Management of water quality in German pit lakes

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ABSTRACT
About 500 pit lakes which result from lignite mining exist in Germany today. Acidification is the most important issue of water quality in these lakes since about 50% of them have been initially acidic. Only few of the acidic lakes became neutral within years or decades due to natural processes without any remediation measure. Other pit lakes remained acid over decades without any progress in neutralization. About 120 of the pit lakes have been filled since 1990. The filling of these lakes had to fulfill modern environmental standards. The main strategy to prevent and overcome acidification of pit lakes is their filling and flushing with water from external sources instead of naturally rising ground water. External sources may be rivers or dewatering of operating mines depending on the availability of such water. Long-term inflow of acidifying groundwater requires additional approaches as well: prevention of acidification during active mining, in-situ ground water treatment and in-lake treatment. These approaches are based on addition of alkaline substances, on microbial sulfate reduction and on electrolysis. An overview over the state of pit lake water quality and the experiences with different strategies of water quality management in German pit lakes is presented.

Additional Key Words: acidification, electrolysis, liming, microbial sulfate reduction, neutralization, remediation, river water

INTRODUCTION
About 500 pit lakes which result from lignite mining exist in Germany today (Nixdorf et al., 2001). Some of them are more than 100 years old. The distribution of the lignite mining districts is shown in Figure 1. Lignite mining ceased completely in the districts of Oberpfalz and of Hessen.

About 120 of the pit lakes have been filled since 1990 or are currently filling (Krüger et al., 2002), all being located in the eastern German mining districts. The filling of these lakes had to fulfill modern environmental standards. Therefore, many research projects have been conducted. They have generated deeper understanding of the limnology of the

pit lakes and improvement of known and development of new approaches to manage water quality in pit lakes (Fritz and Benth us 2000, Zs chiedrich 2005, Zs chiedrich et al. 2007).

Further pit lakes are planned following closure of the currently operating mines after about 2050 including the largest and deepest pit lakes resulting from lignite mining in Germany. Due to geological conditions, the latter will form in the Rhenish mining district. After filling of these lakes, the dimensions of German pit lakes will range between less than a hectare to 40 km$^2$ of surface area, some thousand cubic meters to 5.3x10$^9$ m$^3$ of volume and less than 10 m to 310 m of maximal depth (Nixdorf et al., 2001).

Acidification is the most important issue of water quality in the pit lakes since about 50% of them have been initially acidic (Figure 2; see also Klapper and Schultze, 1995, Peine and Peiffer, 1998, Nixdorf et al., 1998, Nixdorf et al., 2001 and Czegka et al., 2008). In contrast to earlier assumptions (Klapper and Schultze, 1995), eutrophication was found to be usually no risk for water quality of German pit lakes requiring special measures and strategies (Lessmann et al. 2003a, Schultze et al. 2005). The same is true for salinization by geogenic intrusion of highly saline groundwater. Contamination of pit
lakes from waste deposits or highly contaminated industrial sites was restricted to a small number of cases. Therefore, this paper focuses on measures related to acidification.

Figure 2  Overview over the ranges of water quality parameters in German pit lakes (ACY - acidity, measured by titration with 0.1M NaOH to pH 8.2; ALK - alkalinity, measured by titration with 0.1M HCl to pH 4.3; SRP - soluble reactive phosphorus; TP - total phosphorus; Chl-a - chlorophyll-a. Lines within the boxes are median values, box limits 25- and 75-percentiles, and whiskers show the 10- and 90-percentile values, respectively. Data from Nixdorf et al., 2001).

APPROACHES FOR MANAGEMENT

“Doing nothing”

This was the sole approach for many decades. Missing alternative options, missing environmental standards, missing pressure by potential lake users and expected high costs for alternatives were the reasons for its long-lasting dominance. “Doing nothing” is still appropriate where no risk of acidification occurs for a particular lake or where neutralization by natural processes requires only a very short period. Actual examples are Lake Breitenfeld and Ludwigsee (both Central German District) which both were neutral although filled only by groundwater and local runoff. Lake Nenkersdorf and Lake Hauptrestloch (both Central German District) are examples for lakes which became neutral without any special measure within less than 5 years after final filling with groundwater and local runoff. Lake Bergwitz (Central German District) which became neutral about 25 years after its filling with groundwater (Jordan and Weder, 1995) and small lakes of the region Muskauer Faltenbogen (Lusatian District) which still remain acid many decades after their filling clearly indicate that “doing nothing” may also help in the long run but may require really long periods. As environmental standards and the pressure of public use usually require fast neutralization in our days, “doing nothing” is not applicable on new lakes.
Although “doing nothing” sounds very simple, the processes finally causing neutralization are complex. They include: (i) Washout and flushing of the lakes and of the underground where the acidifying substances originate from. The natural water cycle is accompanied by a permanent washout of any water-soluble substance on the long run. This is well known from natural systems and part of the global cycling of substances in geologically relevant periods. The same is true for pyrite and for products of its oxidation in certain mining areas. As the stocks of pyrite and its oxidation products are limited, the time for their wash out is limited as well. (ii) The neutralization by inflow of bicarbonate with naturally neutral groundwater. Groundwater from areas not impacted by mining usually has a natural content of bicarbonate. Its concentration depends on the biogeochemical conditions along the flow path of the groundwater. If such groundwater enters mining impacted areas – aquifers or lakes – it acts as carrier of alkalinity. Its bicarbonate content is consumed by the neutralization of the receiving water bodies. On the long run, the natural groundwater neutralizes the whole receiving water bodies and replaces the initially acid water. Eventually, the initially acid water bodies become neutral and buffered according to the geology and biogeochemistry of their watershed. (iii) Microbial sulfate reduction. The natural occurrence of sulfate reduction was reported for German pit lake e.g. by Peine and Peiffer (1998). They observed neutralization rates of 4-107 meq/m²·a. Hoth et al. (2005) deduced sulfate reduction and even methane formation from chemical and isotopic analysis of water and gas phase in overburden dumps. They reported that such processes are relevant for TOC-levels >1% in the material of overburden dumps. The TOC mainly consists of remaining small particles of lignite. Although lignite is usually assumed to consist of refractory substances, some microbial communities are able to use it as a substrate (Detmers et al., 2001). However, the observed net rates of sulfate reduction are small compared to the acidity to be neutralized in German acid pit lakes. The neutralization rates found by Peine and Peiffer (1998) are somewhat lower than average rates for neutral lakes of relatively low productivity (182.5 meq/m²·a; Koschorreck and Tittel, 2007). Therefore, the contribution to neutralization is limited. The contribution of sulfate reduction to maintenance of neutral conditions may be, however, of relevance if the inflow of acidity with groundwater is small or if the respective pit lake is a productive one. The average potential compensation of acidity load by internal lake processes was estimated to be 1878 meq/m²·a in productive lakes (Koschorreck and Tittel, 2007).

**Prevention**

Prevention is usually the best way to handle problems. In our case, research for the prevention of pyrite oxidation in overburden dumps of open cast lignite mines in Germany and first patented solutions date back into the 1980s (Fischer et al. 1987, 1999). The major approach is the addition of alkaline substances (e.g. limestone, ashes) to the overburden at the time of dumping. This provides buffering substances at the place of acid formation. The formed acidity is not washed out and pyrite oxidation may be even limited as the microbial pyrite oxidation reaches its highest rates under acid conditions which are prevented by the added alkaline substances. However, it took about twenty years and a lot of further research (e.g. Wisotzky 2003) until the full scale application of this approach could start in mine Garzweiler (Rhenish District) (Eyll-Vetter, 2007).
A further approach to prevent or, at least, to limit pyrite oxidation is the technological adaptation of excavation and dumping to the requirements of minimizing pyrite oxidation. It includes minimizing contact time of pyrite and atmosphere and the controlled placement of pyrite rich overburden within the dumps. The application of this approach is common practice in the mines of RWE in the Rhenish District (Eyll-Vetter, 2007) and of VATTENFALL in the Lusatian District (Rolland, 2007). MIBRAG (Central German District) is going to implement this practice (Jolas, 2007).

In-situ manipulation of groundwater

The first tested approach was the construction of reactive barriers. Ashes were used as reactive material between Lake Partwitz and Lake Sedlitz (Lusatian District). The pilot-scale field test failed widely because the ashes formed a sealing wall in the underground (Schöpke et al., 2006a).

The field tests for microbial sulfate reduction in groundwater directly in the aquifer worked well after a relatively long start phase of one year (Schöpke et al. 2006b). Methanol was injected into the aquifer. It caused considerable reductions of acidity, iron and sulfate in the groundwater. Presently, glycerol is tested as an alternative carbon source for the microorganisms together with an improved injection system.

The third approach of in-situ groundwater manipulation tries to minimize groundwater recharge by maximizing transpiration of plants. Optimal selection of plants and intense harvesting shall minimize the formation of groundwater flow and, in this way, the transport of products of pyrite oxidation. The distribution of remaining pyrite in the underground may limit the applicability of this approach because it lowers the groundwater level. This allows, in turn, for deeper intrusion of air into the underground which may result in increased pyrite oxidation under certain circumstances. The first test of this promising approach started on a dump site south of Leipzig (Central German District) at the end of 2008.

In-situ lake manipulation

Diversion of river water

Diversion of river water into pit lakes for primary filling as well as for permanent flushing of pit lakes has been successfully used several times for pit lake neutralization (Lessmann et al., 2003a, Schultze et al., 2005). Lake Senftenberg (Lusatian District) was the first German pit lake filled with river water at the end of the 1960s. It still requires permanent flushing to stay neutral (Werner et al., 2001). The example of Lake Goitsche (Central German District), which was filled with water of river Mulde from 1999 to 2002 (Schultze et al., 2002, Schultze and Geller, 2004) made clear that the diversion of river water is a complex combination of physical, chemical and biological processes which include dilution, chemical neutralization by bicarbonate and microbial sulfate reduction in lake sediment (Schultze et al., 2005). During lake filling, erosion of former side walls and land slides may be decisive for the progress of neutralization (Schultze et al., 2002, Gröschke et al., 2002). Diversion of river water was applied to about 50% of the German pit lakes which have been filled since 1990. Today, flushing is no longer applied if the outflowing water may damage the receiving stream, river or lake. I.e., flushing requires a complete neutralization for application. However, it is still very useful to keep the
alkalinity of lakes on an appropriate level if those lakes receive acidity from groundwater inflows. As groundwater inflow occurs relatively diffuse in most cases, the diversion of river water is usually the simpler and cheaper option to maintain neutral conditions in the lake compared to groundwater treatment.

Important prerequisites for filling pit lakes with river water and for their permanent or frequent flushing are the availability of enough water and an acceptable water quality of the rivers to be used for diversion. In the beginning of the 1990s, the water quality of rivers in the eastern part of Germany improved very fast allowing for the use of river water for pit lake filling. The availability of water is, however, limited due to other water uses and climatic conditions, especially in the Lusatian District (Koch et al., 2006). Therefore, sophisticated systems for water management were developed and applied for filling pit lakes while ensuring other water use and ecological requirements of the rivers (Fritz and Benthaus, 2000, Schlaeger et al., 2003, Koch et al., 2006). In the Lusatian District, 792·10⁶ m³ of river water were used for pit lake filling from 1996 to 2007 and 995·10⁶ m³ of river water were used for pit lake filling in the Central German District from 1993 to 2007. This made up 62% or 57% of the lake volume filled in the respective periods, respectively.

**Filling with water from operating mines**

Water from operating mines has been the major source for filling of the majority of lakes south of Leipzig (Central German District) after 1995 (Jolas, 1998). The good quality and the considerable buffering capacity of the water from dewatering operations of the mines Profen and Vereinigtes Schleenhain (Central German District) made this possible. Lake Copuden at the south-western margin of Leipzig was the first of these lakes to reach its final level in this region in summer 2000. Since then, it has an excellent water quality and is a major destination for recreational activities of the inhabitants of Leipzig (Linke and Schiffer, 2002). Increasing acidification of water from mine Vereinigtes Schleenhain requires the future exclusion of this water from lake filling (Reichel et al., 2008).

In other parts of the Central German District and in the Lusatian District, water from dewatering operation also contributed to the filling of pit lakes but without being the major water source. In some cases, the water had to be treated before use by addition of limestone. The overall contribution of water from operating mines to the filling of pit lakes was 433·10⁶ m³ (31%) in the Central German District from 1993 to 2007 or 228·10⁶ m³ (14%) in the Lusatian District from 1996 to 2007, respectively.

**Addition of alkaline materials and carbon dioxide to the lakes**

Addition of alkaline substances is a common approach to neutralize water. Lake Steinberg (District of Oberpfalz) was the first German pit lake treated with ashes in 1985. After immediate neutralization, the lake re-acidified to pH 4 in 1986 and pH 3.5 within the next two years and remained at this pH until now (Hemm et al., 2002). Still inflowing acidity from the lake’s catchment area caused the re-acidification. Lake Zieselsmaar (Rhenish District) acidified due to groundwater rise after initially being neutral. It has been limed every year to allow bathing and swimming (Hemm et al., 2002). Lake Horstteich (Lusatian District) was neutralized successfully with sodium hydroxide solution and dolomite in 2005 (Rabe and Uhlmann, 2006). Lake Geierswald (Lusatian District) was partially neutralized in 2004 and 2005 by re-excavation and spreading of waste lime formerly deposited at the lake bottom (Benthaus and Uhlmann, 2006).
The re-suspension of ash deposits was tested successfully combined with addition of CO₂ in Lake Burghammer (Lusatian District; Koch C. et al., 2008, Koch T. et al. 2008). The addition of CO₂ considerably improved the reactivity of the ash and lead to a higher gain of bicarbonate in the lake water. CO₂ addition could also improve the alkalinity gain for the lake water from added sludge of the mine water treatment plant Schwarze Pumpe in Lake Spreetal (Lusatian District; Unger, 2007). Addition of the sludge alone contributed about 30% to the overall improvement of the lake water (Uhlmann et al., 2007). This neutralizing activity of the sludge was based on remaining reactive lime.

Lake Bockwitz (Central German District) was neutralized with soda ash from 2004 to 2007 (Neumann et al. 2008). It is the largest German pit lake (18.7x10⁶ m³) completely neutralized by addition of alkaline substances, but it still requires frequent addition of soda ash to prevent re-acidification. Acidity is still transported into the lake via interflow through the underground immediately surrounding the lake, via inflow from acid lakes up-stream, via groundwater and via erosion of the dump sites in the direct catchment area of the lake. 14.595·10⁶ kg of soda ash were added for neutralization to the lake from 2004 to 2007. Another 0.874·10⁶ kg of soda ash had to be added in 2008 to maintain neutral conditions. Inflow of well buffered groundwater also considerably contributes to the stabilization of the reached neutral state. It is expected to become the main contribution on the long run. As field investigations on the remaining acid inventory are still running, the completion of soda addition is not yet known.

Stimulation of sulfate reduction in lake water and lake sediment

The first field test following this approach used swimming bags filled with waste paper in a full scale application in Mining Lake 113 (Lusatian District). It did not result in any measurable increase of pH or any decrease of acidity (Lessmann et al., 2003b). Another approach was based on the addition of Carbokalk and straw on the lake sediment. After promising success in the lab (Frömmichen et al. 2004), the up-scaling in the field (Mining Lake 111, Lusatian District) failed (Geller et al., 2009). The major reason of this failure is the re-oxidation of products of iron- and sulfate-reduction by ferric iron in the lake sediment or by oxygen and ferric iron in the lake water during overturn in autumn and spring (Koschorreck et al., 2007). Diverse lab-experiments indicated that controlled eutrophication might be an interesting approach to stimulate sulfate reduction in pit lakes via production of high amounts of algal biomass (Fyson et al. 2006). However, the amount of biomass required for sulfate reduction was found to be not reachable in the field (Totsche et al. 2006).

Sulfate reduction in swimming reactors

Microbial sulfate reduction in reactors is often used for neutralization. Swimming reactors were tested in Mining Lake 111 (Lusatian District) using methanol as carbon source for the microorganisms (Luther et al., 2003). After several improvements of the reactors, considerable success was reached in 2007 (Preuß et al., 2007a). However, the final reaction rates (0.12 mmol SO₄²⁻/L·h at a flow rate of 200 L/h) were not high enough for further development and full scale neutralization of a complete lake.
Ex-situ treatment of water

Sulfate reduction in reactors

Glombitza (2001) operated a pilot-scale reactor for microbial sulfate reduction near the shore of Lake Kahnsdorf (Central German District) using methanol as carbon source for the microorganisms. He reached relatively high reaction rates (1.396 mmol SO$_4^{2-}$/L·h at a flow rate of 450 L/h). However, no further development and no application in remediation practice were done. The use of autotrophic sulfate reduction was also tested feeding the reactors with hydrogen, but only in the lab (Wagner et al., 2008). The reached reduction rates (0.35 mmol SO$_4^{2-}$/L·h) are still too small for promising field application.

Ex-situ treatment usually includes the necessity to pump water out of the lake and through the respective treatment constructions. Pumping requires much energy. This a considerable disadvantage from an ecological as well as an economic point of view. In addition, the more the neutralization proceeds, the less efficient the treatment becomes: The progress of neutralization becomes smaller and smaller per pumped volume.

Electrolysis and membrane-based techniques

Electrolysis was tested successfully on pilot scale in the field (Zschiedrich et al. 2007). This approach is economically of interest as the by-products (sulfuric acid or ammonium sulfate) can be sold. In addition, the price of electricity is expected to be relatively low if large amounts of electricity are bought for full scale application. Membrane based approaches were tested to remove sulfate and other substances for purposes requiring high quality water (Preuß et al. 2007b, Härtel et al. 2007). However, these approaches are not considered to play an important role in pit lake neutralization in near future. They are of interest for the use of water from pit lakes for purposes requiring special water quality.

Summarizing evaluation

Figure 3 summarizes the management approaches for pit lakes in former lignite open cast mines discussed above. Only three of the above presented approaches have been used for pit lake management broadly in Germany till today: “doing nothing”, diversion of river water and filling with water from still operating mines.
“Doing nothing” is now restricted to lakes which are not acidified. Simply waiting for natural neutralization as in earlier times is no longer accepted by the responsible authorities for new acid pit lakes or acid pit lakes having an outflow. Diversion of river water and use of water from operating mines are by far the cheapest real treatment approaches (Höppner et al., 2006). Therefore they will remain important for the future too. Because water is limited in the river basins impacted by lignite mining, good management of water resources in these river basins is required. Special management strategies and models were developed for this purpose.

Biological approaches were expected to have high potential for sustainable solutions. However, they did not yet reach broad application in German practice of managing pit lake water quality. The only currently expected exception is the stimulation of sulfate reduction in aquifers. The direct application of alkaline substances combined with and without CO$_2$ will play a growing role for the neutralization and its stabilization.
Intense and complete investigations are a necessary prerequisite for successful planning and application of neutralization approaches. Models are important parts of the planning. Special models were developed and applied successfully (Böhrer et al., 1998, Graupner et al., 2005, Müller et al., 2008, Werner et al., 2008, Schultze and Boehrer, 2008). However, their performance is limited by the quality of their input data.

Application of prevention methods should reduce the necessity for neutralization in future pit lakes in currently operating mines. Detailed documentation of applied methods, adequate monitoring of currently treated lakes and publication of results and lessons learned may act as a base for further improvement of methods and of much more efficient application of the treatment methods in future and in other countries.

Type-specific reference conditions were proposed for the application of the EU Water Framework Directive on pit lakes (Nixdorf et al., 2005). They are, however, still under discussion.

When transferring the German experiences to other countries, climatic and geologic settings have to be kept in mind. German lignite seams are imbedded in layers of unconsolidated rock like clay, sand or gravel. The overburden consists of the same types of material. The interaction of the pit lakes with groundwater may be quite different in hard rock environment which contain the coal deposits in other parts of the world and which are typical for open cast ore mining. The excavated overburden is usually dumped inside the mining voids already during active mining in Germany. Dumping of overburden beside the mine may change the efficiency of approaches and may require additional ones. Contamination with toxic heavy metals and arsenic may be of high importance in mining regions of other countries. Such contamination may require additional approaches or may result in another evaluation of particular remediation approaches like artificial eutrophication (Fisher and Lawrence, 2006).

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