

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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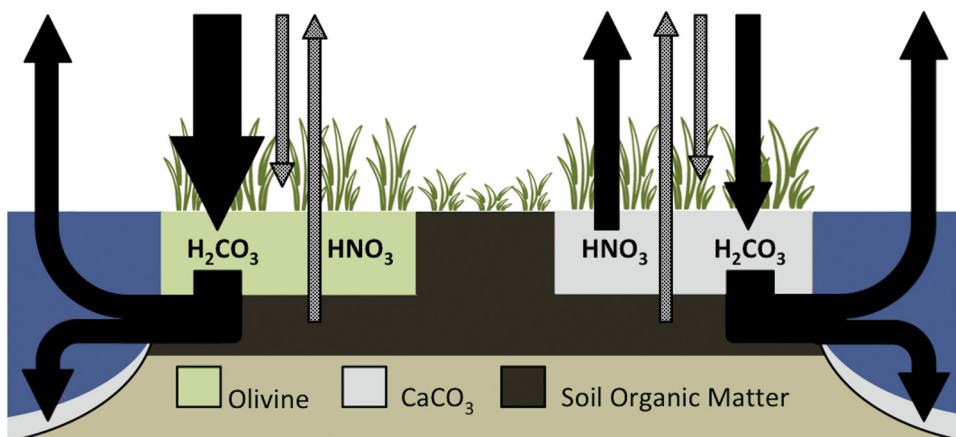
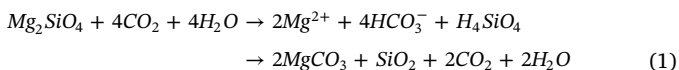


Fig. 1. Carbon fluxes induced by the application of olivine and agricultural lime (CaCO_3) to soils. Black arrows indicate known CO_2 fluxes and are scaled based on the magnitude of flux in moles of carbon per mole of applied soil amendment. Shaded arrows represent unknown CO_2 fluxes that would result 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)



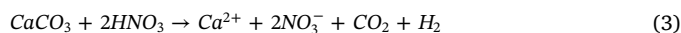
Eq. (1): Weathering reaction of Mg-olivine (forsterite), an abundant silicate mineral, and ensuing precipitation of magnesium carbonate. The weathering reaction results in 4 mol of CO_2 consumed per mole of olivine weathered. The precipitation of carbonate minerals results in more permanent sequestration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

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