



# Potential generation and consumption of carbon dioxide during treatment of mine drainages in South Korea

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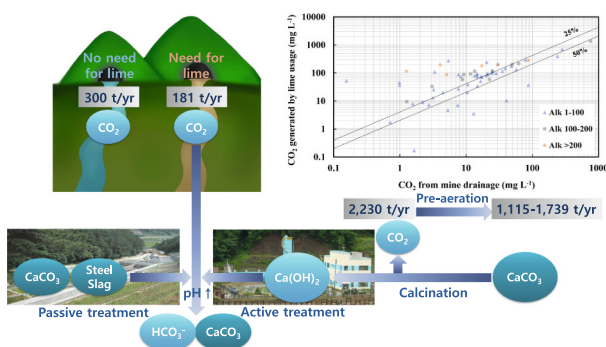
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## HIGHLIGHTS

- Mine drainages with lower pH and higher alkalinity exhibited higher CO<sub>2</sub> emissions.
- Coal mines accounted for 95 % of CO<sub>2</sub> emissions from mine drainage in South Korea.
- CO<sub>2</sub> from potential lime treatment was 12 times the amount of CO<sub>2</sub> from mine drainages.
- Passive treatment is substantially more beneficial than active treatment using lime.
- Particularly mine drainages with high pH and alkalinity preferred pre-aeration.

## GRAPHICAL ABSTRACT



## ARTICLE INFO

Editor: Kuishuang Feng

### Keywords:

Carbon footprint  
CO<sub>2</sub> emission  
PHREEQ-N-AMDTreat  
Hydrated lime  
Pre-aeration

## ABSTRACT

Mine drainage often acts as a source of CO<sub>2</sub>-emissions due to pyrite oxidation and the associated production of H<sup>+</sup>, which promotes the dissolution of carbonate minerals. While the treatment of mine drainage with hydrated lime is a common practice to increase pH, the production of lime generates a considerable amount of CO<sub>2</sub>. In this study, direct CO<sub>2</sub> emissions from mine drainages and indirect CO<sub>2</sub> emissions from the potential consumption of hydrated lime were modeled using PHREEQ-N-AMDTreat based on chemical compositions and flow rates at most mine drainage sites ( $n = 395$ ) across South Korea. The total potential CO<sub>2</sub> emissions from the mine drainages were estimated at 481 t yr<sup>-1</sup>, with 95 % originating from coal mines. Mine drainages with lower pH and/or higher alkalinity generally exhibited higher CO<sub>2</sub> emissions. In contrast, the potential consumption of hydrated lime to treat all sampled mine drainages could generate 2230 t CO<sub>2</sub> yr<sup>-1</sup>, which was >12 times the CO<sub>2</sub> degassed from the drainages, assuming atmospheric equilibrium under surface conditions. Therefore, when considering CO<sub>2</sub> emissions, passive treatment methods are substantially more advantageous than (semi-)active treatment methods using hydrated lime. The estimated CO<sub>2</sub> emissions from most mine drainages were <13 % of the indirect CO<sub>2</sub> emissions attributed to hydrated lime usage. Since the ratio (13 %) is lower than the reported reduction of hydrated lime consumption (22 %–50 %) achieved through pre-aeration in treatment processes, implementing pre-aeration is a preferable approach for most mine drainages from the perspective of CO<sub>2</sub> emission reduction.

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<https://doi.org/10.1016/j.scitotenv.2025.179270>

Received 13 June 2024; Received in revised form 8 February 2025; Accepted 26 March 2025

Available online 1 April 2025

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## 1. Introduction

Groundwater discharged from mines is often oversaturated with carbon dioxide (CO<sub>2</sub>) (Rose and Cravotta, 1998; Kruse and Strosnider, 2015; Brown et al., 2024), leading to degassing into the atmosphere under surface conditions (Kirby and Cravotta, 2005). Thus, mine drainage can serve as an important source of greenhouse gas emissions (e.g., Raymond and Oh, 2009; Vesper et al., 2016).

Elevated dissolved CO<sub>2</sub> in mine drainage generally originates from the dissolution of calcite (CaCO<sub>3</sub>) and other carbonate minerals by H<sup>+</sup> (Eq. (1)).

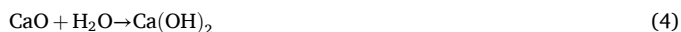


where H<sup>+</sup> is primarily produced during pyrite oxidation, and carbonic acid (H<sub>2</sub>CO<sub>3</sub><sup>\*</sup>) primarily consists of CO<sub>2(aq)</sub>. Additionally, as pH increases, CO<sub>2(aq)</sub> is converted to other dissolved inorganic carbon (DIC) species, such as bicarbonate (HCO<sub>3</sub><sup>-</sup>) (Eq. (2); Kirby and Cravotta, 2005).



Meanwhile, one of the most commonly used alkaline agents for increasing the pH of mine drainage is hydrated lime (Ca(OH)<sub>2</sub>), primarily due to its low cost and high coagulation efficiency in treating metal contaminant ions. Additionally, the removal of CO<sub>2(aq)</sub> can be considered a beneficial side effect of using alkaline agents.

Hydrated lime is produced through the calcination of limestone (Eqs. (3) and (4)).



where 1 mol of CO<sub>2</sub> gas is generated for 1 mol of hydrated lime produced. In summary, dissolved CO<sub>2</sub> can be directly degassed from mine drainage, and increasing the pH of the drainage can help reduce CO<sub>2</sub> emissions. However, if the drainage is treated with hydrated lime, additional CO<sub>2</sub> may be indirectly generated during the lime production process.

Dissolved CO<sub>2</sub> and the corresponding emissions from mine drainage have been reported at both watershed and regional scales. Raymond and Oh (2009) estimated that ~11 Mt CO<sub>2</sub> was degassed due to acid mine drainage in the Susquehanna watershed in Pennsylvania over a century, averaging ~0.11 Mt CO<sub>2</sub> yr<sup>-1</sup>. Similarly, Vesper et al. (2016) reported CO<sub>2</sub> flux of 0.076 Mt CO<sub>2</sub> yr<sup>-1</sup> from 140 mine drainages in Pennsylvania, a value comparable to the annual emissions from a small coal-fired power plant. More recently, Brown et al. (2024) assessed CO<sub>2</sub> emissions from 16 mine drainages in Scotland, estimating a mean CO<sub>2</sub> concentration of 235 mg L<sup>-1</sup> and a total flux of 0.078 Mt CO<sub>2eq</sub> yr<sup>-1</sup> from all coal mines in Scotland. This value represented approximately 0.2 % of Scotland's total contribution to global CO<sub>2eq</sub> emissions.

In 2019, approximately 24 % (14 Gt CO<sub>2eq</sub>) of global anthropogenic greenhouse gas emissions originated from industrial sources, with lime production ranking as the second-largest industrial source after cement production (Shan et al., 2016; IPCC, 2021; Bing et al., 2023). Lime dosages have been studied and modeled in various mine drainage treatment facilities (Cravotta, 2021; Kim et al., 2023), and life-cycle assessment studies have indicated substantially lower CO<sub>2</sub> emissions associated with passive treatment compared to active treatment using lime (Tuazon and Corder, 2008; Hengen et al., 2014). Unlike active and semi-active methods (e.g., automated lime dosing systems that operates without labor, often accompanied by a large settling pond), which require continuous input of chemicals or energy, passive treatment systems function without such inputs (Younger et al., 2002). Specifically, to increase pH, passive systems typically utilize limestone or steel slag within SAPS (Successive Alkalinity Producing Systems), slag reactors, or SLBs (Slag Leach Beds), whereas active treatment systems rely heavily on hydrated lime. Despite the growing awareness of CO<sub>2</sub>

emissions from industrial lime use, the potential effects of different mine drainage treatment strategies on CO<sub>2</sub> emissions—including the conversion of CO<sub>2</sub> to other species due to pH increase and the indirect CO<sub>2</sub> emissions from lime production—remain largely unassessed. To the best of the authors' knowledge, this gap highlights a critical need for further research.

Thus, the objectives of this study are (1) to calculate CO<sub>2</sub> emissions from most mine drainages in South Korea and evaluate the effect of CO<sub>2</sub> removal through pH elevation, (2) to estimate indirect CO<sub>2</sub> emissions associated with the use of hydrated lime for treating these drainages, and (3) to assess the suitability of pre-aeration as a mine drainage treatment strategy, focusing on its potential to reduce lime dosage and associated CO<sub>2</sub> emissions. A conceptual flow diagram illustrating these objectives is presented in Fig. 1.

## 2. Methods

### 2.1. Survey on mine drainages

In 2016, the Korea Mine Rehabilitation and Mineral Resources Corporation (KOMIR) investigated 395 mine drainages across South Korea. Mine drainages with relatively high contamination and/or flow rates were surveyed four times a year, while those with lower contamination and/or flow rates were surveyed twice a year. The majority of the mine drainages were adit discharges, while some were leachates from dumps of waste rock or tailings.

The temperature, pH, and dissolved oxygen (DO) concentrations of the water samples were measured using a portable meter (D-55, Horiba, Kyoto, Japan), calibrated with standard solutions or air calibration (for DO) at the time of each survey or experiment. Water samples were filtered through 0.45-μm membrane filters. Alkalinity was then measured in the field using a digital titrator (16900, Hach, Loveland, CO, USA), and the dissolved Fe<sup>2+</sup> concentration was determined using a portable colorimeter (DR-890, Hach) following the phenanthroline method (APHA, 2017). For cation and anion analyses, samples were collected in 100-mL PE bottles. Cation analysis samples were preserved by adding a few drops of concentrated HNO<sub>3</sub> to maintain a pH of <2. These water samples were stored at 4 °C until further analysis. Additionally, mine drainage flow rates were measured using the bucket-and-stopwatch method, with each measurement repeated three times to calculate an average value.

Cations were analyzed by inductively coupled plasma optical emission spectroscopy (ICP-OES; Varian 720-ES, Agilent Technologies Inc., Palo Alto, CA, USA) at KOMIR, Wonju. Anions were analyzed by ion chromatography (model 850, Metrohm, Herisau, Switzerland) at KOMIR. For both ICP-OES and ion chromatography, the relative standard deviations were <5 % of the measured value.

### 2.2. Data analysis

Seasonal water quality and quantity data of the 395 mine drainages were assessed (Table 1 and Fig. S1). Dissolved CO<sub>2</sub> concentrations and the dosages of hydrated lime required to reach specified target pH values were assessed using the Caustic Titration module of PHREEQ-N-AMDTreat version 1.4.5 (Cravotta, 2020, 2021). As phosphate concentrations were negligible in all samples, and the samples used for alkalinity determination excluded suspended solids, the measured alkalinity values could be used with minimal error to calculate DIC species, including CO<sub>2</sub>. The Caustic Titration tool has three modeling options for computing the treatment to specified target pH values using hydrated lime or other caustic chemicals: (1) Not aerated, (2) Pre-aerated, and (3) Aerated to equilibrium. For this study, the aeration to equilibrium condition with a steady-state partial pressure of CO<sub>2</sub> (pCO<sub>2</sub>) of 10<sup>-3.4</sup> atm was applied. Chemical compositions of the mine drainages, including pH, temperature, alkalinity, and concentrations of cations and anions, were input into the module. The option to compute total

inorganic carbon (TIC) from the input alkalinity, pH, and temperature was selected. When assessing dissolved  $\text{CO}_2$  concentrations in mine drainage, samples with a pH lower than 4.5 or without alkalinity were excluded. Additionally, as the current scope of PHREEQ-N-AMDTreat does not encompass Cu and Zn concentrations, Cu and Zn concentrations exceeding  $1 \text{ mg L}^{-1}$  in this study were converted to equivalent  $\text{Fe}^{3+}$  concentrations by applying different atomic weights and valences. These were then added to the existing  $\text{Fe}^{3+}$  concentrations (Kim et al., 2023). For all simulations, hydrated lime was selected as the alkaline agent, with default purity and mixing efficiency factors of 0.99 and 0.8, respectively. This indicates that the  $\text{Ca}(\text{OH})_2$  content in the hydrated lime was 99 %, and 20 % of the hydrated lime remained undissolved and accumulated with sludge (Kim et al., 2023).

To predict maximum  $\text{CO}_2$  concentrations that could potentially be degassed from mine drainages by natural aeration ( $C_{\text{CO}_2(\text{degas})}$ ), dissolved  $\text{CO}_2$  concentration at the equilibrium  $p\text{CO}_2$  of  $10^{-3.4} \text{ atm}$  ( $C_{\text{CO}_2(\text{eq})}$ ) was subtracted from the modeled initial concentration of dissolved  $\text{CO}_2$  ( $C_{\text{CO}_2(\text{dissolved})}$ ) (Eq. (5)). The  $\text{CO}_2$  flux ( $F_{\text{CO}_2(\text{degas})}$ ) was then computed by multiplying this difference in concentration by the cumulative annual flow rate expressed in  $\text{L yr}^{-1}$  ( $Q_{\text{annual}}$ ).

$$F_{\text{CO}_2(\text{degas})} = C_{\text{CO}_2(\text{degas})} \times Q_{\text{annual}} = (C_{\text{CO}_2(\text{dissolved})} - C_{\text{CO}_2(\text{eq})}) \times Q_{\text{annual}} \quad (5)$$

The mine drainages were classified into two categories: (1) potential (semi-)active treatment with hydrated lime, which includes 11 (semi-) active treatment facilities under operation, and (2) others (indicated as “Others” in the relevant plots), which include (a) mine drainages expected to meet discharge criteria in South Korea (excluding arsenic and fluoride) after aeration, (b) mine drainages being successfully treated by passive treatment facilities, and (c) stagnant mine drainages without surface flow.

To simulate the characteristics of treated effluent, the target pH was varied based on treatment goals. The target pH was determined using the following criteria, based on the composition of mine drainage and reported coprecipitation–adsorption behavior (Kim et al., 2022a): (1) Al: pH of 6, to exceed the minimum pH value of the discharge standard in the Republic of Korea ( $>5.8$ ); (2) Fe: pH of 7.5 ( $\text{Fe} < 20 \text{ mg L}^{-1}$ ), 8.0 ( $\text{Fe} 20\text{--}100 \text{ mg L}^{-1}$ ), and 8.5 ( $\text{Fe} > 100 \text{ mg L}^{-1}$ ), considering  $\text{Fe}^{2+}$  oxidation rate according to pH; (3) Mn: pH of 9.5 ( $\text{Mn} < 10 \text{ mg L}^{-1}$ ) and 10.0 ( $\text{Mn} \geq 10 \text{ mg L}^{-1}$ ) considering oxidation rate according to pH, but coprecipitation–adsorption effect was also considered based on Fe and Al concentrations (Kim et al., 2022a); (4) Zn: pH of 8.5; and (5) Ni: pH of 8.5 or 9.0, considering coprecipitation–adsorption by Fe.

After the geochemical modeling of each seasonal data subset which is described above, annual averages were used for each mine drainage site ( $n = 395$ ).

### 2.3. Calculation of $\text{CO}_2$ emission from hydrated lime consumption

Emissions of  $\text{CO}_2$  from hydrated lime production facilities, categorized as Scope 1 emissions (direct greenhouse gas emissions from sources controlled or owned by the organization), were examined. Approximately 60 % of total  $\text{CO}_{2\text{eq}}$  emissions result from the decomposition (calcination) reaction, 39 % from fuel combustion, and only 1 % from electricity consumption at the plant (European Lime Association, 2019, 2021; Laveglia et al., 2022). To focus on the primary and direct sources of  $\text{CO}_2$  emissions, calcination and fuel combustion were included, while electricity consumption and limestone quarrying were excluded from the carbon budget.

To convert the amount of quicklime ( $\text{CaO}$ ) to  $\text{CO}_2$  generation, an emission factor of 0.75 for lime during thermal decomposition ( $EF_{\text{Lime}(\text{decom})}$ ) was applied. This factor was derived from the stoichiometric ratio (SR) of  $\text{CO}_2$  to  $\text{CaO}$  (0.785), based on Eq. (3), and adjusted for the purity ( $P$ ) of quicklime at 0.95 (IPCC, 2006; IPCC, 2019; GGIRC, 2022). Subsequently,  $EF_{\text{Lime}(\text{decom})}$  was multiplied by a conversion factor (CF) for hydrated lime from quicklime (0.757) to obtain the final emission factor for hydrated lime ( $EF_{\text{HL}(\text{decom})}$ ) of 0.57 during thermal decomposition (Eq. (6); IPCC, 2006, 2019).

$$EF_{\text{HL}(\text{decom})} = EF_{\text{Lime}(\text{decom})} \times CF = SR \times P \times CF \quad (6)$$

Moreover,  $\text{CO}_2$  generation from fossil fuel combustion during the calcination of limestone was added to the total  $\text{CO}_2$  emission (Shan et al., 2016; Laveglia et al., 2022; Wu et al., 2023). Shan et al. (2016) reported mass ratios ( $R_{\text{C-L}}$ ) of coal consumption ( $M_{\text{Coal}}$ ) to lime production ( $M_{\text{Lime}}$ ) ranging from 0.12 to 0.16 in China, with a weighted average of 0.15. The emission factor for coal combustion ( $EF_{\text{Coal}}$ ) was  $1.85 \text{ t CO}_2$  per t coal (Shan et al., 2016), resulting in an emission factor for lime during fuel combustion ( $EF_{\text{Lime}(\text{coal})}$ ) of  $0.27 \text{ t CO}_2$  per t lime (Eq. (7)). By applying the conversion factor (CF) from quicklime to hydrated lime, we calculated the emission factor for hydrated lime during fuel combustion ( $EF_{\text{HL}(\text{coal})}$ ) to be  $0.21 \text{ t CO}_2$  per t hydrated lime.

$$EF_{\text{HL}(\text{coal})} = EF_{\text{Lime}(\text{coal})} \times CF = R_{\text{C-L}} \times EF_{\text{Coal}} \times CF = \frac{M_{\text{Coal}}}{M_{\text{Lime}}} \times EF_{\text{Coal}} \times CF \quad (7)$$

Thus, summing the emission factors for hydrated lime during thermal decomposition ( $EF_{\text{HL}(\text{decom})}$ , 0.57) and coal combustion ( $EF_{\text{HL}(\text{coal})}$ , 0.21) yields  $0.78 \text{ t}$  of  $\text{CO}_2$  directly generated per tonne of hydrated lime produced. When considering the total  $\text{CO}_{2\text{eq}}$  emissions over the entire production process, Laveglia et al. (2022) calculated  $0.94 \text{ t}$  of  $\text{CO}_{2\text{eq}}$  during hydrated lime production in four EU countries. Additionally, Wu et al. (2023) estimated  $0.89 \text{ t}$  of  $\text{CO}_{2\text{eq}}$  during hydrated lime production in the Yangtze River basin in China. Therefore, the  $\text{CO}_2$  directly

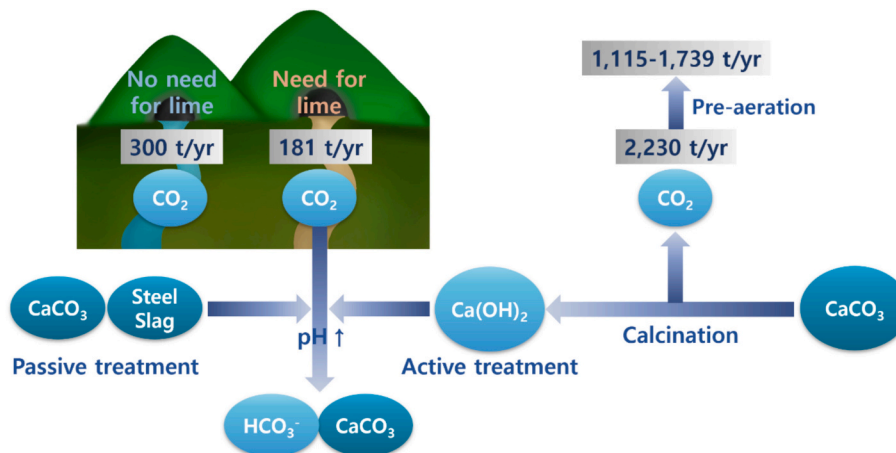


Fig. 1. Conceptual flow diagram illustrating this study.

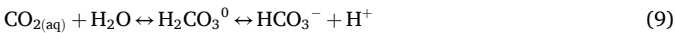
generated during hydrated lime production ( $C_{CO_2(HL)}$ ) was calculated by multiplying the hydrated lime dosage ( $D_{HL}$ ) by the emission factor for hydrated lime ( $EF_{HL} = 0.78$ ) (Eq. (8)).

$$C_{CO_2(HL)} = D_{HL} \times EF_{HL} = D_{HL} \times 0.78 \quad (8)$$

### 3. Results and discussion

#### 3.1. Potential $CO_2$ emissions from mine drainages

Modeled concentrations of potential  $CO_2$  degassing from mine drainages are plotted against the pH and alkalinity of untreated mine drainages in Fig. 2. The logarithm of the potential  $CO_2$  concentrations degassing from mine drainages exhibited a predominantly linear negative relationship with pH within each alkalinity range (Fig. 2a). Similarly, within each pH range, the logarithm of  $CO_2$  concentrations demonstrated a linear positive relationship with the logarithm of alkalinity (Fig. 2b). Equilibrated dissolved  $CO_2$  concentrations after aeration in the model were very low, averaging  $0.80 \text{ mg L}^{-1}$  and reaching a maximum of  $1.36 \text{ mg L}^{-1}$  for the mine drainages (Fig. S1). These results indicate that dissolved  $CO_2$  concentrations in mine drainages are the primary determinant of potential  $CO_2$  degassing. The observed relationships align with the theoretical relationships of dissolved  $CO_2$  with pH and alkalinity in Eq. (9). An increase in  $H^+$  and  $HCO_3^-$  concentrations leads to a corresponding increase in dissolved  $CO_2$  concentrations:



Therefore, mine drainages with low pH and high alkalinity exhibit a higher potential for  $CO_2$  emissions.

Furthermore, the origins of the drainages are classified into coal and metal mines in Fig. 3. In South Korea, drainages from coal mines generally exhibit lower pH than those from metal mines (Figs. 3a and S1a), while alkalinity levels are comparable between the two (Figs. 3b and S1b). This difference in pH can be attributed to the presence of framboidal pyrite—micron- or submicron-sized crystals with very high specific surface areas—which produces acid at much higher rates than larger pyrite grains and is commonly found in sedimentary strata, such as those associated with coal mines (Caruccio, 1975; Kim et al., 2017). In contrast, most metal mines in South Korea primarily excavated gold from ores with low pyrite content. These geological factors contribute to the lower pH observed in coal mine drainages. Consequently, mine drainages with high potential  $CO_2$  concentrations available for

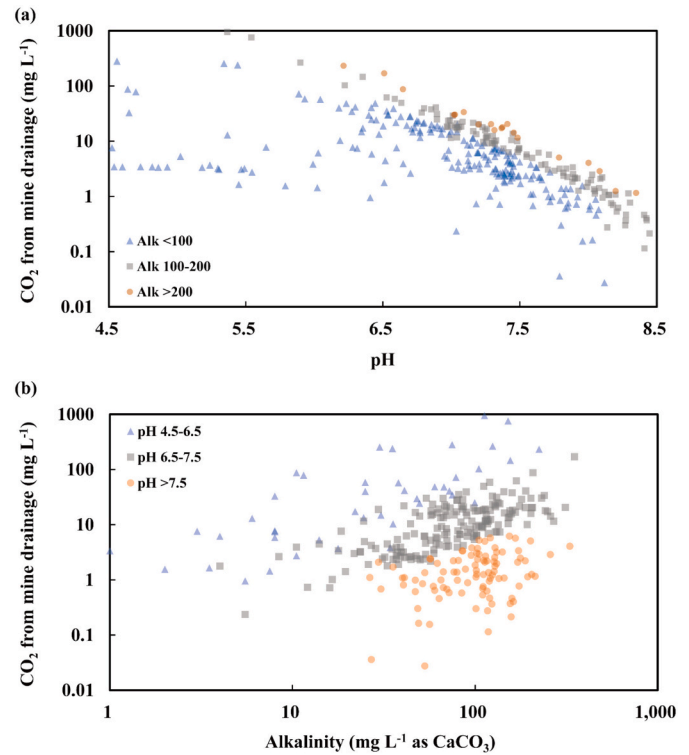


Fig. 2. Relationship between modeled concentrations of potentially degassed  $CO_2$  and (a) pH and (b) alkalinity in untreated mine drainages in South Korea. The samples are classified by alkalinity and pH ranges.

degassing, driven by their low pH, were predominantly classified as coal mine drainage (Figs. 3a and S1c).

Furthermore, the flow rates of coal mine drainages were generally higher than those of metal mine drainages in South Korea. This disparity is attributed to the extensive excavation of underground tunnels and the high hydraulic conductivity of sedimentary strata, such as sandstone and limestone, commonly found in coal mining areas. As a result, the potential  $CO_2$  flux ( $\text{kg d}^{-1}$ ) from drainage became even higher for coal mines than for metal mines (Figs. 4 and S1d). Notably, all mine drainages with potential  $CO_2$  flux exceeding  $30 \text{ kg d}^{-1}$  were from 11 coal

**Table 1**  
Statistics for flow rate and water quality of analyzed mine drainages.

Item	Unit	Statistics	Coal mines		Metal mines		Total
			Potential lime use	Others	Potential lime use	Others	
Number	(ea)		94	138	25	138	395
Flow rate	$(\text{m}^3 \text{ d}^{-1})$	Avg.	286	221	82	25	159
		Min.	0.25	0	1.8	0	0
		Max.	6006	3725	270	430	6006
		Sum	26,844	30,429	2052	3477	62,802
pH	(-)	Avg.	5.0	7.0	6.4	7.5	6.7
		Min.	3.0	3.5	2.8	2.5	2.5
		Max.	8.2	8.4	8.0	8.7	8.7
		Avg.	34	96	70	78	73
Alkalinity	$(\text{mg L}^{-1} \text{ as CaCO}_3)$	Min.	0	0	0	0	0
		Max.	330	350	207	258	350
		Avg.	14	27	52	4.5	18
		Min.	-0.1 <sup>a</sup>	0.04	0.2	-0.2	-0.2
Potential $CO_2$ degassed from mine drainage	$(\text{mg L}^{-1})$	Max.	240	953	759	59	953
		Sum	1329	3790	1298	622	7040
		Avg.	240	- <sup>b</sup>	190	-	230
		Min.	0.2	-	1.7	-	0.2
$CO_2$ generated by lime production	$(\text{mg L}^{-1})$	Max.	3632	-	1354	-	3632
		Sum	22,589	-	4749	-	27,339

<sup>a</sup> Negative value indicates that atmospheric  $CO_2$  dissolves into the mine drainage until equilibrium is reached.

<sup>b</sup> Not applicable.



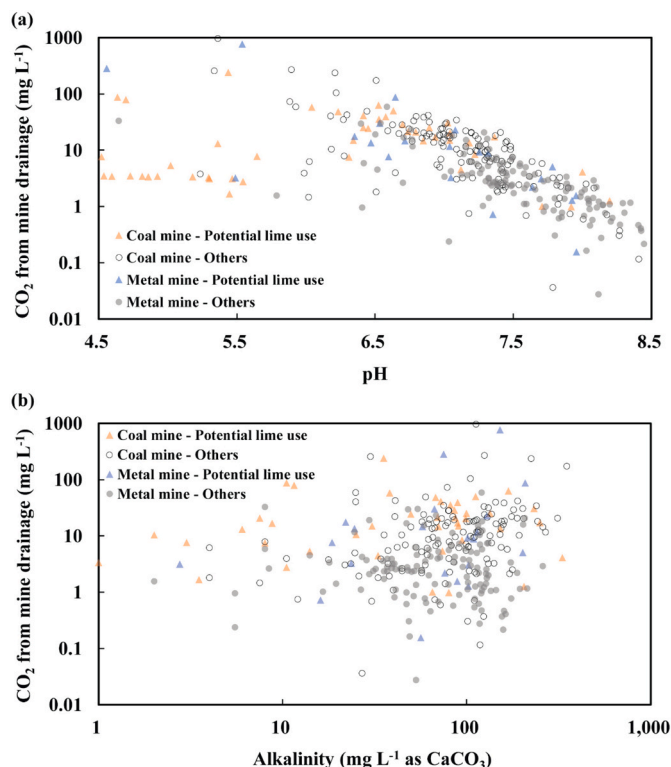


Fig. 3. Relationship between modeled concentrations of potentially degassed CO<sub>2</sub> and (a) pH and (b) alkalinity in untreated mine drainages in South Korea. The samples are classified by mine types and potential of (semi)-active treatment using hydrated lime.

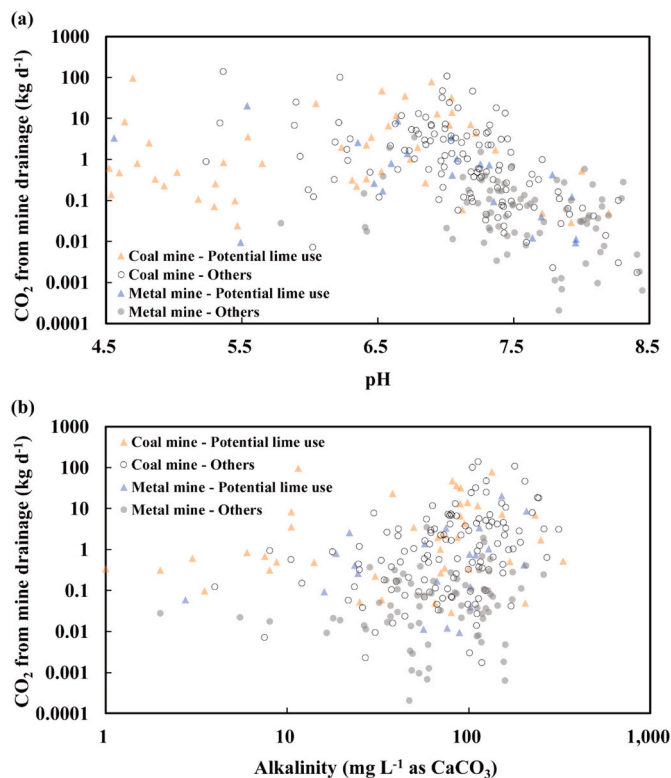


Fig. 4. Relationship between modeled potential flux of CO<sub>2</sub> from untreated mine drainages and (a) pH and (b) alkalinity in mine drainages in South Korea. The samples are classified by mine types and potential of (semi)-active treatment using hydrated lime.

mines (Fig. 4).

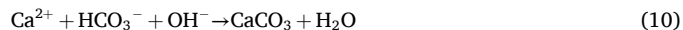
Additionally, mine drainages with the potential for (semi)-active treatment using hydrated lime exhibited lower pH levels compared to other drainages, which was evident in mine drainages with a pH below 5.5 (Figs. 3 and 4).

Furthermore, the pH and alkalinity of mine drainages from limestone strata were compared to those from other geological formations, as these parameters were found to determine potential CO<sub>2</sub> emissions. However, a *t*-test revealed no significant differences in generated CO<sub>2</sub> concentrations between the two groups (Table S1). The F-test for variance indicated a *p*-value much lower than 0.05, suggesting a significant difference in variance (heterogeneous variance) at the 95 % confidence level. Given this heterogeneity, the *t*-test was conducted accordingly, and the resulting *p*-value was much higher than 0.05, confirming that the average potential CO<sub>2</sub> emissions were not significantly different between the two groups. For instance, the Buguk coal mine drainage exhibited a high alkalinity of 330 mg L<sup>-1</sup> as CaCO<sub>3</sub> and a pH of 8.0. Despite of its high alkalinity, this mine is located in strata with only intermittent limestone presence, rather than a regional limestone formation. Similarly, the Okdong metal mine drainage demonstrated a high alkalinity of 151 mg L<sup>-1</sup> as CaCO<sub>3</sub>, even though it is situated far from any limestone strata. This apparent discrepancy between alkalinity and limestone geology may be attributed to localized controls of pH and alkalinity in mine drainages. Specifically, these controls are influenced by the degree of pyrite oxidation and subsequent neutralization by surrounding rocks containing various forms of carbonates, rather than exclusively by limestone formations.

Additionally, potential CO<sub>2</sub> emissions from leachates originating from waste rock or tailings dumps (*n* = 15) were compared to those from adit discharges (Table S2). The F-test for variance yielded a *p*-value much lower than 0.05, confirming heterogeneous variance. Subsequently, the *t*-test was conducted with this consideration, and the resulting *p*-value was also lower than 0.05, indicating that the average potential CO<sub>2</sub> emissions were significantly different between the two groups at the 95 % confidence level. Although the number of leachate samples was limited (*n* = 15), this result suggests that CO<sub>2</sub> degassing during water migration through waste rock or tailings dumps may have reduced the potential for CO<sub>2</sub> emissions upon discharge.

### 3.2. Decrease in CO<sub>2</sub> concentration by increase in pH

Figs. 2a and 3a demonstrate that degassing CO<sub>2</sub> concentrations, which are closely related to dissolved CO<sub>2</sub> concentrations in mine drainage (as discussed in Section 3.1), decrease significantly with increasing pH. This suggests that mine drainage treatment by increasing pH mitigates CO<sub>2</sub> degassing through conversion to HCO<sub>3</sub><sup>-</sup> (Eq. (2)) and/or calcite precipitation (Eq. (10)).



Saturation indices (SIs) for calcite in the treated mine drainages were calculated using the Caustic Titration module of PHREEQ-N-AMDTreat, which assumed a default SI value of 0.3 as the threshold required to precipitate calcite from solution. For mine drainage and groundwater, SI values exceeding zero are mostly distributed between 0 and 0.6, with an average of ~0.3 (Plummer et al., 1990; Nordstrom, 2008; Neogi et al., 2017). This oversaturation reflects the inhibition of calcite nucleation and the relatively slow precipitation rate of calcite (Langmuir, 1997). Furthermore, Langmuir (1997) reported that the SI can exceed 0.3 for calcite nucleation and precipitation in the presence of a few mg L<sup>-1</sup> of DOC which is common in groundwater. Therefore, an SI of 0.3 was selected for calcite precipitation in this study.

The number of samples expected to precipitate calcite was higher than the others for both coal and metal mines (Fig. 5). Thus, calcite precipitation contributed to the DIC consumption and subsequent CO<sub>2</sub> sequestration in most treated samples following the pH increase.

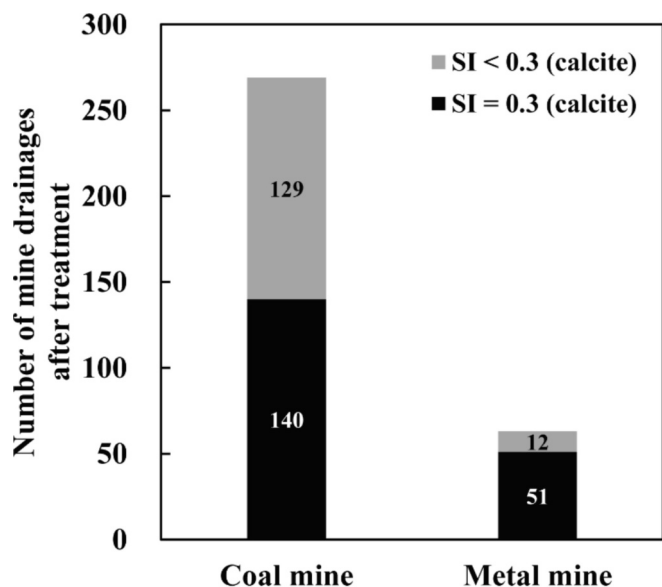


Fig. 5. Number of treated mine drainages with calcite saturation index (SI) of 0.3 or <0.3, which was modeled by the Caustic Titration module of PHREEQ-N-AMDTreat. An SI of 0.3 was assumed as the threshold required for calcite to precipitate from the solution. It should be noted that these numbers represent the number of sampling events and may exceed the number of mine drainages.

### 3.3. Indirect CO<sub>2</sub> emissions by using hydrated lime for treatment

Indirectly generated CO<sub>2</sub> concentrations were calculated based on the modeled consumption of hydrated lime during treatment. The accuracy of predicting hydrated lime consumption using the same model (Caustic Titration module in PHREEQ-N-AMDTreat) was verified against actual measurements from a full-scale treatment facility in South Korea

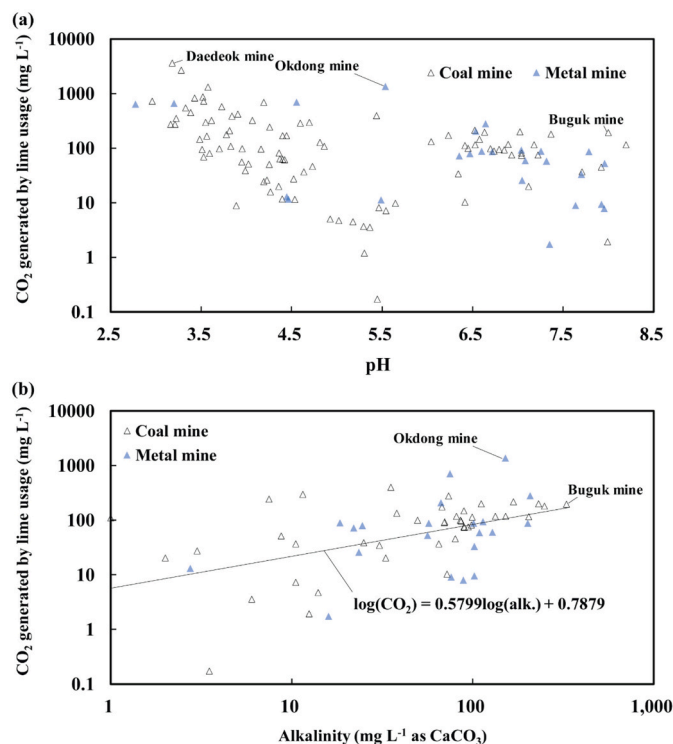


Fig. 6. Relationship between calculated indirect emission (in mg L<sup>-1</sup>) of CO<sub>2</sub> from modeled usage of hydrated lime and (a) pH and (b) alkalinity in mine drainages. The samples are classified based on mine types.

(Kim et al., 2023). The concentrations and flux of generated CO<sub>2</sub> are plotted against pH and alkalinity (Figs. 6–8). A notable trend was observed at pH values below 5.5, where decreasing pH corresponded to increased acidity and associated lime dosages, resulting in increasing concentrations of generated CO<sub>2</sub> by lime usage (Figs. 6a and 7a). Similarly, at a given alkalinity, sample groups with lower pH values exhibited higher potential CO<sub>2</sub> generation from lime usage (Fig. 7b). Additionally, a positive relationship was observed between CO<sub>2</sub> generation by lime usage and alkalinity of mine drainages (Figs. 6b and 7b). Similarly, at a given pH, sample groups of higher alkalinity exhibited greater CO<sub>2</sub> generation by lime usage (Fig. 7a). These results align with the reported role of HCO<sub>3</sub><sup>-</sup> in increasing lime dosage through OH<sup>-</sup> consumption and calcite precipitation (Kim et al., 2023). For example, the Okdong mine exhibited the highest CO<sub>2</sub> generation by lime usage among metal mines, at 1354 mg L<sup>-1</sup> (Fig. 6a). Relatively high alkalinity of 151 mg L<sup>-1</sup> as CaCO<sub>3</sub> at a pH of 5.54 as well as high Mn and Zn concentrations of 20.8–23.5 and 53.4–64.5 mg L<sup>-1</sup>, respectively, may have contributed to the increase in DIC and lime dosage (Fig. 7a). Similarly, the Buguk mine demonstrated the highest CO<sub>2</sub> generation by lime usage (195 mg L<sup>-1</sup>) among samples with a pH of ~8.0, due to its remarkably high alkalinity (330 mg L<sup>-1</sup> as CaCO<sub>3</sub>) compared to all other drainages (Fig. 7b). In contrast, the Daedeok mine exhibited the highest CO<sub>2</sub> generation from lime usage across all drainages, at 3632 mg L<sup>-1</sup>. This was due to its remarkably low pH (3.18) and exceptionally high concentrations of Fe (1016 mg L<sup>-1</sup>) and Al (391 mg L<sup>-1</sup>), which substantially increased lime dosage requirements.

The positive relationship between alkalinity and the CO<sub>2</sub> flux generated by lime usage (in kg d<sup>-1</sup>) was less pronounced compared to the relationship with CO<sub>2</sub> concentrations (in mg L<sup>-1</sup>) (Fig. 8b), as the flow rate influenced the flux calculation.

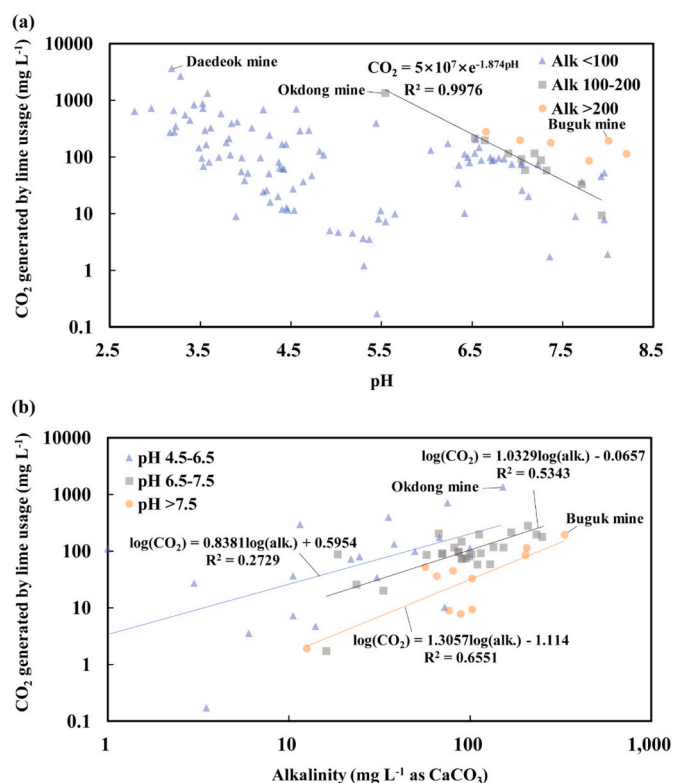


Fig. 7. Relationship between calculated indirect emission (in mg L<sup>-1</sup>) of CO<sub>2</sub> from modeled usage of hydrated lime and (a) pH and (b) alkalinity in mine drainages. The samples are classified based on alkalinity and pH ranges, and samples with pH <4.5 are excluded in (b). Regression lines correspond to the samples with alkalinities of 100–200 mg L<sup>-1</sup> as CaCO<sub>3</sub> in (a) and to samples with three different pH ranges in (b).

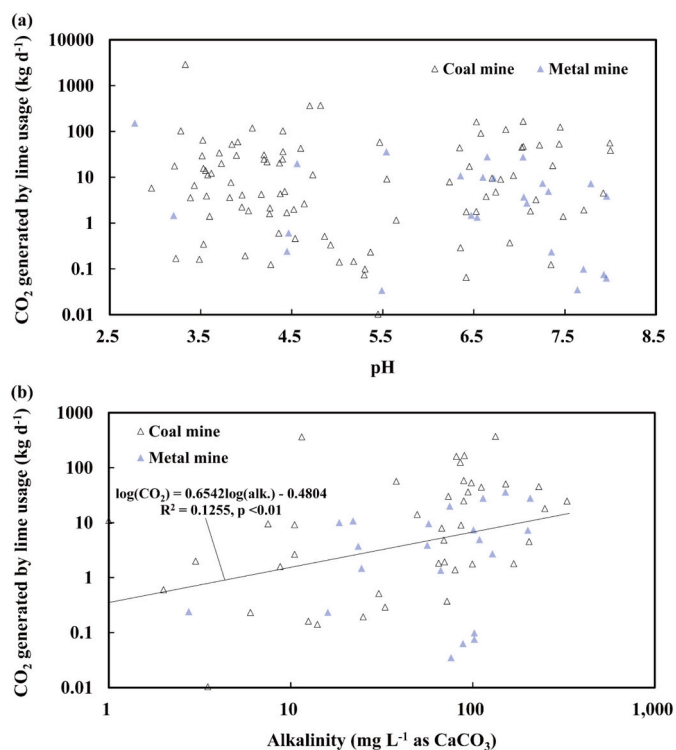


Fig. 8. Relationship between calculated indirect emission flux (in kg d<sup>-1</sup>) of CO<sub>2</sub> from modeled usage of hydrated lime and (a) pH and (b) alkalinity in mine drainages. The samples are classified based on mine types.

Direct CO<sub>2</sub> emissions from mine drainage were compared to indirect emissions from the use of hydrated lime for treatment (Fig. 9a). The total CO<sub>2</sub> generated from mine drainages by aeration to equilibrium was 481 t yr<sup>-1</sup>, comprising 459 t yr<sup>-1</sup> (95 %) from coal mines and 22 t yr<sup>-1</sup> (5 %) from metal mines in South Korea. Moreover, mine drainages requiring hydrated lime treatment generated 181 t yr<sup>-1</sup> of CO<sub>2</sub>, which can largely be mitigated through pH-increasing treatments (refer to Section 3.2). Nevertheless, indirect CO<sub>2</sub> emissions from hydrated lime usage reached 2230 t yr<sup>-1</sup>, which was >12 times the amount of CO<sub>2</sub> removed during mine drainage treatment. This result highlights the much larger disadvantage of using hydrated lime compared to the advantage of CO<sub>2</sub>

removal from mine drainage. Furthermore, constructing (semi-)active treatment facilities requires substantial amount of cement (Winfrey et al., 2015) and operating these facilities—such as agitation and pumping equipment—consumes electricity. Transportation of hydrated lime further adds to CO<sub>2</sub> emissions. Thus, adopting passive treatment methods offers substantial advantages in reducing the carbon footprint. To achieve satisfactory pH levels in passive treatment systems for treating dissolved metals, alternative substrates such as including steel slag and waste concrete, in addition to limestone, could be considered (Kim et al., 2022b, 2022c; Ho et al., 2023).

Additionally, the potential amount of sludge generation during mine drainage treatment was modeled to be 13,846 t yr<sup>-1</sup> (Fig. 9b), which should be disposed of as waste or recycled. The accuracy of predicting sludge amounts using the same model (Caustic Titration module in PHREEQ-N-AMDTreat) has been verified through comparing with measurements from a pilot-scale treatment facility in South Korea (Kim et al., 2023). In South Korea, acquiring sites for waste landfills poses challenges, and most sludge from mine drainage treatment facilities is recycled as a supplementary material for cement production. Of the total potential sludge, coal mine drainage treatment contributed 94 % (13,016 t yr<sup>-1</sup>). Additionally, the sludge generated from coal mine drainage treatment in South Korea typically contains low levels of toxic elements such as As, Cd, Cu, Pb, and Zn (Cui et al., 2011). This makes it a viable candidate for further utilization, such as producing commercial products like arsenic adsorbents (Lee et al., 2018; Kumar et al., 2020) or recovering valuable elements like Al, Mn, and rare earth elements (Vaziri Hassas et al., 2022; Cicek et al., 2023). Meanwhile, for mine drainages that do not require hydrated lime application, sludge generated through natural precipitation was modeled at 3339 t yr<sup>-1</sup> (Fig. 9b). This sludge can accumulate annually in surface water systems, such as streambeds, especially near coal mines in South Korea.

### 3.4. Suitability of pre-aeration step to degas CO<sub>2</sub> and reduce lime consumption

Pre-aeration to degas CO<sub>2</sub> prior to the treatment of mine drainages has been reported to substantially reduce lime consumption, owing to the decrease of H<sub>2</sub>CO<sub>3</sub> (Jageman et al., 1987; Kirby et al., 2009; Kruse and Strosnider, 2015; Means et al., 2015; Hedin and Hedin, 2016; Means and Beam, 2024). This process is typically performed as the initial step in mine drainage treatment facilities and involves bubbling to achieve CO<sub>2</sub> degassing before the addition of alkaline agents (Kirby et al., 2009;

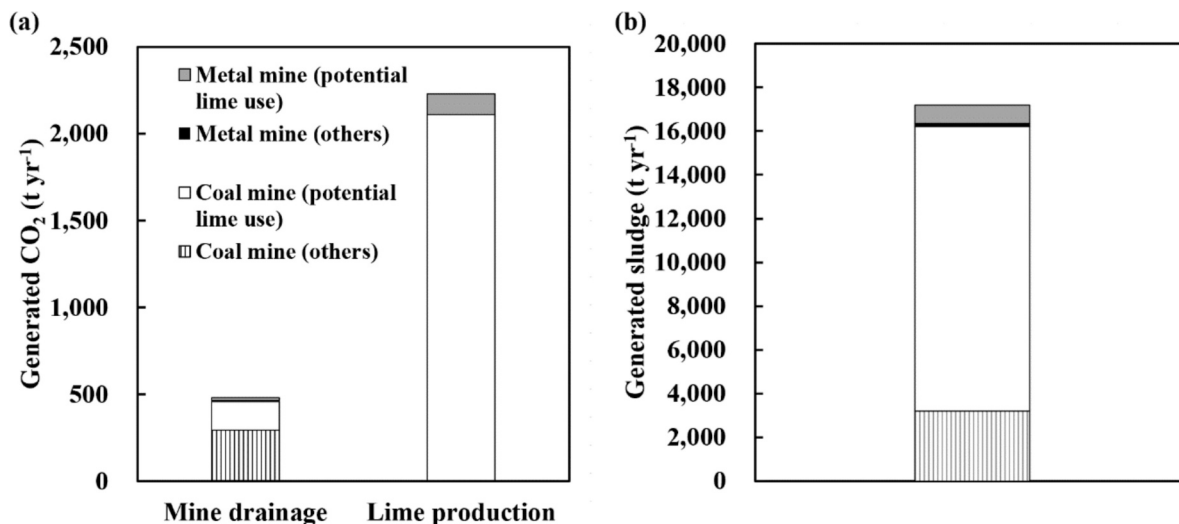


Fig. 9. (a) Modeled total potential flux of CO<sub>2</sub> from mine drainages and calculated indirect emission flux of CO<sub>2</sub> from modeled usage of hydrated lime for mine drainage treatment in South Korea. (b) Modeled total potential amount of annual sludge generation with water content of 75 % after treatment using hydrated lime (for “potential lime use”) and after equilibrium by aeration (for “others”) in South Korea. The samples are classified by mine types and potential use of hydrated lime.

Means et al., 2015). We compared direct CO<sub>2</sub> emissions during pre-aeration with indirect CO<sub>2</sub> emissions resulting from hydrated lime usage. The “Aerated to equilibrium” mode in the Caustic Titration module was applied to simulate pre-aeration to equilibrium conditions. The results, shown in Fig. 10, present the ratios of direct CO<sub>2</sub> emissions from pre-aeration to indirect CO<sub>2</sub> emissions from lime usage. The 1st, 2nd, and 3rd quartiles of these ratios were 2 %, 13 %, and 29 %, respectively. Meanwhile, Jageman et al. (1987) reported that lime consumption was reduced by 27 %, 38 %, and 43 % with pre-aeration. Similarly, Means et al. (2015) reported decreases of 22 % and 28 %, while Means and Beam (2024) noted a net annual lime dosage reduction of 50 %. These ratios of lime consumption reduction are higher than the observed median ratio (13 %) in Fig. 10. Therefore, in most cases, applying the pre-aeration step seems adequate to reduce both lime usage and net CO<sub>2</sub> emissions.

To specifically evaluate the impact of pre-aeration on CO<sub>2</sub> emissions, the CO<sub>2</sub> concentration generated by hydrated lime usage was plotted against the CO<sub>2</sub> concentration generated from mine drainage aeration, grouped by pH (Fig. 11a) and alkalinity (Fig. 11b). As samples with pH > 7.5 exhibited low CO<sub>2</sub> emissions from mine drainage, they are plotted above the 25 % line, indicating that CO<sub>2</sub> emissions from mine drainage are <25 % of the CO<sub>2</sub> emissions from lime usage. For these samples, pre-aeration will be suitable with respect to CO<sub>2</sub> emission when it reduces hydrated lime dosage by >25 %. On the contrary, many samples with pH between 6.5 and 7.5 are plotted near the 25 % line, reflecting higher CO<sub>2</sub> emissions from mine drainage. For those samples with a ratio of 25 %, at least a 25 % reduction of hydrated lime is required for the net CO<sub>2</sub> removal through pre-aeration. Several samples with pH of 4.5–6.5 are plotted below the 50 % line, which is principally attributable to low CO<sub>2</sub> emissions associated with lime usage. The low consumption of hydrated lime despite low pH appears to be related to their low alkalinity (Fig. 11b). Additionally, mine drainages with alkalinity of >200 mg L<sup>-1</sup> as CaCO<sub>3</sub> exhibited higher lime consumption compared to those with alkalinity of 100–200 mg L<sup>-1</sup> as CaCO<sub>3</sub> (Fig. 11b). This is consistent with the role of HCO<sub>3</sub><sup>-</sup> in increasing lime dosage by consuming OH<sup>-</sup> and facilitating calcite precipitation (Kim et al., 2023). In summary, samples

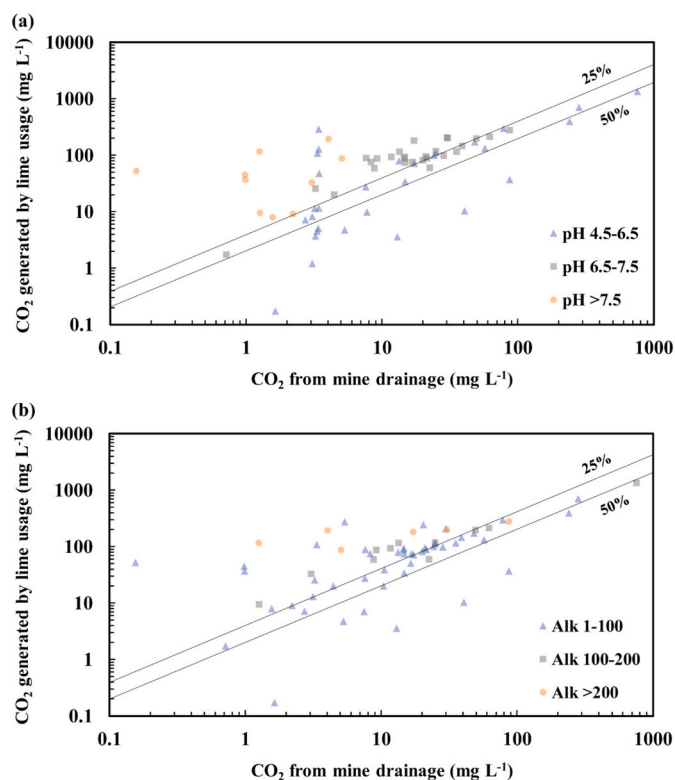


Fig. 11. Plot of modeled indirect CO<sub>2</sub> emission by the consumption of hydrated lime versus modeled direct CO<sub>2</sub> emission from mine drainage by pre-aeration. Ratios of CO<sub>2</sub> emission from mine drainage to CO<sub>2</sub> emission by the consumption of hydrated lime are also indicated as lines. The samples are classified by (a) pH and (b) alkalinity ranges.

with higher alkalinity and pH seem to benefit from pre-aeration in terms of reducing CO<sub>2</sub> emission.

#### 4. Conclusions

Potential CO<sub>2</sub> emissions from mine drainages and from the production of hydrated lime for mine drainage treatment were assessed on a nationwide scale using the CausticTitration model of PHREEQ-N-AMDTreat. The total CO<sub>2</sub> emissions from mine drainages through aeration to equilibrium were estimated at 481 t yr<sup>-1</sup> (Fig. 1), with 95 % originating from coal mines in South Korea. Mine drainages with lower pH and higher alkalinity exhibited a higher potential for CO<sub>2</sub> emissions. Treatment of mine drainage by increasing pH can reduce most of the emissions from mine drainage through conversion of DIC species and precipitation of calcite. Nevertheless, the potential consumption of hydrated lime for mine drainage treatment contributed to CO<sub>2</sub> emission of 2230 t yr<sup>-1</sup>, which was >12 times the amount of CO<sub>2</sub> removed during the treatment process. Furthermore, constructing (semi-)active treatment facilities using hydrated lime involves substantial CO<sub>2</sub> emissions from cement production and operational electricity use. Therefore, regarding CO<sub>2</sub> emissions, applying passive treatment systems, including slag reactors to further increase pH if necessary, is much more beneficial than (semi-)active treatment systems. Passive treatment facilities involving increasing pH will have three advantages: (1) treatment of contaminants, (2) removal of potential CO<sub>2</sub> from mine drainages by pH increase (using alkaline materials), and (3) prevention of CO<sub>2</sub> emissions associated with hydrated lime usage.

The reduction in indirect CO<sub>2</sub> emissions from hydrated lime consumption through the implementation of a pre-aeration step was compared with direct CO<sub>2</sub> emissions from the step. CO<sub>2</sub> emissions from most mine drainages in South Korea were <13 % of indirect CO<sub>2</sub>

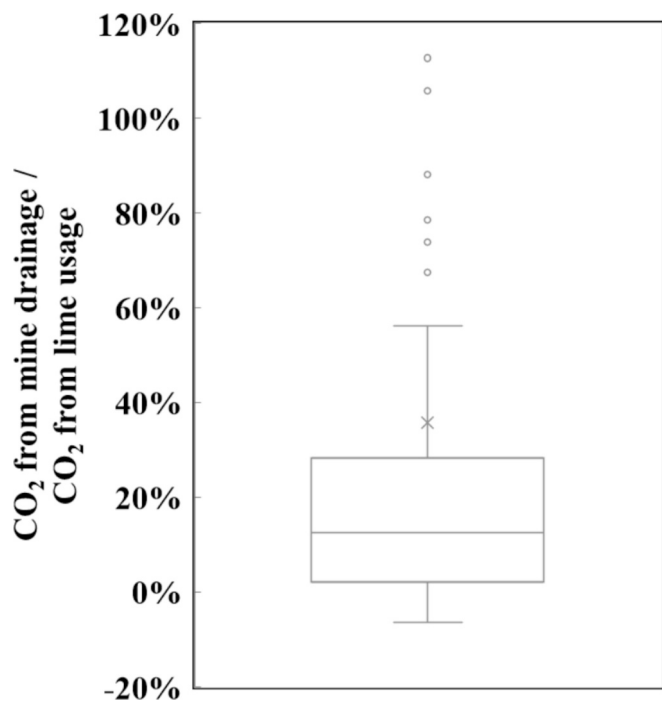


Fig. 10. Box plot representing ratios of modeled direct CO<sub>2</sub> emission from mine drainage by pre-aeration to modeled indirect CO<sub>2</sub> emission by the consumption of hydrated lime.



emissions from hydrated lime. Given that this ratio (13 %) is lower than the reported reduction in lime consumption (22 %–50 %), pre-aeration is preferable for majority of mine drainages in terms of CO<sub>2</sub> emissions. Although mine drainages with pH below 4.5 or without alkalinity could not be assessed, pre-aeration was found to be particularly beneficial for mine drainages with high pH and alkalinity among those with pH above 4.5.

## CRediT authorship contribution statement

**Duk-Min Kim:** Writing – review & editing, Writing – original draft, Supervision, Methodology, Formal analysis, Data curation, Conceptualization. **Ki-Rim Lee:** Formal analysis, Data curation. **Mi-Sun Park:** Project administration, Data curation.

## Declaration of competing interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: Duk-Min Kim reports financial support was provided by Korea Mine Rehabilitation and Mineral Resources Corporation. If there are other authors, they declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

## Acknowledgments

This work was supported by an R&D project of the Korea Mine Rehabilitation and Mineral Resources Corporation from 2022 to 2024 and by the Graduate School of Sangji University. The authors gratefully acknowledge the contributions of the people at Korea Mine Rehabilitation and Mineral Resources Corporation for conducting sampling, pre-treatment, and analyses. We thank Charles Cravotta for creating the PHREEQ-N-AMDTreat model and for his comments on an early draft of the manuscript. Special thanks go to anonymous reviewers for providing constructive comments that helped to improve the manuscript.

## Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.scitotenv.2025.179270>.

## Data availability

Data will be made available on request.

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