Carbon Footprinting of Mine Water Treatment for Future CO_2 Emission Reduction

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Abstract

This study provides an in-depth examination of the carbon footprint associated with neutralization measures implemented in post-mining landscapes, focusing on the activities of the Lausitzer und Mitteldeutsche Bergbau-Verwaltungsgesellschaft (LMBV). The research quantifies CO_2 emissions from the production, transport, and application of neutralization agents, as well as from different application methods. It also investigates and evaluates possible strategies for reduction of the carbon footprint of these essential remediation activities. The findings reveal relevant CO_2 emissions associated with neutralization measures and highlight opportunities for emission reduction through alternative technologies and management practices.

Keywords: Quick lime, chalk, hydrated lime, in-lake treatment, climate neutrality, outgassing

Introduction

Climate change poses one of the biggest global challenges of the century. To mitigate its effects, Germany and the European Union aim to achieve climate-neutrality by 2045 and 2050, respectively. This goal necessitates a drastic reduction in carbon emissions from mining rehabilitation activities, including mine water treatment.

Post-mining landscape rehabilitation is crucial for environmental restoration and sustainable land use. However, the neutralization measures employed to mitigate acidic mine waters contribute to greenhouse gas emissions, particularly CO_2.

Lime products, primarily limestone or chalk powder ($CaCO_3$), quicklime (CaO) and hydrated lime ($Ca(OH)_2$), are used for the majority of water treatment. CO_2 is released in various processes along the path of the lime products from their place of extraction to their place of use in the post-mining landscape. An overview of the CO_2 emitting processes is shown in Figure 1.

The aim of this study is to quantify and evaluate the carbon footprint of the neutralization measures carried out by the LMBV in the Lusatian and Central German mining districts. The objectives of this study are

- to calculate the carbon emissions associated with the production, transport and application of neutralization agents,
- to determine the CO_2 footprint of in-lake treatments compared to mine water treatment plants,
- to compare the CO_2-intensive application of lime products with the use of neutral buffered river water in in terms of carbon footprint,
- to investigate possible strategies to reduce the carbon footprint of neutralization measures.

Methods

Data on neutralization measures was collected from LMBV records for the period 2015-2022. This included qualities and quantities of various neutralization products used in in-lake treatments and water treatment plants.

CO_2 emissions were calculated stoichiometrically, for four main processes using complementary literature data:

1. Production: Carbon emissions from the manufacturing process of lime-based

products were estimated using industry-standard values (Franck & Knop 1979, Guminski *et al.* 2019, Kölling & Schnur 1977).

2. Transport: CO_2 emissions from transporting neutralization agents to application sites were calculated based on average transport distances and vehicle emission factors. (Süßmann & Lienkamp 2015)
3. Application: Emissions from the application process, including energy consumption for in-lake treatments and water treatment plants, were determined using operational data (LMBV 2021) and electricity emission factors.
4. Outgassing: When calcite-based neutralizing agents are added to a water body, the calcite dissolves. Due to a shift in the carbonate equilibrium, this leads to a proportional outgassing of CO_2. This was calculated stoichiometrically, taking into account the reactivity of the neutralizing agent.

Results and Discussion

From 2015 to 2022, LMBV utilized 280,000 t of lime-based neutralization agents, with 77% for in-lake neutralization and 23% for water treatment plants. This resulted in 188,000 t of carbon emission, 63% from in- lake treatment and 37% from conventional water treatment plants (Tab. 1).

In relation to the tonnages used in each case, however, less CO_2 was released by in-lake neutralization than during water treatment in stationary plants. This is due to various

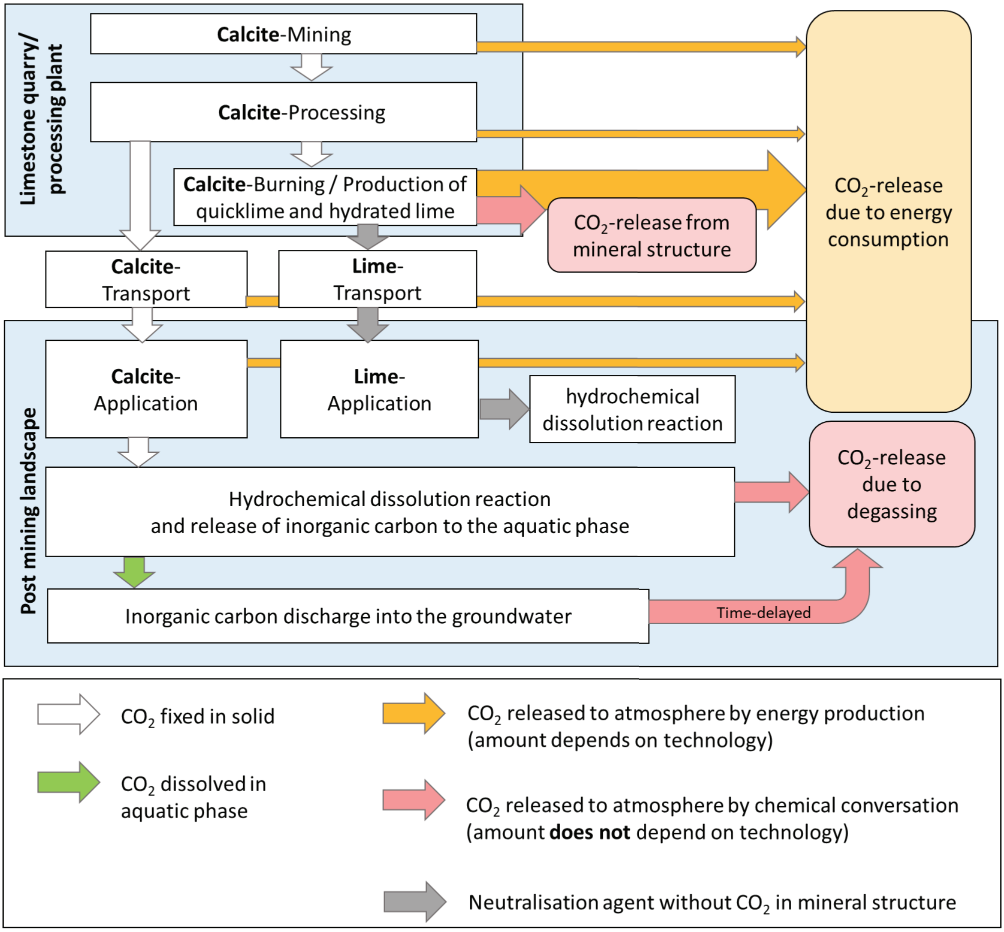

Figure 1 *Flow diagram for CO_2 emitting processes in mine water treatment.*

Table 1 Overall water treatment carbon footprint in the period of 2015 to 2022.

	Production	**Transport**	**Application**	**Outgassing**	**Total**
	t	**t**	**t**	**t**	**t**
in-lake treatment	82,212	2,085	1,280	33,518	119,095
water treatment plant	58,112	376	9,372	1,233	69,093
Total	140,324	2,461	10,652	34,751	**188,188**

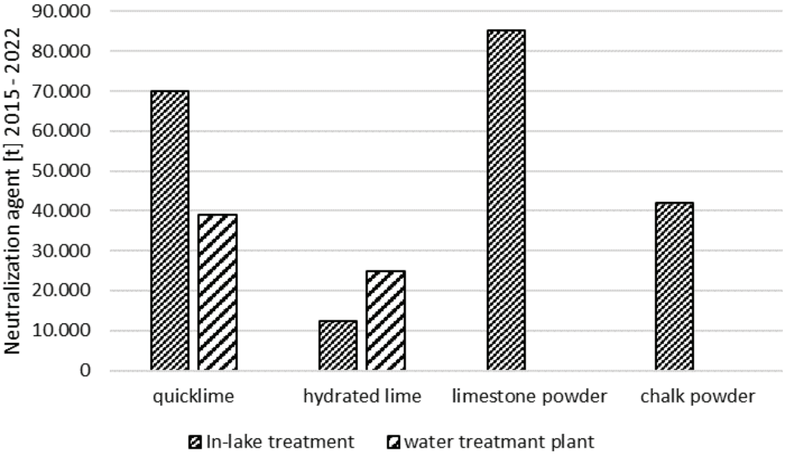

Figure 1 Quantities of neutralization agents utilized by LMBV for in-lake treatments and water treatment plants in the period of 2015 to 2022.

factors. The most important factor here are the differences in the main neutralizing agent used (Fig. 2). While all four neutralization agents are utilized in the in-lake treatment, with a focus on limestone and chalk powder, only quicklime and hydrated lime are used in the water treatment plants due to their better reactivity.

Neutralization Agent Production

The production of quicklime and hydrated lime is much more CO_2-intensive than limestone powder and chalk, with approximately 95% of the total carbon release being attributable to the burning process. The emissions from this sub-step result from the required heat energy (≈ 30%) and the amount of CO_2 separated from the calcite mineral phase (≈ 70%) (BVK 2017).

Transportation

Compared to the production and chemical solution of the neutralization agents in the treated water, the transportation and application of the neutralization agents only release small amounts of CO_2 and therefore offer little potential for savings (Tab. 1). However, the product-specific and absolute shares of transportation in the overall carbon footprint depend on the actual location of the quarries where from the raw materials originate.

Application

The amounts of energy required for discharge into the water (distribution, mixing) were calculated on the basis of ship operating times, fuel consumption and discharge quantities in the case of ship-based in-lake treatments and on the basis of electricity costs and electric power consumption in the case of stationary water treatment plants. Overall, the input of neutralization agents by means of in-lake technology is more favourable in terms of carbon emissions than the input via water treatment plants (Tab. 1).

Outgassing

When limestone and chalk powder are added to a water body, the calcitic part of the solid phase partially dissolves in the treated water, which initially produces carbonic acid under acidic respectively hydrogen carbonate under neutral conditions. Depending on the saturation state of the water in relation to the atmosphere, there is a partial outgassing of CO_2 (Cole & Caraco, 1998; Wetzel, 2001), which is in equilibrium with the aquatic species mentioned. Especially when the pH value decreases, typically in periods without in-lake treatment, there is an increased emission-relevant outgassing of CO_2 into the atmosphere. This process is more intense in fall, when the lakes transition from the stagnation phase into the circulation phase. This is because the increased inorganic carbon species concentrations of the hypolimnion, which have accumulated during summer, approach a steady state with the atmosphere. In spring, on the other hand, neutral lakes tend to incorporate carbonate species into biomass due to the increased biomass growth.

As a result of the calcite solution, a total of 34,750 tons of CO_2 have been released between 2015 and 2022 (Tab. 1), albeit with a time delay in some cases. Compared to limestone and chalk powder, quicklime and hydrated lime release substantially less carbon during application, as the CO_2 has already been released during the production process. Only the very low residual carbon content of the essentially carbon free solids is taken into account for the CO_2-balance.

Exemplary calculation

An examplary calculation for Lake Schlabendorf shows that over 50% more carbon is released when quicklime is used compared to limestone powder (Tab. 2, line 1 and 2). In general, it can be stated that the use of burnt lime products leads to significantly higher carbon emissions than the use of unburnt ones. This is due to the high-energy input for the firing process, but not due to the CO_2 released during firing, as this carbon is also largely released during the dissolution in case of application of unburnt limestone.

Another way of reducing the carbon footprint is to limit water treatment to the outlet of a post-mining lake instead of treating the entire lake. Treatment of the outlet is necessary to avoid harmful effects of the acidic water on the biocenosis in the downstream watercourse.

Comparative calculations were carried out for Lake Schlabendorf. This was based on an average discharge of lake water into the Lorenzgraben of 3.8 m^3/a with a maximum acidity of 10 mol/m^3, which the lake had before its first neutralization in 2013. If an outlet treatment had been installed instead of the initial neutralization, only about half of the acidity would have had to be treated

***Table 2** Comparison of CO_2 emissions with different treatment options of Lake Schlabendorf.*

	Neutralizing Agent	Needed Alkalinity	Input Quantity	Molar Equivalent	Efficiency Factor η	Specific CO_2 release	Total CO_2 release
		Mmol/a	t/a	mol/t	-	kg(CO_2)/t	t(CO_2)/a
in-lake treatment	limestone powder	77.7	8,443	20,000	0,46	258	2,179
in-lake treatment	quick lime	77.7	3,108	35,714	0,70	1083	3,366
outflow treatment plant	quick lime	37.9	1,516	35,714	0,70	1,083	1,642

to date. Although the treatment would have to be carried out with quicklime, the carbon release would be substantially lower than the in-lake neutralization with limestone powder due to the lower quantity required (Tab. 2 line 1 and 3). However, it should not be forgotten that acidic lakes also have restrictions on subsequent use (tourism, fishing).

River water as a source of alkalinity

The discharge of river water into pit lakes is used not only to store water for times of low water supply but also to control the quality of the pit lake itself and its outflow. For this purpose in 2021, 19 million m^3 were withdrawn from the river Lausitzer Neiße, 95 million m^3 from the river Spree and 22 million m^3 from the river Schwarze Elster. As a result of the flushing of the pit lakes with an average alkalinity of the river water of 1.5 mol/m^3, 204 million mol alkalinity have been provided and the use of lime products was reduced accordingly.

Nevertheless using river water as an alkalinity source has its own CO_2 footprint. It results from the construction measures, which are required to build the technical flooding facilities (which are not assessed here), and the energy input for the transfer, in particular the pumping operation.

While the water from the river Schwarze Elster can be used without pumping, the water from the river Spree has to be pumped into some of the lakes. The water from the river Lausitzer Neiße has to be pumped even twice before it can be used.

Assuming an alkalinity of 1.5 mol/m^3, a specific CO_2 release of 45 to 51 g/mol alkalinity can be estimated for the use of Lausitzer Neiße water. This is of the same order of magnitude as the use of quicklime or hydrated lime and does therefore not improve the carbon footprint.

However, this only applies to the specific CO_2-emissions under the conditions of current electricity production. If specific CO_2 emissions of the electricity generation will decrease due to increasing renewable energy production, the CO_2-footprint of pumping will decrease and eventually will become climate-neutral.

Conclusions

The carbon footprint assessment can now serve as a basis for further measures to reduce CO_2 emissions in the LMBV's water treatment operations. There are some measures that can be implemented quickly and easily, while others still need to be developed, planned or installed.

Short-term measures such as the use of chalk and limestone powder for in-lake treatment or the prioritization of river water (without using pumping energy) with respect to the use of lime products have already been implemented.

In the medium term, alternative alkalinity carriers that do not have to be produced specifically but are generated as by-products in industrial processes (Uhlmann 2014) can improve the carbon footprint of the LMBV's water treatment measures. Research into by-products from industrial processes should therefore be intensified. However, extensive testing and approval procedures are to be expected until the by-products are ready for use, as well as initially elaborated monitoring. The LMBV is currently conducting a pilot project for the use of such a by-product (GFI 2022), which has the potential to replace more than 10% of the lime products used in in-lake treatment and the associated CO_2 emissions.

In the long term measures to minimize lake acidification by preventing the inflow of acidic groundwater should be implemented. This can be achieved, for example, by the construction of sealing walls. Another long-term option is the additional transfer of alkaline river water from other catchment areas. Although this can only be done by pumping, the power supply is expected to originate completely from renewable energy in the future. In this case, only the carbon emissions caused by the construction of the infrastructure need to be considered. If the price of electricity falls substantially as a result of renewable energies, electrochemical water treatment processes (Friedrich *et al.* 2007) could also become economically interesting. Such processes have already been tested by the LMBV in pilot projects, but were rejected due to high electricity costs.

The LMBV is a state-owned mining rehabilitation company financed by taxes. On the one hand, the state has the fundamental goal of becoming CO_2-neutral by 2045; on the other hand, strict attention must be paid to the economic use of taxpayers' money. There is a conflict of objectives between these two goals. While carbon neutrality is still a rather generally formulated goal, there are numerous regulations in public procurement law that make it difficult to finance CO_2 savings if they result in higher costs. This conflict of objectives will be defused by the expected increase in the price of CO_2 certificates. The switch to renewable energies will automatically eliminate a large proportion of carbon emissions in the long term.

As long as lime products are used, carbon emissions from quicklime production and the outgassing of CO_2 are unavoidable. However, if we consider not only water treatment, but the entire operations of the LMBV, things look different. While the LMBV's water treatment released an average of around 23.524 tons of CO_2 per year during the examined time period, conservative calculations of carbon storage show that around 86,000 tons of CO_2 are sequestered annually in the soils and forests recultivated by the LMBV since 1995 (with an upward trend). Even if other LMBV carbon emissions are taken into account, the LMBV already is a climate-positive institution.

By addressing the carbon footprint of neutralization measures, LMBV can contribute to broader climate change mitigation efforts while fulfilling its environmental restoration obligations. The implementation of the strategies identified in this study could lead to relevant reductions in CO_2 emissions associated with post-mining landscape rehabilitation.

References

BVK (2017) Bericht zu den 7 Leitlinien der deutschen Kalkindustrie. Kalk - die nachhaltige Lebensgrundlage, Bundesverband der Deutschen Kalkindustrie e.V., Köln Juni 2017

Cole JI, Carac, NF (1998) Atmospheric exchange of carbon dioxide in a low-wind oligotrophic lake measured by the addition of SF. Limnol Oceanogr, 43(4): 647-656

Franck HG, Knop A (1979): Kohleveredlung. Chemie und Technologie, Springer Verlag, Heidelberg

Friedrich HJ, Zaruba A, Meyer S, Kappnik R, Stolp W, Benthaus FC (2007) Verfahren und kleintechnische Anlage zur Aufbereitung schwefelsaurer Grubenwässer (RODOSAN®-Verfahren). Proceedings des Dresdner Grundwasserforschungszentrums e.V., Heft 31: 205-210.

Kölling G, Schnur F (1977) Chemierohstoffe aus Kohle, Thieme, Stuttgart

GFI (2022) Antrag auf ein Pilot- und Demonstrationsvorhaben bei der LMBV: Einsatz des alkalinen Produktes „RENACARB“ aus der Lebensmittelproduktion für die Wasserbehandlung in der Lausitz., Dresden 21. August 2022

Guminski A, Rouyrre E, Wiener M (2019) CO2-Verminderung in der Kalkherstellung. Präsentation vom 27.11.2019. Forschungsstelle für Energiewirtschaft e.V., http://ffe.de/wp-content/uploads/2021/08/CO2-Verminderung_in_der_Kalkherstellung.pdf

LMBV (2022) Nachhaltigkeitsbericht 2021 der LMBV. Lausitzer- und Mitteldeutsche Bergbau-Verwaltungsgesellschaft mbH, Senftenberg. April 2022

LMBV (2023) Nachhaltigkeitsbericht 2022 der LMBV. Lausitzer- und Mitteldeutsche Bergbau-Verwaltungsgesellschaft mbH, Senftenberg. April 2023

Süßmann A, Lienkamp M (2015) Technische Möglichkeiten für die Reduktion der CO2-Emissionen von LKW und Bussen. Fahrzeugtechnik 103, Carl Schünemann Verlag, Bergisch Gladbach

Uhlmann W (2014) Untersuchung des Einflusses der Einlagerung von Eisenhydroxidschlämmen in Tagebauseen auf deren Wasserbeschaffenheit. Dresden, 31. August 2014