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Assessing the long-term carbon balance in mine waste storage facilities and implications for mine closure

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Abstract

The mining industry excavates and processes billions of tonnes of mine wastes per year and has the potential to leverage its infrastructure and capabilities to use suitable mine wastes for carbon dioxide removal (CDR), by enhanced rock weathering (ERW) as part of mine closure planning to offset mine emissions and mitigate climate change. However, before this opportunity can be realised, further development of key methodologies to measure and predict CO_2 *flux (uptake and release) from mine waste over the long timescales related to closure is required.*

While there is an increasing amount of research on the potential for mine wastes to sequester CO_2 *through the mineral carbonation of Mg-rich and Ca-rich silicates, less research into the determination of net* CO_2 *mass flux/balance is being carried out, which also considers the potential of mine waste to emit* CO_2 *due to co-occurrence of reactive sulphides; organic carbon; and carbonate minerals.*

This study expands on prior research into mine waste CDR, using novel laboratory and field methods to calculate and estimate the relative balance between sulphide oxidation (oxygen consumption and acidity production), carbonate dissolution (acid neutralisation and CO_2 *release) and silicate-related carbonation (*CO_2 *sequestration) rates within mine waste. The study describes development of both closed and open system experimental design and utilises data collected to evaluate the carbon balance of various types of mine waste. Findings from the closed-system experiments are used to demonstrate validation of potential testing protocols to estimate site mine waste net* CO_2 *flux.*

While ERW shows great potential in small-scale laboratory tests, several challenges arise when implementing it on a larger scale. ERW large-scale field trials and improved scaler laboratory-based methods can provide valuable tools for mine closure planning, specifically for designing waste storage facilities and assessing opportunities and risks related to CO_2 *net flux.*

Keywords: *carbon sequestration, carbon balance, mineral carbonation, mine wastes, mine closure planning*

1 Introduction

1.1 Background

This research addresses a critical gap in existing studies by proposing a novel and potentially scalable methodology for quantifying the dynamic equilibrium among sulphide oxidation (with associated oxygen consumption), carbonate dissolution (resulting in CO_2 release), and carbonation (leading to CO_2 sequestration) within mine waste storage facilities (WSFs). The project aims to further monitor CO_2 and O_2 flux from mine waste and assess carbon sequestration potential through enhanced weathering and monitoring, reporting, and verification (MRV) frameworks. The study will evaluate net mine waste emissions and explore potential offsets through carbon capture. This research aims to optimise CO_2 removal strategies in mining environments and

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develop robust frameworks for carbon dioxide removal (CDR) under real-world conditions. It investigates the CO_2 sequestration potential of ultramafic mine waste through laboratory-scale experiments while facilitating preparations for field-scale testing. By building on previous research, the methodology integrates established techniques with innovative approaches to advance carbon capture research.

1.2 Gas flux and mineral carbonation reactions

Enhanced rock weathering (ERW) using mafic/ultramafic rocks is a promising CDR technology that has received attention from the mining industry. ERW leverages the natural process of mineral weathering to sequester atmospheric carbon dioxide (CO_2) (Abdalqadir et al. 2024). Mineral carbonation is the weathering of calcium (Ca), magnesium (Mg), and iron (Fe) oxide silicates, which results in CO_2 being stored as chemically stable carbonate materials, or in the dissolved phase (Rackley 2017; Li et al. 2018) – allowing CO_2 to be stored in long-term carbon sinks with a sequestration duration index of 5 (100,000 years).

Aqueous carbonation, the weathering and release of cations from Mg-silicates and Ca-silicates in the presence of water, encompasses two main steps:

1. the dissolution of Mg-silicates or Ca-bearing silicates
2. precipitation of carbonates.

The simplified reaction process of mineral carbonation is shown in Figure 1 and Equation 1 (Li et al. 2018):

$$(Mg, Ca, Fe)\,SiO_4\,(s) + CO_2\,(g) \rightarrow MgCO_3\,(s) + SiO_2\,(s) \qquad (1)$$

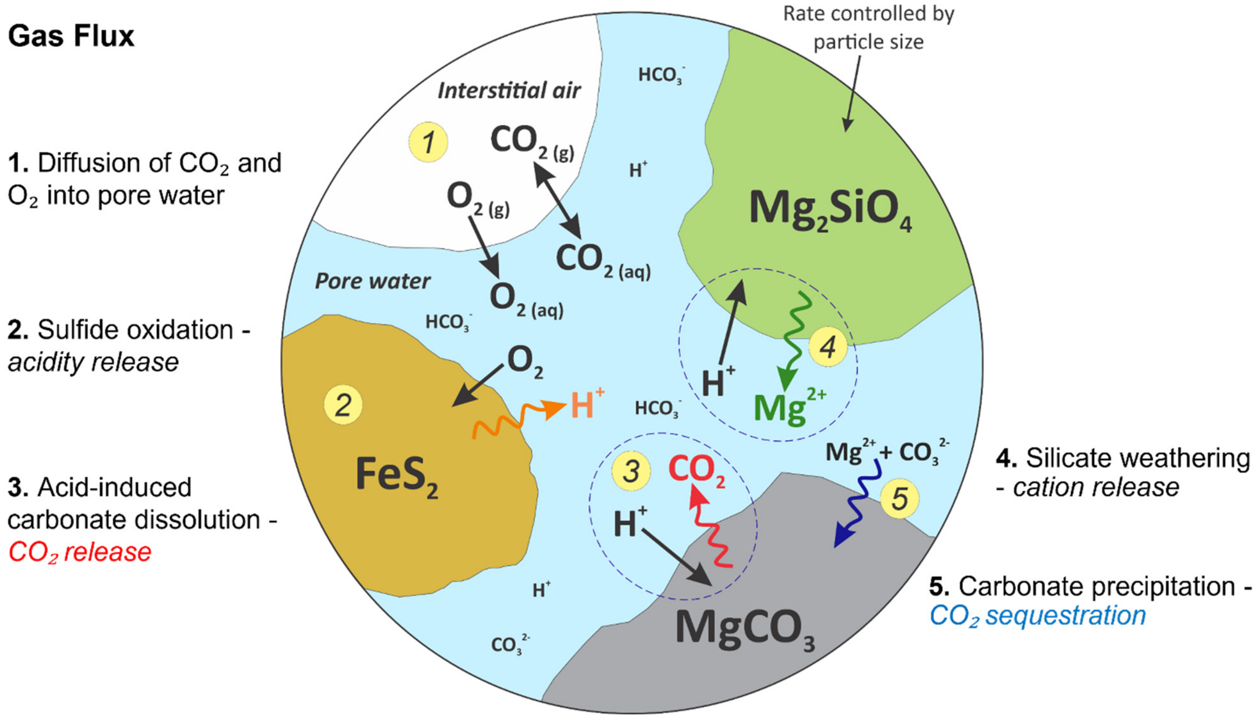


Figure 1 Simplified schematic diagram of pore gas flux in mine wastes (modified from Gras et al. 2017)

In the waste rock and tailings used within this study, the dominant sulphide and carbonate mineral phases are pyrrhotite and dolomite. The oxidation reaction for pyrrhotite (FeS) and the precipitation of iron generates acidity followed by dolomite induced dissolution, and the release of CO_2. The stoichiometric relationship between pyrrhotite and dolomite neutralisation shows that for every 1 mole of O_2 consumed, 0.44 moles of CO_2 is produced (Barnes et al. 2022; Shiimi et al. 2024). The kinetics of this process are controlled by several rate-limiting factors: mineralogy, reactive surface area (related material granulometry), pH, temperature, oxygen and carbon dioxide concentration and water saturation (Stokreef et al. 2022).

2 Methodology

This study expands on previous work by Savage (2019, 2023), Shiimi (2022), Schoen (2022), Cole (2023) and Clancy (2024), and aims to further develop methods to monitor CO_2 and O_2 flux from mine waste and assess carbon sequestration potential through ERW and MRV frameworks. Measuring CO_2 and O_2 flux from mine waste samples was identified as a viable method of measuring the relative balance between sulphide oxidation (and oxygen consumption), carbonate dissolution (CO_2 release) and carbonation (CO_2 sequestration) rates within mine WSFs. This study presents a novel and potentially scalable protocol for measuring rates of the reactions explored above. The experimental framework encompasses distinct methodological approaches:

1. the utilisation of custom-sealed reaction cells known as Xylem WTW OxiTop®-C/B (WTW Soil Biological Oxygen Demand [BOD]) (Platen & Wirtz 2010), designed for controlled oxidation and dissolution experiments
2. large stainless-steel closed-barrel experiments to evaluate short-term to medium-term geochemical interactions occurring within covered mine waste material under varying conditions
3. large open-barrel experiment to simulate geochemical interactions and gas flux in an open system under varying conditions.

2.1 Sample characterisation

Mine waste samples are systematically collected from waste rock storage facilities and tailings storage facilities of a nickel-copper-platinum group metal mine in Northern Europe, to ensure representative characterisation of geological and geochemical properties. All testing was undertaken at the Geochemic Ltd. laboratory facility located in Wales, UK. Selective geochemical and mineralogical characterisation was undertaken at Cardiff University and Petrolab, UK. Sample characterisation adhered to British and European (EU) standards. Analytical geochemical, mineralogical, elemental and geotechnical characterisation techniques (Table 1) were employed to verify and validate the accuracy of instrument-derived datasets, ensuring robust measurement reliability and interpretive precision.

Table 1 Mine waste sample characterisation (adapted from Clancy 2024)

	Waste sample characterisation			
	Mineralogical	**Elemental**	**Geochemical**	**Geotechnical**
Analytical method	Scanning electron microscope energy-dispersive X-ray microscopy (SEM-DEX)	Energy-dispersive X-ray fluorescence (ED-XRF)	Net acid generation, acid neutralising capacity, total carbon and sulphur, pH, electrical conductivity (EC)	Specific gravity, particle size distribution, water content
Objectives	Identify key potential oxidation and carbonation mineral assemblages	Elemental distribution, carbon capture potential (CCP)	Measure of carbon and sulphur, acid base accounting (ABA)	Simulate field conditions, calculate total gas volume, particle size affects surface area and reactivity

From the elemental and oxide data collected, the theoretical CCP can be calculated using a modified Steinour equation (Renforth 2019) (Equation 2).

$$\mathrm{CCP} = 10 \times \left(\frac{\mathrm{CaO}\%}{\mathrm{CaO\ mol}} + \frac{\mathrm{MgO}\%}{\mathrm{MgO\ mol}} + \frac{\mathrm{Na_2O}\ \%}{\mathrm{Na_2O\ mol}} + \frac{\mathrm{K_2O}\ \%}{\mathrm{K_2O\ mol}} - \frac{\mathrm{S}\%}{\mathrm{S\ mol}} - \frac{\mathrm{P_2O_5}\%}{\mathrm{P_2O_5 mol}}\right) \times \mathrm{CO_2\ mol} \tag{2}$$

where:

metal oxide % = mass %

2.2 Closed-system experiments

2.2.1 Xylem WTW OxiTop® (OxiTops and Carbitops)

Laboratory-scale closed experiments utilised established methodologies to evaluate carbon capture. The WTW OxiTop®-C and OxiTop®-C/B pressure measuring heads, designed to determine aerobic respiration rates according to EN ISO 16072:2002 (International Organization for Standardization 2011) were used within this study to determine the carbon balance of waste rock and tailings materials in a closed-system experiment (Shiimi et al. 2024).

The OxiTops experiments (Figure 2a), conducted over 30 days at temperatures ranging from 10–25°C and moisture contents of 7.5–22%, followed the WTW Soil BOD manual (Platen & Wirtz 2010) to measure O_2 consumption and CO_2 produced using a sodium hydroxide (NaOH) solution (Shiimi et al. 2024; Shiimi et al. 2023). The NaOH absorbent media was back titrated to estimate the total CO_2 emitted by waste samples over time and total inorganic carbon (TIC) analysis was carried out. The back titration and TIC methods were carried out to allow validation of the NaOH solution method.

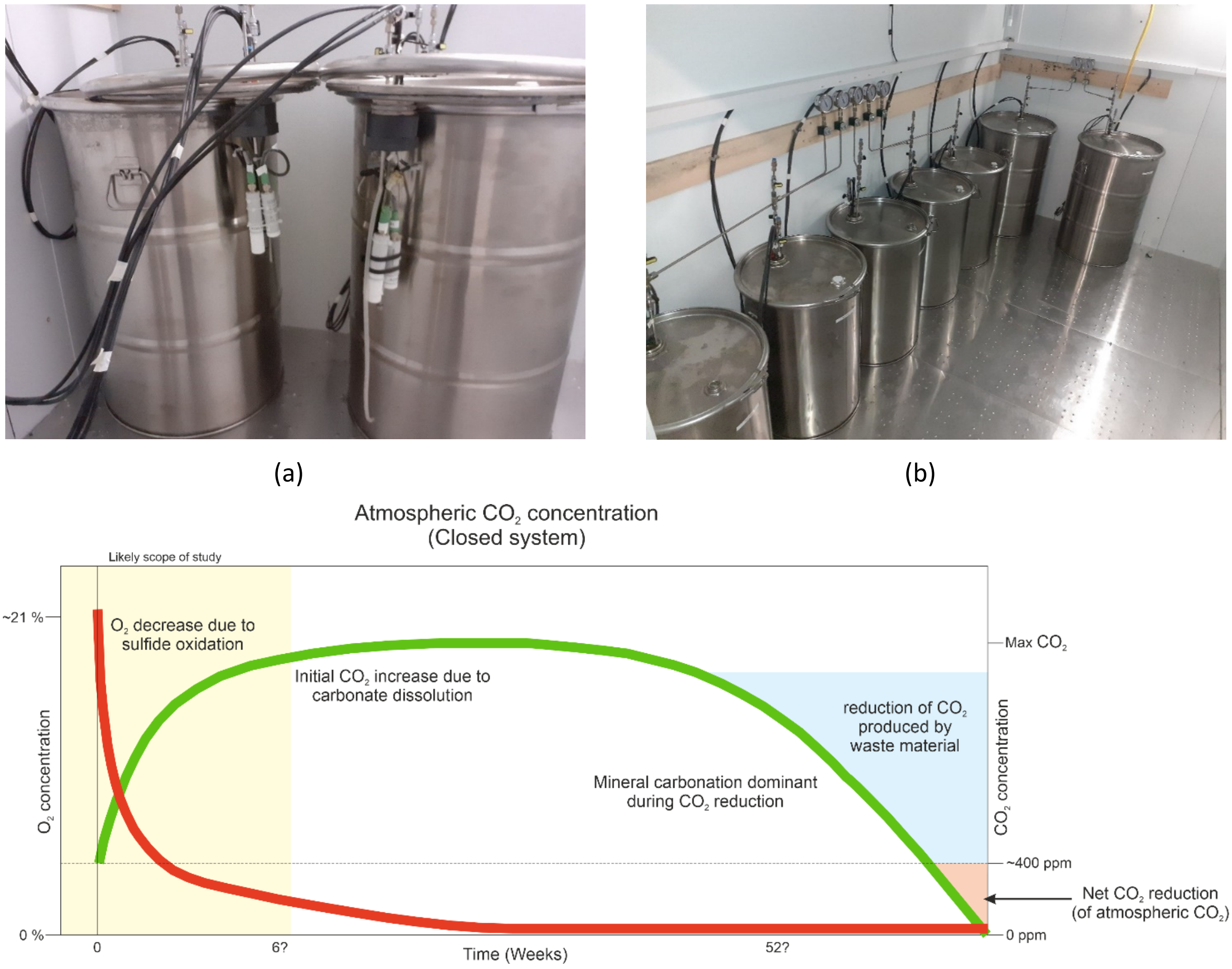
Carbitops experiments (Figureb), also conducted over 30 days, were performed at various CO_2 concentrations (5–10% CO_2), 7.5–22% moisture content and 10–25°C to assess CO_2 consumption under elevated CO_2 conditions (Shiimi et al. 2024; Shiimi et al. 2023).

(a) (b)

Figure 2 (a) OxiTop vessel setup with mine waste material and a suspended NaOH solution at an ambient atmosphere, placed in a temperature-controlled incubator; (b) Carbitop vessel setup with mine waste material at an ambient atmosphere, placed in a temperature-controlled incubator

2.2.2 Closed barrel cells

Sealed barrel experiments (Figure 3b), spanning over 12 months, employed bespoke vessels developed by Schoen et al. (2023), using controlled starting gas compositions (O_2, CO_2) at 8–10°C and ~7.5–15% moisture content. The barrels were partially filled with mine wastes (crushed waste rock or tailings) and fitted with CO_2 and O_2 probes (Figure 3a) within the available air space of cells, to log gas flux over time. Throughout these experiments, pressure and gas concentration changes were monitored to evaluate CO_2 production and consumption rates, enabling detailed carbon balance calculations.



(a) (b)

(c)

Figure 3 (a) Barrels setup and assembled probes pre-sealing; (b) Barrels placed in a cold room (about 8°C); (c) Idealised plot showing expected results in atmospheric CO_2 barrel without considering biological reactions

The anticipated variations in gas flux within the headspace of the sealed barrels were expected to differ throughout the logging period, due to differing experimental conditions and dynamic geochemical interactions. As biological reactions are not considered, barrels with atmospheric gas composition (Figure 3c) are expected to result in an initial rise in CO_2 concentration; coupled with an O_2 decrease due to the oxidation of sulphides leading to acidity-induced carbonate dissolution (Stokreef et al. 2022). Barrels with elevated CO_2 compositions are expected to see a decrease in CO_2, with the negligible O_2 supply leading to the rate of CO_2 sink reactions (silicate dissolution) being dominant over CO_2 source reactions (sulphide oxidation and subsequent carbonate dissolution).

2.3 Open-system experiment

An open barrel test (Figure) was designed to simulate mine site field conditions in Northern Europe, providing a platform to evaluate sensor performance before upscaling to field trials (Clancy 2024). The experiment was conducted under atmospheric O_2 and CO_2 conditions at a temperature range of 8–10°C, with the primary objective of designing and establishing an open-barrel equipment test; to facilitate the monitoring of gas flux and geochemical interactions in mine tailings within a semi-controlled open system. This development introduces a novel approach to advancing the understanding of mineral carbonation processes in mining environments. As a component of a broader EU-funded carbon sequestration initiative, open system experimentation is integrated into the strategy for upscaling to field trials, aiming to assess feasibility and effectiveness in real-world applications.


(a) (b)

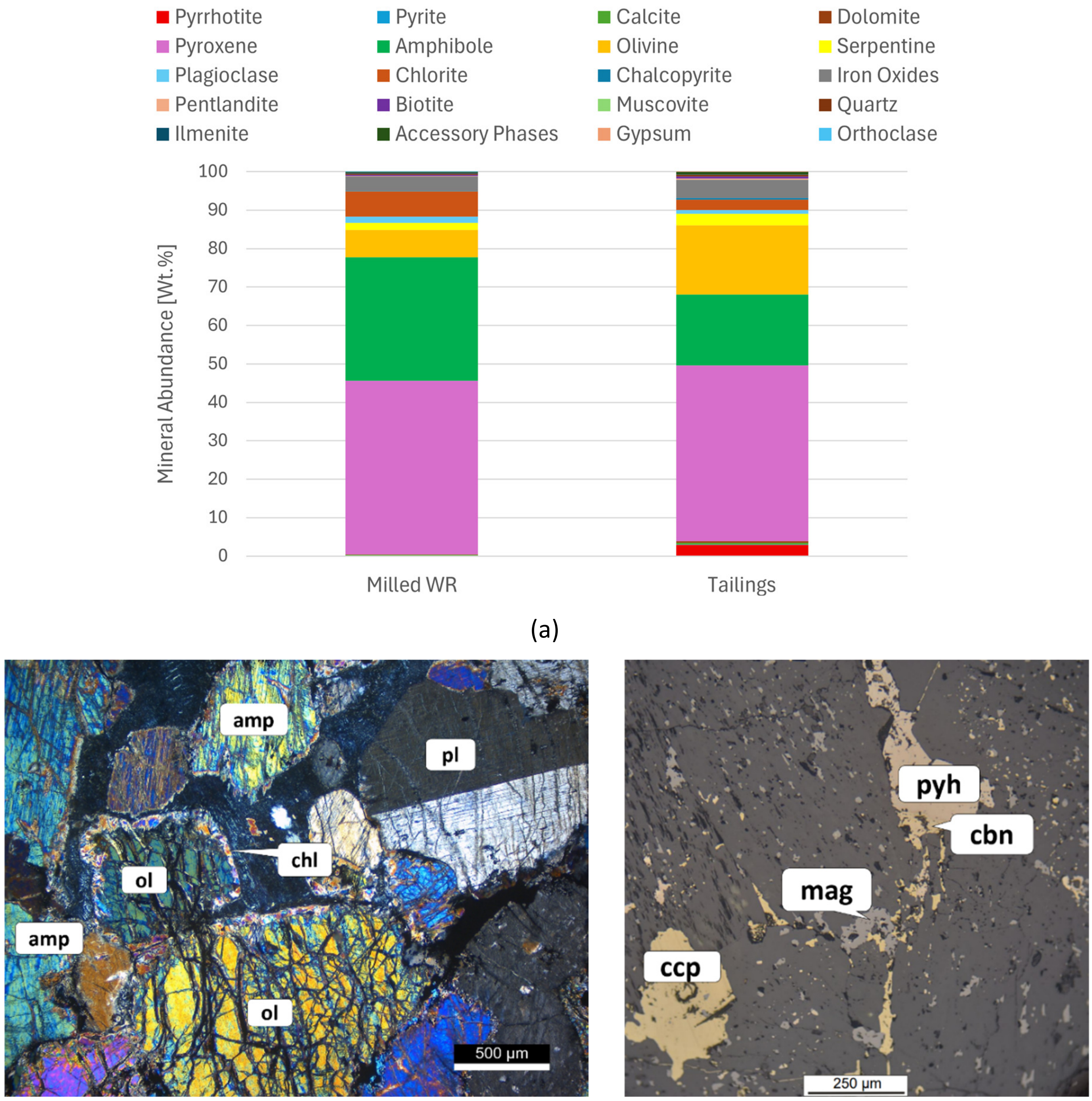
Figure 4 (a) Open barrel partially filled with tailings material and buried probes; (b) Open barrel place in a cold room (about 8°C)

3 Results

3.1 Characterisation of ultramafic mine waste

The tailings and waste rock materials analysed in this study consist of a low-sulphide mafic/ultramafic lithology (Figure 5), characterised by its potential to undergo carbonation weathering reactions upon exposure to ambient environmental conditions. Automated mineralogy data (Figure 5a) show that the mineralogy is similar between the waste rock and tailings. Both samples have a broadly mafic mineral assemblage dominated by pyroxene (~45 wt.%), amphibole (~20–30 wt.%) and olivine (~10–20 wt.%). Diopside and enstatite are the primary Mg deportment minerals (and minor tremolite), while Ca deportment is predominantly in diopside. Mineral carbonation is mainly due to the dissolution of diopside, enstatite, olivine and tremolite to precipitate secondary carbonate minerals. Mineralogical analysis identified pyrrhotite (~0.1–2.9 wt.%) and dolomite (0.1–0.4 wt.%) to be the main sulphide and carbonate phases respectively. Therefore, sulphide oxidation within the mine wastes is primarily due to pyrrhotite, and neutralisation of sulfuric acid is due to dolomite and silicate weathering (Figure 5b and Figure 5c).

The CCP of mine waste material can be calculated from the elemental analysis results. The waste rock recorded higher CCP, ~370 kg CO_2/tonne, compared to the tailings, which recorded a CCP ~330 kg CO_2/tonne. The waste rock reported higher Mg concentrations than tailings, which might explain the reduced CCP in tailings – as Mg plays a critical role in carbon capture.



(a)

(b)

(c)

Figure 5 **(a) Phase abundances chart for the waste rock tailings samples; (b) Photomicrographs of waste rock showing fracture olivine, chlorite interstitial areas, plagioclase and amphibole alteration; (c) Photomicrographs of waste rock showing pyrrhotite, cubanite, chalcopyrite and fine disseminated magnetite**

3.2 The carbon balance in mine wastes

The OxiTop and Carbitop results (Table 2) can be utilised to estimate a carbon balance, enabling the quantification of a net CO_2 flux within a system (Figure 6). The net carbon balance (Net CO_2) can be calculated using the equation below (Shiimi 2023) (Equation 3).

$$\text{Net } CO_2 \text{ (Sequestration/Emission)} = CO_2 \text{ Release} - CO_2 \text{ Consumption} \quad (3)$$

where:

net CO_2 sequestration = <0

net CO_2 emission = >0

Under high-temperature conditions (25°C), the tailings have the potential for net CO_2 sequestration, while the waste rock has the potential for net CO_2 emissions (Figure 6a). The estimated average net CO_2 sequestration rate in tailings is 0.75 kg (CO_2)/tonne/year, the average net CO_2 emissions in waste rock is 0.31 kg (CO_2)/tonne/year.

Under low-temperature conditions (10°C), both waste rock and tailings have the potential for net CO_2 sequestration (Figure 6b). The estimated average net CO_2 sequestration rate is 1.21 kg (CO_2)/tonne/year for tailings and 1.14 kg (CO_2)/tonne/year for waste rock.

Table 2 Summary of estimated average of oxidation rate, CO_2 released estimated from the back titration and TIC analysis and estimated net CO_2 released and sequestered (carbon balance) in mine waste rock and tailings

Material	Temperature (°C)	Moisture content (%)	Oxidation rate (kg (O_2)/ tonne/year)	Average CO_2 release (between back titration and TIC)	CO_2 consumption	Net CO_2 sequestration (<0) or emission (>0)
					(kg (CO_2)/tonne/year)	
Tailings	25	15.6	1.43	0.56	1.30	−0.75
Waste rock	25	7.5	2.87	1.28	0.98	0.31
Tailings	10	20.8	3.64	0.80	2.01	−1.21
Waste rock	10	7.5	0.61	0.06	1.21	−1.14

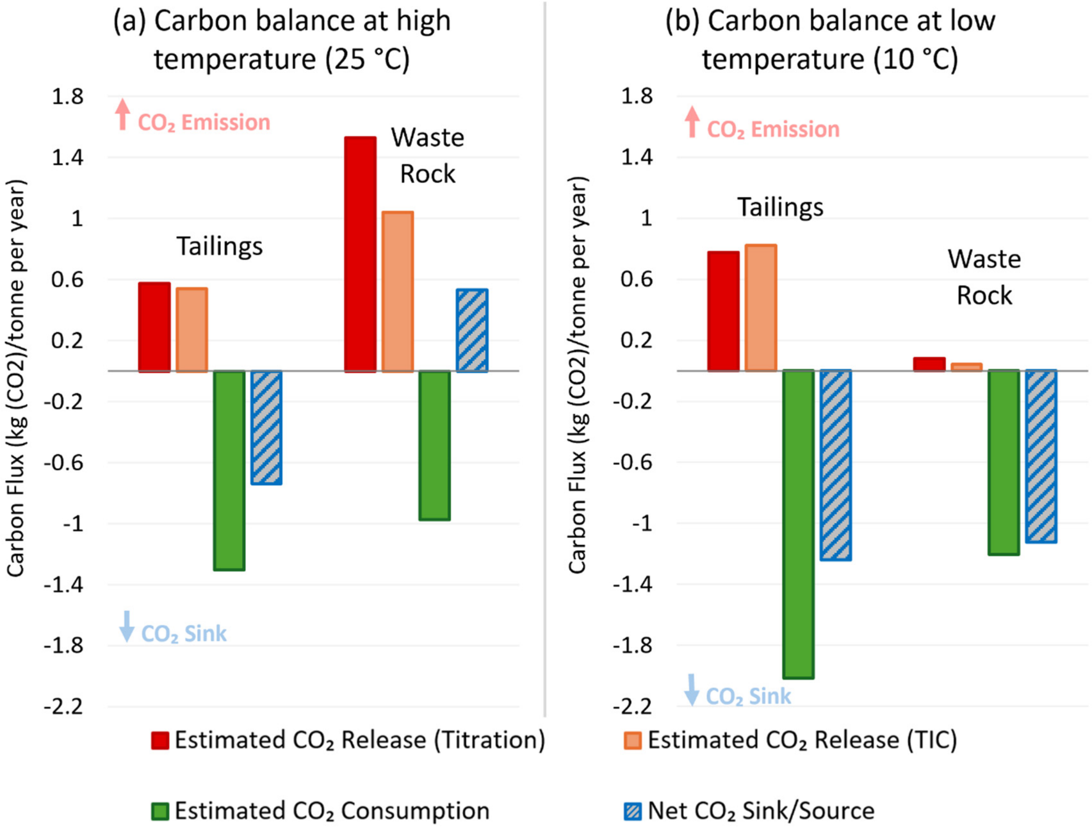


Figure 6 **Net CO_2 emissions and sequestration results estimated from the difference between CO_2 consumption (estimated from Carbitops) and CO_2 release (estimated from titration and TIC methods) (a) at high temperatures and (b) at low temperatures**

3.3 Passive carbonation in waste rock

Closed Barrel 6 (filled with waste rock) and the control closed Barrel 7, both started at atmospheric CO_2 concentrations (419 ppm). In Barrel 6, an initial decrease in CO_2 was observed after 5 days of logging before increasing and reaching a maximum of 1.52% CO_2 over the subsequent 18 months (Figure 7). A corresponding decrease in O_2 of 19.16% over 18 months was observed in Barrel 6 (the starting O_2 concentration was 19.16%). From April 2024 (month 19) the CO_2 has gradually decreased to ~1.19% in early February 2025. Barrel 7, containing quartz chips, serves as a control; maintaining stable O_2 and CO_2 concentrations throughout the experimental period. To date (April 2025) Barrel 6 and 7 experiments are ongoing. The O_2 and CO_2 concentrations (%) in Barrel 6 are quantitatively converted to their respective mass and molar values, which are subsequently used to determine the flux rate expressed in kg/tonne/year (Table 3).

Table 3 Summary of estimated O_2 consumption rates and CO_2 sequestration/release rates in Barrel 6

Barrel 6	O_2 consumption	CO_2 consumption (<0)/release (>0)	Remark
	(kg (O_2/CO_2) per tonne of waste rock per year)		
Rate 1	0.240	0.009	O_2 consumption rate from month 1–18. CO_2 production rate from month 3–18
Rate 2	0.001	−0.018	O_2 consumption rate from month 19–28. CO_2 Consumption rate from month 19–28

During the initial 18 month period of CO_2 release (Figure 7), the net CO_2 production rate (estimated from month 3–18) within the waste rock is estimated to be 0.009 kg/tonne/year. Following the depletion of O_2 within the system, CO_2 consumption became the predominant process starting in month 19 (Figure 7). The estimated net CO_2 sequestration rate in the waste rock is 0.018 kg/tonne/year.

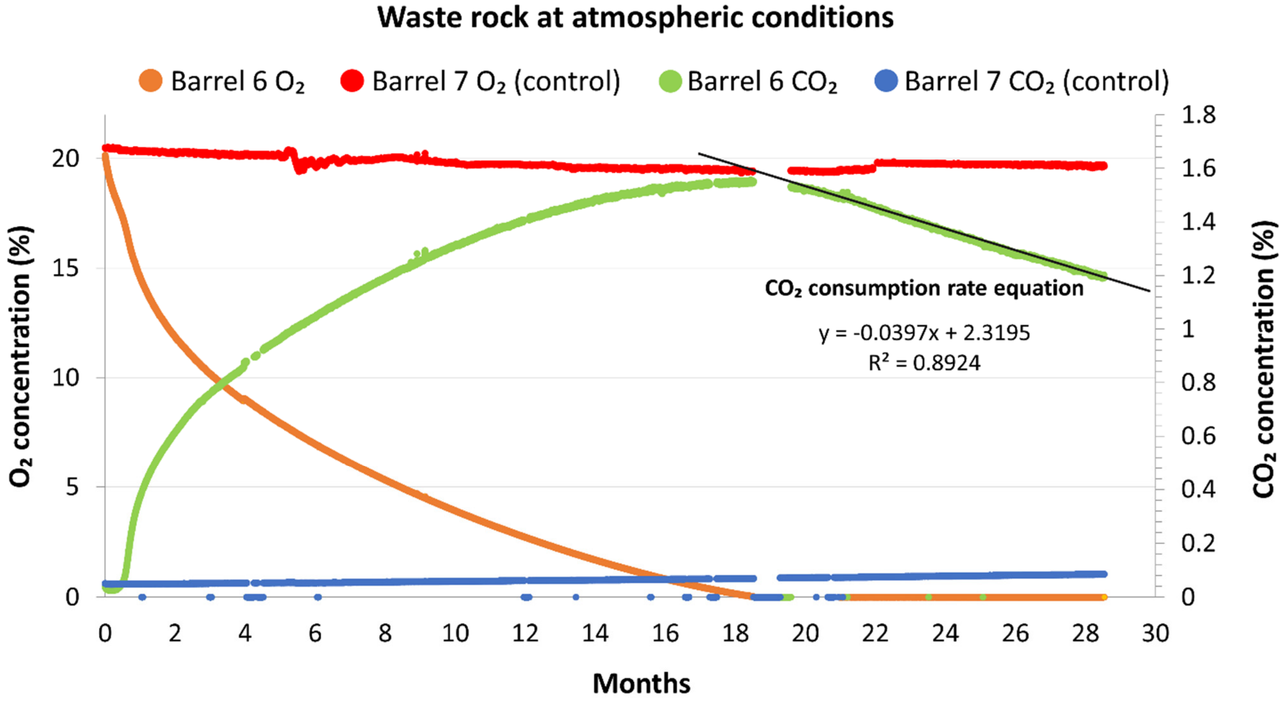


Figure 7 CO_2 and O_2 concentrations within Barrels 6 and 7 at atmospheric conditions. The CO_2 consumption rate equation is presented, exhibiting a linear relationship with a coefficient of determination (R^2) of 0.89 for N = 7,850 (where N is number of data), indicating a strong correlation

4 Discussion

4.1 Nature of the closed-system experiments

O_2 and CO_2 flux in mine waste materials was evaluated using straightforward and cost-effective methodologies, providing valuable insight into gas exchange dynamics. The Xylem OxiTop® (OxiTop) method is used to estimate oxygen consumption rates, providing insight into sulphide oxidation processes. When coupled with carbonate dissolution, this oxidation is directly linked to CO_2 production. Oxidation was due to the presence of sulphides (pyrrhotite) within mine waste material. Consequently, findings from this study show that mine wastes generate CO_2 emissions, which must be included in overall CDR quantification. The Carbitop setup was used to estimate the CO_2 sequestration rates of the material. Reactive Mg-bearing silicate minerals such as olivine, diopside and enstatite in the samples were able to sequester CO_2 through mineral carbonation.

The carbon balance quantifies the equilibrium between CO_2 emissions and sequestration within mine waste materials. This process is primarily governed by sulphide oxidation and carbonate dissolution, which contribute to CO_2 release, while silicate weathering promotes carbonate precipitation through mineral carbonation; facilitating long-term carbon storage. Under atmospheric gas conditions, waste rock material may produce more CO_2 due to sulphide oxidation and carbonate dissolution than it can consume, making waste rock material a net ‘carbon source’ (overall CO_2 emission). However, the results also indicate that under conditions of reduced O_2 availability, such as in an encapsulated waste storage facility following closure, CO_2 production from waste rock decreases due to the suppression of sulphide oxidation processes. As such, the results suggest that the carbon balance of the system may reverse and become net ‘carbon sink’ (sequesters more CO_2 than it emits). Overall, results indicate that both tailings and waste rock have the potential to sequester CO_2 and store it, as a long-term sink for carbon.

It was hypothesised that the waste rock Barrel 6 would observe a steady decrease in O_2, and an increase in CO_2, due to sulphide oxidation and carbonate dissolution, respectively. Once the O_2 is depleted, mineral carbonation becomes the predominant reaction, leading to a progressive reduction in CO_2 concentrations, potentially resulting in net-neutral or negative CO_2 emissions. This hypothesis has remained valid thus far and once the O_2 was depleted within the barrel, a continuous decrease in CO_2 concentrations has been observed. The results from Barrel 6 indicate that the system initially functioned as a net carbon source. However, following the complete depletion of O_2 through sulphide oxidation processes, the closed system transitioned to a carbon sink.

4.2 New emerging technique

Initial sensor readings from the open barrel equipment test are currently being recorded; however, the timeline for carbonation development remains uncertain, and comprehensive trends will only become apparent as data collection progresses. Long-term monitoring will be essential to characterise the kinetics of mineral carbonation and identify key environmental and geochemical factors influencing mineral carbonation and CO_2 sequestration efficiency in mine wastes. Future field trials will provide further validation, enabling the refinement of mineral carbonation process insights and the optimisation of carbon sequestration strategies for large-scale applications.

4.3 Implications for mine operations and mine closure

Research has demonstrated that mine wastes (waste rock and tailings) of suitable mineralogy have the potential to sequester large amounts of CO_2 through ERW (Herzog 2002; Rackley 2017; and Savage et al. 2019). While ERW shows great potential in laboratory and small-scale field experiments (Schoen et al. 2023; Shiimi et al. 2023), several challenges arise when trying to implement it on a larger scale in an open system. Limited studies of CO_2 emissions from mine WSFs are another key challenge that deters the upscaling of carbon capture and storage (CCS) in mine wastes (Savage et al. 2024).

Assessing pore gas fluxes and the carbon balance within mine waste facilities will likely be an aspect to consider for long-term environmental impact assessment of mine waste management and storage and identifying opportunities for large-scale CDR. Measurements of pore gas flux in mine wastes will aid in planning waste management strategies and decision-making prior to, during and post waste deposition. The results suggest that mineral carbonation process can enhance acid mine drainage or metal leaching/acid rock drainage (ML/ARD) risk management in WSFs by increasing carbonate mineral mass, thereby improving the acid buffering capacity of waste materials. The integration of CCS into mining operations will play a crucial role in the movement towards greener and more sustainable mine practices, while also having the potential to aid valorisation of mine wastes through enhance metal recovery and other potential co-benefits.

5 Conclusion

The experimental methods and findings demonstrate a promising, cost-effective approach for directly measuring CO_2 and O_2 flux within mine wastes. This research has advanced carbon sequestration strategies through the utilisation of mine waste materials, demonstrating their potential for effective CO_2 capture and storage. The OxiTop method effectively quantified CO_2 emission rates from mine waste materials, while the Carbitop system measured CO_2 sequestration rates within mine wastes, providing a comprehensive assessment of carbon flux dynamics. The barrel method enabled a larger-scale, long-term assessment of pore gas evolution, simulating site-specific conditions. In parallel, the open barrel equipment test evaluated sensor performance in an open system, replicating site conditions before scaling up to field trials.

Given that most tailing storage facilities are saturated (whereas experiments whose results are presented in this article were done in unsaturated conditions), the CO_2 diffusion rate into the facility is slower than O_2 from the atmosphere. As such, the likely scenario within the tailings is that CO_2 production from sulphide oxidation is taken up by silicate minerals through carbon sequestration. Therefore, the tailings facility has the potential for net negative or neutral CO_2 emissions.

Results from closed-system experiments suggest that under conditions where pore gas is close to atmospheric composition, CO_2 emission rates from waste rock are likely higher than CO_2 uptake. However, results also indicate that where pore gas conditions are O_2 limited, waste rock has the potential to become CO_2 neutral/sequestering; for example, mine waste rock stored within a facility that has an engineered oxygen limiting design such as low permeability cover system have the potential to become neutral.

The testing results obtained from the outlined methods enable both relative and quantitative evaluation of pore gas flux in mine waste under defined scenarios, providing insights into gas exchange dynamics and geochemical interactions. Additionally, the methods provide valuable tools for operational and closure mine planning, specifically for the design of WSFs and assessment of opportunities and risks related to ML/ARD and CO_2 emissions/sequestration from the short-term to long-term storage of mine waste. Future research will look to further assess pore gas flux of wastes from other mafic/ultramafic mine sites (at variable mineralogy, geochemistry, moisture, temperature, etc.), by utilising this study's methodology.

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