (up to 6.6 g/l), dissolved NH_4^+ (up to > 20mg/l), and of electrolytic conductivities of up to 7.5 mS/ cm. The pH partially is decreasing below a value of 3 (see Fig. 1). The measured data correlated well with previous long-term monitoring data of the coal spoil groundwater in the investigated area. They demonstrated well the weathering state and high environmental impact of this site onto the ground and surface water in the vicinity.

4.2 Results of the microbial investigations of sulphate reducers

The acidification in the tertiary coal spoil substrate also inhibits the microbial sulfate reduction (see Fig. 2). At sampling points with only flat topsoil layer and spoil substrate near the surface, a fast decrease of microbial counts of sulfate reducers could be observed (see Fig. 2, Lucerne I and Orchard Grass I).

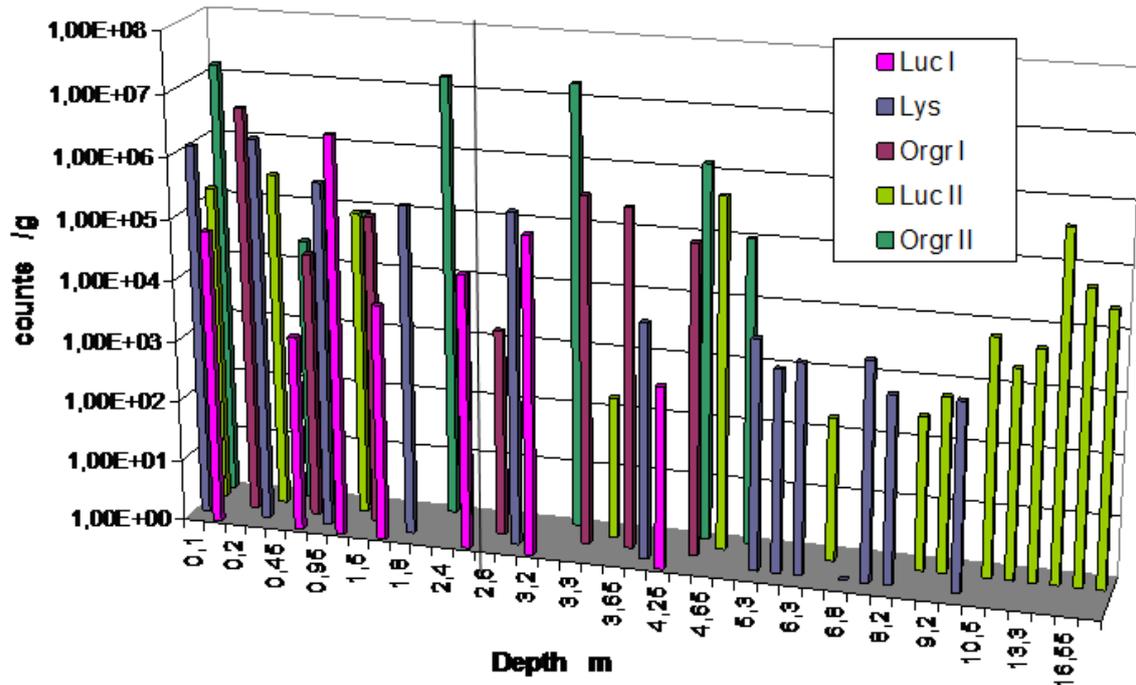


Fig.2. Counts of sulfate reducers at several sampling points and in different depths (Lys –area besides lysimeter; Luc – Lucerne; Orgr – orchard grass; I – test field I; II – test field II). (Every stack column is a result of 96 single measurements)

In the covering layers, the counts of sulfate reducers were constant at nearly neutral pH conditions (see Fig. 2, Orchard Grass II, 0 – 4m) and decreased dramatically to $10^2 - 10^3$ counts /g after transition to the coal spoil substrate with a falling pH (see Fig 2, Orchard Grass II below 5m, and see Fig 1). Underneath a depth of 15m, growth of sulphate reducers is rising again to $10^4 - 10^5$ / g (see Fig. 2), but these counts are still too low for a substantial activity of sustainable sulfate reduction in the coal spoil layers and the ground water zone, respectively.

4.3 Results of the microbial investigations of nitrification and denitrification and environmental impact

As a result of the investigations of the contained N species (NH_4^+ , NO_3^- , NO_2^- , N_{total} , $\text{N}_{\text{mineral}}$) and the the microorganisms participating in the N cycle (nitrificants, autotrophic and heterotrophic denitrificants), an increased fertilization (e.g. a depot fertilization) results in raised concentrations of NH_4^+ in the underground. The biogeochemical degradation of lignite coal substrate (humic matter) in combination with the weathering of the mineral sulfides also contributes to the increased NH_4^+ concentrations in the ground water (Willscher et al. 2012b; Ihling et al. 2013).

Furthermore, the cultivation of lucerne as remediation plant (legume) results in increased N-accumulation in the underground. Therefore an adapted plantation concept of remediation plants, e.g. of legumes with grass (“clover grass”) is recommended.

4.4 Influence of the dumping technology and presence of cohesive substrates

Investigations were carried out at several sampling points with covers of different thickness (0.6m top soil cover; 1m top soil cover; 2.5 – 5.0m of clay partially with calcareous drifted clays).

As a result of these investigations, a top soil layer of only 0.6 – 1.0m thickness is not sufficient to prevent effectively the weathering and acidification of the coal spoil substrate in the underground. Due to the technical dumping, tertiary coal spoil substrates can be located near the surface; hence they can be exposed to an intensified weathering and acidification finally resulting in a contamination of the ground and surface water with the dissolved weathering products (acidity, salinity, dissolved Fe and sulfate, NH_4^+ , heavy metals and As). A reinforcement of the covering layers by cohesive substrates (clay,

calcareous drift clays of 2.5 – 5.0m thickness) result in a stabilization of the geochemical conditions of the coal spoil substrate which shows lower weathering and acidification and its weathering parameters. The neutralization by covering layers acts stabilizing over the following 2 – 3m; underneath after the transition into the coal spoil substrate the biogeochemical weathering again is acting due to the former oxygen contact of the fine grained spoil material (see Fig 1 and subsection 4.1)

4.5 Influence of the rise of the ground water level after the ending of the mining water management

The rise of the ground water level currently results in no substantial reduction of the oxic conditions in the coal spoil substrate (even not in the saturated, but oxidized zone). This result is in accordance with previous experimental results in lysimeters at the site of Witznitz as well as at the experimental lysimeter station at Brandis (Haferkorn 2011). This result implies that even 13 years after completion of the final ground water level near the surface (2 – 4m below top ground surface) no substantial reduction of oxic conditions in the coal spoil substrate could be detected.

As an impact of the high acidification process in the coal spoil substrate, the microbial sulphate reduction as a sustainable process for the immobilization of the weathering products is widely inhibited. In the meantime a gradually biogeochemical Fe reduction can be detected generating the high dissolved iron mass transport in the coal spoil aquifer and subsequently into the surface waters in the vicinity. This Fe mobilization process is currently arising and it will presumably continue for the next centuries.

5. Conclusions

Following main conclusions were deduced from the substantial results of the investigations:

- Despite recultivation, amelioration and agricultural utilization over decades on top, there occurs no substantial neutralization of the coal spoil substrates underneath.
- The influence of the cohesive substrates and of calcareous drift clays is detectable, the action range is only limited (up to 2m).
- The final ground water level after the flooding is accomplished since 13 years, but reducing conditions in the upper saturated zone were not achieved within this time.
- An Influence of the covering layers on top onto the coal spoil substrate is detectable (redox potentials and –processes), but there occurs no complete prevention of the weathering processes of the mineral disulfides in the spoil material.
- In the coal spoil material occur biogeochemical degradation processes in the underground, which were characterized. In analogy to the spoil substrate in the Lusatia, also here a degradation of lignite humic substance and a generation of ammonia were observed.

The investigations resulted in new knowledge and insights into the complex long-term biogeochemical processes decades after the remediation of coal spoil sites. Measures for the improvement of the actual and future remediation are concluded.

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