Recent results of the research project OILL (Optimizing In-Lake Liming)

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Abstract From March 2009 until December 2010 the open pit lake Burghammer with a water volume of 36 million m³ was neutralized by in-lake liming using small vessels. Within 58 days the lake water pH was elevated from 3 to 8. The permanent seepage of acid mine water into the lake was controlled by periodic aftercare treatment keeping the pH in an ecological acceptable range of 6 to 8.5 during the above mentioned period (König *et al.* 2010, Scholz 2011, Zschiedrich 2011). Parallel to the pilot project an intensive monitoring project (OILL – Optimizing In-Lake Liming) was performed to optimize in-lake liming. A major goal of the research project OILL was to utilize wind induced lake currents for distributing lime particles in the lake during application of lime by both vessel and floating pipe systems. Wind speed and direction were constantly monitored and lake currents were traced by drift bodies equipped with GPS buoys. Sediment traps were used to investigate the fall out at the lake's bottom during treatment periods. Air photos based on a remote-controlled quadcopter and depth profile water quality monitoring at 23 points at the lake delivered additional information. These data were used to calibrate the numerical model of lake currents (ELCOM) which can now be utilized as an optimization tool for lake liming. Some needs have been identified with respect to the technology applied. Important aspects regarding navigation tracks and new techniques applying pipe distribution systems for in-lake liming have been identified. Furthermore several commercial available lime products have been tested. First results on investigating the dissolution kinetics of several neutralization products, dissolution inhibitors, and utilization of CO_2 to build up the carbonate buffer in the lake water were investigated as well.

Key Words in-lake treatment, liming, acidic mining lakes, ELCOM

Introduction

In Northern European lakes the acidic load originates mainly from the atmosphere and is not due to pyrite weathering. Since more than 30 years, direct liming in catchment areas, or of lakes by boat or helicopter, is applied (Henrikson *et al.* 1995, Sverdrup 1985). Properties of the neutralizing agents and their influence on the success of the remediation strategy were investigated by Nyberg and Thornelof (1988). Positive effects resulted not only in terms of water quality aspects but also for the whole ecosystem (Driscoll *et al.* 1966, Guhren *et al.* 2007, Iivonen *et al.* 1955). Furthermore, there are considerations and experiences in the rehabilitation of mining lakes by liming in the United States (Castro and Moore 2000, Dowling *et al.* 2004) and Finland (Ahtiainen *et al.* 1983). Despite considerable research only little information is available in international literature about acidic lakes in Germany (Bozau *et al.* 2007, Hemm *et al.* 2002, Koschorreck *et al.* 2007a, Koschorreck *et al.* 2007b). In order to optimize direct liming of acidified surface water lab experiments and thermodynamic models were utilized (Merkel *et al.* 2010, Pust *et al.* 2010). Based on these results skepticisms about the feasibility of in-lake liming for treatment of acid mine lakes which are result of an intensive lignite mining in the eastern part of Germany could be reduced. Recently in-lake liming has received a broad acceptance in the remediation industry.

Batch and laboratory investigations

Various batch experiments had been conducted in the laboratory. The experimental layout for determining the solution kinetics included a free-drift batch experiment. Powdered samples were used with regard to the technical application on a remediation ship in liming campaigns. Temperature and p_{CO2} (0.038/0.03/5/30/50/100 vol.-%) were kept constant during all experiments. The necessary amount of neutralization product was modeled before by the help of PhreeqC. A constant particle surface was assumed over the entire course of an experiment. EC, pH and temperature were recorded continuously during the experiments. Calcium concentrations, as well as concentrations of trace metals, were determined by the help of ICP-MS (Thermo Fisher Scientific). TIC was determined with an elemental analysator (liquiTOC, elementar Analysensysteme GmbH).

Soda (Neumann *et al.* 2007), limestone, hydrated lime (König *et al.* 2010, Clauß *et al.* 2010), quick-lime (UIT Info 2011) and limestone deposits in mining lakes (Benthaus and Uhlmann 2066) were utilized with rather different results. According to our research effectiveness and economics of in-lake treatment depends on adequate alkaline products (chemical properties of the lime products, water chemistry with respect to pH and inhibitors (e.g. manganese, cadmium, sulfate), grain size of lime products controlling sedimentation rate) and adequate distribution by boat and lake currents controlling both distribution and mean residence time of particles and thus time for reaction of particles with the acid water. Carbon dioxide may speed up the dissolution rate and buffer capacity.

In our batch and columns experiments, more than 23 different neutralization products (synthetic marble powder and industrial products) were tested and investigated. Based on the elemental contents determined by XRD and SEM-EDX, no significant difference between synthetic marble powder and industrial products appears. Kinetic experiments with marble powder and limestone (KSM Beroun) support this statement.

Ions typical in acid mine drainage (e.g. Mn^{2+}, Cd^{2+}, SO_4^{2-}) have different effects on the kinetic of carbonate dissolution. Manganese concentrations typical for acidic mining lakes inhibit calcite dissolution. Cadmium has as well a significant influence on dissolution and dissolution kinetics. Only about 50 % of the calcium concentration was reached with cadmium as inhibitor compared to the dissolution in absence of cadmium. Increased CO_2 partial pressure might be used to compensate inhibition by material impurities and/or water constituents. Thus, further experiments considering the influence of CO_2 partial pressure and the influence of possible inhibiting ions were performed. Significant differences in reactivity were obvious at $p_{CO2} > 3.8 \cdot 10^{-4}$ bar. Increased CO_2 partial pressures might be used to suppress the effect

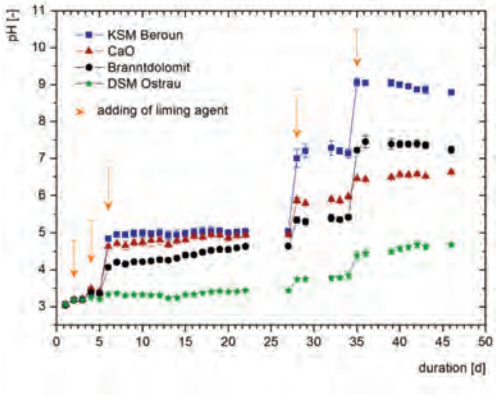

Figure 1 *pH curve using different neutralizing products. Arrows mark adding of the neutralizing agent.*

of inhibition by material impurities and/or dissolved water constituents. Additionally, a possible increased efficiency ratio by using limestone powder during liming campaigns in the pH range > 6 can be achieved.

A variety of column experiments with original water from mining lakes was carried out. Figure 1 shows the behavior of pH during lake water treatment with different neutralizing products. Results showed that a two-stage neutralization, with $CaCO_3$ and then $Ca(OH)_2$ having an optimized grain size distribution, offers neutralization efficiencies close to the theoretical maximum. This treatment scheme was successfully applied in the open pit lake Burghammer.

Field investigation 1: Lake Burghammer

The initial neutralization of the lake Burghammer was carried out between 20th of March and 26th of June 2009. According to theoretical considerations based on primary investigations, modeling and laboratory experiments, a total amount of 36 million m^3 lake water with an acidity of 140 million Moleq had to be treated within the initial neutralization. About 11,010 tons of limestone and 1,123 tons of $Ca(OH)_2$ were distributed on the lake with a medium sized special boat rented from Br. Allerts, Sweden. A total of 58 days for initial treatment was required.

The distribution of the liming products is strongly influenced by wind and lake-internal currents induced by wind. Figure 2 and figure 3 show the distribution of calcite particles at different wind speeds. Wind-induced currents provide an extension of the reaction space within the lake, and thus, increase economics of in-lake treatment. The movement of an applied calcite particle with a diameter of 50 microns is strongly influenced by a wind speed of 4 – 6 m/s as its traveled distance increases by the factor 30 – 40 compared to a no wind scenario. Numerical tracer simulations using the ELCOM model lead to the figures 2 and 3 and were confirmed by GPS drift bodies as will be described in the following. Dissolution processes and thus, decreasing in their particle size and a lower descent velocity, are not considered by this calculation so far.

Lake modeling

ELCOM is a 3D hydrodynamic, thermodynamic and transport model for lake and estuary research (Hodges *et al.* 2000). Besides the use for simulation of variations in water temperature and salinity, it can be useful for the prediction of time and space dependent flow regime in water bodies. Coupling with CAEDYM (Computational Aquatic Ecosystem Dynamics) provides the prediction of scenarios for lake water quality, e.g. Lake Constance (Lang *et al.* 2008, Eder 2007).

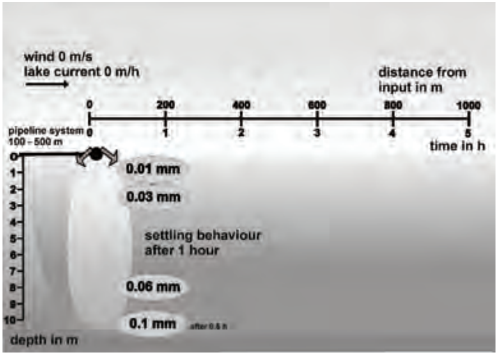

Figure 2 Distribution of calcite particles of different sizes at still air.

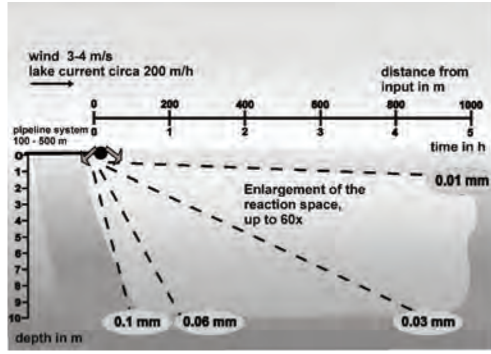

Figure 3 Distribution of calcite particles of different sizes at a wind speed of 3 – 4 m/s and a resulting current velocity of approximately 200 m/h.

Within OILL different flow regimes for the acidic mining lake Burghammer based on different wind and weather conditions have been modeled and compared with measured velocities and directions to verify the numerical model. Besides meteorological data (air temperature, humidity, solar radiation, in-/outflow, cloudiness), prevailing wind speed and direction are important parameters. In addition, the model considers the Coriolis force which might influence the flow direction in particular in large mining lakes.

Results modeled showed flow velocities of 100 – 300 m/h in shallow water depth (0.5 – 4 m) induced by average wind speeds of 3.0 – 6.0 m/s. Figure 4 represents the flow regime in the upper layer of the lake body (depth: 0 – 0.5 m) during westerly wind (3 m/s). Furthermore the model results in vertical currents which are caused by morphological characteristics, but also by density convection and/or temperature differences. To demonstrate the applicability of the numerical model ELCOM for the prediction of lake currents, several test series with GPS drift bodies were conducted in lake Burghammer. A schematic sketch of drift bodies used is shown in Figure 5. The drift body allowed measurements of lake currents in water depths from 1.5 – 8 m at different wind speed and direction. This low cost method was very effective and delivered series of GPS tracks representing lake currents at different depths according to varying wind and weather conditions. These data were used to verify results of numerical modeling lake current and transport of lime particles with regards to wind induced vortices.

For monitoring meteorological parameters especially wind speed and direction a weather station with remote data transmission was installed on the shoreline of the mining lake Burghammer. In addition, wind speed in the center of the lake was measured by the help of a hand anemometer.

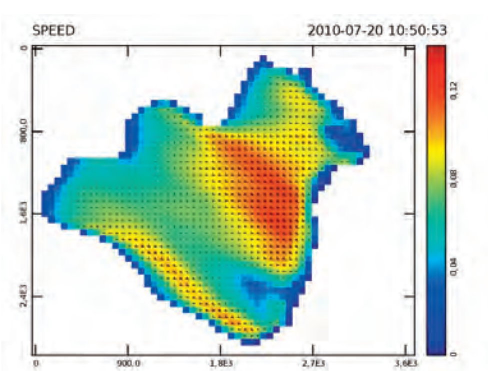

Figure 4 Flow regime in the upper layer of the lake body (water depth: 0 – 0.5 m) in the mining lake Burghammer during westerly wind (3 m/s).

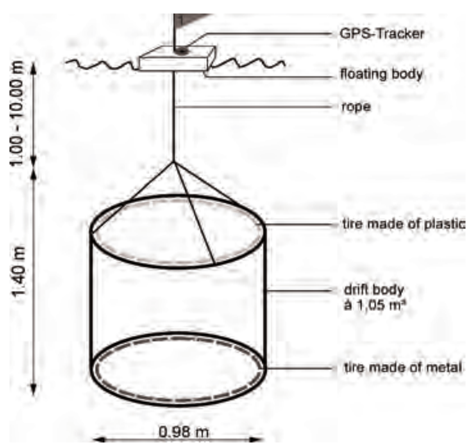

Figure 5 Schematical sketch of a drift body.

Field investigation: Jahnteich, Weißwasser

A further field experiment was conducted at the lake Jahnteich in the city Weißwasser. The initial neutralization of the 6.6 ha large lake was maintained within 3 hours in May 2011. A special sized limestone was distributed by the medium-sized vessel of Br. Allerts (Sweden) and increased the initial pH of 4.3 to 7.5, on average. Table 1 shows selected results of the field experiment.

Aerial photographs were taken by means of a remote controlled electro quadcopter with 4 or 6 rotors carrying a small digital camera flying at an altitude of 50 to 100 m above the lake. This low cost device has been proved as a very efficient tool to control the distribution at the lake's surface in particular along the shore line. Figure 6 shows the distribution of the alkalinity after 10 minutes and 40 minutes after liming (fig.7). The effect of lake internal currents is visible, even with low wind conditions (max. 1 – 3 m/s). Due to high trees on the shoreline only marginal wind energy was induced in this lake body.

Conclusion

The development of further strategies and optimization during lake water treatment by in-lake liming can improve the effectiveness of the method. Using calcite instead of NaOH or CaO as liming product will provide advantages in being more economic and ecological. This has been shown in lake Jahnteich, Weißwasser. If meteorological parameters (wind) and lake specific characteristics (morphology, currents, etc.) are considered efforts and costs for in-lake liming can be minimized. Both vessel and pipeline based methods promise cost effective solutions for initial neutralization and follow-up treatment.

Investigation of the use of CO_2 showed that CO_2 increases the kinetics of dissolution processes. Compared to a multi-stage treatment with limestone and $Ca(OH)_2$ or other alkaline products, more product is needed using CO_2. Due to the developing buffering system less follow-up treatments are necessary. These are more time-consuming; require larger amounts of neutralization product and require the supply of CO_2 which causes additional costs. In particular, if CO_2 cannot be provided free of charge the use of CO_2 to improve sustainability seems to be not economically. Sufficiently precise results in regard to flow direction and speed can be modeled by the help of ELCOM. Thus, a valuable and practical tool for a more effective in-lake treatment is available. Remote controlled quadrocopters are an efficient tool to monitor the lake rehabilitation.

Acknowledgements

The authors thank the Federal Ministry of Economics and Technology (BMWi) for funding the project OILL (Optimizing In-Lake Liming, FKZ KF2383401SA9)

Table 1 *Selected results of the initial neutralization (05/11/2011) of lake Jahnteich, Weißwasser.*

Sampling date	pH	$K_{S4,3}$	$K_{B8,2}$	Fe	Ca
dimension		mmol/l	mmol/l	mg/l	Mg/l
10/15/2010	4.3	0	0.21	< 0.1	46.2
05/05/2011	4.5	0.06	0.25	0.43	44
05/16/2011	7.7	0.5	0.07	0.17	54.4
06/01/2011	6.7	0.45	0.17	-	60

Figure 6 *Aerial photograph showing the distribution of alkalinity after 10 minutes.*

Figure 7 *Aerial photograph showing the distribution of alkalinity after 40 minutes*

within its program "Zentrales Innovationsprogramm Mittelstand (ZIM)".

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