

Effectiveness of enhanced mineral weathering as a carbon sequestration tool and alternative to agricultural lime: An incubation experiment

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ABSTRACT

Applying finely ground silicate minerals to soils could mitigate CO₂ emissions by enhancing the rate of carbon sequestration via silicate weathering. Using these minerals instead of agricultural lime to increase soil pH would also eliminate the dissolution of lime as a major source of agricultural CO₂ emissions. However, dissolution rates of silicate minerals in the soil environment are uncertain and impacts of their application on the decomposition of soil organic matter have yet to be determined. A 3-month soil incubation was performed to investigate the effects of olivine, a highly weatherable silicate mineral, at two application rates (OLIV_{low}, OLIV_{high}) on soil CO₂ flux, available Mg and Al, and pH in comparison to control and lime-amended soils. There was no difference in cumulative net CO₂ flux between the olivine-amended soils and the control though total flux from the limed soils was 221% higher than the control. Heterotrophic respiration was also greatest in the lime-amended soils. The weathering rate of OLIV_{low} (26.7%) was higher than of OLIV_{high} (7.1%), but both treatments increased soil pH to a level sufficient to overcome aluminum toxicity. Our results suggest that olivine amendments are an effective tool for carbon sequestration and a suitable replacement for lime.

1. Introduction

Global agricultural productivity is limited by soil acidity, which impacts over 50% of the world's potentially arable land (von Uexküll and Mutert, 1995). The use of agricultural lime (CaCO₃) to raise soil pH has therefore increased the amount of land available for cultivation and greatly improved crop yields (Sumner and Noble, 2003; Yamada, 2005). However, the dissolution of agricultural lime is a major source of atmospheric CO₂ (West and McBride, 2005). Alternative methods of increasing soil pH that do not result in high rates of CO₂ production would be preferable from the standpoint of reducing global warming. Continually rising atmospheric CO₂ concentrations have rendered agricultural practices that result in CO₂ sequestration rather than production particularly desirable. Replacing agricultural lime with silicate minerals could be a viable strategy for raising soil pH while simultaneously sequestering carbon.

Silicate minerals have long been applied to both agricultural soils in the tropics and forest soils in Europe as a mineral fertilizer that has the added benefit of increasing soil pH (Anda et al., 2015, 2009; Gillman et al., 2002; Van Straaten, 2006). Uptake of CO₂ through the weathering of silicate minerals is the primary regulator of atmospheric CO₂

concentrations on geologic time scales (Gislason and Oelkers, 2008). However, the natural rate of uptake by weathering is not sufficient to negate anthropogenic CO₂ emissions, which are produced at a rate over 35 times higher than the natural rate of carbon sequestration via weathering (Amiotte Suchet et al., 2003; Olivier et al., 2016).

Using finely ground silicate minerals as a replacement for agricultural lime would expose these minerals to soil acidity, enhancing their weathering rate to expedite CO₂ consumption and serve as a low-risk method of mitigating anthropogenic CO₂ emissions (Beerling et al., 2018; Hangx and Spiers, 2009; Schuiling and Krijgsman, 2006). The consumption of protons during the weathering of applied silicate minerals would also increase soil pH, though few studies have quantified the magnitude of this effect (Renforth et al., 2015; ten Berge et al., 2012). Model results indicate that sufficient application of such minerals to weathering hotspots could draw atmospheric CO₂ concentrations back down to 350–390 ppm by 2100 (Taylor et al., 2016). However, due to the paucity of experimental data, it is unclear how the application of silicate minerals may affect soil properties, including soil pH and the decomposition rate of soil organic carbon.

The weathering of silicate minerals consumes dissolved CO₂ by neutralizing carbonic acid (H₂CO₃) to produce bicarbonate (HCO₃[−]),

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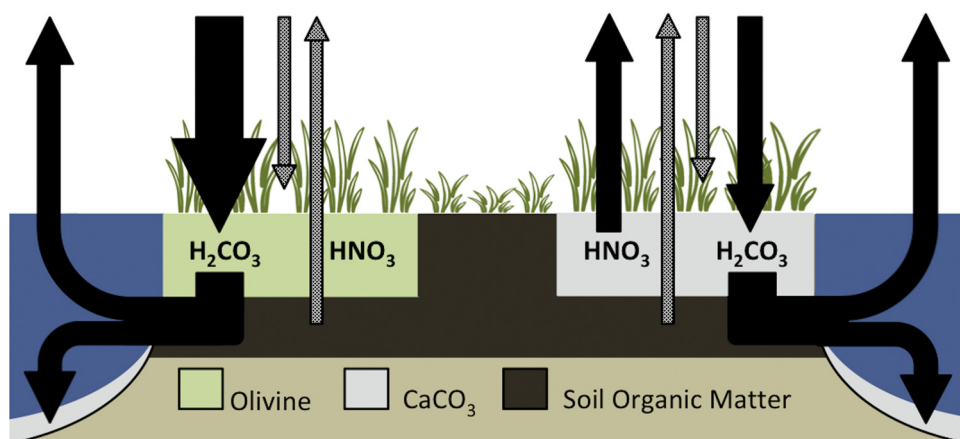
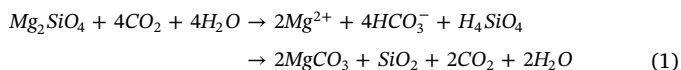


Fig. 1. Carbon fluxes induced by the application of olivine and agricultural lime (CaCO_3) to soils. Black arrows indicate known CO_2 fluxes and are scaled based on the magnitude of flux in moles of carbon per mole of applied soil amendment. Shaded arrows represent unknown CO_2 fluxes that would result if the amendments increase soil organic matter decomposition, which this experiment aims to assess, or net primary productivity. The size of these shaded arrows is not representative of expected flux sizes. Green represents olivine, gray represents CaCO_3 , and brown represents soil organic matter. The reacting acid is indicated within the treatment box, with nitric acid (HNO_3) representing strong acids that may weather applied minerals instead of carbonic acid (H_2CO_3). (For interpretation of the refer-

ences to colour in this figure legend, the reader is referred to the web version of this article.)

silicic acid (H_4SiO_4), and base cations in solution (i.e. Ca^{2+} , Mg^{2+} , Na^+ , and K^+) (Eq. (1)). These reaction products may then be leached out of the soil and transported to the ocean. The resulting influx of bicarbonate contributes to ocean alkalinity, thus counteracting ocean acidification. If the additional bicarbonate precipitates as biogenic carbonates, a portion of this carbon can then be permanently sequestered in geologic formations by forming limestone or dolomite seabeds. (Wallmann and Aloisi, 2012)



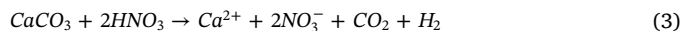
Eq. (1): Weathering reaction of Mg-olivine (forsterite), an abundant silicate mineral, and ensuing precipitation of magnesium carbonate. The weathering reaction results in 4 mol of CO_2 consumed per mole of olivine weathered. The precipitation of carbonate minerals results in more permanent sequestration but reduces the amount of CO_2 sequestered by 50%.

For the consumption of CO_2 by weathering of applied minerals to occur on useful time scales, it would be necessary to select silicate minerals that are easily weatherable. Volcanic rocks with low silica content and poor or glassy crystal structure weather the most quickly (Gislason and Oelkers, 2011). Of the major silicate minerals, olivine, which is common in basic igneous rocks, is the most rapidly weathered (Schuiling and Krijgsman, 2006).

Exposure to soil acidity enhances mineral weathering rates; soil organic acids in particular are dominant drivers of mineral dissolution in the soil (Berner and Berner, 2012). The presence of oxalic acid, for example, has been observed to increase forsterite (Mg-olivine) dissolution rates by as much as 600% (Olsen and Rimstidt, 2008). Additionally, the decomposition of soil organic matter renders the pCO_2 of the soil pore space 10–100 times higher than the pCO_2 of the atmosphere (Bohn et al., 1985). This elevated pCO_2 increases the concentration of carbonic acid in the soil solution and further enhances weathering rates. However, very acidic soils may neutralize a portion of the bicarbonate produced to re-emit CO_2 , reducing net carbon sequestration.

On the other hand, the standard practice of neutralizing soil acidity with agricultural lime adds to agricultural greenhouse gas emissions (West and McBride, 2005), which are the second largest source of greenhouse gasses globally (IPCC, 2014). The United States alone emits between 4.4 and 13.4 Tg CO_2 per year from the dissolution of agricultural lime (West and McBride, 2005). This large range in estimates is due in part to the challenge of determining the relative amounts of weathering carried out by carbonic acid and other, stronger acids. The relative importance of carbonic acid in the weathering process can vary with soil pH and agricultural practices (Hamilton et al., 2007; Perrin et al., 2008; Semhi et al., 2000). Strong mineral acids, such as nitric

acid (HNO_3), which is derived from the dissolution of nitrogen fertilizers or from inputs by acid rain, can also play an important role in carbonate weathering (Hamilton et al., 2007; Perrin et al., 2008; Semhi et al., 2000). Though the dissolution of lime by carbonic acid effectively converts CO_2 to bicarbonate (Eq. (2)), its dissolution by strong acids like nitric acid produces CO_2 (Eq. (3)).



There is therefore some debate as to whether the dissolution of lime is in fact a source or sink of CO_2 . The IPCC assumes that all carbon applied as lime becomes CO_2 upon dissolution (Houghton et al., 1997), but some studies indicate that the net effect of the dissolution of applied lime is to consume CO_2 (Hamilton et al., 2007; Oh and Raymond, 2006). Though the bicarbonate produced when lime is dissolved by carbonic acid serves as a temporary carbon sink, the release of CO_2 during the eventual reprecipitation of carbonate minerals renders this reaction a carbon neutral process in the long term (Eq. (2)). This temporary sink may be relevant to addressing elevated atmospheric CO_2 concentrations in the short term, but more research defining its residence time is needed (Hamilton et al., 2007).

Alternatively, the weathering of olivine by carbonic acid consumes twice as much CO_2 as the dissolution of lime, making it a more effective soil amendment with respect to carbon sequestration (Fig. 1). However, the weathering effect of strong acids also needs consideration when assessing the carbon sequestration potential of silicate minerals, particularly when soil pH is below 5 (Plummer et al., 1979). The dissolution of silicate minerals by nitric acid, for example, does not produce carbonate alkalinity, and would therefore reduce the carbon sequestration efficiency of the process. Nonetheless, given that dissolution of agricultural lime by nitric acid is a source of CO_2 , the replacement of lime with an alternative soil amendment would be still more critical in terms of preventing CO_2 emissions if such acids play a strong role in mineral dissolution in a particular environment.

However, the potential effects of applied silicate minerals on soil organic carbon (SOC) have yet to be addressed by those promoting the use of enhanced mineral weathering as a carbon sequestration technique. It is well established that microbial activity increases with increasing pH, which can result in faster turnover of soil organic matter (Fuentes et al., 2006; Paradelo et al., 2015). Therefore, raising soil pH by the addition of olivine has the potential to increase SOC loss by decomposition, reducing the efficiency of carbon uptake by enhanced mineral weathering.

Before the application of silicate mineral amendments can be considered as a management tool for carbon sequestration, it is critical that any effects on SOC are assessed. This study aimed to determine if

Table 1
Chemical Composition of Olivine.

	Content (%)
Mg	22.30
Fe	3.62
Al	0.70
Ni	0.232
Ca	0.31
Cr	0.157
K	0.065
Na	0.043
S	0.022

increases in decomposition with the application of olivine outweighed the uptake of carbon by enhanced weathering. To do this, we examined the effects of these amendments on heterotrophic respiration and estimated the consumption of CO₂ associated with mineral weathering. As silicate minerals may replace lime, it is also necessary to compare the effects of olivine amendments with the effects of agricultural lime applications and assess differences in net soil CO₂ flux between the two treatments. A liming treatment was included in the study for this purpose.

We also endeavored to assess the effectiveness of olivine as an alternative to agricultural lime. To this end, we compared the effects of these amendments on soil pH. As high levels of exchangeable aluminum at low pH are often the limiting factor for plant productivity in acidic soils (Adams, 1984), we also tested the effects of olivine on aluminum availability.

Though it is known that olivine dissolution proceeds most quickly in warm, moist, low-pH environments (Hangx and Spiers, 2009) and there is general consensus that organic anions and high molecular weight organic acids act to increase the rate of olivine dissolution (Olsen and Rimstidt, 2008; Wogelius and Walther, 1991), only two studies have attempted to quantify weathering rates of applied olivine in the soil environment (Renforth et al., 2015; ten Berge et al., 2012). Uncertainty surrounding dissolution rates has therefore resulted in a wide range of predicted values for the amount of CO₂ the weathering of applied olivine has the potential to sequester (Renforth, 2012). The extent to which olivine application rates may influence dissolution is also unclear, though such information is critical to maximizing the efficiency of this process. Our study therefore included two application rates with the goal of quantifying olivine dissolution as a function of the amount applied.

2. Methods

2.1. Sampling and analyses

Soil samples were taken from Nørholm Hede, a heathland in southwestern Denmark. This site was chosen for its organic-rich, acidic soils and because it had not been managed as an agricultural field since the 1890's, eliminating any risk of the presence of residual applied lime which could impact our results. Though these highly acidic soils are not reflective of typical agricultural soils, the soil properties at this site increased the probability of detecting any effects of olivine applications within the short time frame of our incubation. The low pH of these soils was expected to increase the rate of mineral dissolution, whereas the high carbon content was likely to magnify the effect size of any impacts on the decomposition of soil organic matter. The results of this study therefore represent an approximate upper limit to the impacts of mineral applications.

Soils at the site were developed on glacial sand and are classified as sandy podzols. The undisturbed portion of the heathland has a typical podzol profile, but in the previously cultivated area from which our samples were taken the A horizon has been mixed with the E horizon in

Table 2
Treatment application rates.

Treatment	Amount applied (g g _{soil} ⁻¹)
Control	–
Lime	0.008
OLIV _{low}	0.008
OLIV _{high}	0.04

a plough layer. Our samples were taken from the organic-rich top 0–10 cm of the soil in an area dominated by the perennial grass *Deschampsia flexuosa* (L.). The initial pH was 3.55, and the soil carbon concentration was 5.49%. (Evelien, 2012)

The moist-weight equivalent of 20 g of dry soil was placed into each of forty 110 ml bottles after being sieved to 2 mm. Any roots that passed through the 2 mm sieve remained in the samples. These samples were then divided into four treatment groups: control, lime, high olivine (OLIV_{high}), and low olivine (OLIV_{low}). For our treatments we used a finely ground olivine and a finely ground lime, both of which had a d50 grain size of 20 µm. This lime was determined to be 97% CaCO₃ based on acid digestion according to EPA Method 3050 followed by inductively coupled plasma atomic emission spectrometry (ICP-AES). Our olivine powder was primarily composed of forsterite, containing 22.3% Mg and only 3.62% Fe by mass as determined by ICP-AES following acid digestion according to AOAC Method 957.02. The chemical composition of the applied olivine is shown in Table 1.

To the ten lime-amended samples, 0.16 g of lime (0.008 g_{lime} g_{soil}⁻¹) was added and thoroughly mixed into the soil (Table 2). Similarly, 0.16 g of olivine was added to the OLIV_{low} treatment, and 0.80 g of olivine (0.04 g_{oliv} g_{soil}⁻¹) was added to the OLIV_{high} treatment. All samples, including the controls, were mixed for the same amount of time in order to ensure similar amounts of disturbance. Soils were then packed down in the chambers to a bulk density of approximately 1.245 g cm⁻³.

The higher olivine application rate was chosen to match the 5 kg m⁻² yr⁻¹ of harzburgite, an olivine rich rock, that Taylor et al. (2016) estimated would result in a drawdown of CO₂ sufficient to return atmospheric CO₂ concentrations to approximately 350 ppm by 2100 if applied to weathering hotspots, assuming a 10 cm mixing depth. Without this harzburgite application, atmospheric CO₂ concentrations are otherwise predicted to reach 540 ppm by 2100 under the IPCC's RCP 4.5 medium-level mitigation scenario (Taylor et al., 2016). OLIV_{high} was therefore intended to represent an application rate large enough to substantially impact atmospheric pCO₂. The application rate for OLIV_{low} and lime was selected with several factors in mind. This application rate matched Taylor et al.'s 1 kg m⁻² yr⁻¹ application rate, which under the same conditions as previously described is expected to decrease CO₂ concentrations by ~140 ppm compared to the standard RCP4.5 scenario. It was also not considerably higher than the approximately 0.006 g_{lime} g_{soil}⁻¹ recommended to raise soil pH from 3.5 to 6.5 according to the manufacturer's instructions. These application rates also fall within the range of olivine application rates used as treatments by ten Berge et al. (2012) (0.000727, 0.00364, 0.0182, and 0.105 g_{oliv} g_{soil}⁻¹), allowing for comparison with their results.

$$F_{\text{net}} = F_{\text{resp}} + F_{\text{diss}} - F_{\text{weathering}} \quad (4)$$

The net flux of CO₂ between the incubated soil and the atmosphere (F_{net}) reflects the combination of heterotrophic soil respiration (F_{resp}) and CO₂ produced by the dissolution of lime (F_{diss}), if applicable, less any consumption of CO₂ due to mineral weathering (F_{weathering}) (Eq. (4)). F_{net} was determined by measuring the CO₂ concentration in the headspace of the closed chamber with an SBA-5 infrared gas analyzer (PP Systems, Amesbury, MA), connected via a closed loop. As the CO₂ flux from the soil decreases as the headspace pCO₂ increases, linearly determining the flux rate based on the first and last measurement would

underestimate the rate of CO₂ accumulation in the chamber. Therefore, the Hutchinson-Mosier non-linear exponential regression model was used to calculate the initial F_{net} in $\mu\text{mol CO}_2 \text{ day}^{-1} \text{ g-soil}^{-1}$ from a continuous time series of headspace CO₂ measurements (Hutchinson and Mosier, 1981; Pihlatie et al., 2013). Measurements started from standard initial concentration of 425 ppm. The length of the sampling period ranged from 1 to 3 min over the course of the experiment based on the rate of CO₂ production, but the same measurement period was used for all samples at each time point.

Initial measurements were taken after two days of pre-incubation following the addition of treatments to the samples. Measurements were taken over the course of three months on days 3, 5, 7, 10, 13, 20, 27, 41, 69, and 97 after the addition of treatments. Throughout the experiment, de-ionized water was added after flux measurements to maintain soil moisture approximating field conditions. Samples were stored at room temperature (20–25 °C) and left uncovered between measurements to prevent the buildup of CO₂ from inhibiting microbial activity.

After the 3-month incubation period, samples were oven dried at 60 °C for 48 h, re-homogenized, and split into three sets of subsamples for additional analyses. Soil pH was measured in a 1:1 mixture of soil and deionized water (Burt, 1996). Subsamples of the control and olivine treated soils were subjected to 1M NH₄NO₃ extraction. 30 ml of 1M NH₄NO₃ were added to 3 g of soil, which was shaken over night. After centrifuging for 10 min, the supernatant was retained. This process was repeated two more times with 10 ml 1M NH₄NO₃ and 10 min of shaking before centrifuging. The resulting solution was then vacuum-filtered through a 0.22 μm Millex-GP filter (Millipore) and analyzed for exchangeable cations, including Mg and Al, by ICP-AES.

The fraction of olivine weathered (%O_w) was calculated by subtracting the amount of exchangeable Mg in the control samples (Mg_{control}) from the amount of exchangeable Mg in the olivine-treated samples (Mg_{OLIV}), and dividing by the total amount of Mg applied as olivine (Mg_{applied}) (Eq. (5)) (ten Berge et al., 2012).

$$\%O_w = (Mg_{\text{OLIV}} - Mg_{\text{control}}) / Mg_{\text{applied}} \quad (5)$$

In making this calculation, we assumed stoichiometric dissolution. This is a reasonable assumption as magnesium from weathering olivine has been shown to dissolve congruently with silica in conditions comparable to the soil environment after an initial period in which preferential removal of cations by organic acids results in an silica rich phase developed on the surface (Barman et al., 1992; Olsen and Rimstidt, 2008). However, there is evidence that some organic acids may preferentially remove iron over magnesium, which could result in an underestimation of the amount of olivine weathered when using magnesium as a proxy (Barman et al., 1992). The theoretical upper limit for CO₂ consumption by olivine weathering is 4 mol of CO₂ per mole of olivine to produce 4 mol of bicarbonate (Eq. (1)). This 4:1 ratio was used to estimate the amount of CO₂ that was converted to bicarbonate by weathering of the applied olivine, assuming that carbonic acid was the driver of all weathering.

2.2. Statistical analyses

Differences in F_{net} over time between treatments were compared using a linear mixed effects model (“lme” in R Version 3.2.3) with sample number as a random intercept term to account for repeated measures and treatment and day as categorical fixed effects ($\alpha = 0.05$) (Pinheiro et al., 2017). Heterogeneity of variances was corrected for by allowing variances to differ for each treatment and day. The first four measurements from day 69, one from each treatment, were suspected to be influenced by measurement errors and were therefore removed. Post hoc comparisons using Tukey’s tests (“lsmeans” in R) were used to interpret significant interactions ($\alpha = 0.05$).

The cumulative F_{net} over the course of the experiment ($F_{\text{net,tot}}$) for

each sample was determined by calculating the area under the curve of F_{net} rates over time (Prism 7, La Jolla, CA). We calculated the cumulative soil respiration ($F_{\text{resp,tot}}$) of the olivine-amended samples by adding the amount of CO₂ consumed by olivine weathering ($F_{\text{weathering}}$) to $F_{\text{net,tot}}$. As our methods did not allow us to separate out lime-derived CO₂ from microbially respired CO₂, we were only able to calculate a minimum value for $F_{\text{resp,tot}}$ for our lime-amended samples. To do this we subtracted the amount of CO₂ that would be produced if all of the added lime-C were converted to CO₂ from $F_{\text{net,tot}}$. Though there is significant uncertainty in this estimate, any deviations from our assumed conditions would result in a larger value for $F_{\text{resp,tot}}$ of the limed samples. We are therefore confident in this calculated minimum, which allows for comparison with other treatments. In the control samples, $F_{\text{resp,tot}}$ was equivalent to $F_{\text{net,tot}}$.

Differences in $F_{\text{net,tot}}$, $F_{\text{resp,tot}}$, exchangeable Mg and Al, and pH were tested across treatments by one-way ANOVA followed by Tukey’s tests for multiple comparisons ($\alpha = 0.05$) (Prism 7, La Jolla, CA). We also tested for any correlation between $F_{\text{resp,tot}}$ and sample pH using Pearson’s correlation coefficient (Prism 7, La Jolla, CA) to determine if changes in pH with treatment were responsible for any observed differences in respiration. Straight lines and quadratic curves were fit to the data in order to assess the form of the relationship.

3. Results

We found a significant interaction between the effects of treatment and time on F_{net} ($p < .001$, F-test). Lime had a significantly higher F_{net} than all other treatments until day 97 ($p < .001$, Tukey’s HSD), at which point there was no significant difference between lime and the other treatments (Fig. 2, Table 3). F_{net} of OLIV_{high} was initially significantly higher than the control and OLIV_{low} ($p < .05$, Tukey’s HSD). However, by day 10, OLIV_{high} was no longer elevated relative to OLIV_{low} and the control. Beginning on day 20, there was a trend such that F_{net} from the control was higher than F_{net} from the olivine treatments, but the only significant difference was between OLIV_{high} and the control on day 97, at which point OLIV_{high} was 17.5% lower than the control ($p = .0421$, Tukey’s HSD). At no point was there a significant difference between OLIV_{low} and the control.

Treatment had a significant effect on available magnesium ($p < .001$, F-test). The amount available in the control (52.5 mg kg⁻¹, $SD = 3$) was significantly lower than OLIV_{low} (529 mg kg⁻¹, $SD = 88$) ($p < .001$, Tukey HSD), which was in turn significantly lower than OLIV_{high} (685 mg kg⁻¹, $SD = 131$) ($p = .0022$, Tukey HSD) (Fig. 3). From these values we estimate that 26.7% ($SD = 4.9$) of the applied

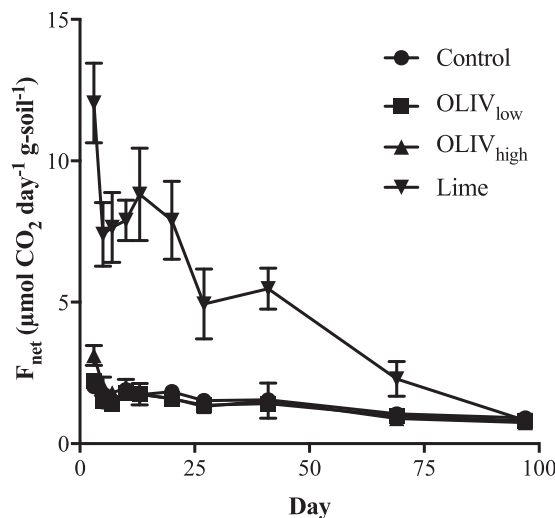


Fig. 2. Net CO₂ flux (F_{net}) by treatment over the course of the incubation. Error bars indicate SD.

Table 3

Mean net CO₂ flux (F_{net}) for each treatment by day (SD). Treatment means from the same day followed by the same letter are not significantly different ($\alpha = 0.05$), as assessed by Tukey's HSD.

Day	Control	Oliv-Low	Oliv-High	Lime
3	2.02 (0.12)a	2.22 (0.23)a	3.12 (0.35)b	12.04 (1.41)c
5	1.47 (0.22)a	1.51 (0.18)a	2.03 (0.33)b	7.40 (1.13)c
7	1.45 (0.26)a	1.39 (0.10)a	1.80 (0.14)b	7.64 (1.24)c
10	1.91 (0.23)a	1.80 (0.24)a	1.97 (0.30)a	7.89 (0.72)b
13	1.73 (0.19)a	1.75 (0.38)a	1.76 (0.17)a	8.82 (1.63)b
20	1.83 (0.17)a	1.59 (0.13)a	1.59 (0.21)a	7.89 (1.38)b
27	1.52 (0.09)a	1.35 (0.16)a	1.32 (0.18)a	4.94 (1.24)b
41	1.55 (0.17)a	1.41 (0.17)a	1.52 (0.62)a	5.48 (0.73)b
69	1.05 (0.17)a	0.98 (0.18)a	0.89 (0.19)a	2.28 (0.62)b
97	0.91 (0.04)a	0.81 (0.12)a,b	0.75 (0.13)b	0.82 (0.20)a,b

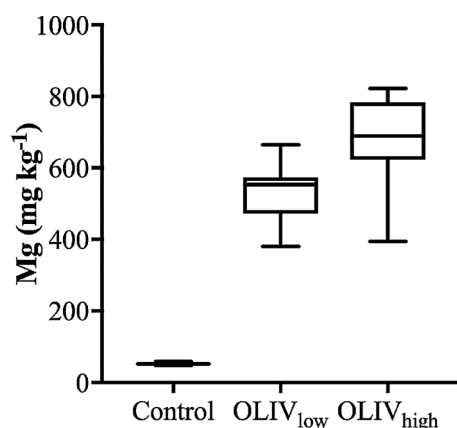


Fig. 3. Distribution of exchangeable Mg values as a function of treatment at the end of the experiment. The central line indicates the median value, the boxes extend from the 25th to 75th percentiles, and the whiskers indicate the minimum and maximum values. All treatments were significantly different from one another ($p < .005$, Tukey HSD).

olivine was dissolved in OLIV_{low}, whereas only 7.1% ($SD = 1.8$) of the applied olivine was weathered in OLIV_{high}.

Based on the amount of olivine dissolved in each treatment, we calculate that gross carbon sequestration was on average 0.050 g CO₂ ($SD = 0.009$) per chamber for OLIV_{low} and 0.067 g CO₂ ($SD = 0.017$) for OLIV_{high}. Scaling these values up with an assumed 10 cm mixing depth, OLIV_{high} application rates would have consumed 4.16 metric tons CO₂ ha⁻¹ ($SD = 1.04$), and OLIV_{low} application rates would have consumed 3.13 t CO₂ ha⁻¹ ($SD = 0.58$) over the course of three months.

Treatment had a significant effect on both $F_{\text{net,tot}}$ and $F_{\text{resp,tot}}$ ($p < .001$, F -test) (Fig. 4). Only the $F_{\text{net,tot}}$ of lime (405 $\mu\text{mol CO}_2 \text{ g}^{-1}$, $SD = 55$) was significantly higher than the other treatments (OLIV_{low}, OLIV_{high}, and control) ($p < .001$, Tukey's HSD). Lime increased $F_{\text{net,tot}}$ by a factor of 3.21 over the control, 3.49 over OLIV_{low}, and 3.58 over OLIV_{high}. There was no significant difference in $F_{\text{net,tot}}$ between the control (126 $\mu\text{mol CO}_2 \text{ g}^{-1}$, $SD = 8$), OLIV_{low} (116 $\mu\text{mol CO}_2 \text{ g}^{-1}$, $SD = 11$), and OLIV_{high} (113 $\mu\text{mol CO}_2 \text{ g}^{-1}$, $SD = 21$).

OLIV_{low} and OLIV_{high} both had significantly higher $F_{\text{resp,tot}}$ than the control ($p < .05$, Tukey's HSD). However, at 174 ($SD = 18$) and 188 ($SD = 29$) $\mu\text{mol CO}_2 \text{ g}^{-1}$, respectively, OLIV_{low} and OLIV_{high} were not significantly different from one another. $F_{\text{resp,tot}}$ from the lime-amended samples (325 $\mu\text{mol CO}_2 \text{ g}^{-1}$, $SD = 55$) was significantly higher than all other treatments ($p < .001$, Tukey's HSD): $F_{\text{resp,tot}}$ with lime was 2.6 times higher than the control, 1.9 times higher than OLIV_{low}, and 1.7 times higher than OLIV_{high}.

Soil pH was significantly affected by treatment ($p < .001$, F -test); the mean pH of all treatments were significantly different from one

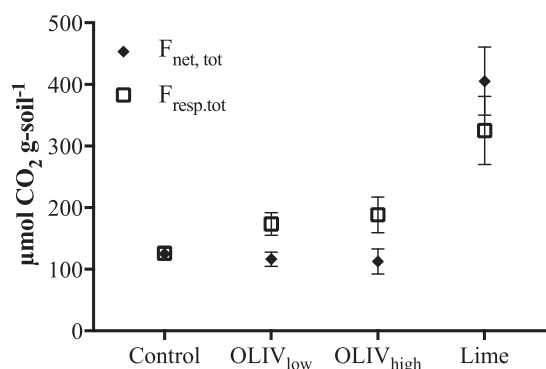


Fig. 4. Mean cumulative net CO₂ flux ($F_{\text{net,tot}}$) and mean cumulative soil respiration ($F_{\text{resp,tot}}$) by treatment. Error bars indicate SD, but are not shown when shorter than the height of the symbol. Lime is significantly higher than all other treatments with respect to both variables ($p < .001$, Tukey HSD). $F_{\text{resp,tot}}$ of OLIV_{high} ($p = .0008$, Tukey HSD) and OLIV_{low} ($p = .0129$, Tukey HSD) are significantly higher than the control.

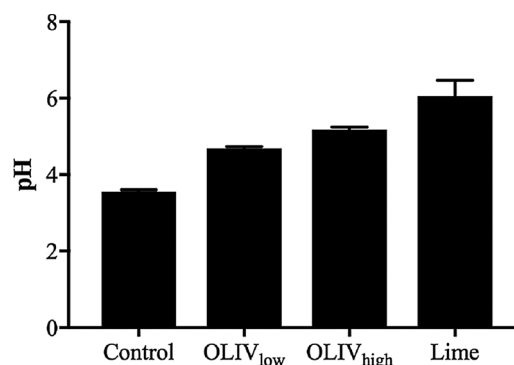


Fig. 5. Treatment effects on soil pH. All treatments are significantly different from one another ($p < .05$). Error bars indicate SD.

another ($p < 0.001$, Tukey's HSD) (Fig. 5). The final pH of the control was 3.56 ($SD = 0.04$), OLIV_{low} was 4.69 ($SD = 0.05$), OLIV_{high} was 5.18 ($SD = 0.06$), and lime was 6.06 ($SD = 0.4$). Lime increased soil pH by 2.51 points compared to the control, whereas the OLIV_{high} and OLIV_{low} increased pH by 1.63 and 1.13 points respectively.

The increase in pH decreased the availability of aluminum ($p < .001$, F -test), which was available in significantly lower concentrations in both OLIV_{high} (2.8 mg kg⁻¹, $SD = 0.6$) and OLIV_{low} (7.7 mg kg⁻¹, $SD = 1.1$) compared to the control (134 mg kg⁻¹, $SD = 9.7$), ($p < .001$, Tukey HSD).

$F_{\text{resp,tot}}$ was positively correlated with soil pH as assessed by Pearson's r ($r = 0.88$, $n = 40$, $p < .001$). This correlation was characterized by a quadratic relationship ($r^2 = 0.85$, Fig. 6).

4. Discussion

Results suggest that the application of olivine to agricultural soils reduces agricultural CO₂ emissions compared to the use of agricultural lime and has the potential to be an effective tool for carbon sequestration. The lime-amended soils were a source of CO₂ compared to the control, but the olivine-amended samples were not. The cumulative net flux ($F_{\text{net,tot}}$) of the olivine treatments was not lower than the control due to the increase in respiration accompanying the application of olivine. However, this does not necessarily imply that olivine amendments are ineffective at sequestering carbon as our experiment did not account for potential changes in plant inputs to soil carbon with treatments. Long term studies on the effects of applying agricultural lime on soil organic carbon often indicate that improved plant productivity associated with higher soil pH results in greater inputs of

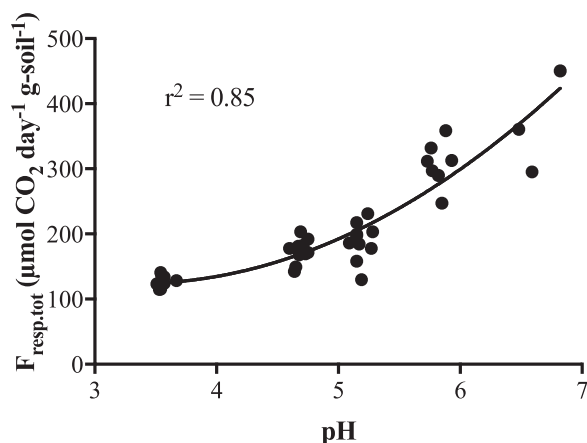


Fig. 6. Relationship of cumulative soil respiration ($F_{\text{resp,tot}}$) to soil pH, fit with a quadratic curve. Each point represents an individual sample.

organic matter to the soil, resulting in increased SOC (Paradelo et al., 2015). It is therefore likely that the improved soil conditions due to the effects of olivine application would likewise result in increased inputs to the SOM pool, negating losses due to increased decomposition rates, and rendering the application of olivine a net carbon sink. However, long-term field studies are needed to assess any differences in effects between lime and olivine on plant inputs to the soil, which would affect the overall carbon balance.

Though we observed a significantly greater increase in total soil respiration in the lime-amended soils, the olivine treatments also increased $F_{\text{resp,tot}}$ over the control. However, as we are unable to parse out any changes in the effects of olivine amendments on respiration or in the rate of weathering over the course of the experiment, it is unclear whether the increase in $F_{\text{resp,tot}}$ with the application of olivine was due to a larger initial increase in respiration, or if soil respiration remained consistently elevated compared to the control throughout the experiment. The small initial increase in the F_{net} of the OLIV_{high} treatments compared to the control was followed by a decrease on the final day. Though our methods did not allow us to determine if this decrease was primarily due a decrease in respiration or increase in consumption of CO_2 by weathering, there was a non-significant trend in which the olivine-amended treatments had lower F_{net} than the control beginning on day 20. We can therefore presume that around this time the amount of CO_2 consumption in these treatments began to outweigh any effects on soil respiration. Had our incubation period been longer and this trend continued, we might have eventually seen a net sink in OLIV_{high} compared to the control, even without changes in plant inputs playing a role.

Though the exact effects of changes in soil pH on soil carbon mineralization can be hard to predict, the increase in pH with the application of our treatments is likely driving the increase in respiration in this experiment. Soil respiration has generally been shown to increase with pH (Kemmitt et al., 2006; Paradelo et al., 2015), and indeed we observed a strong positive correlation between these two variables, but other studies on the effects of liming on soil respiration have produced varied results. Some, like ours, have found soil respiration to increase with the application of agricultural lime (Ahmad et al., 2014; Dumale et al., 2011), while others observed no effect (Bertrand et al., 2007; Biasi et al., 2008), or even reduced decomposition (Wachendorf, 2015). Yet another study found that agricultural lime induced increases in soil respiration for the first three days, likely resulting from increased solubility of SOC, but reduced SOC mineralization after that point, presumably due to greater microbial C-use efficiency under more favorable pH conditions (Grover et al., 2017). Similar mechanisms may be responsible for the changes in $F_{\text{net,resp}}$ of OLIV_{high} over the course of our experiment.

Olivine proved to be effective at increasing soil pH in a relatively short time, though its effect was not as strong as lime. However, a number of studies have indicated that lime requirements should be determined with the goal of correcting the underlying problems associated with low pH, such as toxic levels of aluminum, rather than achieving the near-neutral pH that is typically recommended (Adams, 1984; Farina et al., 1980; Farina and Channon, 1991; Hoyt and Nyborg, 1987). Aluminum toxicity, which develops below a pH of approximately 5.2 (Sumner and Noble, 2003), is often the most limiting factor affecting plant growth in acid soils (Adams, 1984; Sumner and Yamada, 2002). Raising soil pH above the level required to eliminate exchangeable aluminum has not been shown to have to provide additional benefits, and in some cases may even decrease yields (Farina et al., 1980). The application of olivine decreased aluminum availability from levels that were high enough to be toxic to plants to levels that fall within a suitable range for crop growth (Kochian et al., 2005). These results indicate that olivine is a suitable replacement for lime in regards to its ability to correct problems of soil acidity, providing an additional use for this product that would aid in its adoption for carbon sequestration purposes. As existing infrastructure for applying lime to croplands could be repurposed to apply silicate minerals, the cost of applying olivine-rich rock would be comparable to current liming practices (Schuiling and Krijgsman, 2006), and may be further offset by the sale of carbon credits or increases in crop productivity (Beerling et al., 2018), increasing the economic viability of this method of sequestering carbon.

When assessing the suitability of olivine as a soil amendment and determining application rates, it is also necessary to take into account its heavy metal content. Nickel toxicity in particular may pose a problem. However, other forms of silicate minerals, such as olivine-rich basalt, may work well as a more widely available alternative. Though the carbon capture potential of basalt is lower, it tends to have lower concentrations of nickel than olivine and also contains additional plant nutrients. Mining wastes or calcium silicate by-products of cement and steel production are also potential sources of silicate minerals that may be suitable for agricultural applications. (Beerling et al., 2018)

Determining ideal application rates will be necessary to maximize efficiency and cost-effectiveness. A significant amount of Mg was weathered from both OLIV_{high} and OLIV_{low}, which would be of substantial benefit to Mg-limited agricultural soils. However, the O_w was dependent on the rate of application. The percentage of olivine weathered was higher at the lower application rate: 26.7% of the applied Mg in the OLIV_{low} treatment was released into solution, whereas only 7.1% of the applied Mg was released in the OLIV_{high} treatment. Though the olivine application rate of OLIV_{low} was only 20% of OLIV_{high}, OLIV_{low} released 75.4% of the amount of Mg released by OLIV_{high}.

A similar trend in the fraction of olivine weathered was observed by ten Berge et al. (2012). In a pot study, they also found that higher application rates resulted in a lower percentage of olivine dissolved over the course of 32 weeks (ten Berge et al., 2012). However, the proportion of applied olivine dissolved observed in our experiment was markedly higher. The O_w in OLIV_{low} was 80% higher than observed in ten Berge et al.'s (2012) lowest application rate. Only 14.8% of the applied olivine dissolved at their lowest application rate, compared to the O_w of 26.7% in OLIV_{low}, despite the fact that our application rate was almost 50 times theirs and their incubation period was almost three times longer. Renforth et al. (2015) did not calculate the fraction weathered, but found dissolution rates in their column leaching study that were comparable to or slightly lower than those found by ten Berge et al. (2012). The discrepancy between our results and the results of these other experiments is likely due to our use of a highly acidic soil for the incubations.

At the dissolution rates we observed, the annual application rates proposed by Taylor et al. (2016) are too high. Only 28.4% of the applied olivine at the $5 \text{ kg m}^{-2} \text{ yr}^{-1}$ application rate would have

dissolved before the following years application, and the $1 \text{ kg m}^{-2} \text{ yr}^{-1}$ application would have just completed dissolution within one year. However, as pH increases and the soil approaches saturation with respect to olivine, dissolution rates should decrease. It is therefore unlikely that quick dissolution observed in the three months of our experiment would remain constant throughout the year. Further, additional applications would not be likely to weather as rapidly, rendering such high annual application rates an inefficient use of resources.

The fact that %O_w decreased at higher application rates suggests that further work is needed to optimize this process and determine the most cost-effective application rate. It may be preferable to apply smaller amounts of olivine over a larger area to maximize CO₂ consumption, rather than using high application rates on smaller areas. Optimizing the spatial distribution and application rates of silicate minerals with respect to carbon sequestration will require more data across the range of typical site conditions in agroecosystems. Such information will also be critical in order to accurately quantify the amount of CO₂ this process consumes. Accurate accounting of sequestered carbon will be necessary if this process is to be certified for carbon credits, which may offset some of the cost and increase its adoption by farmers.

5. Conclusion

Our results illustrate the potential utility of enhanced mineral weathering as a carbon sequestration tool. Over the course of only three months, on an areal basis the dissolution of olivine at the lower application rate would have sequestered $3.13 \text{ t CO}_2 \text{ ha}^{-1}$. This amount is within the range of expected sequestration rates over the course of an entire year for a number of common land management practices aimed at carbon storage, including conservation tillage, changes in grazing management, reforestation, or the conversion of cropland to permanent grassland (Murray et al., 2005). The use of olivine or other easily weatherable silicate minerals instead of lime to increase the pH of agricultural soils would also prevent the emission of lime-derived CO₂ and may decrease the amount of soil respiration induced by the addition of soil amendments. By consuming anthropogenic CO₂ and reducing agricultural CO₂ emissions, enhanced mineral weathering has the potential to serve as an important part of the solution to the problem of elevated atmospheric CO₂ concentrations.

Declarations of interest

None.

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