

Optimizing the management of quarry fines for on-site carbon removal: Implications of grain size and mineralogy on CO₂ mineralization

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ABSTRACT

Weathering of basaltic quarry fines can enable quarries to remove CO₂ by optimizing the management of underutilized rock fines. In this study, basaltic fines from two quarries in Scotland are used as potential feedstocks for ERW. Using column experiments, fines from both sites were placed into columns as layers with varying thicknesses (1 cm and 5 cm) and grain sizes (bulk and <100 µm). Fines were saturated (~60 % pore water) and exposed to ambient UK conditions (10 °C, 0.04 % CO₂) and accelerated carbonation conditions (50 °C, 20 % CO₂). Quarry site 1 experienced negligible increases in TIC within bulk fines under ambient conditions, yet fines <100 µm experienced carbonation equivalent to 440 g CO₂/m²/yr. However, the total inorganic carbon content (TIC) nearly doubled in the bulk fines from quarry site 2 (5 cm) under ambient conditions, equivalent to 570 g CO₂/m²/yr. In the sieved fines from the same site the TIC content nearly tripled, equivalent to 1330 g CO₂/m²/yr. At site 2, if the bulk fines could be deposited over 0.8 km² of land in 5 cm thicknesses, approximately 460 t CO₂/yr could be sequestered with minimal management practices in place. Using fresh fines that have not previously weathered in stockpiles is important for maximizing the carbon dioxide removal potential. Despite higher carbon offsets within the sieved material, the energy and cost required to crush rock from bulk to <100 µm is not economically feasible, as it exceeds the value of carbon which it could be sold for.

1. Introduction

While society must decrease the total output of global greenhouse gas emissions (GHGs), there is also a need for large-scale deployment of sustainable carbon dioxide removal (CDR) strategies to meet the objectives of the Paris Agreement (IPCC, 2022). Chemical weathering of silicate-rich rocks removes atmospheric carbon dioxide (CO₂) over geological timescales (Sundquist, 1993), but it can also be expedited through the design and implementation of various CDR approaches (Strelfer et al., 2018). These acceleration strategies can effectively remove CO₂ on decadal timescales, contributing to the attainment of global GHG reduction targets. Enhanced rock weathering (ERW) is a recognized CDR strategy that involves speeding up CO₂ sequestration via mineral weathering by spreading out pulverized silicate rocks over different landscapes (e.g., agricultural field, coastal regions, mines;

Renforth, 2012). During this process atmospheric CO₂ dissolves into pore waters, and hydrates to form carbonic acid, which can then be neutralized through the reaction of different Ca- and Mg-bearing silicate minerals (e.g., olivine, wollastonite, plagioclase). Atmospheric CO₂ is first trapped as a dissolved phase but may eventually become mineralized to form a stable sink for CO₂ (Power et al., 2013) as shown in reactions (1)–(5):



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Basalt has gained considerable recognition for ERW as it is highly abundant, contains minerals that are relatively fast weathering, and is rich in cations needed for carbonation (Ca^{2+} and Mg^{2+} ; Lewis et al., 2021). Typically, ERW studies have focused on amending agricultural soils with silicate bearing rocks (Kelland et al., 2020; Wood et al., 2023) to promote crop yield while also storing atmospheric CO_2 (Haque et al., 2019). Although there are many benefits to using basalt including CDR, improved soil health and increased production of crops (Edwards et al., 2017), basalt may also have the potential to be used in the aggregate and mining industry for on-site CDR without needing to be transported to another application site like an agricultural field. In regions where ERW in soil is not currently implemented, this could serve as a potential CDR strategy for the mining and quarrying industry, helping to offset some of their emissions and further contribute to global CO_2 reduction efforts. There is currently limited knowledge about how active or closed quarries may be able to remove emissions on-site at relatively low cost and energy, which is one of the primary goals of this study.

The use of mine tailings as a feedstock for CO_2 sequestration has been well documented and offers promising solutions for safe and long-term storage of CO_2 , as demonstrated in studies by Power et al. (2014), Stubbs et al. (2022), and Wilson et al. (2014). However, mafic quarry fines, despite their abundance and ideal mineralogical and geochemical compositions, have been largely overlooked for on-site CDR. Some earlier research explored the use of basaltic quarry fines mixed with green waste compost to produce artificial soils that are suitable for passive carbon sequestration (Manning et al., 2013). The rate of carbonation in mine tailings under ambient conditions is dependent on four primary factors: 1) the characteristics of the waste material, 2) local climate conditions, 3) process and pore water chemistry, and 4) the design and management of storage facilities (Power et al., 2014). While the first three factors cannot be modified, the design of tailings storage facilities (TSFs) can be changed and optimized for CO_2 sequestration. The TSFs at mine sites are large (i.e., several km²; Wilson et al., 2014) however at quarries, fines are generally stocked in numerous piles as there is typically much less material. If these fines can be dispersed throughout different areas of the quarries and left to weather, there may be potential for reducing CO_2 emissions on site. Various studies have highlighted the influence of material depth (Kandji et al., 2017) or grain size (Rinder and von Hagke, 2021) on rock waste carbonation; however, these two factors have not been explored simultaneously in basaltic fines. The present study aims to address how these two factors may affect the capacity for CDR on-site under ambient conditions.

The United Kingdom (UK) hosts approximately 1750 active quarries (Cameron et al., 2020), many of which produce aggregates from basalt or other rock types with similar compositions, such as dolerite. Of particular relevance to this study, there are around 37 mafic quarries in Scotland: 15 are basalt, 17 are dolerite, and 5 are gabbroic (Cameron et al., 2020). The high abundance of mafic quarries in Scotland makes it a desirable location for sourcing basaltic rock as a CDR feedstock. Notably, these sites contain an abundance of unused quarry fines, often accumulated in large piles. With the recent surge in interest regarding basalt's potential for CDR, there is interest in exploring the utilization of these fines on-site as a strategy to reduce GHG emissions associated with quarry operations.

While there are numerous advantages to incorporating basalt in more conventional ERW scenarios, the availability of nearby land for the application of basalt to soils is not always guaranteed. Furthermore, the prospect of not needing to transport basalt off-site to agricultural or forested areas presents the additional benefit of reducing transportation-related emissions (Edwards et al., 2017). Due to the reactive nature of basalt fines, it is possible that passive (unintentional) carbonation may be occurring. By optimizing the on-site management of quarry fines to enhance mineral dissolution and CO_2 supply, there may be significant potential for reducing atmospheric CO_2 emissions.

The primary goals of this study are to 1) understand how mineralogy, particle size, and sample thickness affect the capacity of CO_2 removal in

basaltic fines, 2) estimate the CO_2 removal rate for both samples under ambient and accelerated conditions, 3) consider how fines could be managed at quarries to optimize CDR, and 4) estimate the approximate monetary value of carbon stored over time. This study focuses solely on the potential of spreading a layer of rock dust at industrial quarry sites to determine if carbonation does occur, without mixing it with other materials like soil. To accomplish these goals, laboratory experiments were performed in an environmental chamber and a CO_2 incubator to accelerate carbonation that may occur.

2. Methods

2.1. Fieldwork and sampling

Two mafic quarries in Scotland were selected for this study based on site accessibility and abundance of available fine material (geological background provided as supplementary material). Fieldwork and sampling were conducted at both quarries (referred to as Quarry 1 and 2) in Scotland in March 2023. Quarry 1 is an operational site which extracts dolerite/gabbro from the Midland Valley Sills complex. In 2023, this quarry produced 97,500 t of dolerite/gabbro fines in stockpiles that are not in use for commercial purposes (Table 1). Other quarries of the same owner in Scotland and Northern England have fines available with untapped potential for CDR, ranging between 401 and 83,108 t.

Quarry 1 is also operational and extracts basaltic rock from the Kinghorn Volcanic Formation. This site began operations in the 1950s and will continue for years to come. At the time of writing, this site was processing approximately 140,000 t of extracted basaltic rock. When making specific single size products from the competent basaltic rock, between 30 and 40 % will result in a 'dust/fines' size by-product, equivalent to 42,000–56,000 tonnes at the time of this study (Table 1). As a result, it is important to find additional uses for this material. This material can be blended with sand for concrete sand products or added to lower grade fill products. However, there is also a benefit to finding a sustainable use for this material. If there is a benefit to using this material for CDR, it can act as a multi-functional product.

Bulk samples (10s kg) of quarry fines were collected from both sites and placed into large buckets. Although these fines are deposited dry from the crushing circuit, they were damp upon collection due to heavy rainfall at the time of sampling. It is unknown how long these fines had been sitting in the stockpile prior to collection. All fines were dried under ambient conditions prior to any analytical or experimental work. After the samples were dried, they were homogenized and sorted according to site. The majority of fines were used for experiments with a smaller subset of samples used for characterization of their mineralogical, geochemical, and physical properties.

2.2. Initial sample characterization

X-ray diffraction (XRD) patterns on the initial samples were collected using a Bruker AXS D8 Advance Diffractometer equipped with LYNXEYE Detector. Initial quarry fine samples were sent to ALS Laboratories in Ireland for whole rock analysis by Inductively Coupled Plasma - Atomic Emission Spectroscopy (ICP-AES). Detailed methods for XRD and whole rock analysis are provided as supplementary material.

The initial and final (post-experiment) TIC (%) were measured for all solid samples (100 mg) using a Model CM5017 Coulometer from UIC Inc. at Trent University. Blanks and calcium carbonate chelometric

Table 1

Site name, rock exploited, and annual production of fine material (minimum and maximum for site 2).

Site	Rock exploited	Annual production (t)
Quarry site 1	Dolerite/ Gabbro	97,500
Quarry site 2	Basalt	42,000–56,000

standards (CaCO_3 , Alfa Aesar, 99.95–100.05 %) were analyzed before running samples (triplicates) and again every ~15 samples. All samples were milled for 10 min using a Retsch Planetary Ball Mill before analysis to ensure complete reaction of carbonates within the samples. Detailed methods for the coulometer are provided as supplementary material.

Representative sub-samples (1000 g) of the bulk quarry fines from site 1 and 2 were produced using the coning and quartering method. Samples were sieved using a Matest A060–01 Auto Sieve Shaker through 13 different apertures ranging from 10 mm to 100 μm . Both samples were sieved for 10 min, and triplicate analysis was done. A N_2 adsorption method was used to calculate the Brunauer-Emmett-Teller (BET) specific surface areas (Brunauer et al., 1938) of both basaltic fines and their respective size fractions. Samples were degassed via heating under vacuum; this was done at 250 °C for 4 h, using a heating ramp rate of 10 °C/min. BET analysis was performed on the Micromeritics ASAP 2420 instrument.

Small white precipitates formed on the surface of dried fines from quarry 2 prior to experiments beginning were selectively sampled using tweezers. To understand the composition of these precipitates, Raman spectroscopy was performed using a Renishaw InVia Raman microscope within the Geoanalytical Electron Microscopy & Spectroscopy Centre at the University of Glasgow. A 514 nm monochromatic light and 1000 grating were used to identify the Raman shifts intensity, which was used for mineral identification. Data were processed using WiRE 4.1 software (Reinshaw) and they were indexed with reference to the RRUFF database (Lafuente et al., 2015).

2.3. Column carbonation experiments

To prepare for experiments, all quarry fine samples were deposited in thin layers (~2 cm thick) in the laboratory for 48 h to ensure the material was completely dry for any sample processing and experimental assembly. For experiments, half the samples were kept at their original grain size from initial sampling and are referred to as bulk fines throughout this study. The other half of samples were sieved to <100 μm using a Matest A060–01 Auto Sieve Shaker. The grain size of the finer fractions was determined using laser diffraction at the University of Glasgow. Replicates ($n = 3$) were thoroughly homogenized using a riffle box. Subsamples were then suspended in sodium hexametaphosphate [$(\text{NaPO}_3)_6$] and mixed for 1 min using a stir plate to prevent the fines from agglomerating. Using a disposable pipette, samples were extracted from the solution and injected inside a Bettersizer 2600 laser diffraction analyzer. During the measurements, the solution was sonicated to continue dispersing particles.

Although previous studies have demonstrated that finer fractions of basalt (<10 μm) are needed to achieve meaningful CDR in certain regions (Rinder and von Hagke, 2021), only a small portion of basalt would be usable if this material were further screened. Using basaltic rock dust <100 μm utilized ~30 % of the bulk quarry fine mass, therefore limiting waste compared to finer fractions. No other modifications were made to the samples.

For each of the columns, a transparent polyvinyl chloride (PVC) pipe (9.5 cm inner diameter) was placed on top of a rubber bung and pushed downwards to ensure there was an airtight seal. The experimental columns had two different heights; the first set were 14 cm tall and the other set of columns were 10 cm tall to allow for different sample thickness. In each of the columns, plastic discs (each were 0.5 cm thick with 9.5 cm outer diameter) were added to separate the quarry fines from any water that might drain. To achieve this, a 1 cm hole was drilled in the centre of the discs and 25 μm nylon sieve paper was cut to cover the entire disc, allowing only water to transfer to the bottom of the column. This sieve paper was placed on the same side the quarry fines would be in contact with to prevent particles >25 μm from passing through. Another plastic disc of the same size was added to the opposite side of the sieve paper and was attached with Araldite epoxy resin to ensure the sieve paper stayed in place. These discs were then placed

inside the column at 5 cm and 1 cm in depth for the taller (14 cm) and shorter (10 cm) experiments, respectively, to allow for the different sample thicknesses of 5 cm and 1 cm (Fig. 1). These depths were selected to test how sample thickness affects basalt weathering with respect to CO_2 sequestration. The rubber stoppers occupied 3.5 cm on the column space from the bottom, allowing for 5 cm of empty space for water to pool in both sets of columns (Fig. 1).

Quarry fines were homogenized and added to the columns in discrete layers (0.5 cm at a time) to avoid any sorting, until a total thickness of 5 and 1 cm was achieved. The mass required was dependent on the sample density (ρ_{sample}) and the particle size; however, every experiment had a thickness of either 5 or 1 cm and an exposed sample area of 70 cm^2 . The experiments with a sample thickness of 5 cm had 354 cm^3 of occupied space and the columns with a sample thickness of 1 cm had 70 cm^3 of occupied space. No other natural materials (e.g. soil or natural substrate) were added to these columns, as only the rock dust was present. After the samples were dried, deionized water was added to the column to achieve approximately 60 % pore water saturation to initiate the weathering cycle.

The bulk volume (V_{bulk} ; Eq. 6) and density (ρ_{bulk} ; Eq. 7) which includes voids was calculated to determine the porosity (Φ ; Eq. 8) of the quarry fines:

$$V_{\text{bulk}} = \pi r^2 \times h \quad (6)$$

$$\rho_{\text{bulk}} = \frac{m_{\text{sample}}}{V_{\text{bulk}}} \quad (7)$$

$$\Phi = 1 - \frac{\rho_{\text{bulk}}}{\rho_{\text{sample}}} \quad (8)$$

where r is the radius of the experimental column (cm), h is the thickness of the sample in the column (cm), m_{sample} is the mass of fines (g), and ρ_{sample} is the sample density (g/cm^3) which was calculated based on the measurements of mass and volume. The volume of solids (Eq. 9) and voids (Eq. 10) were then calculated to determine the volume of water needed to achieve 60 % pore water saturation (Eq. 11).

$$V_{\text{solids}} = \frac{m_{\text{sample}}}{\rho_{\text{sample}}} \quad (9)$$

$$V_{\text{voids}} = V_{\text{bulk}} - V_{\text{solids}} \quad (10)$$

$$V_{\text{H}_2\text{O}} = V_{\text{voids}} \times 0.60 \quad (11)$$

The V_{solids} is the volume (cm^3) of the quarry fines excluding voids, V_{voids} is the volume (cm^3) of voids in the fines, and $V_{\text{H}_2\text{O}}$ is the volume (cm^3) of water needed to achieve 60 % water saturation. This saturation was chosen as previous studies have reported that it is the high end of optimal water content for carbonation (30–60 %; Assima et al., 2013; Harrison et al., 2015; Stubbs et al., 2022). In the experiments conducted in this manuscript, the quarry fines were left to dry from 60 % to ~0 % saturation (except for moisture from humidity), allowing the samples to pass through the optimal water content range. Water was added every month, with a total of three wetting cycles per column, making the total experiment run time 3 months (summary of these properties is provided in Table 2).

The CDR rate (Eq. (12)) is calculated in $\text{g CO}_2/\text{m}^2/\text{yr}$:

$$\text{CDR rate} = \frac{[(\text{TIC}_f - \text{TIC}_i)/100\% \times m] \times m_{\text{CO}_2}/m_c}{A_c \times t} \quad (12)$$

where TIC_f and TIC_i are the final and initial TIC contents (%C), respectively. m_{CO_2} and m_c are the molar masses of CO_2 and C, respectively. m is the mass of material used in experiments (g) and A_c is the area (m^2) of the columns. t is the experiment duration (yr). To determine the amount of CO_2 captured in the fines at a given site (Eq. (13)), the CDR rate was multiplied to the area footprint of the respective

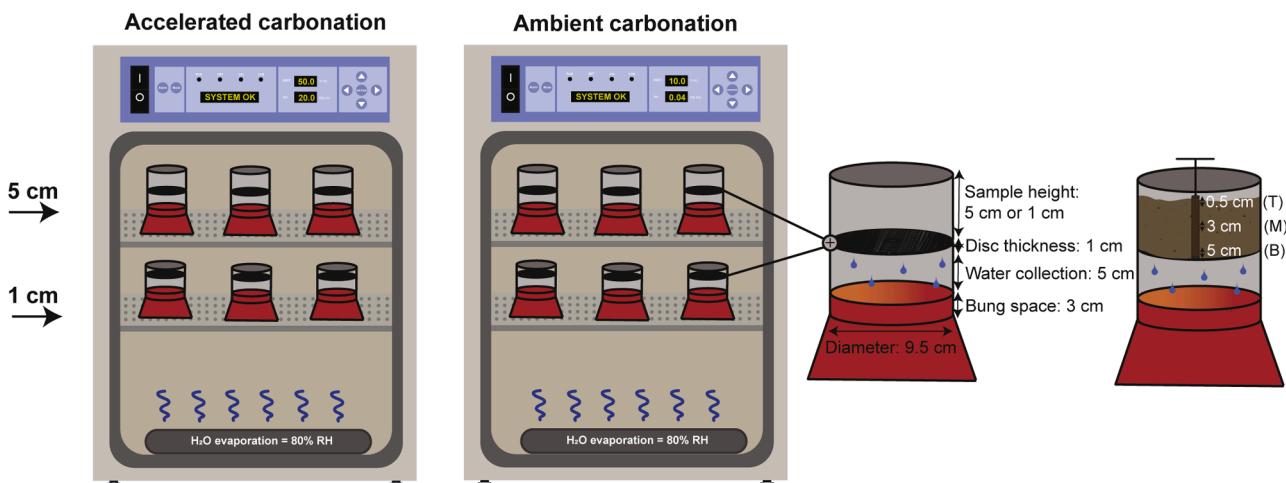


Fig. 1. Experimental setup used for basalt fines in both the CO₂ incubator (left) environmental chamber (middle). Samples in the incubator were exposed to 50 °C, 20 % CO₂, and 80 % RH. Samples in the environmental chamber were exposed to 10 °C, ambient (0.04 %) CO₂, and 80 % RH. The samples were added at a height of either 5 cm or 1 cm. Sample discs (1 cm) were placed below the thickness level to separate any fines from the water. A total height of 5 cm was added in case any water drained. Each stopper took up ~3 cm of space from the column to create an air and watertight seal at the bottom of the column.

Table 2
Physical properties for the experimental columns with bulk and sieved fines from quarry site 1 and 2.

Grain size	Sample thickness	Dry mass (g)	Water additions (count)	Bulk density (g/cm ³)	Sample density (g/cm ³)	Porosity (%)	Solids volume (cm ³)	Void volume (cm ³)	H ₂ O (mL)
Bulk	1	100	3	1.4	2.9	49	35	36	22
	5	500					172	182	110
Sieved	1	70	3	0.99	2.9	34	24	47	47
	5	350					121	234	140

site, and was then divided by the mass of fine occupying that area as follows:

$$\text{Annual CO}_2 \text{ uptake} = \text{CDR rate} \times \frac{A}{m_f} \quad (13)$$

where A is the required area of fines, calculated based on the volume of fines produced and their respective density (Eq. (7)), with an assumed thickness of 5 cm and m_f is the mass of fines produced at the respective site.

2.4. Experimental conditions

Experiments were carried out using two types of chambers to simulate different environments. The first set of columns utilized a Climacell Environmental Chamber to simulate an environment that is reflective of average annual temperature (10 °C) and relative humidity (80 %) in the United Kingdom, at an atmospheric CO₂ concentration of ~400 ppm to reflect global present-day conditions. The second chamber used was a Thermo-Scientific Series 8000 Direct-Heat CO₂ incubator (184 L). In this chamber the temperature was set to 50 °C, which is the maximum temperature that can be set on the instrument. The humidity was kept at 80 % to be consistent with the environmental chamber, and the CO₂ concentration was set to 20 %, more reflective of engineered carbonation conditions. These accelerated conditions were chosen to gain a better understanding of how much carbonation could occur over extended periods of time, given that it can take several months for weathering signals to be detected in certain feedstocks.

2.5. Experimental sampling and post-experiment characterisation

Upon experiment completion, each of the columns were cored (duplicates or triplicates) with subsamples of the fines taken 0.5 cm (T) from

the top, the middle 3 cm (M), and the bottom 5 cm (B) for the 5 cm thicknesses, and only in the top 0.5 cm with the 1 cm thick columns. Each of these samples were analysed for TIC following the methods previously described. Where the surface of the fines had hardened and agglomerated together to the point it was too difficult to core, small Sections (2 by 2 cm) were broken apart using metal spatulas to be examined using scanning electron microscopy (SEM). Samples were laid flat and embedded in epoxy and left to cure at 60 °C for 48 h. After the mounted sample fully hardened, a Logitech CS10 thin section 175 cut off saw was used to reduce the thickness, bringing the surface of the sample close to exposure. P240 grit abrasive paper was used to expose the surface of the sample, followed by P800, P1200, and P2500 to gradually create a polished surface. A Kemet 300 Series polisher coupled with a polishing pad and 1 µm aluminium oxide glycol was used to polish the sample for the final grade required for energy dispersive spectroscopy (EDS) analysis. The polished samples were carbon coated at a thickness of 20 nm. Backscatter electron (BSE) and EDS was conducted using a Zeiss Sigma VP Field-Emission Scanning Electron Microscope equipped with an Ultim Max (170 mm²) EDS detector based in the Geoanalytical Electron Microscopy and Spectroscopy (GEMS) facility at the School of Geographical and Earth Sciences at the University of Glasgow. Observations were done at an operating voltage of 20 kV and a working distance of 8 mm. BSE images and EDS signals were acquired across regions of interest and were analysed using Oxford Instrument AZtec ® 6.0.

To approximate the pH of pore waters from this study, slurries were made from the different basaltic fines. Aliquots (25 g) were mixed with deionized water (10 mL) in 50 mL centrifuge tubes. The solution pH was measured 1 h after mixing and again after 7 days using a VWR ® pHonomenal ® pH 1100 L meter. Results are provided as supplementary material.

3. Results

3.1. Initial quarry fine characterisation

In general, fines from site 1 and 2 have basaltic compositions (Table 3). Samples contain major abundances of desirable cations, with 8.49 wt.% and 8.65 wt.% CaO and 8.69 wt.% and 5.54 wt.% MgO for site 1 and 2, respectively. The rest of the samples was dominated by Al₂O₃, Fe₂O₃, Na₂O, TiO₂, and K₂O. All other oxides present were at abundances of <1 wt.%. The loss on ignition was 6.56 % and 3.51 % for each respective sample.

Bulk fines from site 2 were dominated by plagioclase (21.9 wt.%), pyroxene (43.0 wt.%), chlorite (8.5 wt.%), and olivine (9.7 wt.%). Some smectite was present in these bulk fines (2.5 wt.%). After site 2 fines were sieved to <100 µm, the plagioclase was twice as high, containing 40.0 wt.% plagioclase. Pyroxene, chlorite, and olivine were less than the initial samples at 18.9 wt.%, 6.5 wt.%, and 0.0 wt.%, respectively. Smectite concentrations were nearly 3 times higher in the sieved fines (7.2 wt.%) compared to the bulk fines while biotite was 16 times greater (0.8 wt.% to 13.2 wt.%). No calcite was detected in the bulk fines from quarry 1; however, 4.5 wt.% was present in the sieved material.

Amongst the fines from site 1, similar minerals were identified. Plagioclase was the most abundant phase at 46.5 wt.% followed by pyroxene (23.2 wt.%) and chlorite (13.5 wt.%). In the sieved material, plagioclase remained fairly consistent (47.9 wt.%), yet pyroxene reduced by more than half (13.0 wt.%). The chlorite content was greater (17.5 wt.%) in the sieved fines. Similar to site 2, the smectite content was greater in the sieved fines from site 1 (6.7 wt.%) compared to the bulk fines (4.2 wt.%). The calcite content was greater in the sieved fines by 1.0 wt.%. The full list of minerals identified in each of the samples can be found in Fig. 2 along with their respective abundances.

The carbonation potential (C_{pot}) for both bulk feedstocks was calculated using the modified Steinour formula presented in Renforth et al. (2019), however, the MgO content from chlorite and CaO content present in calcite was deducted from the raw whole-rock data. With these deductions taken into consideration, the C_{pot} for bulk fines from both sites is approximately 100 kg CO₂ y⁻¹.

At quarry site 1, the BET surface area increased with decreasing particle size. The bulk fines are dominated by sand size (0.05 to 2.0 mm) particles and have a surface area of 3.06 m²/g. The surface area of the sieved (< 100 µm) fines which are primarily dominated by silt sized (0.002 to 0.05 mm) particles increased by six times to 18.39 m²/g. The bulk fines from quarry site 2, which are also dominated by sand sized particles, have a surface area of 5.82 m²/g while the sieved (< 100 µm) fines which are dominated by silt size particle experience a decreased surface area to 3.77 m²/g (Table 4).

3.2. Ambient weathering experiments

During the first two weeks, white efflorescences appeared on the surface of bulk fines from site 1 but disappeared by one month, with no further weathering observed. In contrast, bulk fines from site 2 showed more weathering, with small white precipitates visible by two months and increasing after the third water addition (Fig. 3). Detailed information of the visual observations are provided as supplementary material. Subsamples of the sieved fines from site 2 were analysed by EDS, where areas of potential carbonation were identified. EDS elemental maps demonstrate that some pores are rich in Ca and O, while the rest of

the material is dominated by elements including Na, Mg, and Si. The stoichiometric ratio of Ca-O was calculated from target areas in EDS maps (Fig. 4), which matched the ratio of elements in CaCO₃.

Under ambient conditions, the changes in TIC content for bulk fines from quarry site 1 was negligible regardless of sample thickness (Fig. 5A and B), suggesting that sample depth had no effect on ambient carbonation for these fines. Within the sieved fines from site 1, the surface sample (S1-Amb-5cm-T) taken from the 5 cm core experiences an increase in TIC (up to $D + 0.16\%$) compared to the initial sample. Core samples taken from the sieved fines from site 1 at 3 cm and 5 cm saw similar increases in TIC ($D + 0.06\%$).

Bulk and sieved fines from quarry site 1 at both sample depths experienced an overall increase in TIC content by the end of experiments (Fig. 5C and D). Within the 1 cm column containing bulk fines the TIC content increased by 0.02 %, and within the 5 cm column with these fines, the change in TIC content for the surface sample was negligible. Despite limited changes observed on the surface of the bulk fines, the TIC content increased by 0.05 % in the core samples taken at 3 cm and 5 cm. Amongst the sieved site 2 fines a substantial increase in TIC was observed for all samples. The two surface samples taken from the 1 cm and 5 cm experiments both increased by 0.16 % TIC by 3 months, while the core sample taken at 5 cm depth increased by 0.18 % TIC.

3.3. Accelerated weathering experiments

During the experiments, bulk fines from both sites became increasingly cemented and nearly fully hardened, covered in desiccation cracks, with only surface coarse grains easily disturbed (Fig. 3). Detailed visual descriptions are provided as supplementary material.

Under accelerated weathering conditions, the bulk site 1 fines experienced an overall increase in TIC. Within the top sample of the 1 cm thick column, the TIC content increased by +0.08 % (Fig. 6A). Within the 5 cm column of the bulk fines, the bottom sample (S1-Acc-5cm-B) experienced the greatest increase in TIC content (+0.07 %) compared to the other depths (Fig. 6A). Within the sieved fines from site 1, the TIC content increased by +0.09 % in the 1 cm thick column (S1-Acc-1cm-T). However, in the 5 cm column the TIC content nearly tripled, experiencing maximum increases of +0.46 % (Fig. 6B).

In the accelerated weathering experiments the bulk and sieved fines from site 2 at both sample depths experienced an overall increase in TIC content by the end of experiments (Fig. 6C and D). For the bulk fines, the greatest increase in TIC content was in the surface samples of the 1 cm experiment (+0.09 %). Within the 5 cm depth column, the greatest increase was observed at the at the bottom (M5; +0.08 %), consistent with the bulk fines from site 1. In the 1 cm column with sieved material there was 6 times the amount of TIC compared to the initial unreacted sample (+0.31 %). Within the 5 cm column the greatest increase in TIC was observed on the surface (+0.35 %), which was ~ 7 times higher than the initial sample. The core samples taken at 3 cm (M) and 5 cm (B) experienced the same increase in TIC content of +0.31 %.

4. Discussion

4.1. Influence of mineralogy and geochemistry on carbonation

The mineralogical composition of fines from mining and quarrying is one of the most important properties that determines whether a feedstock is suitable for ERW (Power et al., 2024). These feedstocks

Table 3

Whole rock analysis of fines from quarry sites 1 and 2. All values are given as weight percentages (wt.%).

Sample	SiO ₂	Al ₂ O ₃	Fe ₂ O ₃	MgO	CaO	Na ₂ O	K ₂ O	TiO ₂	P ₂ O ₅	MnO	Cr ₂ O ₃	SrO	BaO	LOI	Sum
Quarry site 1	45.00	13.95	12.50	8.69	8.49	2.44	0.52	1.66	0.19	0.16	0.05	0.03	0.08	6.56	100.32
Quarry site 2	48.40	13.25	13.95	5.54	8.65	2.85	1.08	2.46	0.28	0.19	0.01	0.06	0.07	3.51	100.30
Detection limits (%)	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	

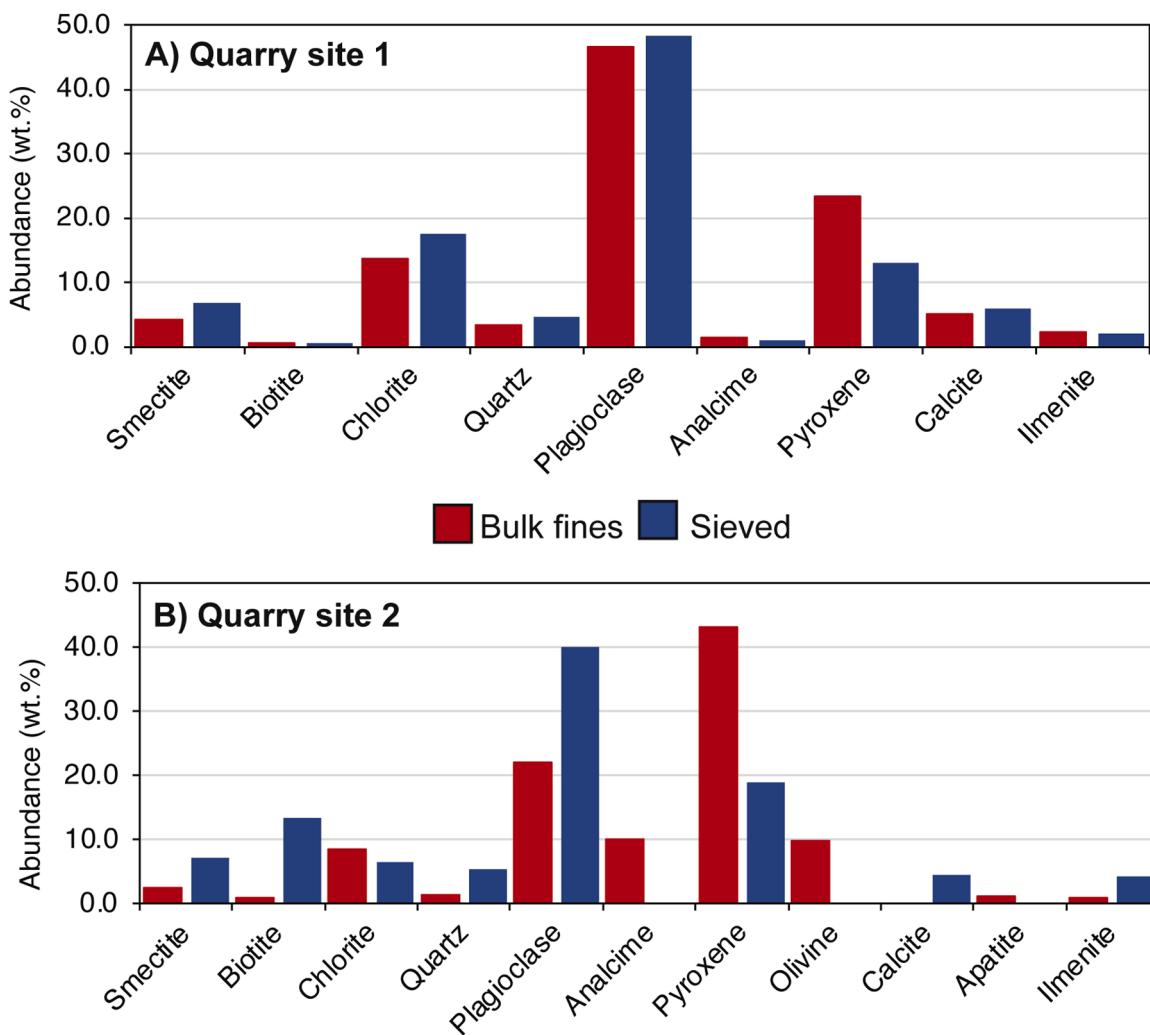


Fig. 2. Quantitative mineralogy (wt.%) of bulk and sieved fines from A) Quarry site 1 and B) Quarry site 2. Measurement errors are approximately 2 wt.%.

Table 4
Physical properties of basaltic fines used in experiments.

Site	Particle treatment	D80 (μm)	BET (m^2/g)
Quarry site 1	Bulk	3200	3.06
	Sieved	62	18.39
Quarry site 2	Bulk	3100	5.82
	Sieved	59	3.77

generally have complex mineralogy and can often be heterogeneous in nature (e.g., Lu et al., 2022; Woodall et al., 2021 and references therein) due to the regional geology where the rock is extracted. Within minerals targeted for ERW, dissolution rates vary by orders of magnitude, which will influence the overall carbon removal ability (Gislason et al., 2010).

The bulk fines from both sites consist of plagioclase and pyroxene, which are target minerals for CO_2 mineralization, with olivine also found at quarry site 2 (Fig. 2). The log dissolution rates for the acid dissolution mechanism is -9.80 , -7.16 , and $-5.17 \text{ mol}/\text{m}^2/\text{s}$ for pyroxene (e.g., diopside), olivine (e.g., forsterite), and plagioclase (e.g., anorthite), respectively (Hermanska et al., 2022). Plagioclase has the quickest dissolution rate of these minerals and is the primary source of Ca in the quarry fines, followed by pyroxene. Multi-oxide minerals, such as the target phases identified here, generally dissolve orders of magnitude slower than single-oxide minerals (Power et al., 2013), which were not detected. However, some multi-oxide minerals may experience rapid initial dissolution due to the presence of labile (i.e., easily

extractable) cations on the surface of minerals (Power et al., 2020; Thom et al., 2013). In this instance, loosely bonded ions from the surface of silicate minerals may be released, causing the initial dissolution rate to be orders of magnitude faster than what is reported here (Vanderzee et al., 2019). The fines collected for this study were deposited in a stockpile for an unknown period and likely underwent limited mineral dissolution due to their burial in tall, wide stockpiles, preventing precipitation from reaching them. In experiments that directly measured CO_2 fluxes into forsterite, CDR rates decreased over time as water continued to be added (Stubbs et al., 2022). With only three precipitation/evaporation cycles in this studies experiment, mineral dissolution/CDR rates may decline over time as the labile component depletes.

Under ambient conditions, bulk fines from site 2 nearly doubled in TIC content by the end of experiments, increasing from 0.06 % in the initial samples, to 0.11 % TIC in the bottom of the 5 cm depth experiment (Fig. 5). Given the difficulty of identifying small changes in carbonate using XRD, the stoichiometric ratio of Ca-O was calculated from target areas in EDS maps (Fig. 4) following the methods previously described by Khudhur et al. (2023), which matched the ratio of elements in CaCO_3 . It is likely that this carbonate would have formed from the dissolution of bedrock minerals identified by XRD such as plagioclase (e.g., anorthite) and pyroxene (e.g., augite; Eqs. (14)–(15)), releasing Ca^{2+} , and subsequently reacting with atmospheric CO_2 (Earle, 2015).

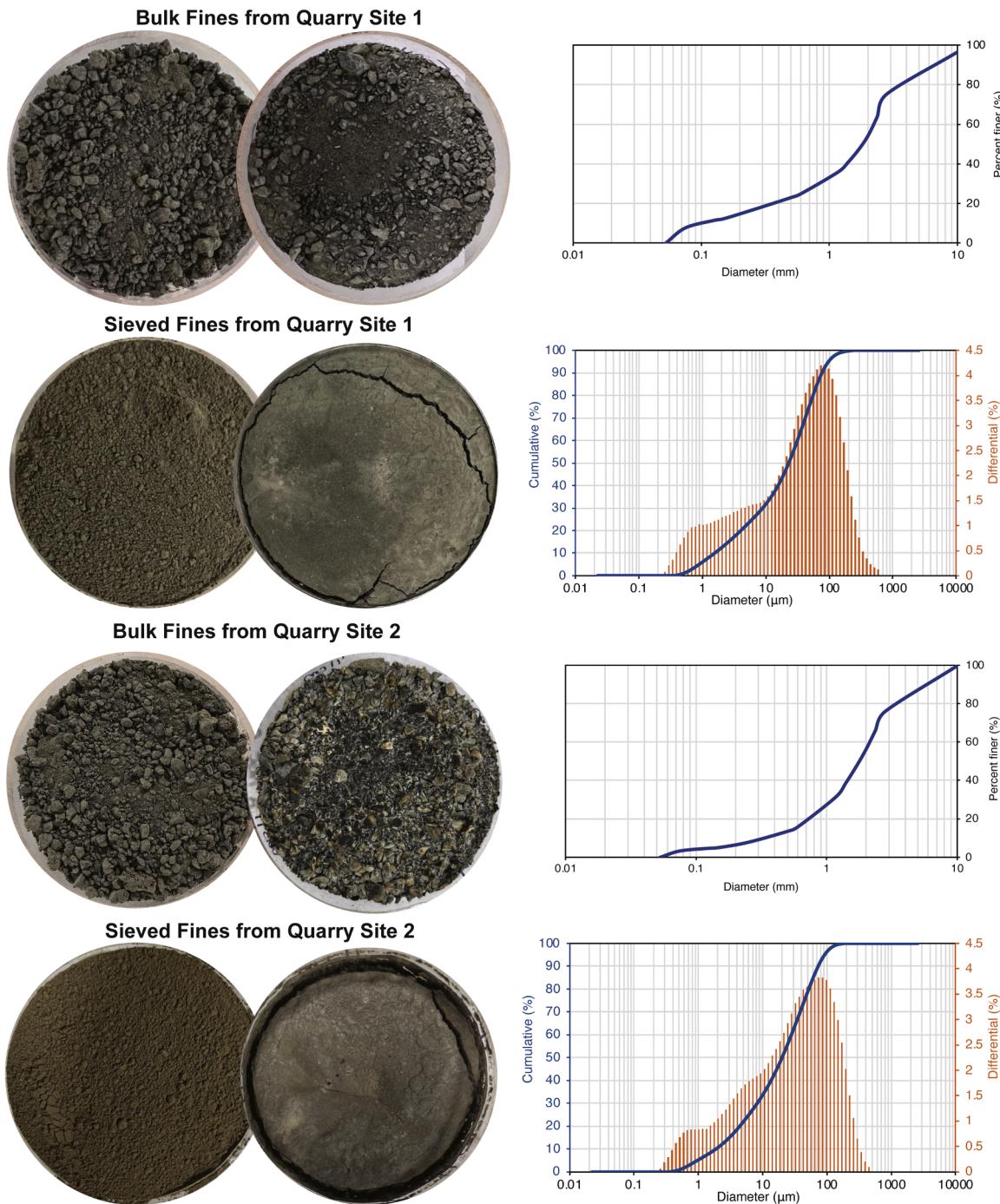
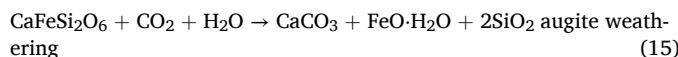


Fig. 3. Visual differences of bulk and sieved fines before and after three months of ambient weathering. Note desiccation cracks on the surface of the sieved material. Particle size distributions on the left reflect the particle size for the samples shown to the left. Differential (%) is shown on the secondary y-axis and represents the percentage of particles from the total that are within a specified range while cumulative (%) shown on the primary y-axis is the total sum of differential distributions. $\text{CaAl}_2\text{Si}_2\text{O}_8 + 3\text{H}_2\text{O} + 2\text{CO}_2 \rightarrow \text{CaCO}_3 + \text{Al}_2\text{Si}_2\text{O}_5(\text{OH})_4 + \text{H}_2\text{CO}_3$



Differences in mineralogy between bulk and sieved fines from site 2 showed decreased pyroxene content offset by doubled plagioclase content, providing cations (e.g., Ca^{2+}) for carbonation; calcite (4.5 wt.%) was detected in sieved fines but not in bulk fines, possibly due to XRD detection limits. Calcite would also be another source of Ca, but if dissolved, could lead to a loss of CO_2 if 1) carbonates with lower carbon content reprecipitate or 2) if non-carbonates precipitate (Stubbs et al.,

2023). More information on non-carbonate precipitation is provided in the supplementary material.

Quarry fines from site 1 showed smaller mineralogical differences between bulk and sieved samples compared to site 2, with increases in smectite (4.2 wt.% to 6.7 wt.%) and plagioclase (46.4 wt.% to 47.9 wt.%), and a halving of pyroxene (23.2 wt.% to 13.0 wt.%). Calcite levels were higher than site 2, at 5.0 wt.% in bulk and 6.0 wt.% in sieved fines. Under ambient conditions, there were no detectable changes in TIC within the bulk fines before and after experiments (Fig. 5A), despite seeing visual evidence of weathering on the surface of samples. Paulo et al. (2021) reports that in samples with greater carbonate abundance,

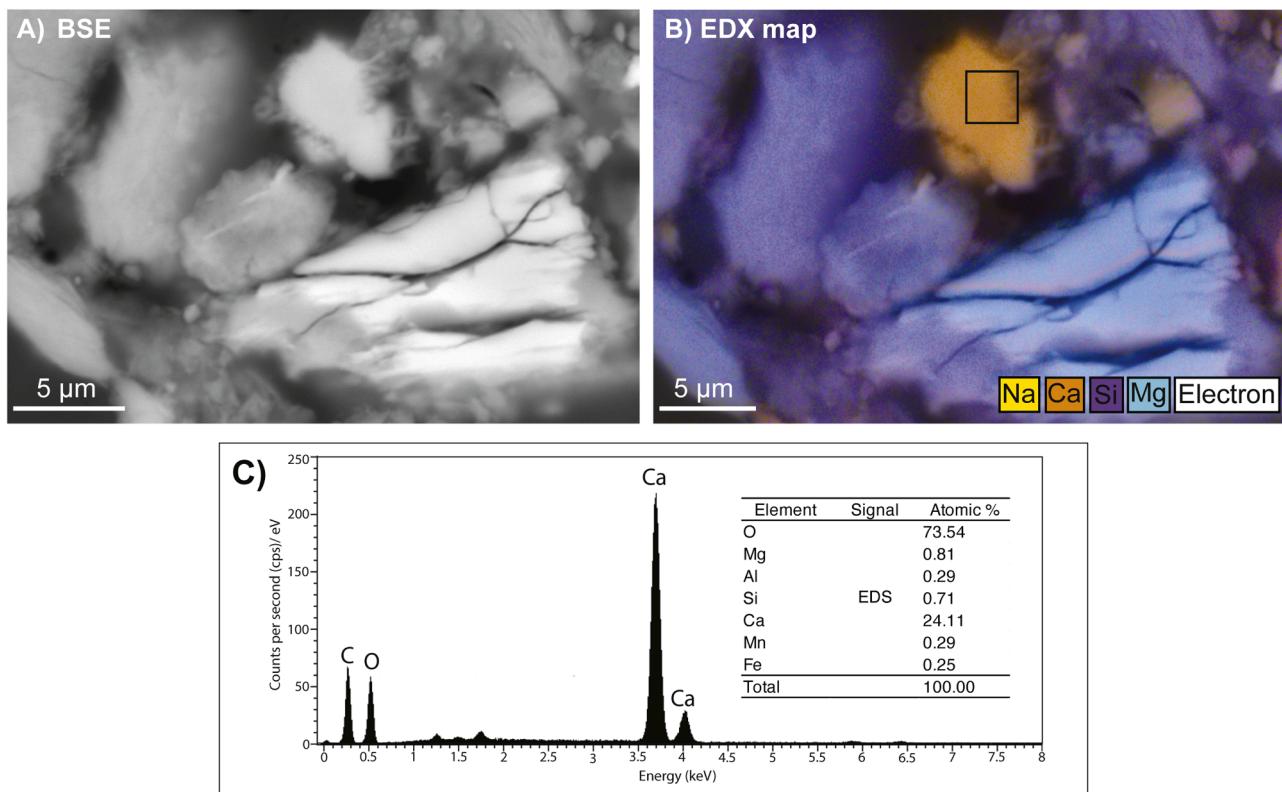


Fig. 4. A: BSE image of sieved S2-Amb-5cm-B. B: EDX map overlaying the same BSE image. Blue areas represent Mg, purple areas represent Si, orange areas represent Ca, and yellow areas represent Na. The black box is the area where the EDS spectra was taken. C: EDS spectra of carbonate paired with the quantification results.

and therefore greater TIC content, the CO₂ mineralization potential will decrease for that feedstock. In the case of fines from site 1, it is unknown how long the samples were left in the stockpiles; therefore, it is difficult to accurately determine how long these samples have been exposed to weathering in the quarry. Some of the calcite detected in initial samples could be a result of calcite on the joint faces, which would eventually make its way to the crushing circuit and into the fines. The calcite in the initial samples may result from quarry weathering rather than the original bedrock, potentially masking mineralization extent.

4.2. Influence of particle size, surface area, and depth on carbonation

The extent of secondary carbonate precipitation in quarry fines is influenced by both particle size and reactive surface area. Under ambient conditions, increases in TIC are equivalent to 570 g CO₂/m²/yr (0.06 % to 0.11 % TIC) for the bulk fines from site 2 and 1330 g CO₂/m²/yr (0.06 % to 0.24 % TIC) for the sieved fines from site 2. The sieved fines from the second quarry site contain twice the plagioclase and half the pyroxene of the bulk sample, with less surface area than fines from site 1, suggesting the presence of faster weathering minerals helps compensate for a lower surface area. Bulk site 1 fines did not experience any measurable increases in TIC under ambient conditions while the sieved site 1 fines sequestered up to a maximum of 440 g CO₂/m²/yr. Within the sieved fines, the plagioclase content remained consistent, and pyroxene was present at approximately half the abundance of the bulk material (Fig. 7). Given the reactive surface area of the sieved fines from quarry site 1 is approximately 6 times higher than the bulk material, the particle size and surface area likely have more of an effect on CDR than the changes in mineralogy.

Other studies have reported that particle size has an important impact on enhanced rock weathering (Amann et al., 2022; Deng et al., 2023), where finer particles accelerate mineral dissolution, leading to greater CO₂ uptake (Stefler et al., 2018), consistent with what is seen in

the present study. Rinder and von Hagke (2021) assessed the potential of ERW using basalt on agricultural land and found that the net drawdown in CO₂ increases as the particle size is reduced. In their study, they considered transportation of materials off-site for application on agricultural land. After accounting for a transport distance of 300 km via a railway, approximately 28 kg CO₂ t/year could be removed in fines that are <100 μm. However, in fines that are <1 μm, 125 kg CO₂ t/year could be removed (Rinder and von Hagke, 2021). In the present study, the CDR rate increased from 8 kg CO₂ t/year in the bottom of the bulk fines from site 2 to 26 kg CO₂ t⁻¹ in the bottom of the sieved (<100 μm) fines from site 2, similar to the findings of Rinder and von Hagke (2021). Transportation of materials is not considered in this study, as there may be scope to find land to spread fines either at, or surrounding, the quarry. Although the finer residues have the capability to remove more CO₂, implementing management practices that involve spreading bulk fines may provide a quick and simple solution for reducing site emissions.

Similarly, changes to particle size are also known to impact specific surface area, where finer particles typically have higher surface areas than coarser particles (Renforth, 2012). However, bulk site 2 fines have a surface area of 5.82 m²/g, but decrease to 3.77 m²/g in the sieved fines; this is likely due to the higher pyroxene content in the bulk fines compared to the sieved fines. Fines from quarry site 1 increased by six times from 3.06 to 18.39 m²/g between the bulk and sieved material. The differences in specific surface areas between the bulk and sieved fines would have implications for the release rate of elements, and subsequent carbonate precipitation, where finer grained samples dissolve faster than coarser material (Amann et al., 2020). For example, in the bulk site 1 fines there was no significant difference in TIC at any sample thickness. However, within the sieved site 1 fines, the TIC content increased by +0.06 % in the bottom 5 cm. Although the initial carbon content in fines from site 1 is higher than that of site 2, the increased reactive surface area of the sieved material may compensate

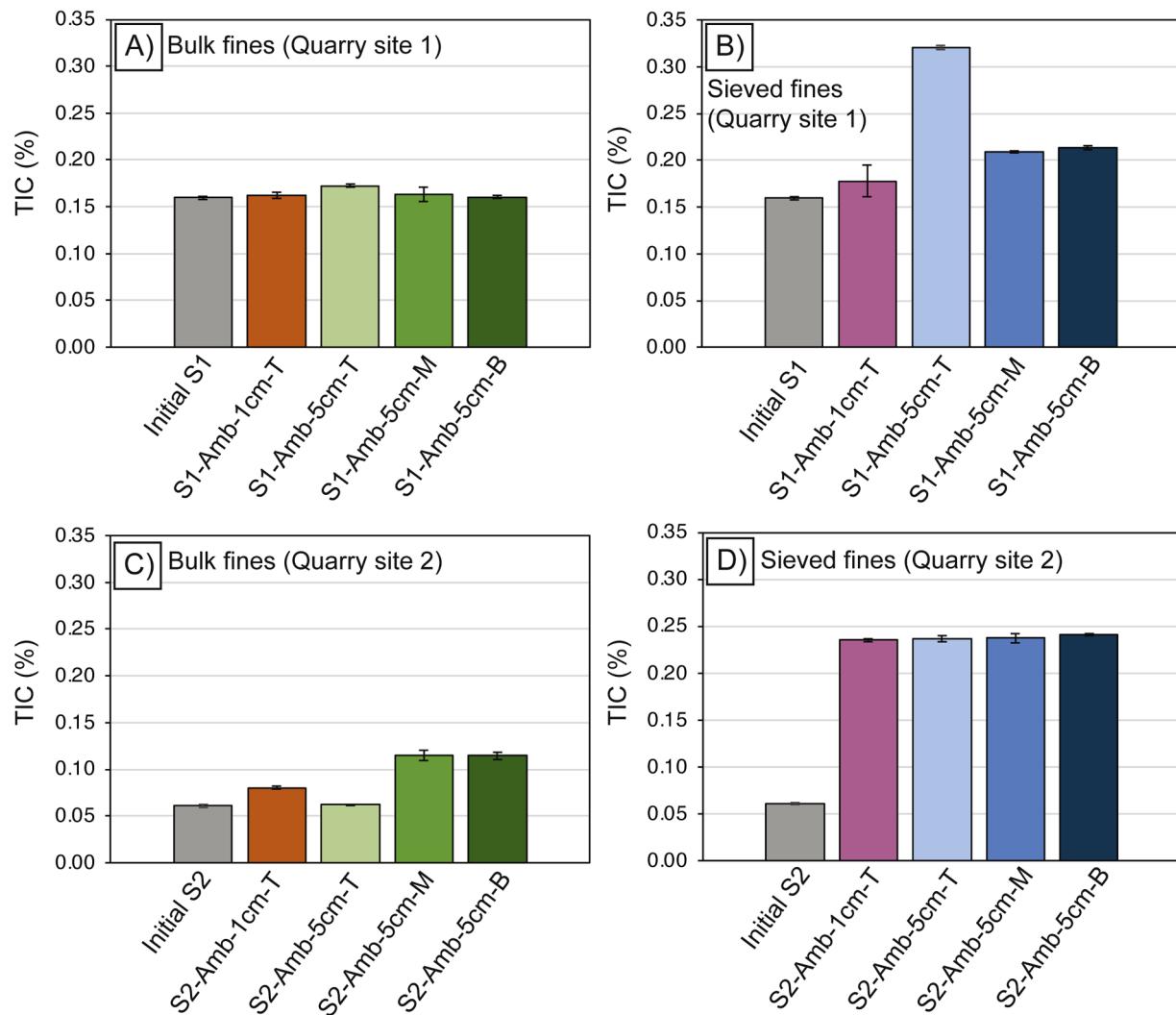


Fig. 5. TIC before and after 3 months of weathering under ambient UK conditions. A: Changes in TIC for the bulk fines from site 1, B: changes in TIC for the sieved fines from site 1, C: changes in TIC for the bulk fines from site 2, and D: changes in TIC for the sieved fines from site 2. Grey bars represent initial TIC data. Amb – ambient conditions, 1 cm or 5 cm – specific column depth, T – top 0.5 cm sample, M – middle 3 cm sample, B – bottom 5 cm sample.

for the higher initial carbon pool.

In this study, the depth of carbonation was impacted by particle size and surface area. The greatest carbon concentrations were measured at the base of columns (5 cm) in the bulk material, and within the sieved fines, the carbon content remained consistent throughout the column (0–5 cm), regardless of depth. In a two-year mine tailings carbonation experiment, Kandji et al. (2017) investigated the effects of mineral carbonation at different depths. The greatest changes in total carbon (TC) were found within the top 2 cm, increasing from 0.15 % to 0.83 % TC. From approximately 5 cm to 24 cm, TC measurements remained fairly consistent, ranging between 0.32 %–0.36 % (Kandji et al., 2017). Although the bulk fines tested in the present study experience the greatest changes in TIC at 5 cm, the particle size is much coarser ($D_{90} = 700 \mu\text{m}$) compared to the tailings used in the reference study ($D_{90} = 206 \mu\text{m}$). Stubbs et al. (2022) measured CO_2 fluxes in different kimberlite residues from a diamond mine and determined that the coarser residues had enhanced permeability, allowing more CO_2 ingress during instantaneous flux measurements compared to finer kimberlite residues.

Bulk and sieved fines from both sites experienced one water addition per month equivalent to 60 % pore water saturation; however, the bulk fines dried almost twice as fast as the sieved material. The purpose of this water addition was to facilitate weathering, as carbonation rates are limited where the water content is too low (Assima et al., 2013; Harrison

et al., 2015), yet flooded environments that have highly saturated pore waters tend to decrease carbonation rates (Wilson et al., 2011). The gas-water interface (GWI) is known to increase as evaporation progresses (Fernandez Bertos et al., 2004; Harrison et al., 2017), suggesting that the bulk fines from these two quarries would have a larger GWI for most of the experiment. It is likely that water in the bulk 5 cm columns pooled at the bottom, interacting with fines for longer than water that was present at the top of the columns. Water present at the top would have evaporated quicker, where there would be an insufficient amount of water for carbonation (Stubbs et al., 2022). Eventually, water at the bottom of the columns would also evaporate, meaning it would not be in contact with the fines for as long as the sieved fines. The TIC content remained consistent throughout the columns with sieved fines, suggesting a thickness of 5 cm is appropriate for management practices. Between the finer grain size, increased water-retention, and increased surface area, it is expected to have higher carbonate concentrations in this material compared to the bulk fines.

4.3. Implications for CDR at active quarries

Although the mining and quarrying industry contributes to global CO_2 emissions (Cox et al., 2022), it can play a crucial role for global CDR as major abundances of rock powder are produced annually that can be

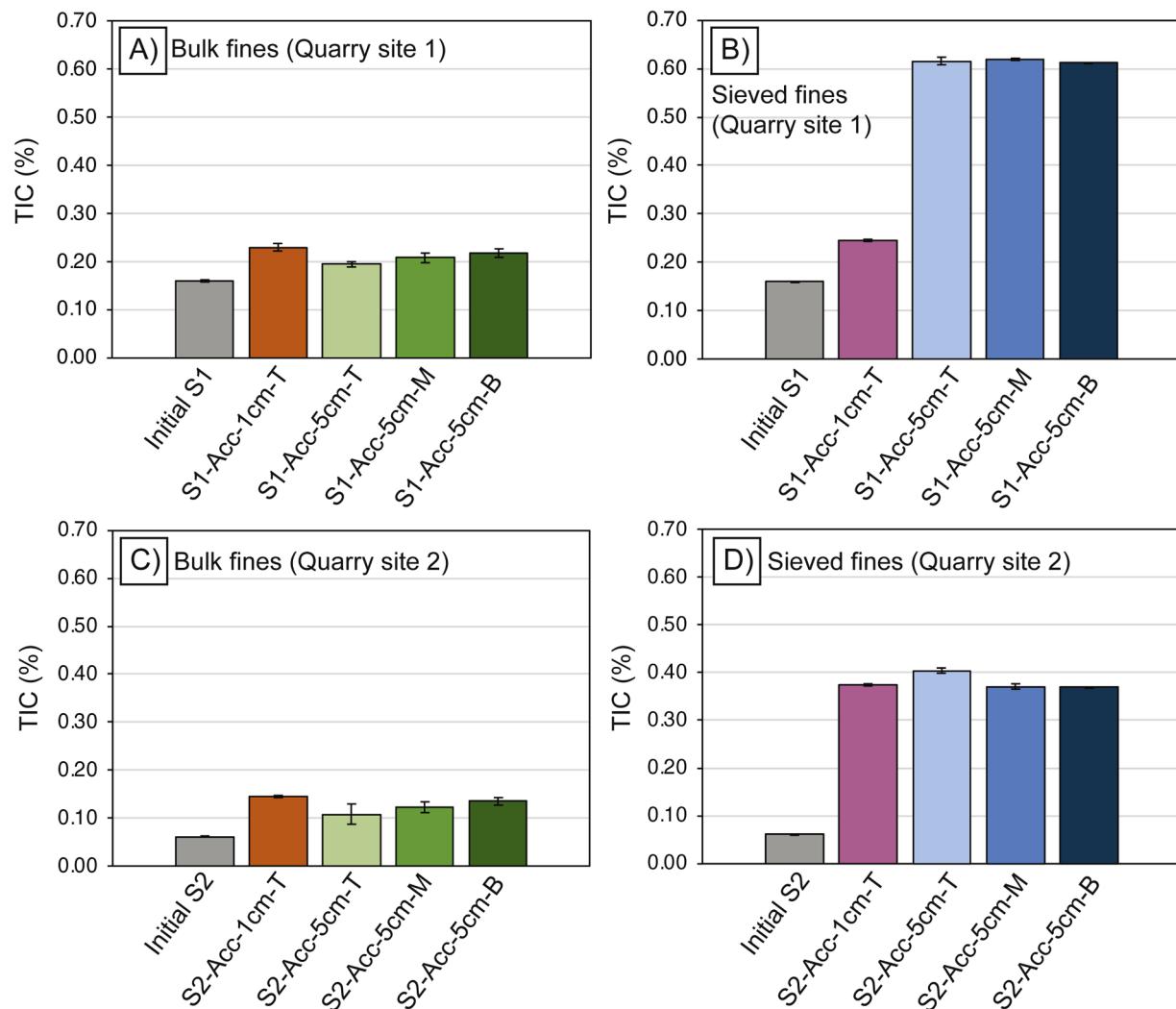


Fig. 6. TIC before and after 3 months of weathering under accelerated conditions. A: Changes in TIC for the bulk fines from site 1, B: changes in TIC for the sieved fines from site 1, C: changes in TIC for the bulk fines from site 2, and D: changes in TIC for the sieved fines from site 2. Acc – accelerated conditions, 1 cm or 5 cm – specific column depth, T – top 0.5 cm sample, M – middle 3 cm sample, B – bottom 5 cm sample.

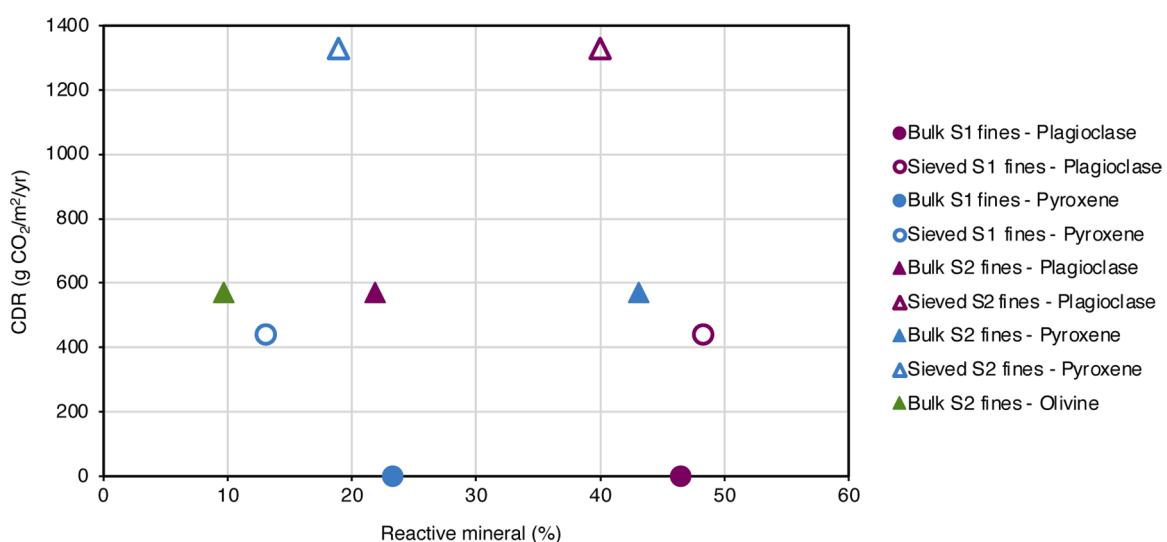


Fig. 7. Effect of grain size (bulk and sieved samples) and reactive mineralogy on CDR. Circles represent site 1 (S1) fines and triangles represent site 2 (S2) fines. Reactive minerals are represented by different colours where purple = plagioclase, blue = pyroxene, and green = olivine. Solid data markers represent bulk material and hollow data markers represent sieves fines.

used for ERW and CO₂ mineralization (Power et al., 2024). In the UK, there have been significant reductions in the annual production of crushed rock since the 1990s (BGS, 2018). Madankan and Renforth (2023) estimate that the total basic silicate rock production in the UK is estimated to be ~8.5 Mt/yr by 2050. Although the overall rock availability is decreasing, there is still ample quantities of potentially suitable rock for ERW (Madankan and Renforth, 2023), with numerous studies showing promising CDR rates (Fig. 8). To remove CO₂ at quarries, it is advantageous that these sites use their own fine materials to reduce the overall cost of CDR.

The two types of quarry fines tested in this study showed differing CDR rates depending on their mineralogy, geochemistry, and physical properties (e.g., grain size and reactive surface area). The bulk fines from site 2 under ambient conditions showed the greatest change in TIC at the bottom of the 5 cm columns. Approximately 570 g CO₂/m²/yr, could be sequestered, assuming the rate of change in TIC remained consistent for an entire year. If these fines were deposited in 5 cm layers for one year, between 0.6 km² and 0.8 km² of land would be needed to deposit between 42,000–56,000 t of basaltic rock, which is the minimum and maximum annual waste production at quarry 2. Assuming homogenous carbonation throughout this entire area, approximately 340–460 t CO₂/yr could be sequestered if the fines were deposited without any modification to the physical properties. In the 5 cm columns containing sieved site 2 fines, CDR rates ranged from 1270 to 1330 g CO₂/m²/yr between the top 0.5 cm and bottom 5 cm. Under the same circumstances suggested above (e.g., 5 cm layers), between 0.8 km² and 1.1 km² of land would be required to deposit the minimum and maximum waste tonnage. If homogenously carbonated, between 1100 and 1480 t CO₂/yr could be sequestered in fines <100 µm, under ambient conditions.

At both quarries, the basaltic rock and dolerite fines are deposited dry, where there is little to no water upon deposition. In the context of carbon removal, this is not ideal as the absence of water prevents any carbonation reactions from occurring. In the present study, the environmental conditions were controlled where pore water saturation started at 60% at the start of each month, eventually drying to ~0% by 30 days, before the next water addition. Although there are no current management practices in place that involve saturating the fines, the extent of carbonation happening in only 3 months within this study

indicates that quarries should attempt to control water content within the fines if these management practices were in place. CDR rates calculated above suggest that the fines should be deposited in 5 cm layers, as carbonation was consistently higher at the bottom of the bulk material yet was homogenous throughout the entire column when sieved fines were utilized. At active quarries where fines are exposed to ambient UK conditions, it is evident that management practices should consider controlling the pore water saturation, grain size of feedstock, and volume of material spread out over a certain area. Annually, Scotland receives >1500 mm of rainfall (MetOffice, 2024) which might pose challenges when trying to facilitate mineral precipitation driven by evaporation. If this rainfall were adequately drained it may enable CO₂ to be captured as dissolved inorganic carbon, potentially doubling the CDR rates.

Under accelerated conditions, the bulk fines from site 2 showed that in the 5 cm column, the greatest change in TIC was at the bottom, consistent with observations made under ambient conditions. Assuming the rate of change in TIC remained consistent for an entire year, approximately 500 t CO₂/yr and 660 t CO₂/yr could be sequestered based on the minimum and maximum production (Table 1) of fines, respectively. If fines were sieved to <100 µm, between 1910 t CO₂/year and 2550 t CO₂/year could be sequestered. However, unlike the ambiently weathered fines, it would not be feasible to spread these residues over large areas of land while also replicating the accelerated conditions (e.g., high CO₂ concentrations, high temperatures). Rather, quarries might consider constructing reactors where fines are deposited with a high pore water saturation (>60%) and injected with high concentrations of CO₂ to accelerate carbonation processes (Snaebjornsdottir et al., 2020). Rather than attempting to carbonate materials at low temperatures, the fines would need to be exposed to higher temperatures (e.g., 50 °C) to 1) increase evaporation time and 2) increase the carbonation rate. Given the high volume of fines produced annually, more frequent depositions should be considered rather than only one deposition per year. It is possible that most of the carbonation occurs before the one-year mark, suggesting the above rates could be overestimated. Further experimental testing would be required to determine if the extent of carbonation eventually stabilizes, and if so, the timeline for fines deposition could be determined.

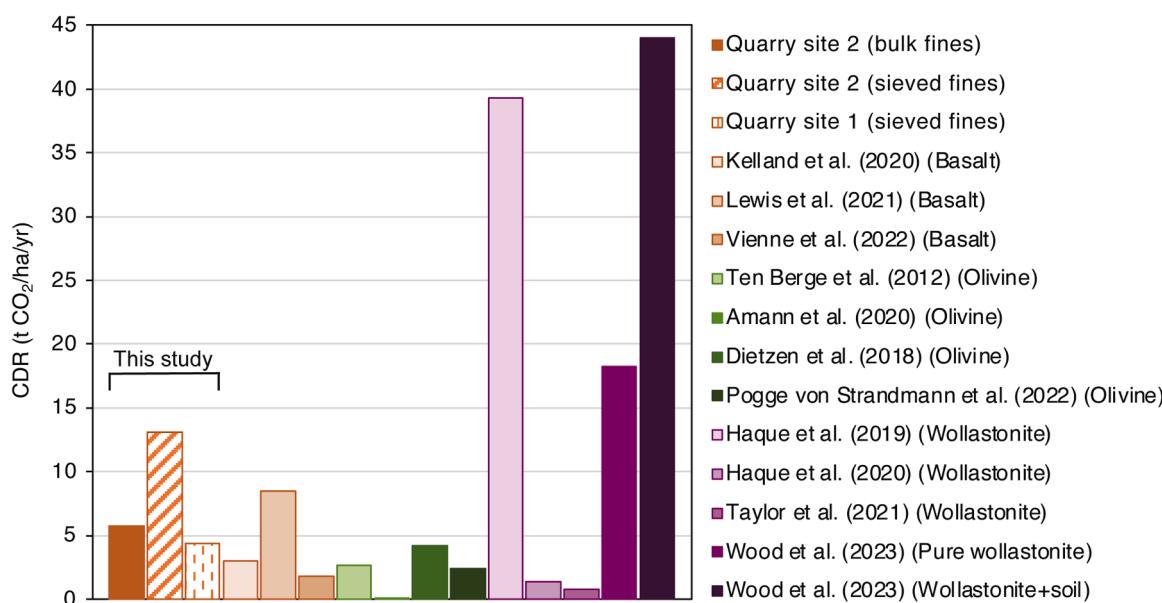


Fig. 8. Comparison of CDR rates (t CO₂/ha/yr) within enhanced rock weathering studies using basalt, olivine, or wollastonite as a feedstock. Rates for this study are labelled accordingly. Accelerated carbonation experiments have not been included here as they don't reflect earth surface processes (Amann et al., 2020; Dietzen et al., 2018; Haque et al., 2019, 2020; Kelland et al., 2020; Lewis et al., 2021; Pogge von Strandmann et al., 2022; Taylor et al., 2021; ten Berge et al., 2012; Vienne et al., 2022; Wood et al., 2023).

4.4. Economic evaluation of spreading ultramafic fines at quarries

The primary cost associated with the weathering of fines at quarries is the crushing/ grinding of rocks to achieve a reduced particle size compared to the pre-existing fine material (e.g., further crushing the feedstock). At quarries, rocks undergo crushing and separation through extraction and processing circuits to access their target commodities (e.g., asphalt for both quarries in this study). This process generates thousands of tonnes of fines annually that are often underutilized. In the context of CDR, this study has demonstrated that whether fines are reacting under ambient or accelerated conditions, basaltic fines with a reduced particle size have the capacity to offset a greater amount of CO₂. However, trying to achieve a smaller target grain size increases the energy demand for rock grinding, subsequently raising the overall cost of CDR (Strelfer et al., 2018). The crushing and grinding requirements for both quarry sites have been assessed to understand the energy requirements and costs associated with achieving a grain size of ≤100 µm, from the original grain size of fines.

To determine the energy required to reduce the grain size from bulk fines to sieved fines, the following Bond equation was used (Eq. (16); Bond, 1960; Rowland, 1998).

$$W = \frac{10Wi}{\sqrt{P}} - \frac{10Wi}{\sqrt{F}} \quad (16)$$

The W is the power requirements (kWh/ tonne), Wi is the work index which is predetermined from comminution testing (18.51 for basalt; Bond, 1960), P is the diameter in µm which 80 % of the product passes through, and F is the size which 80 % of the feed (e.g., finer material) passes through.

At site 2 the D80 of the bulk materials ($\equiv F$ in this equation) is 3100 µm and the D80 of the finer material ($\equiv P$) in this equation is 59 µm. Depending on whether the minimum or maximum quantity of fines was being processed, the energy requirement would range between 873 MWh and 1164 MWh. Based on an energy consumption between 500 and 1999 MWh per year which is considered a small/medium energy band (UKGovernment, 2024a), the energy unit price is 23.85 pence/kWh. This accounts to a total energy cost between £208,197 and £277,595 per year (including climate change levy, excluding VAT) for the minimum and maximum fines production.

At site 1, the D80 of the bulk materials is 3100 µm and the D80 of the finer material is 92 µm. In 2023, the quarry produced 97,500 tonnes of fines, resulting in an energy requirement of 1967 MWh. Based on the small/ medium energy band and the same energy unit price provided above, the total energy cost would be £469,126 (including climate change levy, excluding VAT).

With an understanding of costs associated with the crushing and grinding of rock, the value of CO₂ sequestered can be determined with 1) a calculated CDR rate based on the annual production of fines and a calculated area required to deposit a given mass of fines, and 2) a known carbon price (£83.03 in the UK; UKGovernment, 2024b). The annual income is simply the CDR rate per year multiplied by the carbon price. However, to consider the time-value of money, the Present Worth (PW) analysis is used. Based on discrete compounding payments at an interest rate of 5 %, the present worth (PW) can be determined for a uniform annual cash flow. Using present worth factors (PWF) from Sullivan et al. (2019), the PW was calculated for quarry sites 1 and 2, where the profits are dependent on 1) CDR rates, 2) production rate of fines, and 3) grain sizes. However, it should be noted that the following assumptions were made during this evaluation: 1) a fixed carbon price and interest rate over the projected timeline, 2) a fixed CDR rate every year, 3) a new 5 cm layer of fines is added every year, where the previous pile stops capturing carbon.

At site 2, under ambient conditions, the greatest changes in the value of carbon are based on the grain sizes, where finer particles have a higher monetary value compared to the bulk fines. For example, the bulk material has an estimated PW of £123,250 (if CO₂ uptake occurred for in

5 years at a 5 % interest rate) for the minimum production of fines. The PW of finer material increases to £400,000 in 5 years at a 5 % interest rate for the maximum production of fines. Although both these values increase over time (Fig. 9), the annual cost of crushing and grinding material to the finer fraction would cost more than the profits would be. The same scenario applies to the accelerated conditions, where the cost of crushing does not make the value of carbon profitable. This suggests that reducing the grain size of fines does not equate to more profit for quarry companies. Spreading bulk material over a known area and thickness would be less time and labour intensive, cost effective, and offset a considerable amount of CO₂.

5. Conclusions

At an experimental scale, the distribution of un-weathered basaltic fines is able to sequester CO₂, and if applied to an active quarry, could potentially offer both environmental and economic benefits if carbon were permanently stored. In this study, the geochemical/ mineralogical composition of the feedstocks had the most important effect on CDR, followed by its physical properties and water content.

Weathering of bulk and sieved fines from site 2 under ambient conditions suggest they could potentially sequester up to ~460 t CO₂/yr and ~1500 t CO₂/yr, respectively, if their maximum production of fines was spread out over 0.8 km² in 5 cm thicknesses every year. Under ambient conditions, fines from site 1 did not indicate any weathering had occurred in the bulk fines however the sieved fines suggest up to ~500 t CO₂/yr could be sequestered following the same conditions as mentioned above. However, these estimations are based on controlled laboratory experiments which do not capture the complexities of real-world conditions that may affect the amount of CDR that occurs.

Under accelerated conditions where samples were exposed to 50 °C and 20 % CO₂, CDR estimates substantially increased. Fines from quarry site 2 contained a small initial carbon pool (0.06 %), meanwhile, the initial TIC in fines from site 1 was 2.7 times higher. Furthermore, the bulk site 2 fines featured a greater abundance of target minerals, and a higher surface area compared to the bulk site 1 fines. In the sieved fines, site 2 doubled in plagioclase content while the plagioclase content of the fines from site 1 remained the same and pyroxene reduced by half. Notably, the reactive surface area of the sieved site 1 fines was significantly higher than the sieved fines from site 2, which likely played a role in their differing reactivity.

Under current storage practices for fines, the duration of material deposition is often unknown, making it difficult to account for any carbonation via weathering that may have occurred. However, this study demonstrates that the management of quarry fines could be modified to sequester CO₂ on-site/ near the site, but there must be enough available area to do so. Although reducing the grain size of fines can enhance CDR estimates, the associated energy and cost of crushing fines must be carefully evaluated to ensure that the economic and environmental benefits of CO₂ sequestration outweigh the costs of further processing. To address these considerations under real-world conditions, larger-scale pilot projects (e.g., spanning several square meters) are recommended. Such studies will help verify whether the observed CDR rates in controlled laboratory conditions can be replicated in natural environments.

CRediT authorship contribution statement

Amanda R. Stubbs: Writing – review & editing, Writing – original draft, Visualization, Validation, Supervision, Software, Resources, Project administration, Methodology, Investigation, Funding acquisition, Formal analysis, Data curation, Conceptualization. **Faisal W.K. Khudhur:** Writing – review & editing, Writing – original draft, Methodology, Conceptualization. **Ian M. Power:** Writing – review & editing, Formal analysis, Data curation. **Linzi McDade:** Writing – review & editing, Investigation. **Mark Friel:** Writing – review & editing,

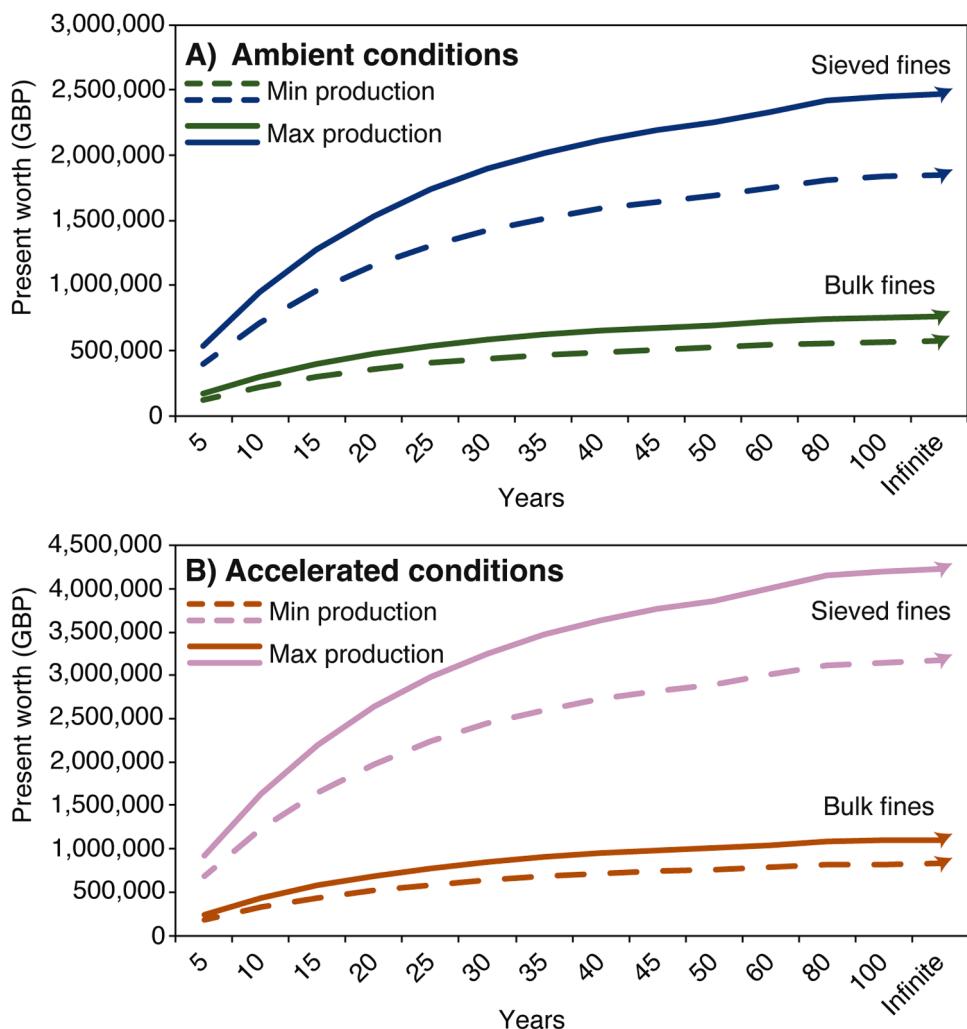


Fig. 9. Present worth of carbon (thousands of GBP) sequestered during weathering of fines from site 2 assuming a constant carbon price of £83.03 in the UK, a steady interest rate of 5 %, and homogenous carbonation throughout the entire distribution of fines. Dashed lines represent minimum production of fines, and the solid lines present maximum production of fines. Data for the bulk and sieved fines are labelled accordingly.

Investigation. **Iain Neill:** Writing – review & editing. **John MacDonald:** Writing – review & editing, Writing – original draft, Supervision.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Supplementary materials

Supplementary material associated with this article can be found, in the online version, at [doi:10.1016/j.ijggc.2025.104344](https://doi.org/10.1016/j.ijggc.2025.104344).

Data availability

Data will be made available on request.

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