Lausitzer und Mitteldeutsche
Bergbau-Verwaltungsgesellschaft mbH

IN-LAKE NEUTRALISATION OF POST-MINING LAKES

IN THE LUSATIAN AND CENTRAL GERMAN LIGNITE MINING REGION

STATE OF THE ART AND TECHNICAL DEVELOPMENT

12/2017

Title page photograph (source: LMBV, Peter Radke):
LMBV water treatment vessel *Klara* discharging lime into Lake Partwitz, left: Lake Geierswalde, top: Lake Sedlitz, and centre: the Barbara Canal

Imprint

LMBV mbH: Eckhard Scholz, Beate Lucke and Dr. Friedrich-Carl Benthaus

Authors: Prof. Dr.-Ing. habil. Ludwig Luckner and Dr. rer. nat. Oliver Totsche, with the support of Dipl.-Ing. Chantal Jentschura and the members of the "Water Management" scientific and technical advisory board, Prof. Dr. rer. nat. habil. Walter Geller, Prof. Dr. rer. nat. habil. Uwe Grünewald, Prof. Dr.-Ing. Holger Weiß and Dipl.-Geogr. Steffen Reußner

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List of abbreviations

Abbreviation	**Explanation**
Aci	Acidity
Alk	Alkalinity
ANC	Acid Neutralisation Capacity
BGD	BGD ECOSAX GmbH
BNC	Base Neutralisation Capacity
CH	Calcium hydroxide
CL	Caustic lime
BTU	Brandenburg University of Technology Cottbus-Senftenberg
GFI	GFI Grundwasser-Consulting-Institut GmbH
GMB	GMB GmbH
GW	Groundwater
HDHc	High Density Hydrogen carbonate reactor
LSchiffV	State of Brandenburg Shipping Ordinance
LUG	LUG Engineering GmbH
Neq	Neutralisation equivalent
$Neq_{CL,eff}$	Effective percentage of the burnt lime neutralisation equivalent
PL	Pulverised limestone
TIC	Total inorganic carbon
UIT	Umwelt-und Ingenieurtechnik GmbH
V	Volume of lake water
η	Efficiency
η_{pure}	Degree of purity
η_{con}	Conversion factor

Preface

German federal law commits the mining industry to rehabilitate the areas exploited by the extraction of raw materials. To comply with this requirement, many of the mined out pits left by the former opencast mines in the Lusatian and Central German lignite coalfields are being flooded. To create artificial lakes is the preferred form of redevelopment. For establishing these post-mining lakes suitable beds must be formed on the exploited surfaces including the mined out and dump areas in and around the former lignite mines. They usually remain unsealed and unconsolidated, and consequently the surface waters of the post-mining landscape will interact with the local ground water and the solids in adjacent ground layers.

The creation and sustainable maintenance of post-mining lakes as artificial surface waters in the former exploitations of the Lusatian and Central German lignite region requires a high standard of cooperation between the Lausitzer und Mitteldeutsche Bergbau-Verwaltungsgesellschaft (LMBV) as former mining enterprise and the licensing authorities operating under the provisions of mining and water legislation [25]. Generally, the establishment of an artificial surface water body and its long term maintenance is subject to water resources planning permission under water legislation. Particular importance must be given to the monitoring of these lakes in terms of quality and quantity, the results are reported in LMBV’s annually updated “hydrological report” [18].

The actual surface water bodies are created in geotechnically secured beds either by the natural rise of the groundwater level or by flooding with water from nearby rivers. The water in the pit lakes formed in this way is sometimes acidic and requires neutralisation. This is necessary because the inflowing mining-affected ground water and leachate is acidic and contains iron and sulphate. To meet the officially mandated qualities for lake and discharge water it must be neutralized.

The objective of mining and water legislation on creating neutral pit lakes, and thereafter maintaining their neutrality, arises primarily because most post-mining lakes

- discharge lake water into receiving water courses, which can detriment surface water bodies
- infiltrate lake water into the groundwater, and so have a negative impact on the latter,
- being in a neutral state, can offer significantly more potential uses (tourism, fishery, low and high water management, source of potable or industrial water).

In the absence of lake water quality specifications by law, neutralisation of outflows by water treatment plants may be an alternative to the treatment of the complete water body (in-lake neutralisation). In the outflow neutralisation process only the surplus water discharged into the receiving water flow undergoes that treatment. In the majority of cases, however, LMBV will opt for in-lake treatment as the more reasonable alternative in terms of ecology and economic feasibility.

The neutralisation of post-mining lakes and their long term maintenance to sustain a neutral pH is quite costly. This is particularly the case for long term neutralisations, as according to our present knowledge, some lakes will still require several decades of treatment to maintain a neutral pH value.

Even though the acidification of surface waters is a typical consequence of mining, the dimensions of the neutralisation in the eastern German post-mining lakes are globally unique. They are only surpassed by the even larger-scale Scandinavian lake liming programme necessitated by the acid rain.

The chemistry of the Scandinavian soft water lakes, though, cannot be compared with that of the hard water lakes in the eastern German lignite area. Moreover, the acidification propensity of the Scandinavian lakes affected by acid precipitations, and therefore their lime requirements, are substantially lower.

Because of the high costs, and the fact that there was no available technique suited to the local conditions, LMBV – with funding from the German government and the four states Brandenburg, Saxony, Saxony-Anhalt, and Thuringia – has made great efforts over recent years to reduce the cost of initial and follow-up neutralisation of the post-mining lakes. This has led to significant progress concerning the state of the art for in-lake neutralisation measures as defined in Section 3 (11), Annex 1, of the German Federal Water Act.

1 Introduction

When lignite coal is mined from opencast mines the surface layers must be dewatered and removed to access the lignite seams. Draining these top stratua in and around the lignite deposits leads to aeration of the subsurface matter. The drained matter is further aerated as it is extracted and moved to dump sites. This aeration oxidises sulphide minerals (such as pyrite and marcasite), which results in sulphuric acid and water-soluble ferrous iron, aluminium, and some heavy metal ions being released into the aqueous phase. The process is generally known as “contamination by aeration” of the soil and groundwater. This results to the formation of acidic mining waters with relatively high concentrations of SO_4^{2-}, H^+, Fe^{2+}, Fe^{3+}, Al^{3+}, Mn^{2+} and NH_4^+. In the Lusatian mining region there are inadequate quantities of neutral water available for flooding the mined out opencast mining pits. For the follow-up maintenance of the post-mining lakes created in the process, technological neutralisation measures must be introduced to achieve and to sustain neutral water bodies in the post-lignite mining landscapes.

The purpose of neutralizing the sulphuric waters caused by mining is to raise the pH-value as well as to remove the metal ions from the water column. The neutralisation causes the precipitation and flocculation of the metals, and subsequently their discharge from the water into the sediment. After neutralisation the mine water’s pH-value is between 6 and 8, with low concentrations of iron, aluminium, and manganese. Under neutral conditions the ammonia is decomposed by microbiological nitrification. Ammonia nitrification is inhibited in acid waters; this is why acidic lakes have relatively high ammonia concentrations. The process of decomposition may take several years and will not commence before the initial neutralisation has been completed. Sulphate concentrations are not significantly reduced by neutralisation They can only be lowered by gypsum precipitation (formation of $CaSO_4$) when present in high concentrations.

2 pH-value, acidity, and buffer systems in post-mining lakes

When defining the acid status of a body of water, pH-value serves as the intensity parameter and acidity as the capacity parameter (here, see e.g. [8]); the pH value shows the concentration of free acids. It is the relevant parameter in terms of ecology, and therefore notified in official water treatment permissions as the target for the relevant neutralisation measure. Acidity takes account of both free acids and any buffer systems present. It is therefore the relevant parameter when determining the neutralisation agent requirement for treatment. In acidic post-mining lakes, it is usually given as the base neutralisation capacity until a pH of 4.3 (BNC(4.3)), i.e. the quantity of base that must be added to raise the pH to 4.3.

Buffer systems can stabilise the pH-value of the water within a specific range by reacting with the added base (or acid). When adding basic materials (or acid), the pH remains unchanged as long as buffer system elements are present. Once this buffer capacity has been exhausted, the pH-value changes in response to the addition of further basic materials (or acid).
As Fig. 1 shows the most important buffer systems for post-mining lakes are the ferric iron and the aluminium buffers. In addition, in the strongly acidic range the hydrogen sulphate buffer, and in the alkaline range, the manganese buffer can be of some relevance.

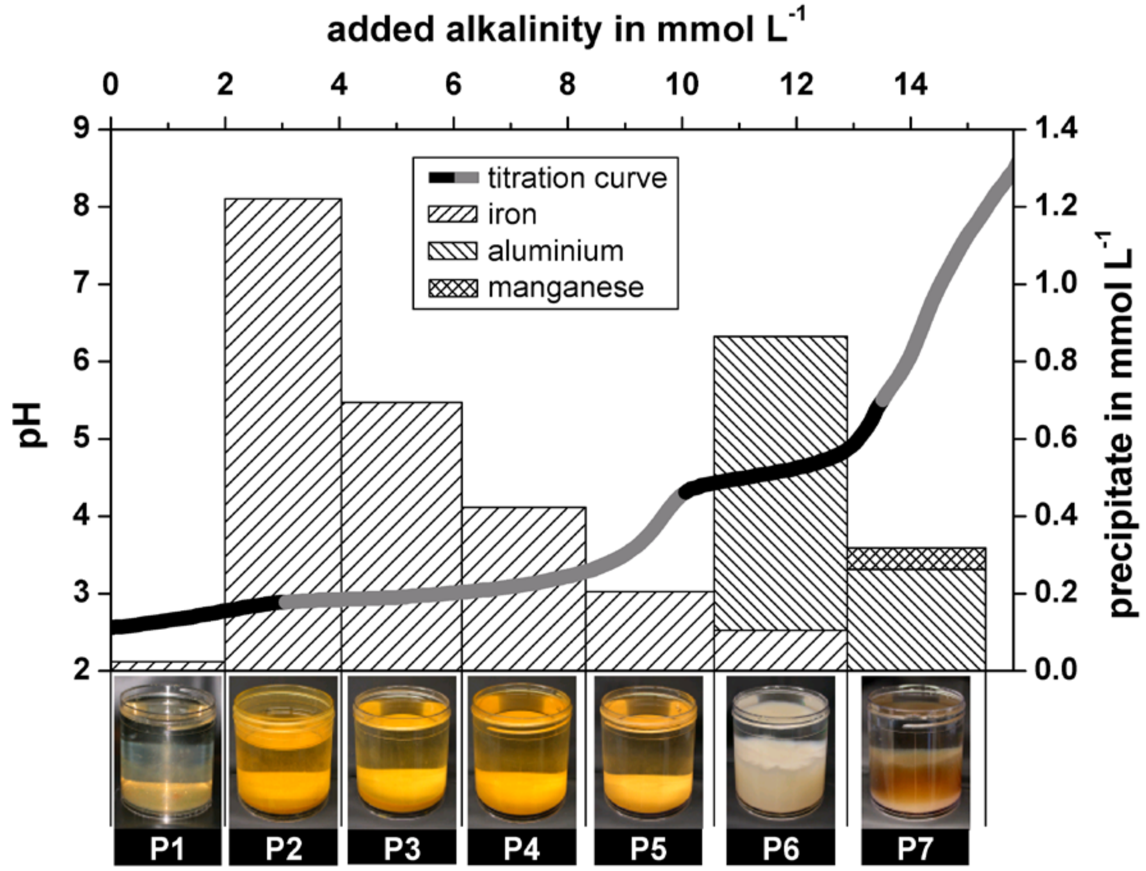


Fig. 1: Titration curve of typical water from an acidic post-mining lake. The columns show the ferric iron, aluminium, and manganese precipitates of lake water samples P1 to P7. The photographs show the freshly-formed precipitates that cloud and tint the lake water, and the formation of sediment in the relevant pH ranges (source: [32])

In the course of the in-lake measures, the lake successively passes the following buffer phases:

- In the case of the **hydrogen sulphate buffer,** in the pH range from 2 to 3, one aqueous species (HSO_4^-) is transformed into another aqueous species (SO_4^{2-}). If performed in a laboratory trial, gradually adding the neutralisation agent, it will not result in any precipitation (see photograph P1 in Fig. 1) and therefore no turbidity of the water. But as the liming of a lake does not entail optimal mixing, iron precipitates are usually identifiable from the very beginning.
- In the case of the **ferric iron buffer**, in the pH range from 3 to 4, dissolved ferric iron is precipitated as orange-brown iron hydroxides (see photographs P2 to P5 in Fig. 1). This leads to the typical orange-brown colour of the water (Fig. 2, left photograph). This buffer is usually the strongest buffer system in acidic post-mining lakes, and consequently the iron turbidity phase continues very long without any decisive change to the pH-value.
- The **aluminium buffer** occurs in the pH range between 4 and 5 and is usually much weaker than the iron buffer. Here, white aluminium hydroxide (see photograph P6 in Fig. 1) precipitates. During this stage, most lakes become turquoise-coloured, similar to the opalescent turquoise colour (Fig. 2 right photograph) that appears when aluminium is added to soft waters being acidified by rain. This colour disappears as phytoplanktonic algae colonisation takes place, turning to the greenish-blue colour that is typical of neutral lakes.
- Above the neutral range, manganese oxide, also known as pyrolusite, may still be precipitated (see photograph P7 in Fig. 1), but because of the low concentration of manganese the impact of

this **manganese buffer** can usually be ignored here. However, as the precipitation occurs spontaneously, in some lakes a dark brown layer of suspended matter can be seen above the sediment (Fig. 2, lower part, see also [30]).

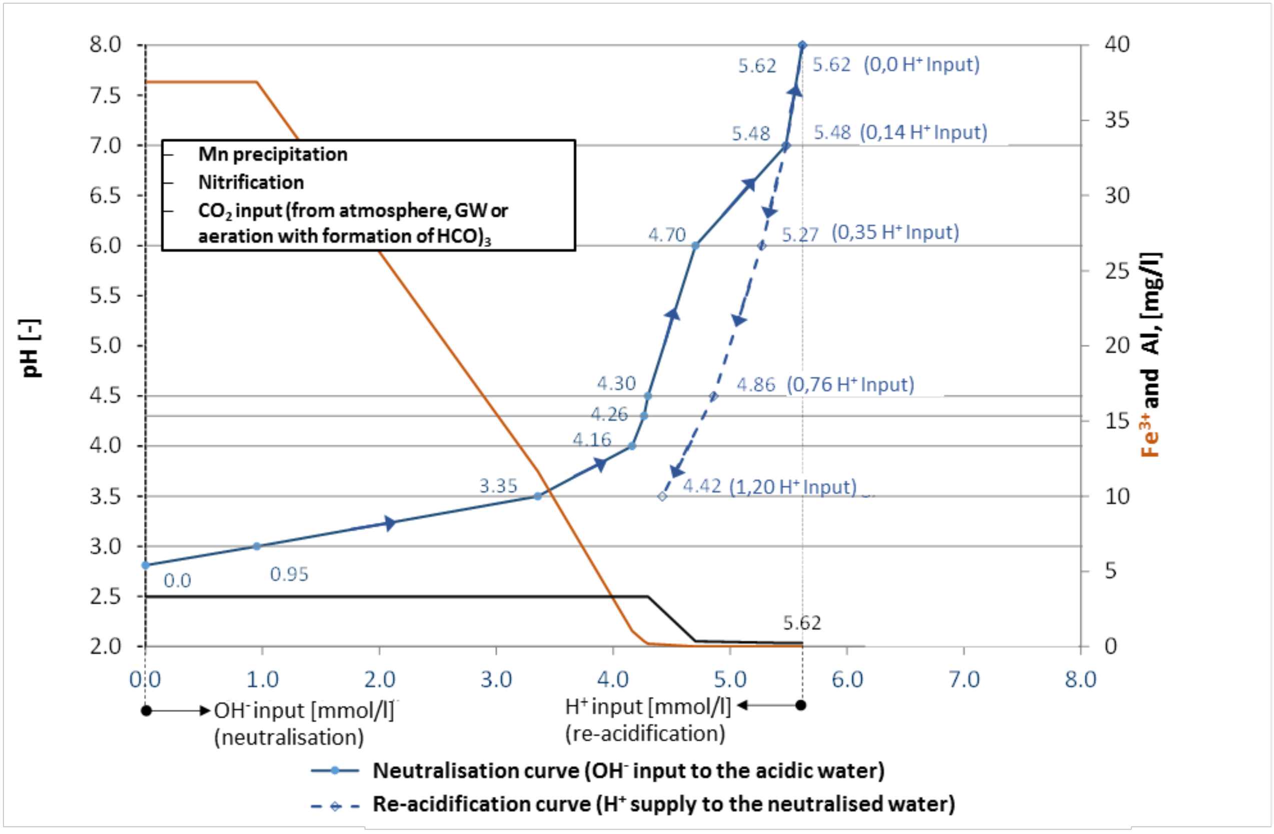
Fig. 2: Lake Partwitz shows water colour of the iron buffer phase, upper left, and the aluminium buffer phase, upper right photograph; below: Lake Scheibe after manganese precipitation (source: LMBV/BTU, Volker Preuß)

In addition to these buffer systems, there is the **hydrogen carbonate buffer**, which is the pH-determining buffer system in neutral lakes by establishing the carbonic acid-to-lime balance. During the initial neutralisation phase its impact is quite negligible, though, as the CO_2 required is only very slowly supplied to the lake by the exfiltrating groundwater and from the atmosphere. However, if a post-mining lake is subsequently maintained in the neutral pH phase by follow-up neutralisation measures, this hydrogen carbonate buffer will become the dominant buffer system. The buffering capacity of the hydrogen carbonate buffer will be shown as acid neutralisation capacity to a pH of 4.3 (ANC(4.3)), i.e. the quantity of acid material that needs to be added to bring the pH-value to 4.3.

Many of the post-mining lakes are prone to re-acidify after initial neutralisation. Usually this is caused by the inflow of sulphuric and ferrous groundwater, leachates, and eroded matter from the shore line (e.g. wind wave erosion, heavy precipitation erosion, deformation of underwater slopes). An acidic sediment at the lake bottom may also contribute to temporary re-acidification.

Only a part of the iron and aluminium hydroxides removed from the lake water through precipitation and sedimentation in the initial neutralizing phase are re-dissolved into the lake water during subsequent re-acidification. This is why the re-acidification curve does not normally match the neutralisation curve; it rather is hysteretic (Example 1).

Example 1:
The titration curves of a lake water sample
Referring to the hysteretic progress illustrated approximately 2.13 (5.48 to 3.35) mmol/l of alkaline matter would have to be added to neutralise the water under consideration from pH 3.5 to pH 7.0. On the other hand, assuming pH = 7, re-acidification to approximately pH 3.5 would have to be expected (broken line) with the delivery of as little as 1.06 mmol/l of acid.



3 In-lake versus lake outflow neutralisation

In contrast to lake outflow neutralisation in-lake neutralisation involves treating the surface water body in the former opencast mining pit by mixing neutralizing agent into the entire water body. This normally requires significantly larger quantities of neutralizing agent than the neutralisation of the surplus waters. This is caused by treatment of the sediment of the lake, erosion material, and the lake water flowing into the groundwater aquifer which must also be neutralized in the process. In the majority of cases, however, LMBV will opt for in-lake treatment as the more reasonable alternative in terms of ecology and economic feasibility. The main advantages it offers usually are

- the option of designing the capacity of the neutralisation technology based on the actual , not the peak, requirement,

- the degradation of ammonia by microbes in open water and at the sediment-water interface. This degradation does not occur in acid water and, where necessary, would need to be achieved through a separate cleaning stage in outflow treatment,
- not having to dispose of the iron sludges formed during neutralisation, because they settle to the lake bottom,
- the input of neutral lake water to, inter alia, the adjacent downstream groundwater aquifer, and
- the wider range of potential uses of a neutral post-mining lake.

Nevertheless, outflow neutralisation continues to be relevant in those post-mining areas where there are numerous minor lakes , often characterised by historic mining activity, and small-size post-mining lakes, as e.g. in the Lauchhammer region.

4 Neutralisation agents

The agents used by LMBV for in-lake treatment across its Lusatian and Central German remediation areas have, until now, been mainly caustic lime (CaO), pulverised limestone ($CaCO_3$), calcium hydroxide ($Ca(OH)_2$), and soda products (Na_2CO_3) (see Fig. 3).

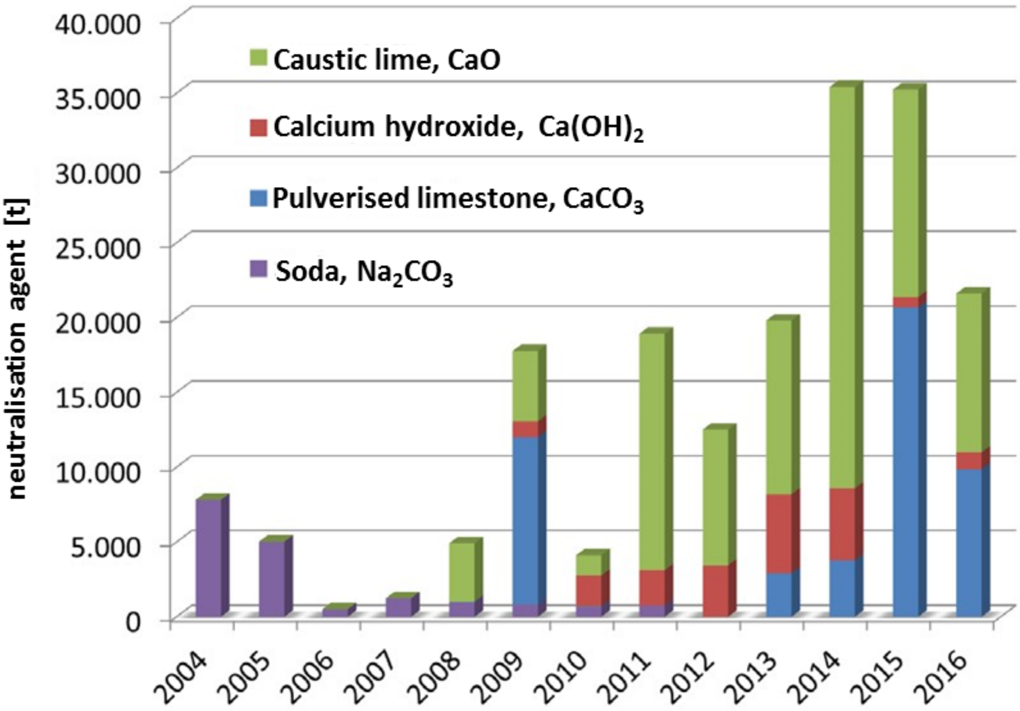


Fig. 3: LMBV's use of neutralisation agent for in-lake neutralisation

To generate a certain quantity of alkalinity, expressed in moles (the substance quantity unit) and capable of neutralizing an equal quantity of acid in each case, different types of these neutralisation agent products are required. Usually, a defined laboratory procedure is used to determine the **neutralisation agent equivalent** in moles of alkalinity (Alk) per kilogram.

To make an initial assessment, a neutralisation equivalent is assumed that is the result of the stoichiometry of the reaction equation of the neutralisation agent's significant component (e.g. CaO in caustic lime) taking certain conditions into account (Example 2). This reaction equation functions as a model that can reflect the heterogeneous neutralisation reaction of a sample of lake water with sufficient accuracy.

Example 2:
The initial assessment of the neutralisation equivalent (N_{eq}) based on the assumed stoichiometric reaction model

Caustic lime (CL):
From $CaO + H_2O \rightarrow Ca^{2+} + 2\ OH^-$ follows $\mathbf{Neq_{CL}} = 2\ mol_{OH-} / 0.056\ kgCaO = \mathbf{35.7\ mol_{Alk}/kg_{CaO}}$.
Moreover, to estimate the actual effective portion, both the degree of the substance's purity and a pH-related conversion rate can be taken into account, such that, for example:
$\mathbf{Neq_{CL,eff} = Neq_{CaO} \cdot \eta_{pure} \cdot \eta_{con} = 27.1\ mol_{Alk}/kg_{CaO}}$
whereby:
Degree of purity $\eta_{pure} = kg_{CaO}/kg_{CL}$ e.g. **0.95** (from manufacturer's data sheet)
Conversion rate $\eta_{con} = kg_{CaO,con}/kg_{CL}$ e.g. **0.80** (i.e. 20 % of CaO do not react in accordance with the reaction model equation)

The neutralisation equivalents for the other lime products are substantially lower, e.g. for
Calcium hydroxide (CH): $Ca(OH)_2 \rightarrow Ca^{2+} + 2OH^-$ => $\mathbf{Neq_{CH} = 27\ mol_{Alk}/kg_{Ca(OH)2}}$
Pulverised limestone (PL): $CaCO_3 + H_2O \rightarrow Ca^{2+} + HCO_3^- + OH^-$ => $\mathbf{Neq_{PL} = 20\ mol_{Alk}/kg_{CaCO3}}$
Soda: $Na_2CO_3 + H_2O \rightarrow 2Na^+ + OH^- + HCO_3^-$ => $\mathbf{Neq_{Soda} = 18.8\ mol_{Alk}/kg_{Na2CO3}}$
The conversion rates (η_{con}) of these neutralisation products can match those of caustic lime only if they are very fine-grained.

Caustic lime is characterised by a high neutralisation equivalent and high reactivity. At the same time, even minor overdosing can rapidly lead to a pH higher than 10. Usually, for ecological reasons, the authorities require that the pH of a neutralized lake be between 6 and 8.5. A high pH arises when the total quantity of neutralisation agent has been overdosed. In the vicinity of the input area can exceed the healthy or ecologically tolerable pH levels. In order to check whether these locally elevated pH levels represent the source of a relevant hazard, LMBV has performed analyses of the pH distribution behind a water treatment vessel adding caustic lime. The tests (Fig. 4) show that in an acidic lake (pH ≈ 2.8) the pH assessed as being potentially hazardous, i.e. greater than 9, only occurs over a short distance approximately 20 metres behind the vessel, spreading approximately 6 metres laterally. It means that any hazard at a certain point would cease to persist just 10 seconds after the vessel has passed by. In contrast, in a neutral lake pH values greater than 9 can still be identified even 700 metres behind the vessel and spreading some 400 metres laterally. This is the equivalent of a time span of ≈ 10 minutes during which there is a potential hazard after the ship has passed.

The main reason for this substantial difference lies in the fact that in acidic lakes up to a pH of approximately 4.5 there is still dissolved iron and aluminium in the lake water. These metals are precipitated as hydroxides when alkalinity is added. This means, that in an acidic environment the pH reduction is primarily brought about by chemical reaction, while in the neutral range, it is mainly due to the commingling with less alkaline water.

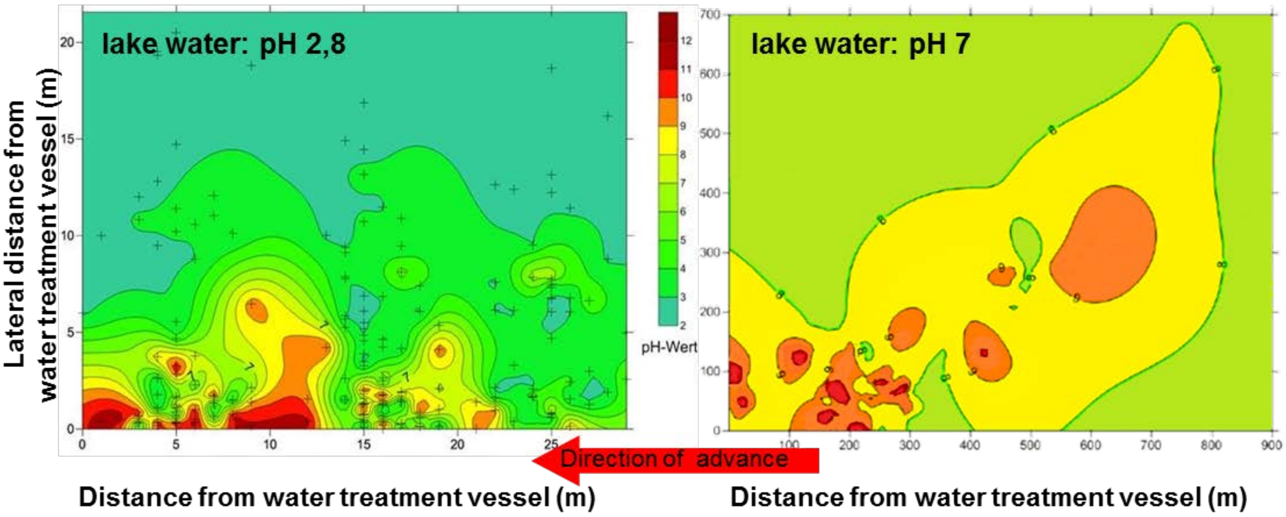


Fig. 4: Comparison of the pH values behind a water treatment vessel adding caustic lime in an acidic and in a neutral post-mining lake (source: GFI/LUG)

Calcium hydroxide has the same characteristics as caustic lime, but its bulk density is lower, and it is less reactive due to the water content as hydroxide.

Pulverised limestone has a significantly lower neutralisation equivalent as well as less reactivity than caustic lime. As a result, approximately twice as many tons of pulverised limestone than of caustic lime is required in order to achieve the same neutralisation effect. On the other hand, an excessively high pH cannot occur using pulverised limestone for neutralisation as it will only react up to a pH of approximately 7.5. Also, because of its carbonate component, pulverised limestone can buffer the lake water against re-acidification. The suspensibility and sedimentation rate of the lime particles depend on their size and density. While lime particles with a diameter of between 20 and 40 µm sink quite rapidly, particles having a diameter of less than 10 µm remain in suspension [24]. One particular form of pulverised limestone is **chalk;** it is fine-grained (3 to 5 µm) and has a large inner surface area.

Because of its carbonate component, **soda** can establish a certain buffer in a similar fashion to pulverised limestone. Unlike pulverised limestone, however, soda remains very soluble even at a high pH so that potentially hazardous pH values may be reached, the same as with caustic lime. On the other hand, already with a pH greater than 8 the previously formed hydrogen carbonate buffer is precipitated as calcite, which limits the buffering characteristics of soda.

The neutralisation agent can be added in a dry, a suspended, or a liquid state. As the neutralisation agent must be transported to the input location in tanks of limited volume over distances, transport costs for dry caustic lime, with its high neutralisation equivalents and bulk densities of approximately 0.9 to 1.0 t/m³, are generally the lowest. For pulverised limestone or chalk suspensions the parameters of relevance to transport costs are, in most cases, significantly less favourable. Calcium hydroxides, but also very finely pulverised limestone, frequently have low bulk densities. The input of lime to neutralize water bodies in post-mining lakes may have only reduced effects for sulphate-rich lake water if protective layers of gypsum form on the surfaces of the caustic lime or limestone particles (calcification), which can inhibit them from dissolving any further into the lake water.

Alkalinity delivered to the lake bed by limestone particles that have remained undissolved cannot generally be dismissed as being disadvantageous. A slightly alkaline lake bed is beneficial to the zoobenthos and can reduce the acidity of any acidic discharges from the lake bed sediments into the lake's hypolimnion layer. The re-dissolution of sedimented neutralisation products can thus contribute to stabilising the lake water quality.

5 First approaches to in-lake procedures

This chapter analyses the in-lake neutralisation processes concerning lignite coalfield remediation of the recent years carried out by LMBV in the Central German and Lusatian lignite-mining regions.

As the first lake neutralisation measures became necessary there was not any standard technology available to ensure an efficient reaction of the neutralisation agents or the best possible spreading of the resultant alkalinity ($OH^-$and/or HCO_3^- ions) across the lakes. LMBV tested several in-lake treatment techniques (see also [8], [21] and [25]).

The techniques available for adding lime particle suspensions to acidic post-mining lakes are either the use of mobile water treatment vessels, which cover most of the lake surface during the liming, or stationary plants, spreading the lime with pipes into the lake.

The early in-lake treatment will be illustrated here with several examples of measures implemented during roughly the first decade following the turn of the millennium. After the first full-scale in-lake neutralisation procedures in Lake Senftenberg (former Niemtsch mining pit) with approximately 11 000 tonnes of lime and 3 000 tonnes of caustic soda put into the lake using hopper barges, it very quickly became clear that any efficient neutralisation of post-mining lakes with solid lime products required relatively long periods of reaction with the lake water and intense mixing processes. The lime fell far too rapidly from the hopper barges to the lake bottom in clumps, without dissolving any further into the lake water. In addition, the lime particles became inert in the sulphate-rich lake water, presumably because of the development of gypsum on the particle surface. This kind of lime deposits on post-mining lake beds were also found in, for example, Lake Geierswalde, and in Lake Südteich around the inflows of technically neutralized mine waters.

5.1 Ash Resuspension in Lake Bernstein

Until the 1990s power station ash, coal dross, and iron hydroxide sludge were being flushed into Lake Bernstein. These alkaline-reacting sediments were taken up by a suction dredger in 2002-2003, suspended, and then redistributed back into the lake through a pressure pipeline. In 2007, this procedure was augmented in the course of a trial by adding CO_2 to the ash suspension, whereby it was caused to react intensively in a tube reactor before being returned to the lake, achieving HCO_3^- concentrations of up to 7.5 mmol/l in the suspension [28]. However, the power station ash resuspension method did not prove to be economic, and thereafter LMBV did not repeat this method in any of the post-mining lakes.

5.2 Lime resuspension in Lake Geierswalde

On the lake bed of Lake Geierswalde a lime deposit was built by the inflow of water from a water treatment plant. In 2004 and 2005 this deposit was collected using a suction dredger, then suspended in a pipeline of 2.4 km length, and subsequently distributed across the lake surface by means of ten large sprinklers. The removal of the lime heaps, formed by water treatment in the past, was necessary to eliminate shoals of < 2 m in the lake. The alkaline suspension brought to the lake surface had a solids content of 2 % by mass. While these solid particles were sinking to the lake's bottom, some part of the lime dissolved causing neutralisation in Lake Geierswalde. As there are hardly any other post-mining lakes with similar lime deposits, this technology is not suitable as a standard technology. The project was able to show, that wind and density driven convection in the lake water contributed to further dissolution of lime particles, as well as for the intermixing and distribution of their reaction products [1]. The follow-up project confirmed this effect which was to become the basis for all further stationary in-lake neutralisation plants.

5.3 Input of soda to Lake Bockwitz

As part of another LMBV project soda was added to the water of the post-mining lake Bockwitz to initiate neutralisation in 2004, and in the follow-up neutralisation from 2007. The initial values of the acidic water body of some 18 million m³ were:

pH = 2.7, BNC(4.3) = 4.5 mmol/l, and Fe_{tot} = 0 55 mg/l.

Fig. 5: Soda input to Lake Bockwitz (source: BGD/LMBV)

The pulverised soda was initially led directly from the silo vehicle into the lake below the surface close to the shore, and later added pneumatically as a mixture of solids and air, at a distance from the shore (Fig. 5). As a result of the input of soda, a temporary pH value in the range up to 7.7 was achieved with weak buffering of up to ANC(4.3) ≈ 0.4 mol/l. Thus, the method proved technically feasible [4]. The final addition of soda took place in 2011 (Fig. 3).

Lake Bockwitz has since been subject to natural succession, and thus re-acidified at a relatively slow rate which will cease at a pH of 3.5, permitting the general use of the post-mining lake for tourism without the need for any technological follow-up neutralisation. In general, though, the input of soda has proved to be economically inefficient because of the rising price of soda on global markets. So far, the LMBV has not repeated this in-lake neutralisation process in any other post-mining lake.

5.4 Adding caustic lime to Lake Hain

From 2008, in-lake neutralisation was carried out using a plant (Fig. 6) on the southern shore of Lake Hain. The caustic lime was stored in silos and the lime suspension produced in a two-stage process. Lake water from the nearby Haubitz area was used to slake the lime and produce the suspension. Input of the caustic lime suspension was through a sprinkler above water at about 2 % by mass.

The lime was distributed across the lake by convective lake water currents. The part of Lake Hain's water body that had filled the former Haubitz mining pit was likewise neutralized [3] by wind and density driven flow processes. As the follow-up neutralisation of Lake Hain was continued from 2010 by adding well hydrogen carbonate buffered water supplied by MIBRAG from the Profen opencast mine, the plant was no longer needed at this site and moved to Lake Zwenkau for further use.

Fig. 6: Upper left: Lake Hain stationary neutralisation plant and its floating unit for above water distribution (source: UIT/LMBV)

5.5 Microbial sulphate reduction in pit lake 111

Over a period of several years LMBV, in collaboration with the Dept. Lake Research / Magdeburg of the Helmholtz Centre for Environmental Research (UFZ), has performed systematic research and development work on **biological alkalinisation** at the small former mining pit lake 111 near Plessa ([26], [27]). Work was based on the model stoichiometric equation for the microbial sulphate reduction in the absence of oxygen

$$SO_4^{2-} + 2C_{org} + 2H_2O \rightarrow H_2S + 2HCO_3^-.$$

The H_2S formed hereby is precipitated iron-bound as FeS

$$H_2S + Fe^{2+} \rightarrow FeS\downarrow + 2H^+.$$

This stoichiometry suggests that sulphate-rich lake water, thanks to the relatively easily available organic carbon, is able produce iron sulphide and hydrogen carbonate when the iron concentration is sufficiently high, and thus to effectively reduce the acidity and sulphur content of sulphurous mining lake water in post mining lakes.

The process engineering relevance of the Pit 111 trials proved to be relatively marginal. The iron sulphide formed in the anoxic lake bottom was repeatedly re-oxidised by the addition of oxygen from the convective currents in the lake water. Forming acidity in the lake, irrespective of a protective cover of straw – is a process that was hardly relevant in the laboratory trial. Fundamentally, LMBV did not succeed in transferring the alkalinity from the anoxic layer on the lake floor into the lake water body without permitting the access of oxygen. There was also a lack of dissolved iron in the water of pit lake 111, because it had already been discharged from the lake water much earlier by the formation and sedimentation of iron hydroxide.

Not least as a result of the pit lake 111 tests, potential uses for biogenic alkalisation were rather identified in the meromictic deep-water zones of the perimeter trench drains of opencast mines. Being below the hypolimnion, they are not involved in the thermally driven lake water circulation. Groundwater rich in sulphate and iron often flows into these sections of lake water bodies. Particulate organic carbon (such as foliage, dung, and plant residues) sinking under the force of gravity can pass the epilimnion-to-hypolimnion boundary to finally reach the meromictic zone of the water body. The gases formed there (CO_2, H_2S, CH_4, etc.) can, after having formed bubbles, leave the reaction zone through the interface with the hypolimnion and thus return to the epilimnion and ultimately into the atmosphere.

5.6 Using Swedish liming techniques

The liming process of lakes has long been practised in those regions in Scandinavia that have been affected by acid rain. As concerns acidity, though, the Scandinavian soft water areas are far less acidic than the Lusatian post mining lakes (see [8, Chapter 7.2]). The surfaces of Scandinavian lakes are limed by helicopter or aeroplane, as well as by vessels that have been purpose-built for treating water bodies. One such Swedish water treatment vessel was used for the first time on an eastern German post-mining lake in 2008. The “*Brahe* type” vessel was charged with the neutralisation agent from a silo vehicle by hose. The dry-bunkered pulverised limestone on the vessel was mixed on board with a flow of lake water generated by the vessel’s pumping system. Then suspended in the pipeline towed by the vessel it had been distributed through nozzles. The lime-water suspension was spread across the lake surface by two water guns. Fig. 7 illustrates how this extensive distribution of neutralisation agent is typically carried out. 2008, though, only small quantities of pulverised limestone (37 t) were needed for Lake Haselbach to prevent reacidification of the already neutral water body [2]. In the following year, Lake Bernstein was the first post-mining lake to be neutralized using this type of water treatment vessel. 11 000 tonnes of pulverised limestone were used to reach pH = 5, which was followed by 1 000 tonnes of calcium hydroxide [5].

The advantage of these Swedish water treatment techniques is that they are easily transported by road on a trailer. From this they can be launched, and onto which they can be recovered from a lake with relatively ease. They are therefore very flexible and can be rapidly deployed in various post-mining lakes. On the other hand, one disadvantage was that the coarse-grained pulverised limestone being used acidic mine water was much less effective than in the soft water lakes of Scandinavia. The formation of basic (pH > 10), and therefore potentially hazardous, aerosols in the air above the lake surface when the slaked lime suspension is being atomised can be detrimental from a health perspective. Another ecologic drawback is the risk of the fine lime particles drifting into the reed beds along the lake shore as they are unable, or only slowly able, to pass the air-water boundary.

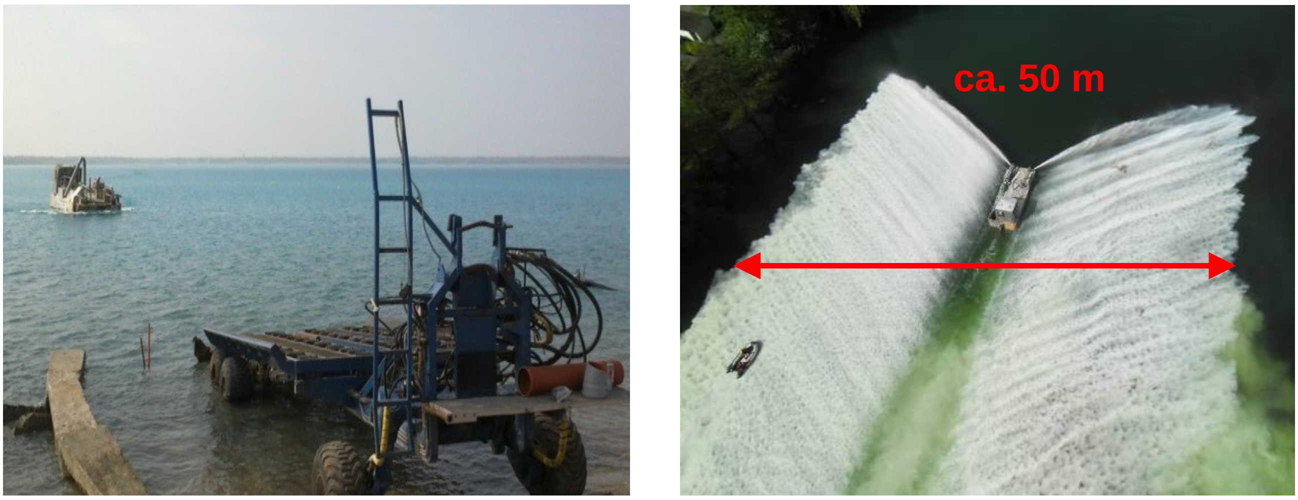


Fig. 7: **Left: *Brahe II* vessel and its trailer (source: LMBV), and right: spraying the lime suspension (source: [29])**

5.7 Interim conclusions on in-lake procedures

During the initial phase of in-lake-neutralisation over the first decade, LMBV has gathered a great deal of experience. It was confirmed at field scale that treating the water body in former mining pits with neutralisation agents can reduce the metals, iron and others, to ecologically and legal acceptable levels so that no further metal recovery processes need be carried out. Moreover, it can be expected that over a period of one to two years following initial neutralisation significant microbial ammonium decomposition will take place. These implemented neutralisation techniques have proven their ability to work economically.

In 2012/2013 it was therefore possible to state, as of an interim conclusion, that the chemical neutralisation of acidic post-mining lakes was state of the art [25, Chapter 5.9.2] (see also [8] and [21]).

With respect to the initial neutralisation measures still to be carried out, and particularly to the follow-up neutralisations that will be necessary for long periods as a consequence of mining, further needs for development have been identified, mainly with regard to

- **efficiency and cost,**
- **transferability to other sites** (infrastructural requirements), and
- **compatibility with intended uses**.

Two adverse processes affecting the efficiency of in-lake neutralisation must be reduced effectively.

They are firstly:

- the decreasing speed at which calcite dissolves as the pH increases, and secondly
- the likelihood of the neutralisation particles becoming inert because of $CaSO_4$ layers forming on their surfaces when there are high sulphate concentrations.

On the other hand, no efficient process engineering technology has yet been developed for the microbial sulphate reduction in post-mining lakes.

6 Advancements of in-lake technologies

Considerable efforts were made to enable development of in-lake technologies while rehabilitation of mining induced water budget in the recent years. LMBV carried out various pilot and demonstration projects for in-lake neutralisation using water treatment vessels and stationary installations. LMBV spearheaded the provision of engineering support to and the assessment of rehabilitation measures.

Many proposals for pilot and demonstration projects were developed by engineering firms. These were reviewed by the LMBV's scientific and technical advisory board for water management, an independent committee of recognised experts. The pilot and demonstration projects (see [20]) intend to test promising processes and transfer them to the remediation practice. This involves verification and up-scaling of the results of research entities. In order to assess their efficiency, sustainability, and transferability to other sites these projects are given scientific support and subjected to comprehensive monitoring.

The engineering of remediation measures normally involves two stages: an evaluation of the actual work including monitoring the measure, and the creation of a hydrological and geochemical modelling of the neutralisation measures following their completion. At the same time **efficiency assessments** are made. They show that the degree of efficiency of a neutralisation measure primarily depends on three factors:

- the quality of the lake water, and above all the pH value, ionic composition and sulphate concentration,
- the material properties of the neutralisation agent, above all its chemical composition, particle size, and inner surface,
- the implementation technology, above all the concentration of the suspension and the turbulence that is generated.

Although, because of the large number of influencing factors, it is impossible to assign each procedure an unambiguous degree of efficiency, general qualitative comparisons can certainly be made, and the results can then be taken into account for subsequent procedures.

There are various approaches to determining the **degree of efficiency** and therefore the quantified impact of a defined amount of neutralisation agent on the lake water to be neutralized. The focus is on considering two quite different approaches characterise efficiency:

- the stoichiometric analysis of a relevant lake water sample (Example 3), and

- the physiographic based assessment of the post-mining lake under review (Example 4).

Example 3:

Stoichiometric assessment of a sample

Input of neutralisation agent, e.g. CaO, to the lake water sample

Determining the decrease in acidity of the lake water sample
Reaction model equation: $CaO + H_2O \rightarrow Ca^{2+} + 2OH^- = Ca^{2+} + 2$ moles of alkalinity

- Here the efficiency η is 100 % if 1 kg CaO results in precisely 35.7 moles of alkalinity (the reaction product quantifying the efficiency being assessed).
- If there were, for example, only 25 moles of alkalinity, the efficiency would be $\eta = 25/35.7 \approx 0.70$ i.e. 70 %.
  (Basing the neutralisation efficiency on the quantity of OH^- would be counterproductive, because the OH^- ions formed enter subsequent reactions in the lake water sample so that they are no longer included in the pH value.)
- An assessment of the degree of efficiency must always be accompanied by a reflection on what has likely occurred to any CaO raw material that has not entered a reaction to form the target product, in this case the alkalinity.

The η value estimated through a stoichiometricciency assessment using the above reaction model equation is a function of

- the lake water quality, i.e. the lake water sample's pH, ionic strength, and sulphate concentration,
- the material properties of the neutralisation agent, i.e. its chemical composition, its granularity and the granules' inner surface, and
- the mixing energy delivered during testing in the laboratory.

A comparison and assessment of the η values thus derived is therefore not trivial.

Example 4:
Physiographic assessment of the water-body in a pit lake

Here, a balance sheet is produced over a set period (e.g. over the initial neutralisation period or over a single day) of all inputs of alkalinity/acidity together with the neutralisation agent and all other quantifiable inputs or outputs of alkalinity/acidity, and the change in the water's acidity inventory. Here, the η value as a rate of efficiency is the factor by which the stoichiometrically possible conversion of the neutralisation agent must be multiplied to settle the model balance.

Each of the elements entered in the balance sheet as a daily, monthly, or annual value therefore affects the resultant η value. Consequently, the η value so arrived at is primarily determined by the values entered and/or taken into account for the model balance equation, and their inherent errors.

This physiographic/balance-based assessment of the efficiency η must itself be supplemented with an assessment of any disregarded, or wrongly regarded, effects that should be assigned to the input of neutralisation agent. The efficiency rate as determined by this physiographic balance-taking is still quite imprecise if derived from just a single neutralisation campaign but will become more and more robust with an increasing data density and number of neutralisation cycles.

The impact of the actual input and mixing technology on the overall efficiency can only be determined by performing this form of balance-based efficiency calculation.

6.1 Vessel-based in-lake neutralisation

In the recent years, the mobile in-lake-neutralisation, water treatment vessels, rehabilitation vessels, or liming vessels, were consistently improved and adapted to the conditions prevalent in the post-mining lakes, whether acidic, or prone to re-acidification.

The Scandinavian ***Brahe* type** water treatment vessel that was already used during the initial neutralisation of Lake Bernstein and in the follow-up treatment of **Lake Haselbach** has been redesigned step by step to meet LMBV’s requirements. The first new feature was an underwater delivery system used in **Lake Bernstein** in 2013, to prevent the hazards to health and nature arising from the above-water input technique. This was accomplished by employing a system of pipes to divert the jets from the water guns on either side of the vessel vertically into the water. In shallow lakes a bend has been added to the piping ends so that the neutralisation agent is not injected directly into the sediment.

Fig. 8: The *Brahe II* water treatment vessel with underwater discharge (source: [33])

The relatively large number of inefficient empty trips that had to be made because of the *Brahe II*’s low bunker volume (11 m³) has been reduced since 2017, as the new *Brahe III* then introduced has a 15 m³ bunker.

To improve the somehow ineffective input technique of coarse grain pulverised limestone used for the initial neutralisation of Lake Bernstein a more finely grained limestone was used during the initial neutralisation of the surface water body of Lake Geierswalde. Although this grade of pulverised limestone is actually more expensive and has a lower bulk density, it proved to be sufficiently effective even with pH values in the neutral range if added to the lake using the underwater input system [9, 11].

A vessel of the ***Puma* type** was fitted out for in-lake-neutralisation with above-water application based on the *Brahe*'s technology. It possesses a greater bunker capacity (30 m³) than the *Brahe II* class. Already on the lake shore is the application substance being pre-suspended to approximately 30 % by mass using lake water, and then filled into the vessel's bunker. On the lake this suspension is further diluted to 3 % with additional water drawn from the lake and sprayed across the lake surface. Because of the greater volume of the suspension, and the resultant reduced rate of delivery (252 m³/h), the spraying time and the distances travelled are greater. On the other hand, the surface area to which the agent is delivered during one trip is also greater, and the agent application per unit area (in this case 0.12 kg/m² of calcium hydroxide) is lower than in the case of the *Brahe II* class. [7].

So far though, the *Puma* has only been deployed once, in 2012. Because the LMBV's requirements had changed, and given the experience gathered during the first order, the discharge approach was completely revised. As in the old vessel, the new vessel carries 30 m³ of suspension prepared in advance in its bunkers. This suspension can be prepared either directly on board or in a separate, shore-based mixing container. It can then be diluted to 3 % by mass in a second stage of the process and injected into the water through nozzles according to the turbulent-jet principle. With the originally used above-water input by sprinkling there was no horizontal spreading momentum, which is much more effectively achieved with the new, submerged high-pressure turbulent-jet input technique.

Fig. 9: Pre-mixing container and *Puma* water treatment vessel using the submerged turbulent-jet input technique (source: BGD)

The effectiveness of the modified *Puma* vessel showed itself to be improved, presumably due to the extreme dilution and the horizontal spread caused by application of the turbulent-jet principle. The ability to load the neutralisation agent already in a suspended state has the advantage that suspensions can be transported through hoses faster and over greater distances than can a pulverised neutralisation agent, so that the vessel's down-times are reduced. [11]. Negative, however, is the fact that the use of caustic lime must be restricted because of the heat developing while a suspension is made inside the vessel. There is a definite need here for further engineering progress.

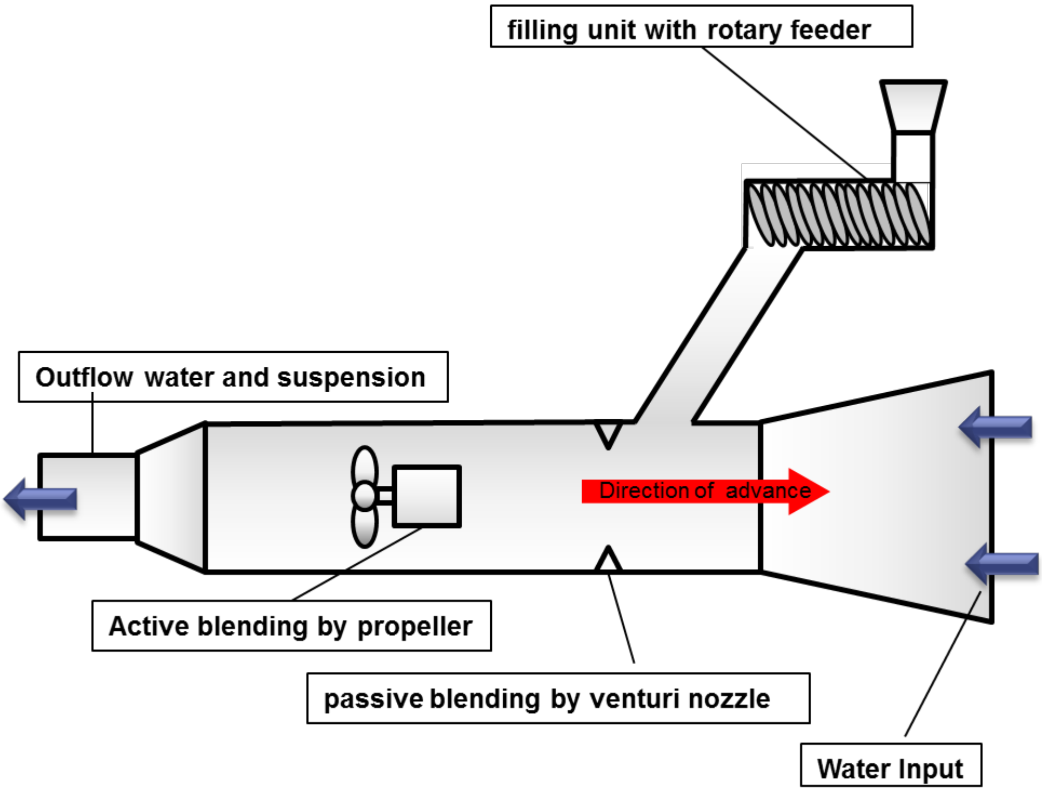


Fig. 10: Under water input system (modified pursuant to letter patent DE 10 2010 019 510)

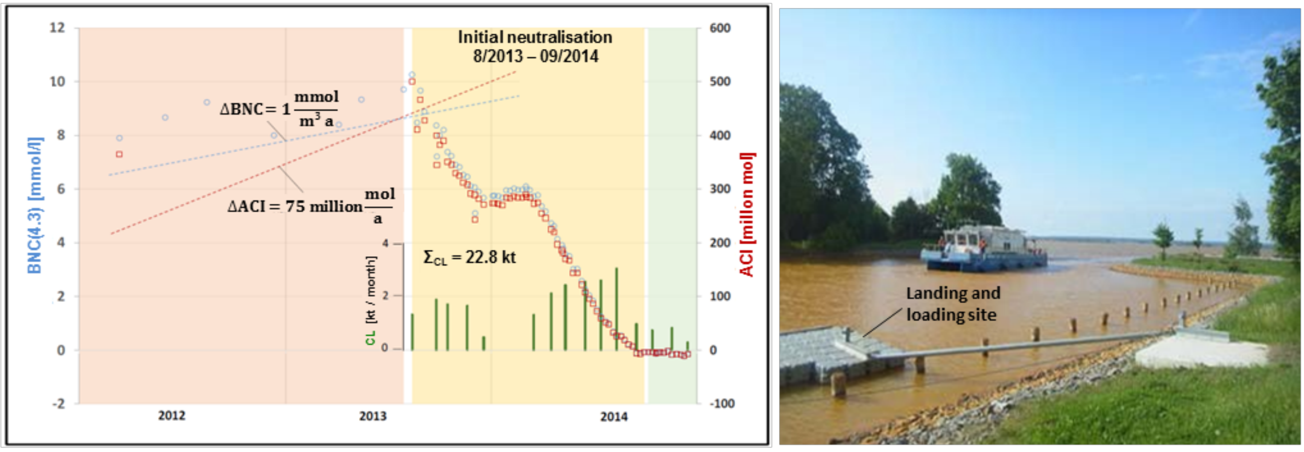
Simultaneously with the continued redesign of the above-water distribution vessels, a new input system tailored to conditions in the acidic opencast mining lakes has been developed. It is based on **dry liming** using a submerged tube mixer as technical reactor and highly turbulent delivery of the suspension (Fig. 10).This research and development has been funded by the state of Brandenburg and the European Union. Together with two Lusatian firms LMBV has filed a patent application (DE 10 2010 019 510) for this input system.In this context, one of the engineering companies in Lusatia contributing to the patent built the **water treatment vessel *Barbara*** (Fig. 11).

Fig. 11: Water treatment vessel *Barbara* in operation, and (bottom right) being lowered – showing the delivery system between the vessel's hulls (source: LUG/LMBV)

The vessel is a catamaran with two tube mixers mounted between the hulls. This permits additional mixing by the vessel's two ship propellers at the ends of the twin hulls. The vessel has two lime bunkers, each with a capacity of 12 m³, allowing material to be bunkered according to the bulk density of the lime. It has been approved for use on navigable waters (pursuant to the state shipping ordinance/LSchiffV).

LMBV tested the water treatment vessel *Barbara* on **Lake Lichtenau** within a pilot and demonstration project [10]. Positive aspects were above all the good handling of the vessel and good effectiveness with slaked lime and pulverised limestone. Because of the high throughput of water, the percentage of solid matter in the suspension, 0.6 % by mass for pulverised limestone and approx. 0.2 % by mass for calcium hydroxide, was significantly lower than that of the *Brahe* and *Puma* vessels' technology. Although the material is spread over a width of than some 12 m, the quantities needed per unit area, 0.07 kg calcium hydroxide per square metre, were relatively low [11]. The subsurface application of the agent resulted in rapid intermixing with a greater reaction volume of lake water.

Example 5:
The initial neutralisation of Lake Schlabendorf [13] [17]



Initial neutralisation with caustic lime (CL) by the water treatment vessel *Barbara* (right) during Fe precipitation and flocculation (courtesy: IWB)

Initial values	pH = 2.8	BNC(4.3) = 10.2 mmol/l	V = 48.8 million m³
Final values	pH = 6.9	ANC(4.3) = 0.5 mmol/l	V = 46.0 million m³

The following neutralisation impact was achieved with the input of 22.8 kt of caustic lime to the water:

Acidity reduced by	≈ 500 million moles	618 million moles
Alkalinity raised by	≈ 23 million moles	
Acidification neutralised over 13 months	≈ 81 million moles	
Lake water infiltration	≈ 14 million moles	

i.e. approximately 76 % (=17 328 t) of the caustic lime discharged achieved the above impacts. 24 % (= 5 472 t) of the caustic lime had the primary effect of neutralising the lake floor and erosive material from wind wave and heavy precipitation erosions as well as slope movement (in particular of the underwater dump slope) or proved to be non-reactive.

The long loading down-times of approximately 80 minutes, which is longer than the actual application span of about 75 minutes, were one negative aspect. Altogether a complete cycle, including mooring and casting-off manoeuvres, took three hours. Neither was it possible to deliver caustic lime, as the conveyor screws were not powerful enough [10]. Because of LMBV's requirement to discharge caustic lime, this defect was rectified by fitting new motors and gearboxes to the conveyor screws. This allowed the initial neutralisation of **Lake Schlabendorf** to be carried out by the *Barbara* vessel almost exclusively with caustic lime (Example 5) [12].

Further development of particular relevance to LMBV is the **water treatment vessel *Klara*** (Fig. 12); it has been designed and built specifically for use in what is known as the Lausitzer Seenland mining lake chain.

Fig. 12: LMBV water treatment vessel *Klara* during operation (source: LMBV)

The Lausitzer Seenland mining lake chain in Saxony and Brandenburg (Fig. 13) is made up of nine lakes between Senftenberg and Spremberg that are connected by navigable canals. Although not included by default, Lake Senftenberg is nevertheless connected to the chain by the Koschen Canal. The water surface area of the entire lake chain is greater than 55 km², and it has a volume of more than 800 million m³.

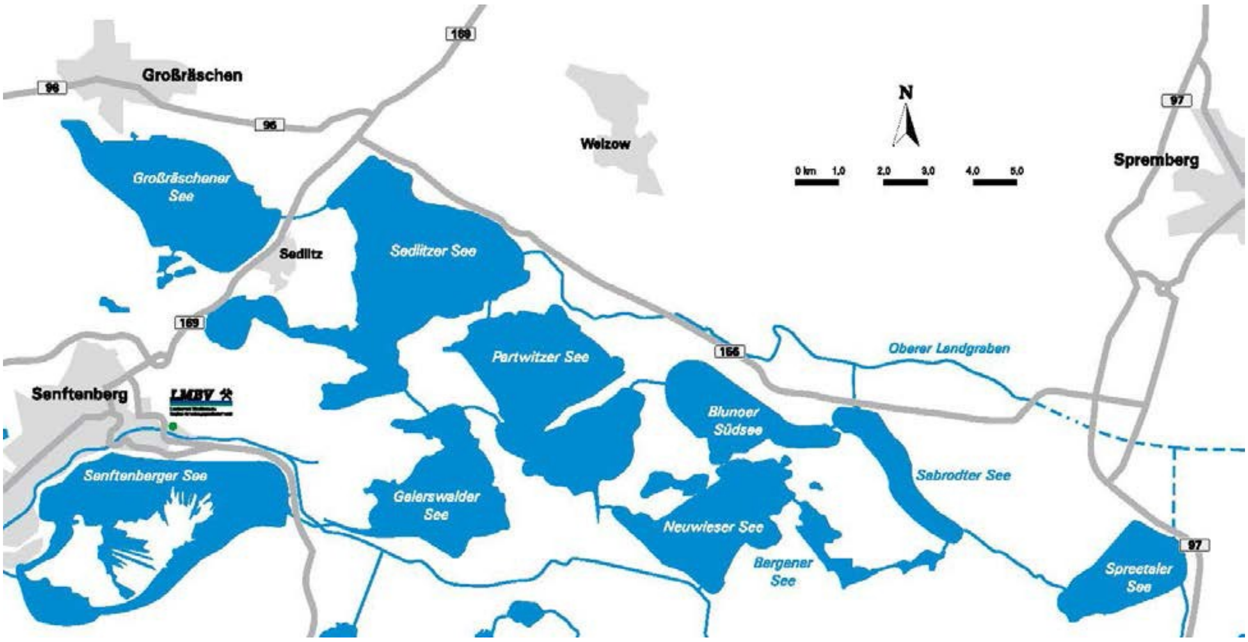


Fig. 13: Lausitzer Seenland mining lake chain and Lake Senftenberg – 10 interconnected post-mining lakes (source: LMBV)

The *Klara*'s dimensions were defined to pass through/under all the canals, bridges, and locks of the mining lake chain. It was built as a push boat with two barges (Fig. 14). The engine, generator, and the bridge are on the push boat. On the barge, there is the lime silo, and between the catamaran's twin hulls of the barges the discharge units have been mounted.

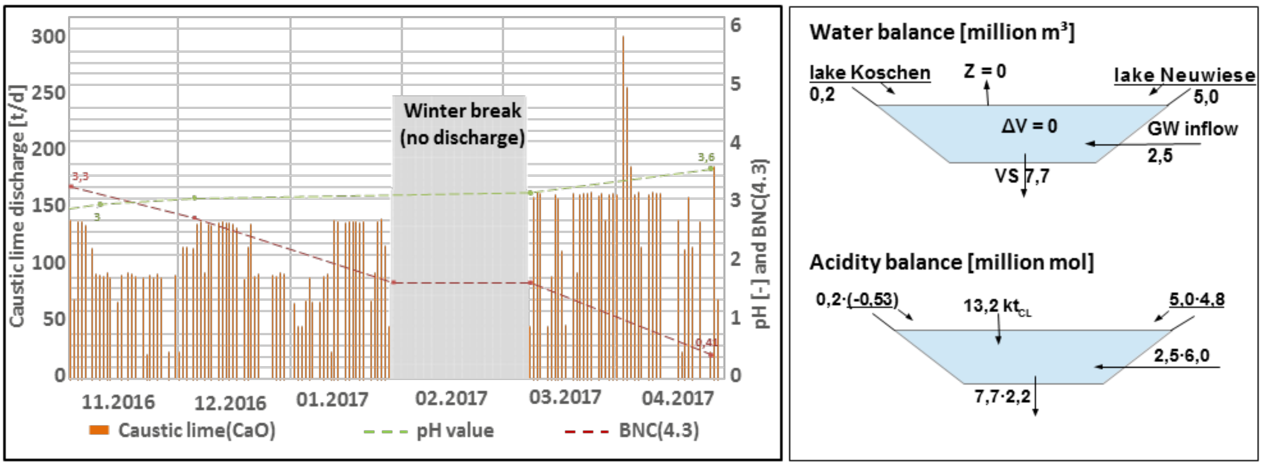
Fig. 14: LMBV water treatment vessel *Klara* coupled to Lighter 1 (right), and Lighter 2 (left) at the landing and loading site (source: [33])

This two-barge arrangement has the advantage, that the vessel does not have to tie up for the lime loading, but must merely exchange barges to continue its work. While the first barge is on the lake discharging neutralisation agent, the second can be loaded. It reduces the cycle time to between 50 and 60 minutes.

The discharge unit operates on the same principle as that of the water treatment vessel *Barbara*. It can use caustic lime, pulverised limestone, or slaked lime, and is able to deliver neutralisation very efficiently because of the high level of dilution in lake water, which is further boosted by the action of the vessel's propellers. The vessel is dimensioned for the discharge of approximately 40 000 t of neutralisation agent a year. However, initial experience shows that on some days the proposed capacity can be significantly exceeded. *Klara* is therefore able to carry out both the initial neutralisation as well as the follow-up treatment for the entire mining lake chain.

Example 6 describes the initial neutralisation of Lake Partwitz. The left-hand part of the example box illustrates the Klara discharging caustic lime during the hydrological winter half-year 01.11.2016 to 31.04.2017. Delivery had to be interrupted between 29.01.2017 and 07.03.2017 because the lake was covered with ice. This did not result in any severe discontinuity in the pH or BNC(4.3) values.

Example 6:
Initial neutralisation of Lake Partwitz



Initial neutralisation using caustic lime performed with the *Klara* water treatment vessel and alkalinity balance during the hydrological winter half year 2016-17 (November 2016 – April 2017)

Initial values: pH = 3.0 BNC(4.3) = 3.3 mmol/l V = 123.0 million m³

Final values: pH = 3.6 BNC(4.3) = 0.4 mmol/l V = 123.0 million m³

The following neutralisation impact was achieved with the input of 13 200 t of caustic lime to the water body during the hydrologic winter half year 2016-17:

Acidity reduced in lake Partwitz	357 million moles	413 million moles
Neutralisation of acidic ground water inflows	+15 million moles	
Neutralisation of surface water inflows from lake Neuwiese	+24 million moles	
Exfiltration of alkaline lake water	+17 million moles	

i.e. approximately 87 % (= 11 500 t) of the caustic lime discharged achieved the above effects and 13 % (= 1 700 t) of the caustic lime had the primary effect of neutralizing the lake sediment and erosive material from wind wave and heavy precipitation erosions as well as slope movement (in particular of the underwater dump slope) or proved to be non-reactive.

6.2 Stationary in-lake neutralisation plants

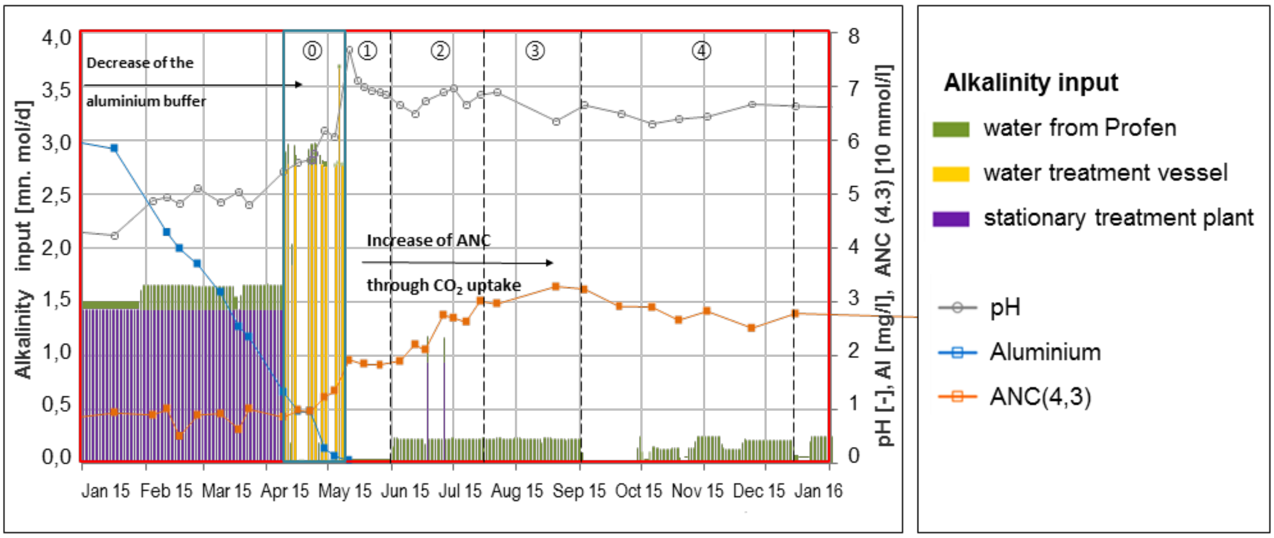
The Lake Hain stationary neutralisation plant was moved to **Lake Zwenkau** in 2011, whereby only the land-based section of the plant continued in use. There was no need for a special discharge system, as the lime suspension could be added to the flooding water. Here the feed to the lake functioned as a tube reactor in which the lime suspension was effectively mixed and diluted with the flooding water from the active Profen open-cast mine (Fig. 15)

This form of lime input to the lake exploits the kinetic energy already present in the flooding water to distribute the lime suspension across the adjacent area. Subsequently, the suspension current descends into the deep area of Lake Zwenkau. Further distribution in the lake is then caused by wind and density driven currents. [15].

Fig. 15: Stationary neutralisation plant on Lake Zwenkau and the lime plume spreading in the flooding water (source: LMBV)

Example 7 shows the development of the surface water quality in Lake Zwenkau in the context of its initial neutralisation, whereby the input of alkalinity was delivered both by the stationary neutralisation plant and, primarily, the hydrogen carbonate fraction of water from the active pit Profen, and in the final phase also by the *Brahe I* water treatment vessel.

Example 7:
Initial neutralisation of Lake Zwenkau [14] [15]



Initial neutralisation with stationary plant using caustic lime and water from Profen until 09.04.2015, followed by delivery of caustic lime from a water treatment vessel until 08.05.2015, and follow-up neutralisation with highly bufferd water from Profen containing an acid neutralisation capacity of ANC(4.3) = 4.5 to 5.0 mol/m³

The initial neutralisation of the lake was achieved by 35 million m³ of water from Profen with an alkalinity of 168 million moles, and 35 million m³ water from the river Weisse Elster (2013) with an alkalinity of 78 million moles, plus the input of 34 300 t of caustic lime.
The diagram above shows that the pH value during the initial neutralisation (phase ⓪) did not increase above pH = 5.5 until overcoming the aluminium buffer in the lake water (i.e. with an aluminium concentration of less than 0.5 mg/l). After completion of the caustic lime input by water treatment vessel (phase ①), the pH rapidly sank from pH 7.5 back to 6.5, but the ANC(4.3) showed a more or less constant value. Subsequently, the ANC rose as expected because of the addition of water from Profen well buffered with hydrogen carbonate, accompanied by a somewhat diminishing pH value (phases ② and ③), before the neutralisation state became largely stable without any lime input, as ≈ 35 000 m³/d water from Profen at pH ≥ 6 and ANC(4.3) ≈ 0.3 mmol/l was supplied (phase ④).

A LMBV pilot and demonstration project comprising another type of stationary plant with a subsurface input system, known as a ***GSD* plant,** was tested at Lake Scheibe (Fig 16). This plant’s special characteristic is that the lime suspension is made on land, and that its mixing into the lake water body makes use of the physical **principle of the turbulent jet** [31]. At first, water is pumped from the lake, followed by the addition of the neutralisation agent to the water volume flow and its suspension in a section of pipe. In contrast to the Lake Zwenkau plant there is no caustic lime slaking. The caustic lime suspension is then jetted into the sub-surface water of the lake by pumps.

Input to Lake Scheibe is at a speed of 6.9 m/s, at a depth of 0.5 m, the jet thereafter sinking deeper and becoming diluted until it reaches the layer of water with the same density. In the model calculation the jet became stratified at a horizontal throw of 13.5 m, above or in the thermocline. At the same time the suspension is diluted in the stratified jet by a factor in excess of 1:100. In the full circulation phase at a depth of 33 m below the water surface, the jet throw distance is 40 m until it stratifies at a dilution ratio of ≈ 1:400.

Fig. 16: *GSD* plant on Lake Scheibe with a submerged input system (close-up) and distribution of the lime suspension in the lake (source: GMB)

With suspension input of 256 m³/h, and a jet dilution factor of 150, a one metre deep strata volume of Lake Scheibe is built up in about one week. This strata glides onto the lake's thermocline (a density-driven horizontal intrusion [24]). Within several days, an uniform horizontal distribution across the lake follows as a result of the vertical-horizontal mass balancing. The distribution undergoes additional deformation and modification caused by the input of external, weather-related energy (wind, heat). The influence of wind on particle distribution in the epilimnion of the stratified Lake Scheibe was limited. The plant used was efficient and able to discharge each week more than 1 000 t of caustic lime into the lake [6].

In Lake **Ferdinandsteich**, a plant operating on a similar principle, but smaller-sized, has been deployed since 2016. The initial neutralisation and the first follow-up neutralisations were still accomplished using *Brahe II* water treatment vessels. However, the problem arose that the official outflow threshold requirement for dissolved iron (1 mg/l) and total iron (3 mg/l) was not consistently met. In the case of dissolved iron this was primarily due to the inflow of ferric groundwater, which was relatively high in relation to the small volume of the lake. Particularly when pH values were between 6 and 7, and temperatures were low, iron oxidation and its precipitation as ferrous hydroxide were too slow to meet the outflow threshold limit. The iron parameter is still more critical, because ferrous hydroxide that had already sedimented was resuspended under windy conditions because the lake is so shallow.

Based on the foregoing it was decided to replace periodic liming by water treatment vessel with a stationary plant which would be able to carry out the pulverised limestone or calcium hydroxide follow-up neutralisation, either constantly or at more frequent intervals, maintaining the lake water pH in the target range of $6.5 < pH < 7.5$. This pH range accelerates iron oxidation, and results in larger flakes being formed, which settle more rapidly, and are more difficult to resuspend.

6.3 Buffering post-mining lakes with CO_2

As previously described in Chapter 4, the use of pulverised limestone can result in a certain buffering effect. This is because part of the alkalinity delivered by the neutralizing agent does not result in an increase of the pH or the precipitation of metal hydroxides; it rather is retained in the water as dissolved hydrogen carbonate (HCO_3^-), which has a buffering effect during the re-acidification phase.

Buffering is quantified on the basis of the acid capacity determined by titration to the reference pH value of 4.3 (ANC(4.3)). While using caustic lime or calcium hydroxide and limiting the pH value to < 7.5 to 8.0 can usually only achieve an efficient acid neutralisation capacity of 0.2 to 0.3 mmol/l. But using pulverised limestone alkaline buffering can often be increased to 0.5 mmol/l. To achieve still greater buffering, CO_2 must be added to the water. This takes place both as a natural process from the atmosphere, and where applicable, from inflowing ground and surface water. If the HCO_3^- buffer must be built up rapidly e.g. to ANC(4,3) ≈ 1.0 mmol/l, it can also be achieved through the technological input of CO_2. To achieve the latter, LMBV has tested three different techniques in the framework of pilot and demonstration projects.

The first pilot project to **develop a hydrogen carbonate buffer** in **Lake Lichtenau** was launched in 2013, starting with a neutral pH following the successful completion of initial neutralisation in 2012, and two follow-up neutralisations. Over the period of some six months, during which the pilot and demonstration project was run, 1 750 tonnes of carbon dioxide and 2 100 tonnes of hydrated limestone were discharged to the water body. They caused an alkalinisation of the surface water of approximately 19 million Moles of alkalinity. The CO_2 gas – liquefied under pressure – was delivered by lorry into a tank (Fig. 17, left). Taken from there it was gasified in an evaporator, and piped via hoses to the **gas delivery frames** anchored to the lake floor (Fig. 17, right) [10].

There are micropores in the gas delivery frames that allow the gas to escape in very small bubbles, and to rise towards the surface of the water. The pressure applied keeps the bubbles so small that they largely dissolve in the lake water before reaching the surface. This gassing continues 24 hours a day.

Fig. 17: At left: Vertical tank being filled with liquefied CO_2, and the water treatment vessel (behind lorry) being loaded with calcium hydroxide; and right: gas delivery frame with micro pores before installation on the lake bed (source: LMBV)

Simultaneously to the gassing 2 100 tonnes of calcium hydroxide were discharged by the water treatment vessel *Barbara* during daytime. The following reaction was targeted:

$Ca(OH)_2 + CO_2 \rightarrow Ca^{2+} + OH^- + HCO_3^-$.

This resulted in raising the buffer capacity from ANC(4.3) = 0.15 to 1.1 mmol/l, meaning that a buffering 4 to 5 times greater than that without CO_2 was achieved.

Model calculations show that approximately 62 % of the carbon dioxide, which was dosed through the gas delivery frames, were found to remain in the lake as additionally dissolved carbonic acid. The loss of 38 % can be assigned to diffusive outgassing to the atmosphere (17 %), to the calcite precipitation (12 %), to the outflow of ground and surface water from the lake (5 %), and bubbling during the adjustment phase (5 %). Of these losses the calcite precipitation and the bubbling can be influenced. The diffusive outgassing and export via groundwater and surface water are, though, unavoidable [10].

A second example refers to **Lake Scheibe** where from June 2015 a **hydrogen carbonate buffer was developed using a *GSD* plant** [16]. After the stationary plant had been modified, the input of pulverised limestone and carbon dioxide began in the context of another LMBV pilot and demonstration project. In the alkalinisation with pulverised limestone half the alkalinity was delivered as a free base, and half already as hydrogen carbonate:

$CaCO_3 + H_2O \rightarrow Ca^{2+} + OH^- + HCO_3^-$.

When ground limestone is used, less CO_2 input then using calcium hydroxide or caustic lime is required to develop the alkaline buffer.

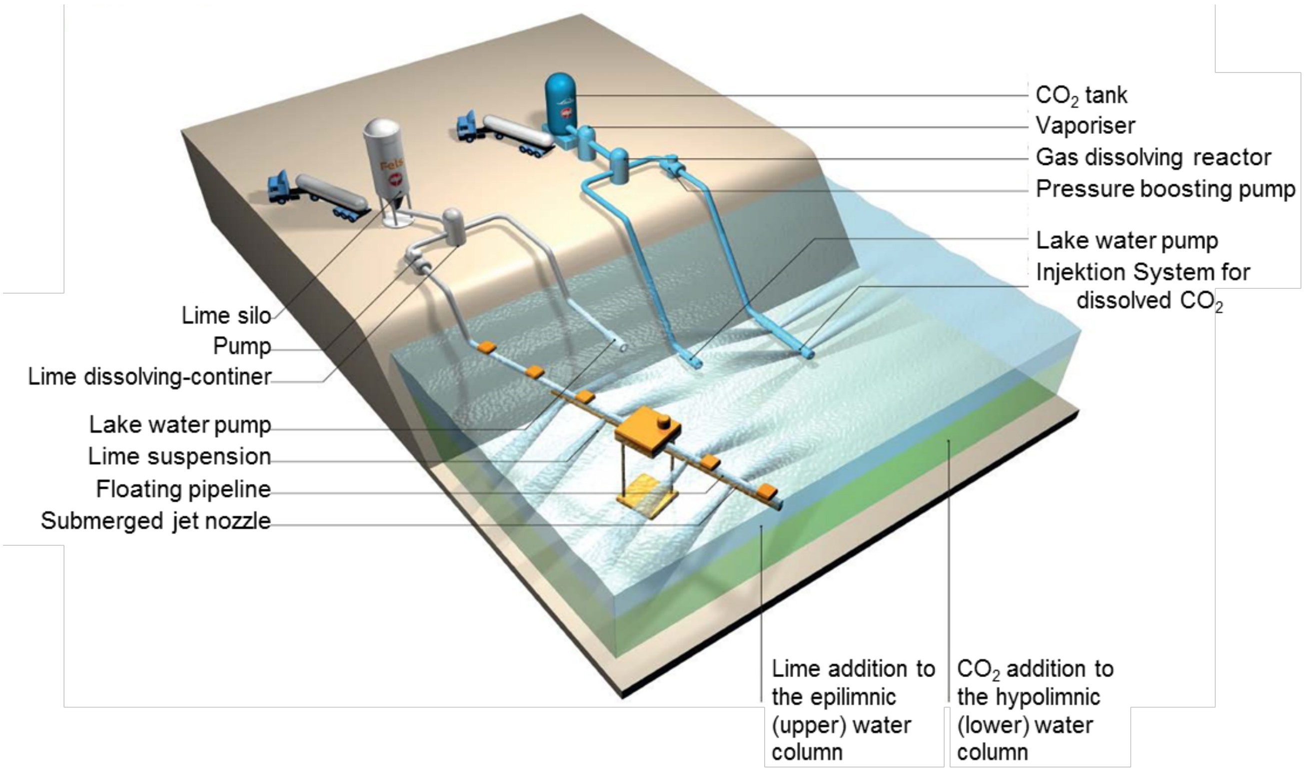


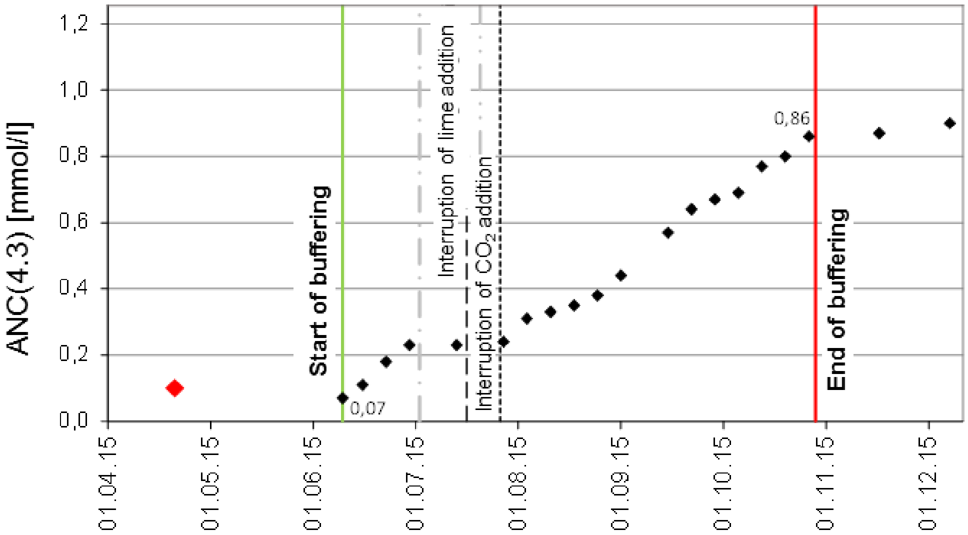
Fig. 18: Stationary neutralisation plant; left: for the input of pulverised limestone, and right: for the input of CO_2 (source: GMB)

Fig. 18 shows the neutralisation plant employed. The right-hand section of the plant delivers the CO_2 into the hypolimnion of the lake water after it has been dissolved in water drawn from the lake in the shore-based dissolved gas reactor. The left-hand section is in congruence with the plant that had been used for the initial neutralisation with caustic lime.

Fig. 19 shows the measured impact of the delivery of pulverised limestone and CO_2 during the pilot project. This project has been the basis of a discussion of the **potential risks** of gas input to the lake's waters. During the summer of 2015, at T = 7.7 °C and at a depth of 10 – 12 m, a CO_2 concentration of 3.7 g/l was dissolved into the lake water at the interface of hypolimnion and epilimnion [16], which according to pH value speciates $CO_{2(aq)}$, HCO_3^- and H_2CO_3.

The concentration of CO_2 in the intermixed epilimnion is therefore determined by the transfer of the gaseous CO_2 from rising gas bubbles as well as of dissolved CO_2 caused by transverse dispersion, the CO_2 used to form HCO_3^-, and the diffuse loss of CO_2 to the atmosphere via the lake surface. The latter can be expected when there is a concentration > 1 mg dissolved CO_2 per litre.[1]

The input of pulverised limestone to the epilimnion only makes sense, therefore, if the resultant concentration of dissolved CO_2 near the lake surface does not exceed 1 mg/l [Fig. 19]. Furthermore, the physiological impact of CO_2 on legally protected subjects such as humans (threshold limit value in the air above the lake) and aquatic fauna must be appropriately observed. Potentially at hazard are above all the zooplankton in the hypolimnion and the zoobenthos on the lake floor.



Input of 11 094 tonnes of pulverized limestone and 5 044 t of CO_2 with an alkalinisation effect of some 87 million moles alkalinity

Fig. 19: Results of the development of an alkaline buffer in Lake Scheibe using pulverized limestone and CO_2 (source: [16])

The third buffer-related pilot and demonstration project using CO_2 took place from 2014 to 2016 in Lake Drehna. There a closed technical reactor that generated the target product of hydrogen carbonate from lake water, caustic lime, or pulverised limestone and CO_2 gas, and delivered it to the water of Lake Drehna at a hydrogen carbonate concentration of 25 to 30 mol/m³ and a pH < 7, was built. Intermixing into the waterbody again relied on the turbulent jet technology. As the reactor operates with a high density of the reactants CO_2 and caustic lime or pulverised limestone in a fluidised bed, it was designated a **High Density Hydrogen Carbonate Reactor (HDHC Reactor)**.

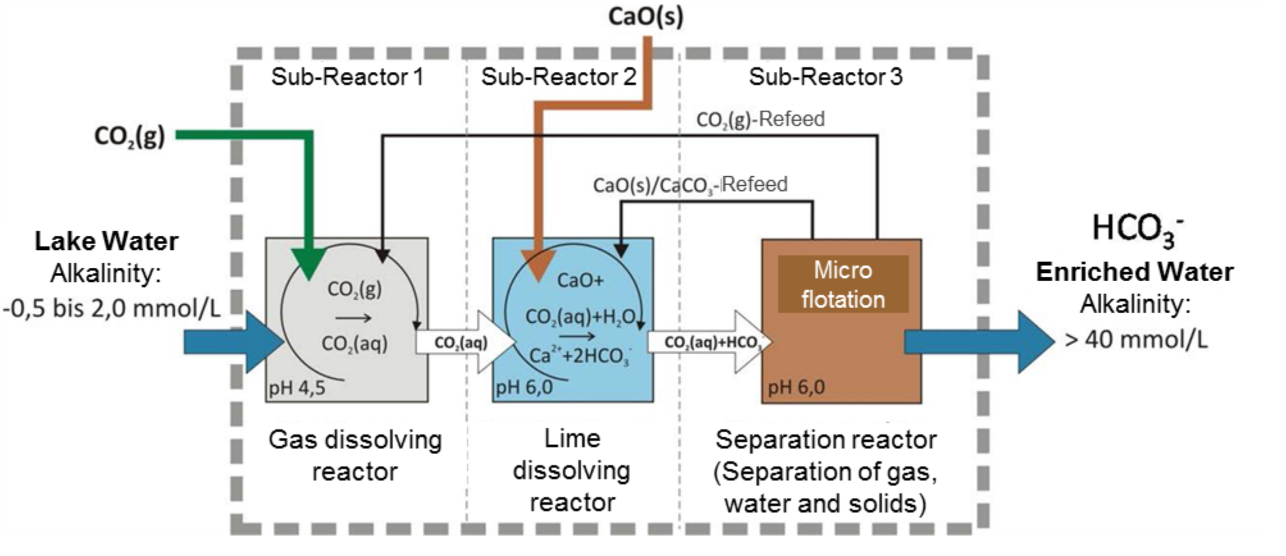


Fig. 20: Block diagram of operation of the High-Density Hydrogen Carbonate (HDHC) Reactor in the context of the 2014-2016 LMBV pilot and demonstration trials (source: GFI)

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[1] The saturation value of CO_2 versus the partial air pressure of 0.0314 percent by volume is equivalent to 5 °C 0.84 mg/l, and at 20 °C to 0.54 mg/l at the surface of the water. When using CO_2 gas (100 percent by volume) the saturation value is greater by a factor of 3000. This is the equivalent of a CO_2 concentration of 2.7 to 1.7 g/l at 5 to 20 °C on the water surface. In the hypolimnion at a depth of 10 m the value is still greater by approximately 1 g/l.

Figure 20 shows the HDHC reactor with its three sub-reactors for dissolving gas and solid matter, and the separation of the fluid into its components (i.e. into yet unconsumed CO_2 gas and the yet undissolved lime, both of which are recirculated to the relevant sub-reactors), and water enriched with dissolved hydrogen carbonate (HCO_3^-), that can be discharged to the lake via the underwater turbulent jet. The objective of the development of this reactor had been to enable high conversion rates by returning still undissolved CO_2 (gas) to the gas-dissolving sub-reactor and still undissolved lime into the solids sub-reactor inside the closed reactor, so avoiding degassing and sedimentation losses in the HDHc-reactor. Another objective was to discharge only the dissolved HCO_3^- into the lake water body to be neutralized and buffered, and to maintain the pH of this buffered discharge water in the region of pH ≈ 7.

Technical implementation was achieved by breaking the plant units down into onshore and offshore plant components.

In the context of this pilot and demonstration project, three trials with three intermediate re-acidification phases were carried out and assessed during 2014 to 2016. The following buffering was achieved in the lake water body:

1. using caustic lime and CO_2 12/2014 $K_{S4.3} = 0.7$ mmol/l at pH = 7.3,
2. using pulverised limestone and CO_2 12/2015 $K_{S4.3} = 0.9$ mmol/l at pH = 7.1
3. using caustic lime, pulverised limestone and CO_2 10/2016 $K_{S4.3} = 1.1$ mmol/l at pH = 7.13.

With an annual alkalinisation to ANC(4.3) ≥ 1 mmol/l it is expected that the follow-up cycle in the surface water body of Lake Drehna can be limited to three months alkalinisation and nine months re-acidification.

7 State of the art achieved

LMBV is able to carry out efficient and effective in-lake neutralisation measures using mobile water treatment vessels or stationary neutralisation plants. Therefore a number of tried and tested techniques are available. Which technology and which neutralisation agent is actually used depends on technological and economic considerations. Decisions are usually made on a case-by case basis.

7.1 Choice of neutralizing agent

With regard to the neutralizing agents most frequently tested so far, the following statements can be made. These are based on experience gathered over the past few years, regarding their efficient and effective use:

Caustic lime (pulverised calcium hydroxide):

- high neutralisation equivalent (approximately 30 to 35 mol/kg),
- reasonable price (e.g. a specific price of approximately 0.3 Euro cents per mol of alkalinity),
- high reactivity ensures a high kinetic rate of reaction, but can also cause locally high pH values in lake water

Calcium hydroxide, main ingredient: ground $Ca(OH)_2$

- neutralisation equivalent of approximately Neq = 25 to 29 mol/kg,
- relatively high specific price due to the additional process steps involved in its production,

- the availability of prepared lime water suspensions is an advantage for small amounts needed

Pulverised limestone, consisting of ground $CaCO_3$ (limestone or chalk)

- relatively low neutralisation equivalent of Neq ≤ 20 mol/kg,
- efficient use, it needs to be finely pulverised (e.g. d90 < 40 µm),
- low specific costs, approximately 0.3 Euro cent per mole of alkalinity,
- ability to reach higher buffering than using caustic lime, developing hydrogen carbonate buffer up to ANC(4.3) ≈ 0.5 mmol/l,
- ecological advantages due to the fact that over neutralisation to pH-values of more than 8 are not possible.

Soda, main component: ground Na_2CO_3

- easily soluble in all pH ranges,
- does not require any complicated technology,
- comparatively expensive, and
- ecologically not ideal as it increases salinity (i.e. elevated sodium level).

Although the specific prices per mole of alkalinity of caustic lime and of pulverised limestone are very similar, caustic lime is usually the more cost-effective option, as significantly more mass (i.e. tonnages) of pulverised limestone must be discharged to achieve the same neutralizing effect. The input costs thus are inevitably higher for pulverised limestone than they are for caustic lime. Caustic lime would be preferred wherever neutralisation does not require alkaline buffering.
The focus of initial neutralisation will therefore be on the use of caustic lime, while follow-up neutralisation of post-mining lakes will focus on pulverised limestone.

Figure 21 illustrates how increased knowledge and the evolution of the lakes are reflected by changing trends regarding the selection of neutralizing agents. In coming years, when the initial neutralisation has been completed and the lakes are being put to touristic use for the greater part of the year, this proportion will be shifting still further towards finely pulverized limestone. In certain circumstances the use of chalk products as a specific form of pulverised limestone may become increasingly relevant.

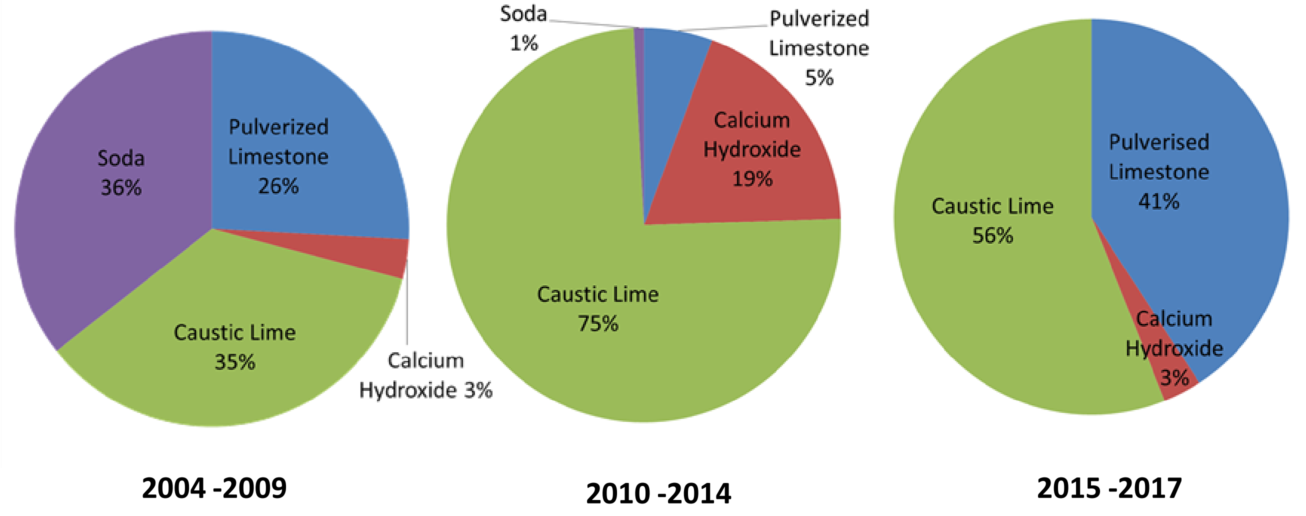

Fig. 21: **Alkalinity-related proportions of neutralizing agents used by LMBV for in-lake neutralisation over three different periods**

Use of gaseous CO_2

The use of CO_2 to develop a **hydrogen carbonate buffer** decreases the number of necessary follow-up treatments significantly. Although this can reduce plant costs, the expenses for the materials and agents used increase, as the costs of CO_2 which are currently some 120 €/t must be included. Neither should it be forgotten that CO_2, in the scope of the pH ≈ 7 reference point (and thus the target state of follow-up neutralisation), is an acidity reserve (as is e.g. iron at pH ≈ 3) and that this acidity must also be neutralized if the carbonate buffer is to be established at pH = 7. The size of this "latent" acidity requiring neutralisation is a function of the pH-dependent speciation of the total inorganic carbon (TIC) [23, p. 40].

The use of CO_2 will remain of interest in the future, especially to prevent the pH value from sinking during the winter if the lakes freeze over. This had been the case at the Lake Drehna pilot project. Here classic liming technology requires eight liming campaigns with caustic lime per year, and a pH-drop during the ice cover could not be avoided for sure. Using CO_2, a three-month campaign is sufficient to keep the pH-value stable for more than a year. The Lake Drehna pilot project is now being continued as a standard operation. From an economic point of view the pilot plant was at a par with classic liming due to the strong acid inflow but has the advantage of avoiding a pH-Drop under ice cover.

Buffering with CO_2 can likewise be economic where acidity flow into the lake is small. Then, with a single treatment a neutral pH value can be ensured for some years.

Generally, it can be stated that hydrogen carbonate buffering is a relevant follow-up treatment as especially from the ecological point of view. It avoids the rapid neutralisation during the treatment phase, and slower re-acidification during intervals without treatment. The pH fluctuations from $5 < pH < 9$ is assessed to be disadvantageous for the lake ecosystem.
If the cost of CO_2 would drop significantly it is also conceivable that over the long term a vessel-based hydrogen carbonate reactor could be installed on the barges of the *Klara* water treatment vessel.

7.2 Choice of in-lake procedure

Technically, not all treatment procedures are equally suitable for all types of lakes. Lake shore-based stationary water treatment plants would appear to be the preferable solution when continuous treatment cycles are required. They can be automated to a large extent so they can be both operated and monitored remotely. The lime should be introduced into the lake water body at an appropriate distance from the shore using pipes and subsurface turbulent jet technology. The required lime suspension is produced with lake water on the shore. Stationary plants of this kind are well-suited for relatively compact water bodies where good mixing as a result of the convective lake water currents can be expected.

Some German mining lakes are heavily segmented and made up of several sub-basins. Here it is more likely that mobile plants (i.e. water treatment vessels) are capable of distributing the neutralisation agent across the lake as required. The water treatment vessels are usually loaded just-in-time directly from silo lorries.

A relevant example in this context is LMBV's water treatment vessel *Klara*. It was custom designed and built for the initial and follow-up neutralisation of the Lausitzer Seenland mining lake chain in

Saxony and Brandenburg. The *Klara* is the most powerful water treatment vessel built to date; it can efficiently discharge 150 to 300 t neutralisation agent per day. Thanks to the two lighters that are loaded alternately the vessel has hardly any down-times while loading, which is another plus in efficiency. Because there are navigable canals between the water bodies of the mining lake chain, the vessel need not be hauled across land.

Other large water treatment vessels such as the *Barbara* and the *Puma* are most suitable for, post-mining lakes with relatively high demands for neutralisation agent. They have proved their usefulness and require fewer empty trips because of their bunker capacities. Hauling these large water treatment vessels is, though, complicated. They also require suitable surfaces for setting up cranes at their sites of deployment. Small water treatment vessels such as the *Brahe* type can easily be launched into the Water without a crane and can be moved by road without requiring a heavy-load transport licence. Small water treatment vessels are an attractive choice when there are extended periods of time between in-lake treatments. In the interim the vessel can be moved by road from one lake to another for further deployment.

7.3 Estimated cost of in-lake neutralisation

The cost of in–lake neutralisation depends on a number of factors. The most important one, without doubt, is the neutralisation agent, which accounts for about 60 % of the total cost. For initial neutralisation, the amount required is mainly determined by the acidity of the lake water and the water body volume. In follow-up treatment the acid influent from the mining affected groundwater, as well as the lake’s own internal acid formation processes are the dominant factors. In addition to the alkalinity required, which must usually be taken for granted, the cost is dependent on the type of neutralisation agent selected (see Chapter 7.1) and the effectiveness of the process.

In addition, infrastructure costs such as those for preparing the foundation for setting up a stationary plant and/or the water treatment vessel launching and loading points must be taken into account. Although there may be instances where such costs are very high, as a rule, they are only incurred once.

Increased efficiency and competition have sharply reduced the specific treatment costs. Whereas the first stationary in-lake neutralisation of Lake Bockwitz in 2004/2005 cost 1 euro cent per mole, the initial Lake Scheibe neutralisation was accomplished at not more than 0.8 ct/mol in 2012. The picture is similar for the water treatment vessels. While the initial neutralisation of Lake Bernstein in 2009 still ensued specific costs of 1.2 ct/mol, the initial Lake Schlabendorf neutralisation only came to some 0.7 ct/mol in 2014/15.

Generally, the following average specific costs for the in-lake-neutralisation of post-mining lakes can be quoted at present:

Initial neutralisation using caustic lime for pH < 5

• with stationary plant	0.6 cent/mole
• with large water treatment vessels	0.7 cent/mole
• with small water treatment vessels	0.8 cent/mole

Follow-up neutralisation using pulverised limestone for pH > 5

• with stationary plant	1.0 cent/mole

• with large water treatment vessels	1.2 cent/mole
• with small water treatment vessels	1.35 cent/mole

Due to the peculiar characteristics of each individual case these figures display a relatively large spread. The specific costs fall with the quantity to be discharged during each in-lake campaign, as engineering-related one-time costs, such as the transport to the lake, are then of less bearing.

8 Prospects

In the next few years, LMBV has the obligation to implement the follow-up treatment of four post-mining lakes in the Central German coalfield: Lake Zwenkau, Lake Störmthal, Lake Hain, and Lake Haselbach. The focus here is on using water from the White Elster river in Lake Zwenkau, de-watering effluent from an active mine pit in Lake Haselbach, and lime in both Lake Störmthal and Lake Hain, and in Lake Zwenkau if necessary.

A total of 11 lakes have been neutralized in the Lusatian coalfield, so far. They will be followed by a further five lakes of the Lausitzer Seenland mining lake chain presumably by 2022. There are three other lakes where the need – or otherwise – for neutralisation will be clear after the completion of studies, which are currently in progress.

In East Saxony the focus is on the initial and follow-up neutralisations of the yet untreated post-mining lakes of the Saxon part of the mining lake chain, followed by presumably temporary follow-up neutralisations of Lake Bernstein, the Lohsa II reservoir, and Lake Scheibe.

In addition to the mining lake chain in Brandenburg, the focus is on the Seese/Schlabendorf region where the follow-up neutralisation of Lake Drehna and of Lake Schlabendorf have priority. In the Lauchhammer area, where abandoned mines are a prominent feature, and which alone counts more than 30 – mostly small – post-mining lakes, regular in-lake liming would be too expensive. Consequently, taking recourse to the principle of outflow neutralisation, a water treatment plant is being built at the area's main outflow near Plessa. Whether the area's largest lake, Lake Bergheide requires in-lake neutralisation is now being reviewed. Outflow neutralisation by the Pößnitz mine water treatment plant is also in operation for the Meuro-Süd area.

Prognoses for the follow-up treatment of the various lakes differ considerably in quality. Estimates of the quantities of neutralisation agent required for those lakes which have already undergone several follow-up cycles are in general fairly reliable. For lakes that are still acidic, the estimates remain to be relatively uncertain. The assumption is that re-acidification in the lakes will gradually slow down, and that the lakes' own internal alkalinity production will gradually increase. A prognosis of the time span over which follow-up neutralisations will be necessary cannot be made at present. The assumption is that such measures will be needed for some decades yet.

The foregoing shows that in-lake neutralisation of post mining lakes first created and thereafter to be maintained in the former opencast lignite mining regions has resulted in comprehensive, innovative and cost-relevant new developments and advances. LMBV has itself driven key developments of the state of the art, particularly within the framework of the LMBV-funded pilot and demonstration projects. Today, as a result, there are various stationary and mobile technologies available for initial and follow-up neutralisation of the post mining lakes that are to be created and maintained. In this context [33] states:

“The in-lake neutralisation tool box is well filled. LMBV has at its disposal the appropriate tools and service providers needed to implement the impending measures, which must continuously be enhanced and extended.”

The knowledge gained is, of course, equally relevant for in-lake neutralisation at home and abroad outside the LMBVs scope of responsibility.

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