Investigation of a Pit Lake Acting as a Large-scale Natural Treatment System for Diffuse Acid Mine Drainage

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Abstract Natural anaerobic biochemical processes used for passive treatment of AMD were observed in the extensive shallow water zone of a pit lake in the former German lignite district of Upper Palatinate. Although continuously fed by acidic metalliferous groundwater, lake-pH increased from 3.5 to circum-neutral over a 15-year-period. The natural attenuation processes were studied and quantified using a regional groundwater flow model linked with geochemical calculations and sediment core analysis. The results indicate that full-scale pit lakes could be used for passive treatment of both diffuse and point source AMD on a much larger scale if the determining environmental conditions were identified.

Key words Acid Mine Drainage, pit lake, passive treatment, diffuse pollution, NAPA

Introduction

Mining activities, especially open-pit coal and lignite mining, are usually associated with the extensive formation of Acid Mine Drainage (AMD). Diffuse AMD is predominantly generated by contact of sulphide-bearing minerals with oxygen and water below the surface. The resulting acidic and metalliferous groundwater can cause extensive pollution and environmental impairment downstream of the actual pollution source. Although there is still a great knowledge gap, studies by Mayes et al. (2008) indicate that diffuse contamination can account for as much as 50 % up to nearly 100 % of surface water pollution and influx of potentially toxic metals depending on mine and catchment characteristics. Whereas point source AMD can easily be collected and treated, the effective control and treatment of diffuse AMD is considered to be almost impossible. Consequently, diffuse AMD is often the main cause for both surface- and groundwater pollution.

It is well known that natural biochemical processes such as microbially mediated sulphate and iron reduction can lead to de-acidification and metal immobilisation (Geller et al. 2009). Sulphate reducing bacteria (SRB) require anoxic conditions, absence of ferric iron, a sulphate concentration > 100 mg/L, a carbon source and thrive under circum-neutral pH (Younger et al. 2002):

$$4FeOOH + 16H^+ + 8SO_4^{2-} + 15CH_2O \rightarrow 4FeS_2 + 25H_2O + 15CO_2$$

Microbial sulphate reduction was observed to contribute to the overall water quality of natural wetlands and small pit lakes (e.g. in the German lignite district of Lusatia). The process is emulated in passive treatment systems such as anaerobic wetlands and Successive Alkalinity Producing Systems (SAPS) (Skousen et al. 2017; Younger et al. 2002). Furthermore, laboratory- and field-scale experiments were conducted to actively induce sulphate reducing processes by introducing external sources of alkalinity and carbon (e.g. lime and straw) into acidic pit lakes (e.g. Fyson & Nixdorf 2006; Geller et al. 2009; Koschorreck et al. 2011). However, to date there is no report of a full-scale pit lake self-neutralising predominantly induced by natural biochemical processes and hence attendant decreasing of the relevant contaminations. The study at hand describes a formerly acidic pit lake which self-neutralised over the course of 10 to 15 years. Furthermore, the study addresses potential perspectives as well as challenges and limitations regarding technical adaptation of pit lakes for mine water management.

Background and Site Description

Lake Knappensee is a pit lake in the former lignite mining district of Upper Palatinate in southeast Germany. The lake is the upstream link in a cascade of two pit lakes surrounded by former opencast segments and overburden dumps. Flooding of the 550,000-m^2 pit lake ended in 1982. Most of the formerly 50 m deep pit was backfilled, resulting in an unusually shallow, polymictic pit lake with a maximal depth of only 9 m (avg. 5 m). As a natural consequence, the northern part of lake Knappensee represents an extensive shallow water zone of approximately 250,000 m^2, which is densely vegetated by submerged bulbous rush (*Juncus bulbosus*). Additionally, the western and northern shores are lined with reed and cattail belts (*Phragmites australis*, *Typha latifolia*). In 1995, a mine water treatment plant was installed in close vicinity to lake Knappensee for chemical treatment of seepage water from an adjacent former pit by adding lime slurry and flocculants. The treated water is discharged into lake Knappensee at its north-western end.

Methods

Investigation of lake Knappensee included, but was not limited to:

1. Continuous monitoring of inflow and outflow volume and water quality as well as monitoring of surrounding groundwater observations wells;
2. Limnological and environmental investigation (vegetation, stratification, depth profiles);
3. Investigation of the regional geology, mining history and mined land morphology with a special focus on waste rock mineralogy and consequential acidification potential;
4. Development of a regional groundwater flow model;
5. Geochemical modelling of regional acid generating and neutralising processes (spatial Acid Base Accounting);
6. Sampling of 15 sediment cores and analysis in a glove box lab under anoxic conditions.

By combining the results, especially of (4) and (5), a regional transport model was developed which was used for the quantification of acidity fluxes to and from lake Knappensee (Schäfer et al. 2016). The sediment cores were analysed for typical mine water contaminants and different sulphur compounds to investigate contaminant and acidity deposition in the sediment of lake Knappensee (Schäfer et al. 2016).

Results and Evaluation

Lake Knappensee is well characterised in terms of water balance and quality since the end of flooding. Additional monitoring of groundwater observation wells shows that up to the present day most of the mined and backfilled land is heavily affected by pyrite oxidation, resulting in highly acidic (acidity up to 48 mmol/l) and metalliferous (total Fe up to 1,500 mg/L) groundwater discharging to lake Knappensee. Nevertheless, between 1999 and 2012 the initially acidic pH of lake Knappensee increased from about 3.5 to circum-neutral. Simultaneously acidity as well as iron and aluminium concentrations decreased substantially from peaks in 1992 and 2001 at up to 1.4 mmol/l and up to 7.0 mg/L respectively to +/- 0 mmol/l and < 0.5 mg/L (fig. 1).

Since 2010 lake Knappensee displays circum-neutral pH and a stable acid-base equilibrium. Only groundwater leakage and lake water in very confined areas directly adjacent to pits and dumps display acidic pH of 2.5 – 4.0. In addition, contamination of lake Knappensee by influent diffuse AMD is still apparent in elevated sulphate (+/- 1,000 mg/L, fig. 1) and manganese (+/- 1 mg/L) concentrations.

Quantitative monitoring and modelling of inflow and outflow show that the total throughflow of lake Knappensee (avg. 1 Mio. m³/year) can be divided into three categories:

1. Rainwater (ca. 50 %): Surface runoff, ditch influx, floodway, precipitation on lake surface less evapotranspiration of lake surface;
2. Discharge from the active-chemical seepage treatment plant of an adjacent landfill for lignite ashes (ca. 25 %);
3. Net-groundwater inflow (ca. 25%): Groundwater inflow from surrounding pits and dumps less groundwater outflow.

A rough qualitative and quantitative water balance of lake Knappensee is outlined in table 1. The first category is of little interest as it is predominantly comprised of unaffected rainwater. As stated above, groundwater discharge to lake Knappensee from surrounding pits and dumps is still highly acidic (Schäfer et al. 2016). In contrast, the effluent of an adjacent chemical treatment plant is circum-neutral and buffered due to addition of lime slurry.

Table 1 *Rough water balance of lake Knappensee*

Category	Water quality	Mean net acidity	App. Volume
1. Rainwater	Neutral, low salinity	+/– 0 mmol/l	500,000 m³/a
2. Treatment plant discharge	Neutral pH, net-alkaline, low Fe/Al, high Mn/SO_4	– 2 mmol/l	250,000 m³/a
3. Net-groundwater inflow	Low pH, high acidity, high Fe/Al/Mn/SO_4	> 6 mmol/l	250,000 m³/a

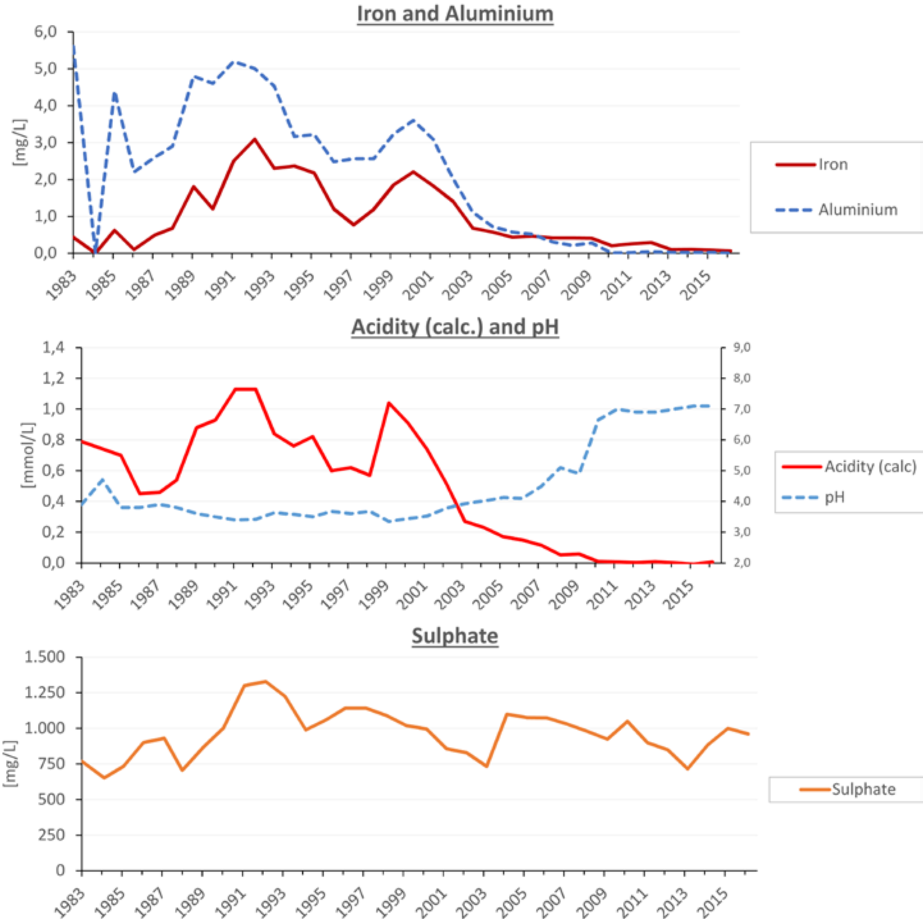

Figure 1 *Development of typical mine water analytes (calc. annual median) in lake Knappensee*

The balance in table 1 shows that since the installation of a chemical treatment plant in the mid-1990s lake Knappensee received substantial inflow of net-alkaline water. Nevertheless, inflowing acidity still exceeds inflowing alkalinity with a net-surplus of approximately 500 – 1,000 kmol acidity per year. This is consistent with the findings of Schäfer et al. (2016), who estimated the cumulative missing acidity surplus in the time interval of 2001 – 2015 to be about 8,400 kmol by assuming a mean acidity of 0.8 mmol/l for lake Knappensee as derived from the transport model mentioned above.

Laboratory analysis of 15 sediment cores sampled across lake Knappensee shows distinct accumulation of typical mine water analytes such as iron (avg. 77 mass-%), aluminium (avg. 7 mass-%), sulphur and organic carbon (avg. 4 mass-% each) in the sediment. Sulphur was predominantly found to be in a reduced state as sulphide (S^{2-}) and pyritic sulphur (S_2^{2-}) plus small amounts of elementary sulphur (S^0). Schäfer et al. (2016) estimated that about 6,400 kmol of acidity are stored in the sediment of lake Knappensee.

Discussion

Firstly, a rough water balance of lake Knappensee shows that there is a large acidity surplus missing in the pit lake. Secondly, sediment core analysis shows that plenty of acidity is stored in (iron-) sulphides in the sediment of lake Knappensee. As both estimations for the relevant time interval from 1999 to 2012 are roughly in the same order of magnitude, the results of this study strongly indicate, that well-known geochemical mechanisms such as natural microbial reduction processes are the driving force of in-lake neutralisation of lake Knappensee.

Furthermore, there is a good cause to believe that continuous discharge of net-alkaline water to lake Knappensee since the mid-1990s might have been a vital trigger and/or prerequisite by creating a local environment suitable for growth and distribution of both hygrophilous plants and SRB. In this case increasing accumulation of dead plant material provided a continuous carbon source whilst living plants stabilised and covered the sediment. Limiting access of oxygen or oxidants to the sediment is a vital prerequisite for the development of a reducing and circum-neutral environment ideal for SRBs (Geller et al. 2009; Younger et al. 2002).

This hypothesis is supported by the fact that the discharge point of the chemical treatment plant at the shallow northern end of the pit lake coincides with the highest intensity of hydrophytes. Presumably over time continual dissimilatory sulphate reduction in the littoral and shallow water zone of lake Knappensee resulted in increasing acidity consumption and eventually in full neutralisation of the pit lake. However, the precise nature and scope of influent alkalinity and hydrophyte distribution as a trigger for biogenic de-acidification remains uncertain and is a subject of follow-up investigations. Additional contribution of further processes is possible but less likely and unverifiable (Schäfer et al. 2016).

Lake Knappensee covers a groundwater drainage area of approximately 3 – 4 km², which corresponds to roughly 50 % of the mining district's diffuse AMD. In addition to extensive acidity consumption, sediment core analysis shows that the sediment of the pit lake is a natural sink for typical mine water contaminants such as sulphate and iron. Since dissimilatory sulphate reduction and subsequent precipitation of iron sulphides developed naturally in an artificial post-mining environment because of advantageous circumstances, major parts of lake Knappensee can be regarded as a large-scale anaerobic passive treatment system or "Natural Attenuating Pit lake Area" (NAPA).

Prospects

Both dissimilatory microbial sulphate reduction and discharge of river water have been investigated in terms of their potential as a tool to manage water quality in acidic pit lakes (e.g. Fyson et al. 2006; Geller et al. 2009; Schultze et al. 2005, 2009), yet no stable and self-sustaining system was discovered or created so far. Previous studies show that a multitude of factors affect the development of acidic pit lakes:

- Morphology (especially water depth and stratification);
- Regional climate, geology and hydrogeology;
- Limnology and ecology (especially vegetation, microbial activity and nutrient cycles);
- Exchange processes at boundary layers between groundwater, sediment and lake water.

Lake Knappensee is one of the very few well-documented examples of successful semi-natural in-lake neutralisation. The case study indicates that the development of natural biochemical processes, already utilised in small-scale passive treatment systems, is possible in full-scale post-mining waterbodies. If the results can be verified in following investigations and if the necessary environmental conditions could be identified, the concept of NAPAs might be transferable to utilise post-mining waterbodies for attenuation of both diffuse and point source AMD on a much larger scale (fig. 2).

Using pit lakes for enhanced natural attenuation of mine water would be much more effective as well as cost-, resource- and energy-saving compared to active measures such as in-lake liming and technical or chemical treatment plants. A stable and self-sustaining system such as lake Knappensee would require only low expenditures for maintenance and monitoring. In addition, lake sediments would provide a stable and spacious long-term sink for mine water contaminants (Junge & Schultze 2016). The comprehensive management of not only point source, but most notably also diffuse mine drainage would be a significant contribution to water protection in post mining landscapes, since so far treatment of diffuse AMD required complex and costly subsurface activities interfering with the groundwater balance (Gast et al. 2010).

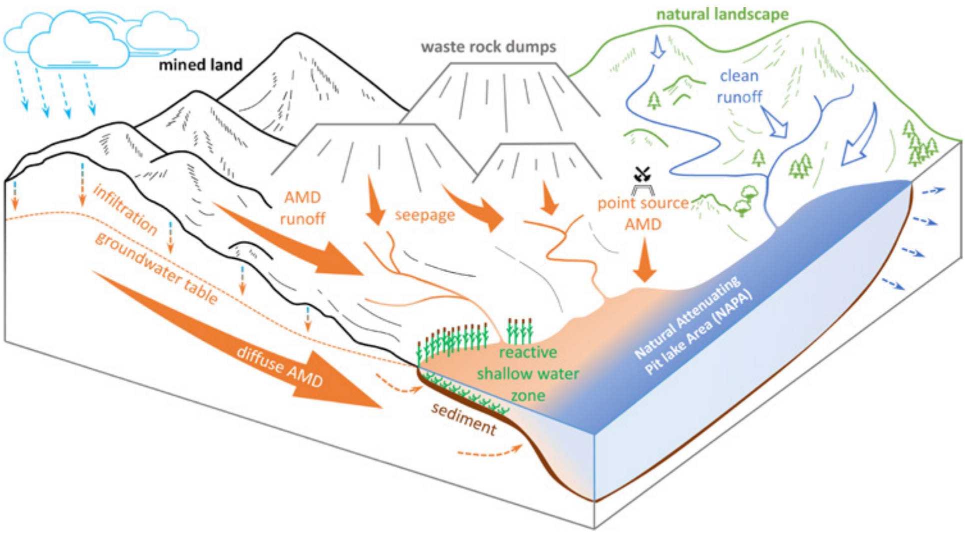

***Figure 2** Scheme of a Natural Attenuating Pit lake Area (NAPA)*

Emulating and augmenting natural attenuation processes in NAPAs could take passive mine water treatment to a new level with great potential regarding water pollution control and cost saving as well as conservation of resources and energy.

Challenges and Limitations

Various studies and experiments, especially in the lignite fields of Eastern Germany, have tried to determine and/or reproduce the environmental conditions necessary for natural in-lake neutralisation. Although lake Knappensee makes a difference in this regard, the identification and sizing of the essential environmental factors and boundary conditions remains the major challenge before artificial reproduction is conceivable. Implementing a NAPA only works as a long-term measure, since in contrast to normal-scale passive treatment plants it takes years or decades until the respective processes get fully established in full-scale. Consequently, appropriate strategies are needed for both short-term pollution control and long-term developments such as water level fluctuations and sediment accumulation. Moreover, research should focus on ways to initiate and accelerate the determining biochemical processes, especially if water authorities are to accept the principle of sacrificing a pit lake for pollution control.

Using pit lakes as large-scale treatment systems for diffuse AMD is only feasible in hydrogeological settings where most of the groundwater discharges to a central void. Furthermore, the results of this study indicate, that extensive shallow water zones are necessary. Since pit lakes are usually created due to the lack of backfill, creating shallow pit lakes contradicts the original purpose.

In summary, as in all cases where natural systems are adapted for technical purposes, several challenges must be met and the exact outcome and chance of success is virtually unpredictable.

Conclusions

- Spatial Acid Base Accounting based on a groundwater model was successfully used for the prediction of geohydrological conditions, yet caution must be taken using geochemical models such as PHREEQ-C for pit lakes, as vital biochemical processes such as microbial sulphate reduction are not considered unless respective kinetics are specially (pre-)defined.
- Lake Knappensee is the first case of natural in-lake neutralisation, although follow-up investigations are necessary to identify and quantify the decisive prerequisites, boundary conditions, sizing criteria and limitations as a basis for potential artificial emulation.
- Contrary to numerous studies postulating a deep meromictic or holomictic pit lake as prerequisite for stable immobilisation of acidity in the anoxic monimolimnion or hypolimnion, lake Knappensee is an example of a very shallow polymictic lake where acidity is immobilised in the epilimnion, presumably due to dense vegetation covering and stabilising the sediment.

- In a best-case scenario, the adaptation of pit lakes for AMD attenuation (especially diffuse AMD) could revolutionise mine water management in opencast mining, away from active or end-of-pipe treatment to integrated, sustainable large-scale NAPAs.
- Demand for novel approaches is evident from the large number of acidic pit lakes worldwide.
- Since the basic principle has already been proven in small-scale anaerobic wetlands, the next step is the adaptation of pit lakes for passive treatment purposes in a large-scale field trial to reproduce the environmental conditions already observed in lake Knappensee.

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