Assessment of AMD Potential and Prediction of a long-term Sulfate Plume of a Tailing Storage Facility Decades after its Decommissioning

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Extended Abstract

The once famous mining of lead and zinc ores in the Stolberg and Mechernich regions (western Germany) left behind former mine sites most of which were remediated but still affect the ecosystem. The Beythal tailing storage facility was located at one of these sites, and is classified as potentially AMD releasing. Mineralogical and geochemical investigations showed that the tailings remains neutral and has a positive neutralization capacity. However, it has a potential of approx. 2,100 t of sulfate and concentration in the seepage is well above threshold value. A water balance model was established, and a numerical transport model was developed in order to test various possible remediation measures.

Keywords: Assessment, AMD potential, tailing storage facility, sulfate plume, remediation

Introduction

The elongated Beythal tailing storage facility (TSF) covers approx. 45 ha and is surrounded by an up to 35 m high ring dike. From the mid 1950ies to 1969, the TSF received around 5.4 million m3 of tailing from lead and zinc ore mining. Until 2002, around 1.7 million m3 of sand had been recovered from the TSF. The remaining tailings are around 15 to 20 m thick. They contain arsenopyrite, bravoite, galena, pyrite, and sphalerite as well as a small amount of chalcopyrite. The sulfide minerals add up to a total concentration of 0.7–3.6 wt% with sphalerite being the most common (up to 1.9 wt%). The ring dike is now forested, pioneer vegetation has spread in some areas on the tailings surface and a temporary pond has developed on silty areas in the north-western section of the facility. The TSF is under mining supervision by the District Council of Arnsberg, Department 6, Mining and Energy, State of North Rhine–Westphalia, Germany, and has been recognized as a nature reserve in the year 2017.

A trout farm is located at the northwestern foot of the TSF, where a discharge of leachate caused a fish kill in 1999. Investigations at the time revealed tailings' pore water with high concentration of zinc up to 54 mg L^{-1}, nickel up to 10 mg L-1, lead up to 2.5 mg L^{-1} and arsenic up to 1 mg L^{-1} (monitoring data 2011). Although pH was only slightly acidic (arithmetic mean 6.8), the TSF was classified as potentially acid generating. In 2001, a 2 m deep seepage trench was constructed in the area of the trout farm to reduce this immediate hazard and the contaminated seepage water has since been pumped back to the TSF. Concepts for the remediation of the TSF were developed and initial pilot tests were carried out including lime injection into the tailings and active treatment of the collected seepage, all with inconclusive results. In 2015, the Association for Land Recycling and Remediation of Contaminated Sites (AAV) assumed responsibility for the remediation. Since 2017, detailed monitoring, mineralogical and geochemical analyses were conducted and thereafter modelling of the site water balance and groundwater transport modelling were done in order to test various remediation measures.

Methods

Fig. 1 schematically illustrates the comprehensive approach of the various investigations. A monitoring network of existing observation wells records both the tailings' water and groundwater. New hollow-stem auger drillings with complete coring from the TSF plateau were conducted and provided samples for mineralogical and geochemical investigations. The PVC sample tubes were kept in inert gas and frozen until processing in the laboratory.

In the laboratory, the PVC tubes were cut in half under inert gas in a glovebox. The pore water was extracted from a partial sample by centrifugation. In addition to the determination of pH and specific electrical conductivity, filtered samples were analyzed using ion chromatography for major ions, alkalinity titration and ICP-MS and ICP-OES for the metals. Kinetic net acid generation tests according to MEND (2009) and the net neutralization potential according to DIN EN 15875 (2012) were carried out on samples dried under inert gas. Scanning electron microscopy (SEM) and QEMSCAN (Quantitative Evaluation of Minerals by Scanning Electron Microscopy) were used for a quantitative analysis of minerals.

For an in-depth investigation of the water balance of the TSF, the seepage water was calculated with a soil water balance model (BOWAHALD, Dunger 2007), differentiated by area, and the resulting groundwater recharge was used as an upper boundary condition for a numerical groundwater model. According to the soil types found on the TSF

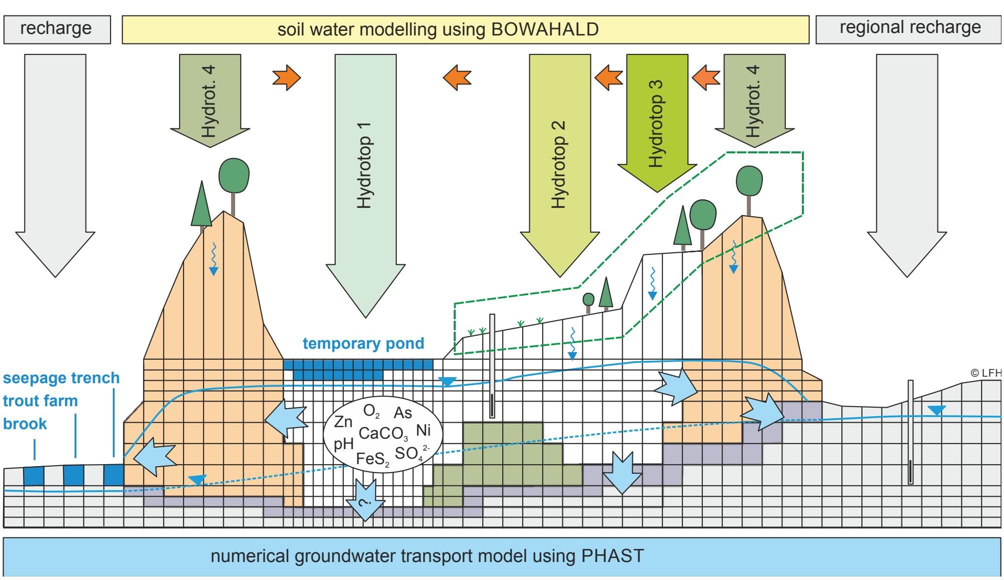

Figure 1 *Concept of sampling and modelling*

(sandy soil, soil with a silty-clayey component, etc.), the type of vegetation (grass, deciduous, coniferous, mixed forest), the slope inclination and exposure, hydrotopes were delimited on the TSF. These one-dimensional hydrotopes were coupled with each other in quasi-two dimensions if required.

Borehole data and geological maps were gathered in a detailed conceptual model. The conceptual model describes the complex geological subsurface under and around the TSF. In NW–SE direction, a tectonic graben with Triassic sandstone and Tertiary and Quaternary sediments extends beneath the TSF. Devonian siltstone and claystone form the graben shoulders. Main axis of the TSF is approximately parallel to the graben's one and main seepage from the TSF is to the NW.

The PHAST software (PHREEQC and HST 3D) was chosen to model the chemical processes taking place in the tailings. In addition to modeling the groundwater flow (program code of HST3D), PHAST models the mass transport with a comprehensive representation of the geochemical processes (PhreeqC). PHAST is a finite difference approach for modeling under saturated conditions (Parkhurst et al. 2010).

Results

The vadose zone in the tailings is approx. 3 m thick. Even after several decades, the tailings remain mostly saturated. This aspect is in agreement with data reported by Rodríguez et al. (2021). Reported values of the tailings' hydraulic conductivity are 10^{-5} – 2×10^{-4} m s^{-1} with smaller values for the silty sections. The calibrated numerical model gives 8×10-5 for sandy and 2×10^{-5} m s^{-1} for silty sections. Vertical seepage is limited by a loamy base layer with a calibrated hydraulic conductivity of 8×10^{-6} m s^{-1} in the south-eastern to 10^{-8} m s^{-1} in the north-western part of the TSF.

In contraction to the previous rating as potentially acid generating, the pore waters from the newly drilled cores were predominantly alkaline, with many samples around pH 8. Only four samples had pH values between 6.7 and 6.9. The carbonate-carbonic acid buffer is decisive for the pH value. Increased metal concentration, especially of zinc, occurs mainly in the capillary fringe and towards the base of the tailings. Data from pore water do not indicate an oxidation front beneath the capillary fringe.

Only minor NAG reactions were observed in the majority of the samples. Maximum temperature increases and the integrated temperature divide the samples into three groups: temperature always below 30 °C, moderate temperature increase to 30–35 °C, and substantial, rapid temperature increase to 45–58 °C (fig. 2). The pHNAG is consistently neutral to alkaline. Group no. 3 is located in the north-western section of the TSF but distributed in all depths of the saturated tailings.

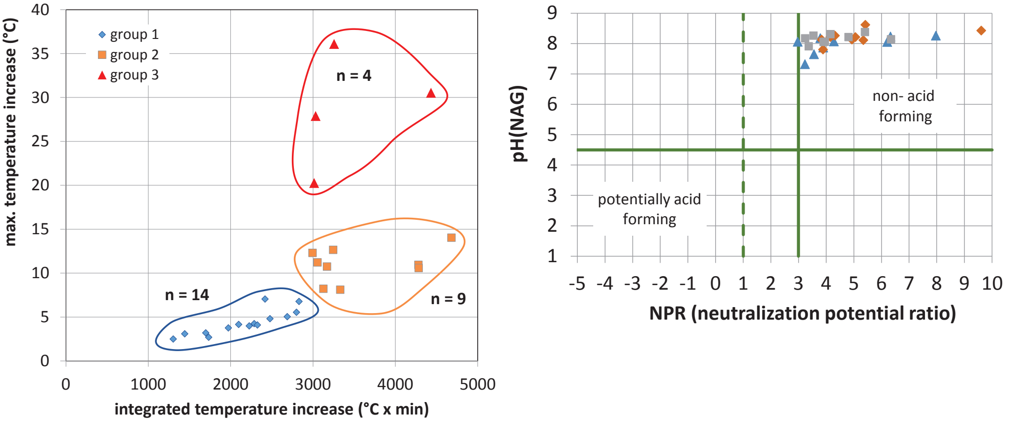

Figure 2 Results of static lab tests: temperature of kinetic NAG tests (left) and ARD rock type classification plot based on ABA and NAG test (see www.gardguide.com) (right)

Total sulfur content was between 0.3 and 0.9 wt%. The acid release potential (AP) was calculated as 0.2–0.6 mol H^+ kg^{-1} from the total sulfur content as sulfate-sulfur averaged only 0.04 wt-%. The neutralization potential (NP) was 0.9–2.2 mol H^+ kg^{-1}. The resulting neutralization potential ratio (NPR) was 2.9 to 9.6. The tailings examined are "non-acid forming" (fig. 2).

Mineralogical results show that dolomite (arithmetic mean 6.6 wt%) and ankerite (arithmetic mean 1.1 wt%) are the buffering carbonates in the tailings. Calcite was hardly detectable. The modal composition of the sulfides was in accordance with the hydrochemical-geochemical data. Bravoite, pyrite, sphalerite, and some galena are well preserved in the tailings (fig. 3). Arsenopyrite was only observed as intergrowth. By QEMSCAN the concentration of iron hydroxides is 0.6 wt% but these minerals were not observed in SEM images. Not surprisingly, jarosite was not detected in the samples due to the pH range.

Sulfate concentration in the pore waters ranges from several hundred up to approx. 1,800 mg L^{-1}. High concentration is not limited to the capillary fringe but was analyzed in all depths. This aspect is in good agreement with the high sulfate concentration in the tailings' water monitoring wells and groundwater monitoring wells. Considering only the actual concentration of sulfate minerals of 0.04 wt% (arithmetic mean), the TSF has a potential of 2,100 tons of readily soluble sulfur. Additionally, sulfidic sulfur of 0.3–0.9 wt% is retained in the TSF in current pH and redox conditions.

Water balance of the TSF determined in the numerical model suggests a groundwater recharge of 81,000 m^3 a^{-1}, an infiltration of 16,000 m^3 a^{-1} and a groundwater inflow below the TSF area of 31,000 m^3 a^{-1} as positive balance elements. These values balance with a seepage at the TSF base of 119,000 m^3 a^{-1} and a small seepage of 9,000 m^3 a^{-1} by the embankments.

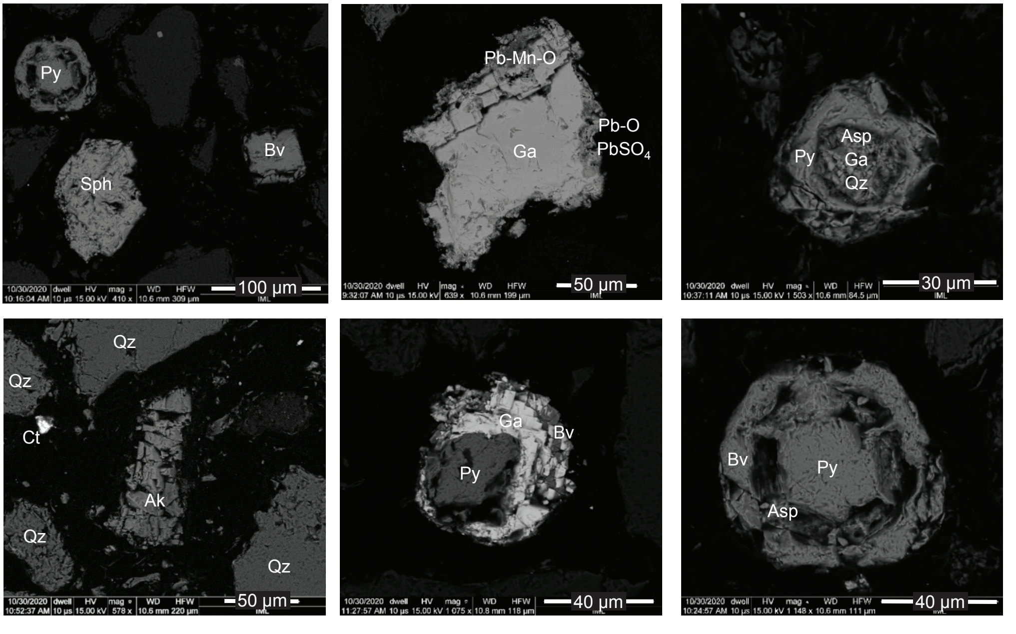

Figure 3 SEM images of sulfides in the tailings. Shape of the grains resemble minerals in the deposits as described in the literature except galena. Pyrite grain of the upper left image is given in more detail as the lower right one. Galena shows some oxidation products (upper center and small Ct grain lower left). Lower left image shows an ankerite grain. Ak: ankerite, Asp: arsenopyrite, Bv: bravoite, Ct: cerrusite, Ga: galena, Py: pyrite, Qz: quarz, Sph: sphalerite

From the groundwater model, the simulation results in a steady-state sulfate flux around 12 t a^{-1} in the outflow of the TSF. This is around seven times the load compared to the surrounding, unaffected groundwater and substantially above threshold values. Data of observation wells were used to calculate sulfate flux of each sedimentary rock by the numerical model. Assuming the modelled seepage and flux, sulfate could potentially leak from the TSF for approx. 170 years.

One possible measure to reduce sulfate flux is to reduce seepage water by increasing evapotranspiration on the plateau of the TSF. A combination of sealing the flat areas around the temporary pond, extensive afforestation of the central sandy areas with coniferous forest and afforestation of the northern and northwestern edge areas with coniferous forest results in a reduction of around 25% (19,000 m^3 a^{-1}) in the seepage water produced. Planting coniferous forests was successfully used for some dumps in the region to reduce seepage. Yet, for the TSF Beythal the formation of preferential flow paths by tree roots and enhanced soil gas convection can enhance sulfide oxidation as the vadose zone is only a few meter thick.

At around 3.5 t a^{-1}, the seepage trench constructed in 2001 only captures part of the sulfate flux. Measurements downstream the TSF suggest the presence of a sulfate plume in the groundwater. An extension with an 8 m deep drain would have to be constructed in front of the toe of the TSF (fig. 4) in order to capture the seepage effectively. The deep drainage reduces the sulfate flux in the groundwater downstream by around 4.7 t a^{-1}.

The leachate collected in the seepage trenches is pumped to the TSF plateau after treatment. Technical sealing could increase the pond area on the TSF plateau and thus increase the evaporation of the captured leachate. An enlarged lake area with technical sealing would reduce sulfate mobilization in the TSF. In the transport calculation, this leads to around 1.4 t a^{-1} less sulfate in the leachate collection and around 0.3 t a^{-1}

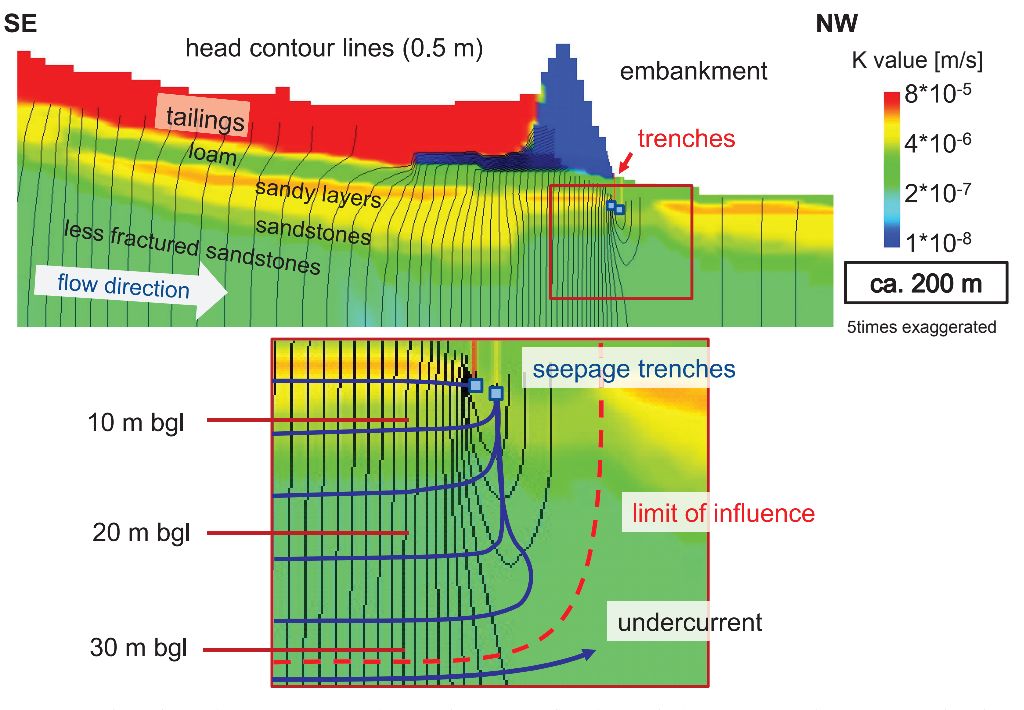

***Figure 4** Effect of 8 m deep seepage trenches on the seepage flow beneath the TSF using the PHAST code. The section shows the northern tip of the TSF and sediments and rocks of the graben*

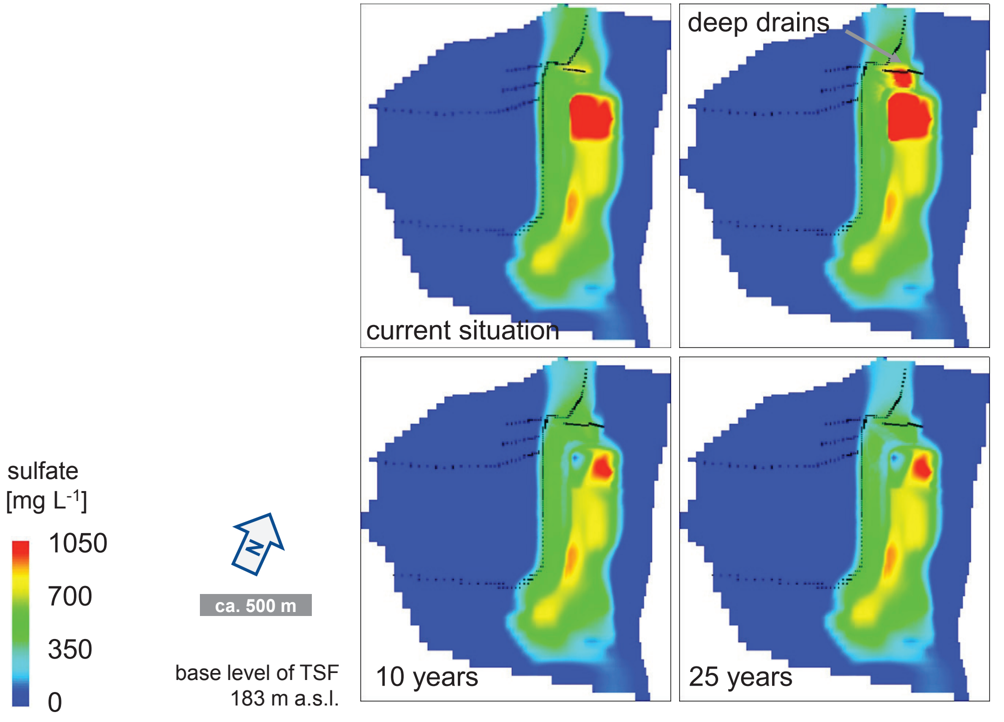

Figure 5 *Simulation of the sulfate plume downstream the northern rim of the TSF. Transient flow and conservative sulfate transport. Current situation is 50 years after the start of simulation in 1969. Expansion of the pond with technical sealing; representation of sulfate in the current situation also with expanded drainage, forecasts for 10 and 25 years*

less sulfate in the groundwater discharge compared to a scenario without extended sealing. A simulation started in 1969 indicates a sulfate plume north of the TSF after 50 years (current situation, fig. 5). All the measures discussed, with the exception of changes to the vegetation, would lead to a considerable reduction in concentration and the retreat of the plume.

Conclusions

This study demonstrates the importance of mineralogical and geochemical analyses for comprehensive and accurate assessment of the potential for tailings to produce AMD. The study included water balancing of a tailing storage facility (TSF) and the modeling of its long-term sulfate release to adjacent groundwater. The research expands understanding of the environmental effects of storage facilities several decades after their decommissioning.

Detailed mineralogical and geochemical investigations have shown that the tailings remain neutral and have a positive neutralization capacity, with dolomite being the main buffering carbonate. The neutralization potential ratio is three to ten. Metal concentration is locally high but remain below threshold values in the seepage more than 20 years after damage to some fish farming ponds. However, the TSF has a potential of approx. 2,100 t of readily soluble sulfate, as the sulfidic sulfur is not mobilized in the prevailing pH and redox conditions. The transport modeling indicated a sulfate flux of 12 t a^{-1} into groundwater. Concentration was seven times higher than regional values and well above threshold values. Sulfate concentration in a small brook and in a water supply well downstream of the TSF is in a similar range, but the model was not calibrated using these data.

The comprehensive approach of the study supported the testing of different remediation measures and enabled decisions about effective approaches to mitigate the influences to downstream ecosystems. Three measures proved to be the most promising. Altering the vegetation on top of the stored material can reduce infiltration and seepage by 5 to 25 %. Along the storage facility's toe an up to 8 m deep drain would be required to capture the sulfate plume. In addition, infiltration of collected and treated leachate on top of the TSF could substantially reduce sulfate concentration and the potentially 170 years of sulfate flux from this tailing storage facility, assuming the actually modelled seepage and flux.

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