The enhanced weathering potential of a range of silicate and carbonate additions in a UK agricultural soil

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HIGHLIGHTS

- A core study assesses enhanced weathering potential of natural & industrial silicates, & lime.
- Industrial silicates draw down CO_2 quickly, but application will be limited.
- Ten years of basalt application at 50 t ha^{-1} could remove 1.8 ± 0.9 $MtCO_2$ yr^{-1} over UK cropland.
- Liming in some agricultural UK soils is a net sink of C rather than source.

GRAPHICAL ABSTRACT
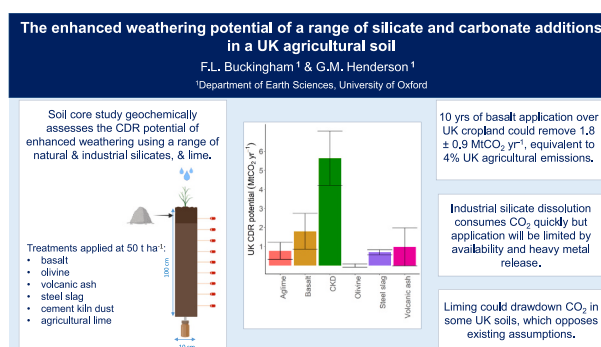
ABSTRACT

Enhanced weathering (EW) is a carbon dioxide removal (CDR) technology which aims to accelerate silicate and/or carbonate weathering in agricultural land. At present, the rate and magnitude of CDR from EW remains uncertain. In this study, soil cores extracted from a typical UK agricultural site in Oxfordshire were used to geochemically assess the efficacy of EW while simulating field conditions. Six material "treatments" were applied to soil cores at a rate equivalent to 50 t ha^{-1}: agricultural lime (aglime), basalt, cement kiln dust (CKD), olivine, steel slag, and volcanic ash. A range of chemical measurements were used to constrain the rate of dissolution, fate of dissolution products, and the CDR potential and environmental impact of treatment.

After a single application, the CDR rates were, in decreasing order: steel slag (20 ± 4 $kgCO_2$ ha^{-1} yr^{-1}) > CKD (16 ± 4 $kgCO_2$ ha^{-1} yr^{-1}) > basalt (5 ± 3 $kgCO_2$ ha^{-1} yr^{-1}) > volcanic ash (3 ± 3 $kgCO_2$ ha^{-1} yr^{-1}) > aglime (2 ± 1 $kgCO_2$ ha^{-1} yr^{-1}) > olivine (0 ± 2 $kgCO_2$ ha^{-1} yr^{-1}). Despite its drawdown potential, steel slag addition is not advised because application raised the dissolved concentration of heavy metals in the soil. CKD application will be limited due to availability of this material. Liming of agricultural soil is normally considered a source of CO_2, but this study indicates liming could cause CDR in some UK soil conditions.

Extrapolating from this site to a wider scale supports the conclusions of recently published research which suggests 10 years of basalt application over UK cropland could remove 1.8 ± 0.9 $MtCO_2$ yr^{-1}. Given the low rates of CDR observed in this study, EW is only likely to be worthwhile, at least in soil and climate conditions common to SE England, where there are co-benefits beyond carbon uptake.

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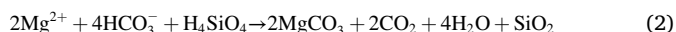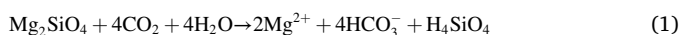*E-mail address:* fbuckingham.earth@gmail.com (F.L. Buckingham).

1. Introduction

To limit global average temperature rise to 2 °C and meet decarbonisation targets, existing research suggests we need to remove CO_2 from the atmosphere at rates as high as ~25 % of the present emission rate; equivalent to several hundred $GtCO_2$ over the 21st century (IPCC, 2018; The Royal Society, 2018). One route to such removal is by dramatically accelerating the removal of CO_2 caused by natural weathering of silicate and carbonate minerals. This natural mineral weathering controls climate on long-timescales, with present natural consumption of atmospheric CO_2 by weathering of ≈0.35 $GtCO_2$ yr^{-1} (Moon et al., 2014). This compares to present anthropogenic emissions of 49 ± 4.5 $GtCO_2$eq yr^{-1} (IPCC, 2014).

Enhancing the rate of terrestrial weathering as a CO_2 removal technology involves the application of crushed rocks/minerals to agricultural land to accelerate the weathering process via the release of alkalinity and compensating removal of CO_2 to natural waters or secondary minerals (Seifritz, 1990; Hartmann et al., 2013 and references therein). A critical factor for assessment of EW is the rate of dissolution of fresh mineral surfaces in natural soil conditions. An extensive body of past work has used laboratory experiments to assess weathering rates in the lab and field. Olivine dissolution recorded in laboratory experiments typically give high rates (≈10^{-14} mol(Olivine) cm^{-2} s^{-1}, 19 °C, pH 7; Palandri and Kharaka, 2004) but oversimplify the array of variables which influence mineral-fluid interaction in the soil environment. These lab dissolution rates are up to three orders of magnitude faster than olivine dissolution recorded in more natural configurations in pot ($10^{-17.12}$ to $10^{-17.75}$ mol(Olivine) cm^{-2} s^{-1}, Amann et al., 2020) and core studies ($10^{-15.8}$ to $10^{-16.7}$ mol(Olivine) cm^{-2} s^{-1}, Renforth et al., 2015). However, these pot and core studies underestimate olivine dissolution in the natural soil environment as they only account for dissolution products in soil solution. In contrast, Kelland et al. (2020) measured the dissolution rate of basalt ($10^{-15.3}$ to $10^{-16.2}$ mol(Ca) cm^{-2} s^{-1}) in a comprehensive biotic mesocosm study which also considering the accumulation of dissolution products in the dissolved and exchangeable pool and by plant-uptake.

Although these pot and core studies are more representative of the soil environment than purely laboratory-based studies, they do not simulate the pattern or flux of natural rainfall. A recent core study that sought to do so (Buckingham et al., 2022) identified water flux as a critical control which limited CDR due to basalt weathering in soil cores extracted from cropland in SE England. Furthermore, existing pot and core studies are based on soil solution measurements at only a few depths (≤4) in the soil which conceals changes in soil-water chemistry with depth. Existing pot and core studies have also only measured dissolution of one or two mineral additions in the same mesocosm set-up. As the experimental set-up can greatly influence dissolution rate, this prevents a direct comparison of the EW potential of multiple proposed treatments.

The release of alkalinity into soil solution drives CO_2 uptake by converting atmospheric CO_2 to dissolved HCO_3^- (e.g., for release of Mg^{2+} in Eq. (1) below). If dissolved cations are re-adsorbed to mineral surfaces or by secondary silicates it reverses this CO_2 uptake, while formation of secondary carbonate partially reverses the uptake (e.g. as in Eq. (2)).

$$Mg_2SiO_4 + 4CO_2 + 4H_2O \rightarrow 2Mg^{2+} + 4HCO_3^- + H_4SiO_4 \qquad (1)$$

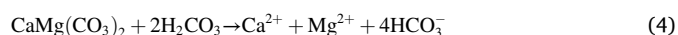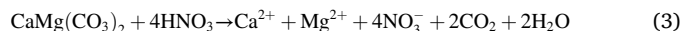$$2Mg^{2+} + 4HCO_3^- + H_4SiO_4 \rightarrow 2MgCO_3 + 2CO_2 + 4H_2O + SiO_2 \qquad (2)$$

A number of recent modelling and theoretical studies have estimated promising drawdown potential for EW (Smith et al., 2015, 2016; Renforth, 2019; Lewis et al., 2021; Kantzas et al., 2022). Kantzas et al. (2022) used a vertical reactive transport model to estimate that basalt application at 40 t ha^{-1} yr^{-1} for multiple years over UK cropland could remove 6–30 $MtCO_2$ yr^{-1} by 2050. This magnitude of CDR would compensate for a considerable proportion of UK agricultural emissions (which were 46.3 $MtCO_2$e yr^{-1} in 2019; DEFRA, 2021). However, a recent soil core study by Buckingham et al. (2022) (subsequently commented on in West et al. (2023), and rebutted in Buckingham et al. (2023)) identified water-limitation as a critical factor and suggested that the rate of drawdown in UK cropland could be as low as 1.9 ± 0.1 $MtCO_2$.

In addition to the CDR potential, the possible co-benefits and deleterious consequences of EW on the soil-water environment should be robustly evaluated prior to extensive EW deployment (see Swoboda et al., 2021, for a full discussion). Of particular importance is the release of heavy metals from a soil treatment. Elevated Ni was measured in soils after olivine application in a pot study by ten Berge et al. (2012), while Renforth et al. (2015) estimated that >99 % of dissolved Ni and Cr accumulated within olivine-treated soil, and Amann et al. (2020) measured elevated dissolved Ni and Cr in soil pore-water after addition of 220 t ha^{-1}. These results suggest olivine is not appropriate as a soil addition. In contrast, basalt is considered as a promising enhanced weathering treatment with low heavy metal content (Renforth, 2012; Beerling et al., 2020).

In this study, the EW potential of six different mineral treatments is evaluated in relatively dry UK agricultural conditions over 16-months. Three of these treatments are agricultural lime (aglime) and alkaline waste materials (steel slag, CKD), which presently have poorly constrained EW potential in agricultural soils. Aglime is formed predominantly from $CaCO_3$ and is regularly applied to cropland to maintain an optimal soil pH for crop growth. Aglime spreading is currently considered a source of greenhouse gas emissions based on the assumption made by the IPCC that aglime reacts with strong acid resulting from fertiliser use and that all C in lime is eventually released to the atmosphere (Eq. (3) below) (IPCC, 1997). Importantly, these calculations do not consider the reaction of aglime with carbonic acid in moderately acidic soils. In such conditions, released cation alkalinity is balanced by bicarbonate resulting in net drawdown of CO_2 from the atmosphere (Eq. (4)) (Hamilton et al., 2007). This means that spreading of agricultural lime could lead to greenhouse gas removal rather than addition to the atmosphere in some settings. In addition, recent research suggests alkaline-rich silicates produced from industrial processes, such as steel slag or cement kiln dust (CKD), could provide an alternative source of cations for EW without the need for extensive crushing (Renforth et al., 2011; Renforth, 2019). To date, the enhanced weathering potential and geochemical impact of aglime and industrial silicate dissolution in soil has not been evaluated experimentally.

$$CaMg(CO_3)_2 + 4HNO_3 \rightarrow Ca^{2+} + Mg^{2+} + 4NO_3^- + 2CO_2 + 2H_2O \qquad (3)$$

$$CaMg(CO_3)_2 + 2H_2CO_3 \rightarrow Ca^{2+} + Mg^{2+} + 4HCO_3^- \qquad (4)$$

By comparing the chemistry of soils in cores treated with each of the additions to that in untreated control cores, the geochemical impact of treatment dissolution on the soil and freshwater environment was isolated and novel insights into rates of dissolution and the fate and distribution of dissolution products were learned. These data provide a comprehensive empirical assessment into the efficacy of enhanced weathering application on UK arable land.

2. Experimental design

A detailed description of the experimental set-up is provided in Buckingham et al. (2022) and summarised more briefly here. Soil cores of 1.0 m length and 0.1 m diameter containing lime-rich, loamy soils were extracted from UK agricultural land (FAI Farm, N Oxfordshire, 51.781°, −1.3141°; Aug'18). Soil pore-water was sampled at 10 depths in each core using Rhizon CCS samplers with a 0.15 μm pore size (Rhizosphere Research Products) inserted at 10 cm intervals, and from effluent which drained from the base of the core. Soil solution was sampled on 13 occasions in 16-months (Feb'20–Jun'21), including twice

prior to treatment addition (Mar'20) and then monthly from Aug'20 to Jun'21, inclusive – see S2 for further details.

Six different treatments (agricultural lime, cement kiln dust, crushed basalt, crushed olivine, crushed steel slag, volcanic ash – see Table 1 and SI for details including chemical analysis) were added to three soil cores each at a rate equivalent to 50 t ha^{-1} (5 kg m^{-2}) with an untreated triplicate control (Mar'20). To apply each treatment, the top 13 cm of soil was extracted from each core, combined with the treatment and returned to the core. This process was repeated for three control cores but without treatment addition. In all cases the remainder of the cores were not disturbed and were exactly as recovered by coring from the field-site. The application rate of each treatment is higher than those of UK liming guidelines (0.5–10 t ha^{-1}; Goulding, 2016), similar to the modelling study of Kantzas et al. (2022, 40 t ha^{-1}), and lower than the application rate used in recent pot and core studies (100 t ha^{-1}; Kelland et al., 2020; Buckingham et al., 2022).

Natural rainfall was the only source of irrigation throughout the majority of the time-series to represent the on-site water balance in UK cropland. The exception to this is 330 ml locally-collected rainwater which was applied to each core 24 h before and again immediately before each set of pore-water samples were extracted. This rainwater addition increased pore water saturation and permitted sample collection. Without such irrigation, there was insufficient soil solution in the pores for sampling. Transit times for this rainwater through the core is ca. two days (as constrained by fluorescent dye tracing) and it is assumed that, while the rapid passage of water flushes dissolution products from the core, it does not play a significant role in fresh dissolution during its rapid transit. The impact of dilution of dissolution products in waters taken from the core is fully accounted for using dye tracing (see Buckingham et al., 2022).

The infiltration flux (1.3 ml day^{-1}, 60 mm yr^{-1}; 1.5 × 10^{-5} g s^{-1}) was calculated from natural rainfall, recorded directly at the core location, minus evapotranspiration, recorded at Chimney Meadows, Oxfordshire, (UKCEH, 2021) over the study period (Feb'20–Jun'21), and is consistent with measurements made in Chimney Meadows, and at the experimental site (Buckingham et al., 2022) (57 mm yr^{-1}). The flux of water measured in this study is representative of agricultural soils in areas of SE England and is one to two orders of magnitude lower than the infiltration rates applied in existing pot, core and modelling studies which include regular irrigation (Renforth et al., 2015; Amann et al., 2020; Kelland et al., 2020; Kantzas et al., 2022; Vienne et al., 2022).

The concentration of dissolved cations was measured with ICP-MS as in Buckingham et al. (2022). A series of additional measurements further constrain the geochemistry of the core system in this study, including dissolved anion concentration, soil solution pH and titrated alkalinity, and solid soil analysis. Soil samples were collected at the beginning and end of the study from a representative core from each treated and untreated subset. Soil samples were oven dried, crushed and collated into subsections representative of the top (0–30 cm), middle (40-60 cm) and base of the core (70-100 cm). Samples were analysed for soil mineralogy (XRD), elemental composition (XRF), exchangeable cation concentration (ammonium extraction), cation exchange capacity (Co-Hex extraction and ICP-OES analysis) and total (in)organic carbon content. A full description of analytical techniques is provided in the supplementary information.

This study aims to address the limitations of existing research by providing direct comparison and quantification of different treatment dissolution, including industrial silicates and agricultural lime; replication of low CDR rates for basalt; and analysis and costing of possible routes for UK greenhouse gas removal. The experimental approach used in this study facilitates the collection of a high-resolution, extensive dataset which is more representative of the soil environment than laboratory studies. An essential factor which can influence EW, water flux, was represented by natural rainfall between sampling periods to develop the understanding of EW in dry, arable UK soils.

Although the core approach used in this study has a number of advantages in enabling detailed characterisation of the weathering process, there are some limitations of this approach. By its nature, this study is limited to the chalky soil type used, and to the low rainfall conditions typical of SE England; other empirical studies will be required to assess EW in other soil types and climate conditions. Extraction of the cores from the field site leads to a small change in the precise temperature and environment of the cores, and the reasonably short duration of the experiment means that the build-up of secondary minerals or undissolved treatment cannot be quantified. These limitations do not prevent this study from providing a realistic empirically constrained assessment of the potential for CDR by EW in conditions typical to SE England.

3. Results

3.1. Soil solution chemistry

3.1.1. pH and titrated alkalinity

The titrated alkalinity and pH of soil solutions was averaged over the full length of triplicate cores across 11 sampling events after treatment addition (Aug'20–Jun'21). Titrated alkalinity significantly increased in all treated cores, except those treated with olivine, and increased most markedly in cores treated with steel slag (+1.5 meq L^{-1}) (Fig. 1). Soil solution pH significantly increased in cores treated with basalt and steel slag by 0.11 pH and 0.20 pH units, respectively. This is similar to the

Table 1
Description of six applied treatments.

| Treatment	Type of treatment	Source	Key cations (wt % as oxide)	Use in previous EW studies	BET surface area (m^{2}g^{-1})
Agricultural lime (aglime)	Carbonate	Pigdon Quarry, Cornwall, UK (E & JW Glendining Ltd.)	CaO (91.9 %)		0.475 ± 0.002
Cement kiln dust (CKD)	Silica-rich industrial by-product	(Tarmac Blue Circle Ltd.)	CaO (76.8 %)		5.279 ± 0.008
Crushed basalt	Naturally occurring silicate	Cascade Mountain Range, Oregon, US (Central Oregon Basalt Products)	Al$_2$O$_3$ (12.5 %), CaO (8.9 %), Fe$_2$O$_3$ (15.5 %)	Same region as basalt applied in Kelland et al. (2020) & Buckingham et al. (2022)	15.2 ± 0.2
Crushed olivine	Naturally occurring silicate	Western Norway (Minelco Ltd.)	Fe$_2$O$_3$ (14.2 %) MgO (37.1 %)	Applied to soil cores in Renforth et al. (2015)	1.435 ± 0.005
Crushed steel slag	Silica-rich industrial by-product	West Glamorgan, Wales, UK (Port Talbot Steelworks)	CaO (50.8 %), Fe$_2$O$_3$ (28.3 %),		3.513 ± 0.008
Volcanic ash	Naturally occurring silicate	Lampung, Indonesia (S 6.08392°, E 105.45415°). 2018 eruption of Krakatau Kecil. Samples collected in Jul'2019.	Al$_2$O$_3$ (14.9 %), CaO (11.6 %), Fe$_2$O$_3$ (15.7 %)		0.660 ± 0.005
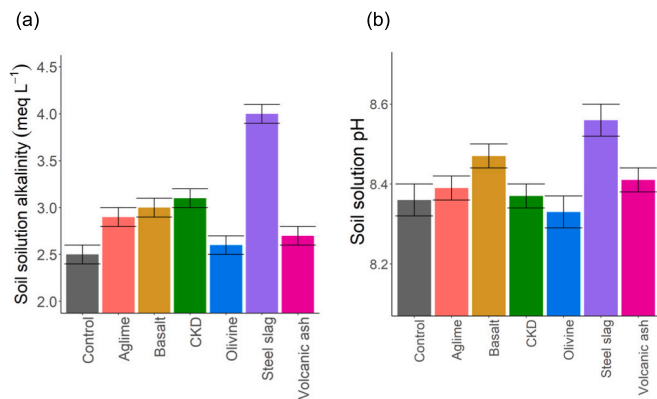
Fig. 1. Soil solution (a) titrated alkalinity and (b) pH of treated cores (coloured) and control cores (grey). Soil solution was averaged after treatment addition across the whole length, for all sampling periods, and for the three cores. Uncertainty was calculated from the standard error of observations in the three cores.

increase in leachate pH measured in basalt-treated cores by Kelland et al. (2020). Changes to soil solution alkalinity and pH were most marked in the top and middle sections of treated cores (see supplementary data).

3.1.2. Dissolved cations and anions

The concentration of dissolved cations and anions was averaged over the top (0–30 cm), middle (40–60 cm) and bottom (70–100 cm) and whole length (0–100 cm) of three cores – see SI for details. The dissolved concentration of major cations (Ca, K, Mg, P, Si) was most markedly elevated at the top of treated cores (Fig. S3). This indicates dissolution of treatments added to arable soil will increase the bioavailability of nutrients localised to the area of plant/root growth and could relieve nutrient limitation. The dissolved concentration of cations was also elevated in the effluent solution of all treated cores except olivine. This suggests EW will increase the land-ocean alkalinity flux and could alter the saturation index of carbonate and clay phases in the wider freshwater environment.

The dissolved concentration of Fe reduced in all treated cores except those treated with volcanic ash (Fig. S3) and is thought to reflect secondary mineral formation (Section 4.2). Dissolved Al reduced in cores treated with aglime, basalt, and steel slag; and dissolved Mn reduced in cores treated with basalt and steel slag. These findings are consistent with reduced [Al], [Fe] and [Mn] in basalt-treated soils in Buckingham et al. (2022) and with reduced Al and Mn toxicity following basalt addition to weathered soils (Anda et al., 2013). The dissolved concentration of Cd, Co and Pb increased in all treated cores and is associated with an increase in metal hydroxide solubility at high pH (van der Sloot and Kosson, 2010; Król et al., 2020). Although EW is often discussed as a means of reducing heavy metal toxicity in the soil-plant system, these findings suggest heavy metal toxicity depends on the initial pH of arable land.

The total dissolved positive and total dissolved negative charge of solution was calculated from the concentration of dissolved major cations (Ca^{2+}, K^+ and Mg^{2+}) and anions (F^-, Cl^-, Br^-, NO_3^-, SO_4^{2-}) and is shown in Fig. 2a (see also SI; Eqs. (S3)–(S4)). The combined positive charge largely derives from Ca release (Fig. S4a); and is significantly elevated at the top of all treated cores, except those treated with olivine and basalt (Fig. S4b). The combined positive charge averaged across the whole length of three cores after treatment is elevated relative to the control cores for all treatments except olivine (Fig. 2a). Increased dissolution of negatively charged ions in treated cores, relative to the control cores, is smaller and only significant for Aglime and CKD. Where observed, increases in anions is associated with elevated concentrations of dissolved nitrate and sulphate ions, and dissolved chloride ions in CKD-treated cores. Anion release is largely attributed to treatment
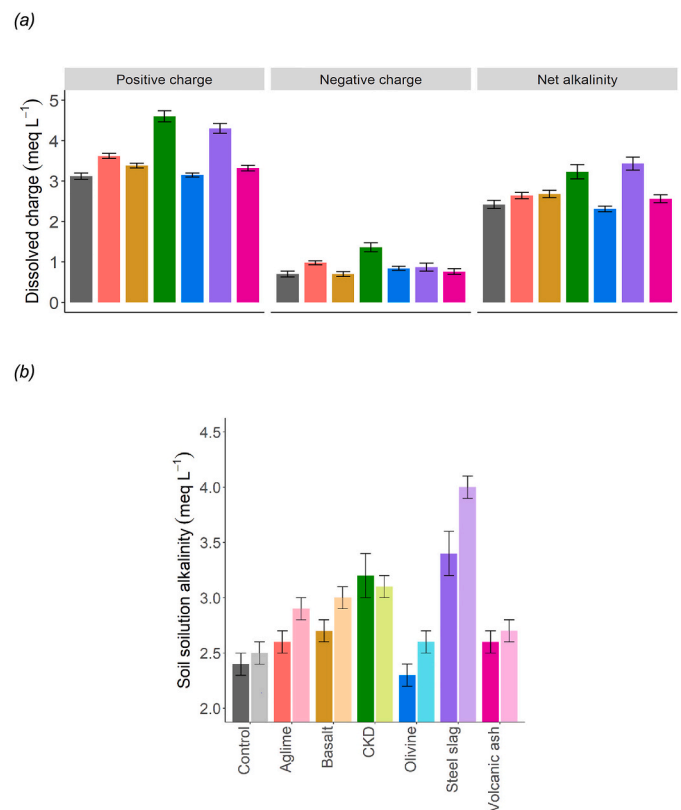

Fig. 2. (a) The combined dissolved positive and negative charge associated with major cations and anions, and the net conservative alkalinity calculated from the difference. Values are averages across three cores for each treatment type. (b) Comparison between conservative alkalinity (dark colours, left bar) and titrated alkalinity (light colours, right bar), averaged after treatment across the full length of three treated and untreated cores.

composition, because CKD contains comparatively high levels of chloride (0.4 ± 0.5 %) and sulphide (0.28 ± 0.01 %) ions, but is also expected to be due to increased dissolved nitrate associated with indirect changes to soil-water chemistry (Amatya et al., 2011).

Increases in conservative alkalinity due to treatment addition is an indicator of CO_2 drawdown and is calculated from the net charge difference between the combined dissolved positive and negative charge, averaged after treatment addition across the whole length of triplicate cores (Eq. (S5)). Conservative alkalinity is compared with titration alkalinity measured on the same samples (Fig. 2b). Titration alkalinity is similar to or higher than conservative alkalinity, with the higher values likely due to presence of dissociated organic acids which are not expected to contribute to CO_2 drawdown. Conservative alkalinity is used in all further analysis presented here.

3.2. Exchangeable fraction & XRD

Treatment made little or no change to the bulk mineralogy of the soil cores (see supplementary data). Given the detection limits of XRD are normally no better than 5 %, the absence of additional phases 15 months after treatment application is unsurprising. In most cases, no statistically significant changes to exchangeable cation loading are observed after treatment addition (Fig. S5), with the exception of exchangeable Mg which increased by on average 40 ± 10 % across the length of olivine-treated cores, and most notably at the top of those cores (70 ± 10 %). However, uncertainties in the relative change in exchangeable Ca are large across all treated cores (~±500 %), and the potential for uptake of dissolved products into the exchangeable soil fraction remains, particularly on first application of treatments to soils with lower background

levels of sorbed Ca. The extent that the exchangeable pool acts as a sink of dissolved cation remains a poorly quantified aspect of the impact of EW treatment and worthy of further research.

4. Discussion

4.1. Calculating treatment dissolution rates from cation release rate

Quantifying the rate at which added treatment materials dissolve in the soil environment is important for understanding how addition will influence soil composition via the accumulation of residual treatment. The rate of mass reduction of the added treatment material per year (% yr^{-1}) was calculated from the rate of cation release (mol cm^{-2} s^{-1}, Eq. (S2), see SI) of the dominant cation for each treatment (Mg in olivine, Ca in all other treatments) into the dissolved phase, relative to the mass of that cation added in the treatment material. This approach assumes stoichiometric dissolution, and assumes Mg and Ca are not significantly removed into secondary minerals or into the exchangeable pool. If these assumptions are not met, the rate of dissolution of treatment will be increased relative to that calculated here, but products from that additional dissolution will be retained in the soil.

In the relatively dry conditions of this experiment, and with the grain size of treatment used, <1 part in 1000 dissolves in a year for all treatments, and the majority of each treatment is retained within the soil undissolved, or possibly as secondary minerals or adsorbed on mineral surfaces (Table 2).

4.2. Implications for the soil

4.2.1. Accumulation of undissolved treatment

The shrinking core model (SCM) described in Hangx and Spiers (2009) was used to calculate the extent of dissolution integrated over the particle size distribution of added treatments. In this study, the cation release rate of the dominant cation (mol cm^{-2} s^{-1}, see supplementary data) was used as a proxy for treatment dissolution. The SCM indicates >99.99 % all six treatments remain undissolved one year after application and that treatments undergo complete dissolution within 10^6–10^7 years. Treatments therefore remain in the soil for very long periods, adding to the soil material. A single application at 50 t ha^{-1} is the equivalent of 0.2–0.3 cm of soil thickness when accounting for soil porosity, therefore a coating equivalent to the depth of the ploughed layer (20 cm) will accumulate after ≈100 years of annual addition. Consecutive application at 50 t ha^{-1} may therefore be restricted to ≈10 years to ensure agricultural soil remains viable.

4.2.2. Heavy metal release and retention

The impact of treatment addition on the soil-water environment was assessed by comparing trace-metal concentrations, measured in the dissolved and solid soil phase, to recommended safety thresholds for freshwater (Environment Agency, 2009; Gautam et al., 2014; WHO, 2022) and soil (Environment Agency, 2009a) respectively.

The dissolved concentration of heavy metals (As, Cr, Cu, Hg, Ni, Pb, V, Zn) averaged over the whole length of all triplicate treated cores stayed within the safety threshold for drinking water and freshwater with the exception of cores treated with steel slag (Environment Agency,

2009; Gautam et al., 2014; WHO, 2022) (see supplementary data). Steel slag addition increased the concentration of dissolved vanadium and arsenic at the top of the core to 230 % and 87 %, respectively, of the safety threshold for V in freshwater (60 ppb; Environment Agency, 2009) and As in drinking water (10 ppb; WHO, 2022). This suggests multiple steel slag additions could pose an environment risk to plant uptake and the surrounding freshwater environment. Unlike olivine-treated soils in Amann et al. (2020), the dissolved concentration of Ni and Cr was not significantly elevated at the top of olivine-treated soil cores and is thought to reflect slow release of trace metals in water-limited soils.

Heavy metals in the applied treatment will accumulate in the soil environment as a result of slow dissolution. It is therefore important to consider the maximum number of years that treatment can be continuously applied to agricultural soils before soil guideline values are surpassed. Findings in this study suggest olivine addition will be limited by Cr toxicity within 1 year, and steel slag addition will be limited by Cr toxicity within 6 years. In contrast, the low heavy metal content of the remaining treatments permit application of aglime, basalt, CKD and volcanic ash over the 10-year period suggested above.

These findings suggest olivine and steel slag should be applied with caution and restricted to limited amounts of addition to account for the most stringent toxicity limits in the soil and dissolved phase. It is likely extensive monitoring systems will be needed to assess the impact of applications on ground- and fresh-water environments.

4.2.3. Secondary mineral formation

A geochemical model, PHREEQC (Parkhurst and Appelo, 1999), was used to assess the saturation state of mineral phases in soil solutions. The saturation state was averaged across samples taken from the entire profile of three cores at the end of the study (Fig. S6).

Solutions sampled from treated cores were oversaturated with respect to calcite and dolomite. If these minerals do form in the cores, it would represent an additional and unquantified amount of CDR. Precipitation of carbonates is generally kinetically limited, however, and the very presence of oversaturation indicates some such limitation in this soil setting. The presence of secondary carbonate formation could not be identified by XRD in this study, and would likely require a longer study, or one using targeted geochemical tracers, to elucidate and quantify.

Fe-bearing minerals reach saturation in all treated cores (Fig. S6) and are expected to precipitate. This is consistent with the absence of appreciable Fe in the dissolved phase and elevated Fe_2O_3 measured in the middle and/or base of soils treated with steel slag and CKD (see supplementary data). Accumulation of these secondary phases should be considered, with respect to soil quality, before long-term soil treatment is pursued.

4.3. Alkalinity flux

Alkalinity fluxes (eq ha^{-1} yr^{-1}) were calculated by multiplying the net alkalinity release (eq ml^{-1}, Section 3.1) (treated minus control) by the natural infiltration rate (ml yr^{-1}), and normalised to the land-area of treatment addition (ha) (Eq. (S6)). Note that these alkalinity fluxes exclude cations removed from solution into the exchangeable pool and by secondary mineral formation. This approach means that the alkalinity flux provides a direct assessment of CO_2 removed from the atmosphere and discharged from the soil into rivers or groundwater, but does not include CO_2 removed via any carbonate precipitation.

Alkaline-rich industrial silicates (CKD, steel slag) were most effective at releasing alkalinity into solution, whereas olivine addition resulted in negligible alkalinity flux (Fig. 3). This differs from elevated dissolved Mg measured in existing olivine-treated mesocosm studies (Renforth et al., 2015; Amann et al., 2020). The difference between the olivine response in this study and existing studies is likely to be dominantly due to differences in water flux, and may also reflect the preferential substitution

Table 2
Rate of mass reduction of added treatment material (% yr^{-1}). For aglime, for instance, only 1 part in 10,000 of the treatment dissolves per year.

Treatment	Rate of mass reduction of added treatment material (% yr^{-1})
Aglime	0.010 ± 0.001
Basalt	0.031 ± 0.004
Cement kiln dust	0.053 ± 0.006
Olivine	0.001 ± 0.001
Steel slag	0.064 ± 0.008
Volcanic ash	0.020 ± 0.002

of exchangeable Ca in lime-rich soils with dissolved Mg, with potential secondary formation of dolomite and calcite phases which reduces alkalinity (Section 4.2). The extent to which exchange and secondary mineral formation influence the dissolved concentration of cations is likely to differ under different soil landscapes depending on soil and treatment composition, cation exchange capacity, hydrological conditions and soil-mineral-water interaction. In light of this, the CDR potential of enhanced weathering will vary across different agricultural environments and consideration should be given to the compatibility of a mineral treatment to the soil environment to maximise drawdown from EW while maintaining soil quality.

4.4. Carbon dioxide removal potential

In this section, the rate of CO_2 drawdown ($kgCO_2$ ha^{-1} yr^{-1}) was calculated following a single addition of treatment to arable soil at 50 t ha^{-1} assuming additional released conservative alkalinity is balanced by HCO_3^- ions provided by drawdown of CO_2 from the atmosphere (Eq. (S7)). To align with common C-cycle reporting, moles of HCO_3^- assessed from this experiment are converted into $kgCO_2$ and normalised to land area quoted in ha (Table 3). The drawdown potential (in $kgCO_2$ ha^{-1} yr^{-1}) was calculated by scaling the CDR potential over multiple annual additions. This was capped at 10 years for all treatments to prevent undissolved treatment becoming >10 % of the soil plough layer, and to a single year for steel slag and olivine to prevent soil toxicity (Table 3).

The drawdown potential of six treatments applied in this study are detailed in Table 3. Steel slag and CKD were the most efficient treatments at sequestering CO_2 after a single application. This reflects the high flux of Ca into solution from fast-dissolving industrial silicates. Of the three naturally occurring silicates applied in this study, basalt was the most efficient at consuming carbon. In comparison, olivine addition resulted in negligible CO_2 drawdown; this is associated with the high proportion of dissolved Mg retained by sorption on soil surfaces which were dominated by exchangeable Ca (Section 3.2). Sorbed ions do not contribute towards dissolved alkalinity, therefore this process constrains C drawdown in olivine-treated lime-rich soils in this study. These findings demonstrate how treatment composition and soil type influence the drawdown potential of EW and highlights the importance of assessing the impact of EW in different soil landscapes.

The monetary value of carbon credits from pursuit of CDR in arable land is likely to be the major incentive driving the activity. Monetary value is calculated, for soils similar to those of this study, by multiplying
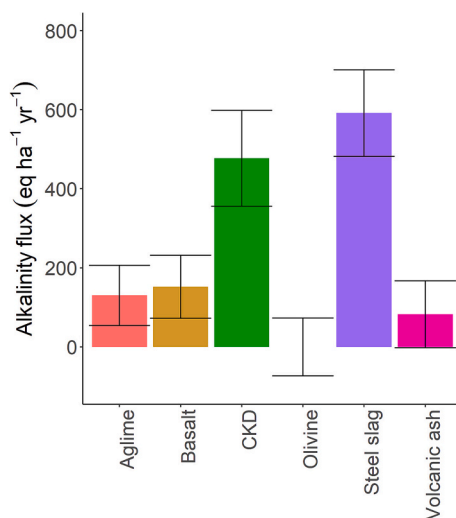

Fig. 3. Additional alkalinity flux following treatment application at 50 t ha^{-1}. The overall uncertainty was calculated in quadrature by summing the errors from each component in Eq. (S6).

Table 3

The carbon dioxide removal potential of six treatments following annual application at 50 t ha^{-1}. The single and maximum CDR potential are calculated from the alkalinity flux normalised per land area (eq ha^{-1} yr^{-1}) multiplied by the molecular mass of CO_2 (44 g mol^{-1}), and by a factor of 0.75 to account for the molar ratio of CO_2 removed from the atmosphere to alkalinity released during enhanced weathering (following Renforth, 2019). The CDR potential of aglime was reduced by 50 % to account for CO_2 released during carbonate dissolution. The maximum drawdown potential was scaled over ten annual additions and capped at a single addition for olivine- and steel slag-treated soils to prevent toxicity in arable land. The UK CDR potential was calculated by scaling the maximum CDR potential across UK cropland (6.1 Mha; DEFRA, 2020) and corrected for variation in hydrology using methods described by Buckingham et al. (2023).

Treatment	Single application ($kgCO_2$ ha^{-1} yr^{-1})	Maximum application ($kgCO_2$ ha^{-1} yr^{-1})	UK CDR potential ($MtCO_2$ yr^{-1})
Aglime	2 ± 1	20 ± 10	0.8 ± 0.4
Basalt	5 ± 3	50 ± 30	1.8 ± 0.9
CKD	16 ± 4	160 ± 40	6 ± 1
Olivine	0 ± 2	0 ± 2	0.0 ± 0.1
Steel slag	20 ± 4	20 ± 4	0.7 ± 0.1
Volcanic ash	3 ± 3	30 ± 30	1.0 ± 1.0

the CDR potential of each treatment after a single application by a price for CO2 (this study applies the present Emission Trading System rate of £83 per tCO_2; DEFRA, 2022). Based on findings in this study, the financial benefit of enhanced basalt weathering on agricultural land (£0.41/ha) is equivalent to £34 per field per year for the UK (assuming an average UK field size of 81 ha; DESNZ, 2022). This is considerably lower than the return from conventional agricultural practices; for example, typical wheat yields of 4 t ha^{-1} might return £1000/ha at £250/t. This comparison demonstrates that EW can provide a minor, but additional, financial return from cultivated land. That application may not be deemed worthwhile, however, given costs of the application itself, and considering the long-term impact to soil composition which is not yet fully understood.

4.4.1. UK CDR potential

Experimental drawdown is extrapolated to give an estimate of maximum potential CDR for the UK. This extrapolation from a single soil and hydrology does not reflect the diversity of UK conditions and is necessarily simplistic, but it still gives some indication of possible efficacy and potential for CDR by EW in the UK. Further experimental assessment will be needed to fully evaluate the nationwide CDR potential of EW.

The extrapolation from soil cores in this study assumes spreading of treatment on all UK agricultural land. Drawdown was scaled to the average UK river flux, normalised to arable cover in catchment areas (345 mm yr^{-1}; NRFA, 2022), to account for variation in hydrology across key UK agricultural areas in Buckingham et al. (2023). Such methods have been followed here to calculate the maximum CDR potential of EW in the UK ($MtCO_2$ yr^{-1}) (Fig. 4b). The CDR potential calculated in this study is the gross drawdown and does not account for emissions released during the life-cycle of EW from mining, transport and comminution. The reduction in net CO_2 drawdown from these processes is expected to be greatest in crushed, naturally occurring silicates whereas life-cycle emission are expected to be lower for fine-grained volcanic ash and industrial silicates. The maximum drawdown potential of each treatment was scaled over UK cropland to consider the contribution of each treatment to national carbon accounting.

Scaling the CDR potential of basalt over UK cropland, and accounting for national variations in hydrology, suggests application of basalt at 50 t ha^{-1} for 10 years could consume 1.8 ± 0.9 $MtCO_2$ yr^{-1}. This rate of drawdown is equivalent to 4 % of current greenhouse gas emissions from UK agriculture (which were 46.3 $MtCO_2$e yr^{-1} in 2019; DEFRA, 2021), and is consistent with a similar soil core study following application at
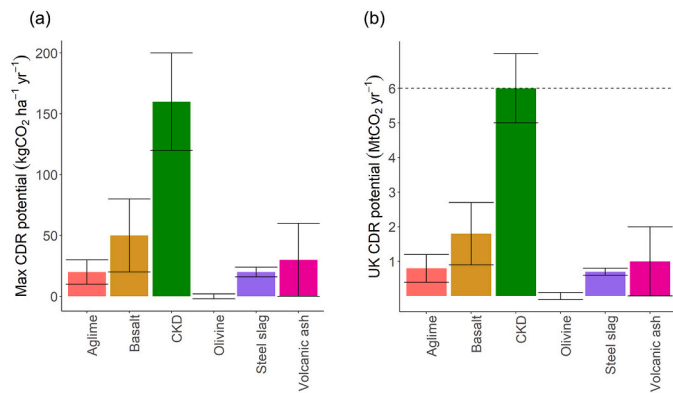
Fig. 4. The maximum carbon dioxide removal potential of six treatments following annual application at 50 t ha^{-1} (a) normalised per ha, per year (b) scaled over UK cropland and corrected for variation in hydrology across the UK. The horizontal dashed line illustrates the lower bound of CDR calculated in a vertical reactive transport model by Kantzas et al. (2022) considering basalt application at 40 t ha^{-1}.

100 t basalt ha^{-1} (1.9 $\pm$ 0.1 MtCO$_2$ yr^{-1}; Buckingham et al., 2022, 2023).

Cement kiln dust presents the most promise as an enhanced weathering treatment with the potential to remove 6 $\pm$ 1 MtCO$_2$ yr^{-1} over UK cropland assuming an infinite supply of CKD (Fig. 4a). However, historical CKD stockpiles (131 Mt., Renforth, 2012) and annual production rates (2 Mtyr^{-1}, Renforth, 2012) fall short of the amount of treatment required for EW over UK cropland for ten years at the high application rate used in this study (50 t ha^{-1}). Instead, CKD is a promising EW treatment when applied on a small scale or at a low application rate. For example, existing stockpiles and annual production of CKD is sufficient for addition at ~3 t ha^{-1} for ten years across 6.1Mha UK cropland (DEFRA, 2020). In comparison, extensive mafic rock formations in the UK (Renforth, 2012) indicate basalt is a favourable treatment for nationwide EW application.

The rate of carbon drawdown discussed in this study is considerably slower than CDR reported in other mesocosm and model-based studies. After a single application, the CDR potential of basalt was ~400 fold slower than estimated by Kelland et al. (2020) (2000 kgCO$_2$ ha^{-1} yr^{-1}), and the nationwide drawdown potential of basalt is 3–17 fold slower than CDR predicted in a model-based study (6–30 MtCO$_2$ yr^{-1}; Kantzas et al., 2022) which applies basalt at 40 t ha^{-1} with a high water flux (461–849 mm yr^{-1}). This is largely associated with components of the experimental set-up in other studies which promote dissolution, such as a high water flux, regular irrigation regime and soil type.

4.5. Impact of liming

Conventional carbon accounting assumes lime is dissolved by strong acids, particularly nitric acid derived from fertiliser use, and that all the C in the lime is therefore released as CO$_2$ to the atmosphere (Eq. (3)). Results from this study allow testing of this assumption in a UK soil, and assessment of the extent to which lime dissolution is caused by the weak carbonic acid with an accompanying removal of CO$_2$ from the atmosphere (Eq. (4)) (IPCC, 1997).

Approximately 7 % of the total tilled cropping area in Great Britain (4.4 Mha) was limed in 2020 at an average rate of 4.4 t ha^{-1} (The British Survey of Fertiliser Practice, 2021). When applying the IPCC assumption, this equates to ~0.6 MtCO$_2$ emitted per year from lime dissolution in Great Britain.

In this study application of aglime at 50 t ha^{-1} causes dissolution and a flux of Ca^{2+} into solution. Combining measurements of additional Ca^{2+} and HCO$_3^-$ in pore waters following this dissolution allows an assessment of the relative role of nitric and carbonic acid in the dissolution

(Hamilton et al., 2007). During nitric dissolution, additional positive charge from Ca^{2+} is balanced by NO$_3^-$, while in carbonic dissolution it is balanced by HCO$_3^-$. Data for this study are shown in that context in Fig. 5. The increase of positive charge from additional Ca^{2+} and Mg^{2+} in aglime-treated cores is 0.93 of the increase of negative charge from HCO$_3^-$. This is within error of the rate of change expected for pure carbonic acid dissolution, which has a gradient of 1 (represented by the dashed line in Fig. 5), and indicates that dissolution is dominantly caused by carbonic acid which will be associated with a drawdown of CO$_2$.

The role of carbonic acid in dissolving aglime in this study is perhaps unsurprising given the high soil pH and lack of N-fertiliser use on the field the cores were extracted from. Nevertheless, this result indicates that the IPCC assumption that liming is always associated with CO$_2$ release to the atmosphere is incorrect and that, in some UK soils with properties similar to those evaluated in this study, it will have the opposite effect and cause net CDR. In such conditions, the application of a carbon penalty for liming would be inappropriate as there is a small carbon benefit associated with drawdown. The soil in this study is carbonate rich and unlikely to be typical of soils where liming is normally applied to control acid conditions, but assessment of the net impact on carbon fluxes in such soils would be merited to test assumptions of CO$_2$ release. Results from this study also indicate the potential for liming to be used on higher pH soils for CDR, although this treatment appears less effective than silicate treatments investigated in this study.

5. Conclusion

The experimental design used in this soil core study closely simulates EW in a UK agricultural setting. Complete geochemical analysis and high-resolution sampling along the profile of soil cores reveals a significant proportion of added treatment is retained in the soil one year after application which could limit the number of additions while maintaining soil quality. Of the six treatments applied in this study, alkaline-rich industrial silicates (cement kiln dust and steel slag) were most effective at removing CO$_2$ after a single application. However repeated, wide-scale addition will be limited by availability (CKD) and
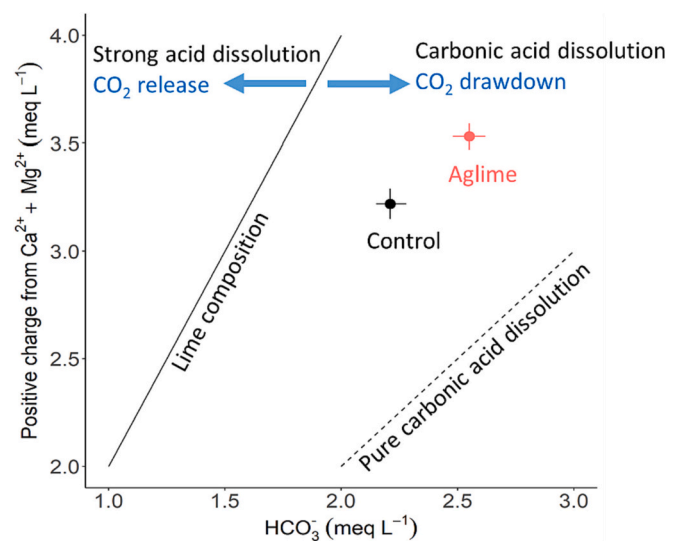

Fig. 5. Assessing C flux during dissolution of lime in soils (following the method of Hamilton et al., 2007). Positive charge from Ca^{2+} and Mg^{2+} plotted against negative charge in HCO$_3^-$. The solid line indicates the stoichiometry of the added lime. Increases of HCO$_3^-$ relative to that line indicate dissolution by carbonic acid and net uptake of CO$_2$ from the atmosphere. Dissolved soil solution data from this study are shown for the control (black) and aglime-treated (pink) cores and indicate that increases in Ca^{2+} are dominantly due to carbonic acid dissolution.

heavy metal toxicity (steel slag). Basalt presents promise for large-scale EW deployment and had minimal impact on the soil-water environment; however, the monetary value of basalt addition (£0.41/ha) is considerably less than the return from conventional agricultural practices. Liming in alkali soils was shown to act as a sink of C and should be considered in UK C budgets.

Conclusions from this core study are sensitive to the experimental design; however, assuming findings are representative of field-scale application, this research suggests spreading basalt to UK cropland land for 10 years could remove 1.8 ± 0.9 MtCO$_2$ yr^{-1} when considering variation in hydrology. This rate of CDR supports the conclusions of recently published research (Buckingham et al., 2022, 2023).

Together, these findings demonstrate CO$_2$ drawdown during enhanced weathering will not contribute substantially to UK net-zero emission reduction targets, at least in the soil and hydrology conditions common to SE England. Future work might focus on understanding the EW potential of basalt and selected industrial silicates in a range of agricultural settings with different soil types and hydrological conditions, the dynamics of sorption into the exchangeable fraction and the extent to which dissolved cations are removed via secondary carbonate formation.

CRediT authorship contribution statement

F.L. Buckingham: Conceptualization, Methodology, Formal analysis, Investigation, Data curation, Writing – original draft, Visualization, Project administration, Funding acquisition. **G.M. Henderson:** Conceptualization, Resources, Writing – review & editing, Supervision, Funding acquisition.

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.

Data availability

Data will be made available on request.

Acknowledgments

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2023.167701.

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