A feasible way to increase carbon sequestration by adding dolomite and K-feldspar to soil

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Abstract: In recent years, many researchers have explored various possible ways to slow down the increase in atmospheric CO2 concentration as this process poses a serious threat to mankind’s survival. Mineral weathering is one possible way. Silicate weathering, for example, causes net carbon sequestration and carbonate weathering occurs relatively rapidly. In this study, dolomite and K-feldspar were added to soil to investigate if these minerals can increase carbon sequestration and also improve the available potassium content. The carbon content of amaranth, the organic and inorganic carbon content of the soil, two kinds of enzymes (polyphenol oxidase and urease), and the available potassium content were all tested. The experimental results show that the minerals accelerate the fixation of organic and inorganic carbon in the soil and also promote amaranth growth. Moreover, the available potassium content was increased when K-feldspar was added. Taken together, adding moderate amounts of carbonate and silicate minerals into the soil is found to be an attemptable way of accelerating CO2 fixation and improving the potassium content of soil.

Subjects: Agriculture & Environmental Sciences; Environmental Sciences; Soil Sciences

Keywords: soil; mixed mineral; mineral weathering; carbon fixation

1. Introduction

It is widely accepted that human activity has caused the atmospheric CO2 concentration to rise continually. The average global atmospheric CO2 concentration reached 395.31 ± 0.10 ppm in 2014 (Le Quéré et al., 2014). Methods of reducing the rate of atmospheric CO2 enrichment, and investigating what kind of practices are feasible and effective in blocking the trend of increasing atmosphere greenhouse gas (GHG) concentrations, have been attracting more and more attention (Liping &
Notwithstanding the temporary nature of the release of anthropogenic CO₂, it should be noted that large quantities of carbon are also exuded through the roots of plants in the form of organic matter which degrade to gaseous form and ultimately return to the atmosphere (Ryan, Delhaize, & Jones, 2001). This process acts as a dominant conveyor in the global carbon cycle, accounting for ~120 Gt Ca⁻¹ which clearly overshadows the 6 Gt Ca⁻¹ produced by anthropogenic activities (Renforth, Manning, & Lopez-Capel, 2009). Many GHGs are sequestered by the agricultural and terrestrial ecosystems, and plant biomass and soil, for example, are the major sinks of atmospheric CO₂ (Liping & Erda, 2001). Therefore, carbon sequestration in soil is something that we should value (Lal, 2004).

Schlesinger and Andrews (2000) showed that the CO₂ released from the soil should be considered to be one of the largest sources of flux in the global carbon cycle, and can thus have a large effect on the atmospheric CO₂ concentration. Plants and the soil are the principal parts of the agricultural and terrestrial ecosystems. Thus, it is highly appropriate to consider the importance of the coupled plant–soil system in carbon capture, and to develop ways to enhance these natural processes (Renforth et al., 2009). For example, adopting restorative land use and using recommended management practices with agricultural soils can mitigate the negative impact of elevated atmospheric CO₂ concentrations. In temperate climates, soil organic carbon (SOC) is always a major carbon sink in agricultural soil (Smith, 2004). By adopting reasonable practices, the global SOC sequestration potential is about 0.9 ± 0.3 Pg Ca⁻¹, which offsets one-fourth to one-third of the annual increase in atmospheric CO₂ (Lal, 2004). In arid climates, the role of soil as a carbon sink is often associated with the accumulation of soil inorganic carbon (SIC). The maximum capacity of SIC capture technology has been shown to be limited by the availability of Ca-rich minerals (Renforth et al., 2009). Seeking cost-effective means of increasing SOC and SIC is therefore very significant, although this is a long and arduous process.

It has been demonstrated that weathering occupies an important position in present and future carbon cycling, and involvement of microbes can accelerate the weathering process of carbonate (Burford, Fomina, & Gadd, 2003; Xiao et al., 2014) and silicate (Xiao, Lion, Dong, & Liu, 2016; Xiao, Lian, Hao, Liu, & Wang, 2015). However, many researchers ignore the accumulation of weathering products in the soil and thus underestimate their contribution to the global climate (Goudie & Viles, 2012). It is also worth considering methods that can reduce atmospheric CO₂ by reacting silicate minerals to form carbonate minerals (Lackner, 2003; Manning & Renforth, 2013; Seifritz, 1990). This typically involves dissolution of the silicate minerals and subsequent precipitation of stable carbonate minerals (Power, Harrison, Dipple, & Southam, 2013). Manning and Renforth (2013) showed that the pedogenic carbonate should now be considered as a consequence of reactions between plant root exudates and calcium liberated by the silicate dissolution. However, it should be noted that the rate of silicate weathering is limited due to the slow kinetics of the CO₂-silicate reaction process (Oelkers, Gislason, & Matter, 2008). In general, the process is over an order of magnitude slower than that of the carbonate process (Mortatti & Probst, 2003; Wu, Xu, Yang, & Yin, 2008). According to calculations, it will take more than one million years to stabilize the atmospheric CO₂ level through silicate weathering (Goudie & Viles, 2012). Adding 1-2 tons of crushed olivine (grain size < 300 micron) to one hectare of soil will last approximately 30 years in a temperate climate (Schuiling & Krijgsman, 2006). Therefore, it appears to be difficult to ease the increase in atmospheric CO₂ by only employing silicate weathering in the short term. The uptake of CO₂ by atmospheric or soil respiration by carbonate rock dissolution has an important effect on the global carbon cycle and serves as one of the most important sinks (Cao et al., 2012). Liu, Dreybrodt, and Wang (2010) showed that dissolved inorganic carbon is an important but previously underestimated sink for atmospheric CO₂. This contribution to carbon sequestration reaches up to 0.8242 Pg Ca⁻¹, which amounts to 10.4% of the total anthropogenic CO₂ emission (Liu et al., 2010). Dissolution of calcite and dolomite can transform soil-generated CO₂ into alkaline form ([HCO₃⁻, CO₃²⁻]) (Macpherson et al., 2008).
Agricultural operations affect the carbon cycle through uptake, fixation, emission, and transfer of carbon among different pools (Lal, 2004). It should be feasible to change the carbon content distribution among these pools by anthropogenic manipulation. Recently, several studies (Fan et al., 2014; Mahmoodabadi & Heydarpour, 2014) have shown that application of manure is generally considered to increase carbon sequestration, although Schlesinger (1999) pointed out that net carbon sequestration does occur. Schuiling and Krijgsman (2006) mentioned that spreading finely powdered olivine on farmland could be extensively used to fix CO₂. In this study, we use soil as a substrate to explore if carbonate and silicate minerals, when added to the soil, can cause an increase in net carbon sequestration.

2. Materials and methods

2.1. Minerals

Dolomite, CaMg(CO₃)₂, for the study was provided by the Institute of Geochemistry, Chinese Academy of Sciences (Guiyang, China). Analysis using X-ray diffraction (XRD) showed that the samples were doped with small amounts of calcite and sanidine. Analysis of the K-feldspar (KAlSi₃O₈) used using XRD showed that quartz, muscovite, and clinochlore were present as impurities. The two kinds of mineral were both crushed and specific-sized particles (100–200 mesh) used in the study.

2.2. Plants

The study is mainly aimed at investigating whether carbon sequestration can be increased by adding carbonate and silicate to the soil. At the same time, we also explored if using K-feldspar was able to provide potassium for plant growth. Therefore, amaranth is used in this study because it is a common vegetable that is planted widely in China and has a strong ability to become enriched in K ions.

2.3. Summary of the experimental method

The experimental pot employed is illustrated in Figure S1. XRD analysis of the soil showed that it is composed of quartz, muscovite, albite (Na(Si₃AlO₈)), orthoclase (KAlSi₃O₈), and kaolinite-1A (Al₂Si₂O₅(OH)₄). Each pot contains 1,500 g of soil and 500 g of mineral powder. Our primary purpose is to detect the effectiveness of mineral, so the addition was relatively large. The different amounts added to each pot are shown in Table 1. Every treatment has three replicates. To increase the permeability of the soil, 450 g glass beads were added to the pots. The amaranth was watered timely according to the soil moisture. Water (500 ml) was regularly added to the pot every six days, and the soil infiltration water (SIW) collected the next day. The experiment was continued for about two months.

2.3.1. Determination of the organic carbon content of amaranth

Whole amaranth plants were collected and dried overnight at 105°C. The dry weight was then measured. The percentage of carbon present was measured using an elemental analyzer (Elementar Vario MACRO, Germany).

2.3.2. Determination of soil parameters

At the end of the experiment, the soil moisture, pH, and activity of polyphenol oxidase and urease were determined. Portions of the naturally air-dried soil samples were used to determine the soil’s organic carbon and microbial biomass.

| Number | Composition | Abbreviation |
|--------|-------------|--------------|
|        | Soil (g)    | Dolomite (g) | K-feldspar (g) |
| 1      | 2,000       | -            | -              | all-s |
| 2      | 1,500       | 500          | -              | d-s  |
| 3      | 1,500       | 375          | 125            | d-p-s|
| 4      | 1,500       | 125          | 375            | p-d-s|
| 5      | 1,500       | -            | 500            | p-s  |
To measure the soil’s moisture content, an aliquot of moist soil (about 5 g) was dried at 105°C. After 5 h, the sample was placed in a desiccator to cool for 30 min. Samples were dried and weighed repeatedly until the weight no longer decreased. Soil pH was determined using CO₂-free deionized water and a 1:1 (w/v) soil-to-water ratio. Samples were shaken for 15 min, left to settle for 30 min, and then the pH measured as in previous reports (Fierer & Jackson, 2006).

Polyphenol oxidase is known to play an important role in carbon cycling in the soil (Sinsabaugh, 1994). The analysis mirrored the work of Carney, Hungate, Drake, and Megonigal (2007). Briefly, (1) litterbags were removed as possible and approximately 2 g of wet weight placed in a blender mini-jar; (2) acetate buffer (60 ml, 50 mM, pH 5) was added and the mixture blended on “whip” for 1 min; (3) homogenate (0.750 ml) was mixed with an equal volume of substrate in a 2-ml tube—tubes were placed in a shaker and incubated for 2 h; (4) the reaction mixture was centrifuged for 2 min at 10,000 g and the absorbance of the supernatant at 460 nm immediately measured on a microplate reader.

Previous work has shown that urease activity has a positive correlation with organic carbon and total nitrogen (Zantua, Dumenil, & Bremner, 1977). For this test, toluene (1 ml) was mixed with 5 g of natural air-dried soil sample for 15 min. Then, urea solution (10 ml, 5%) and citrate buffer (20 ml, 0.96 M, pH 6.7) were added. Meanwhile, as a control, a repeat experiment was performed using an equal volume of distilled water instead of the urea solution. After incubation for 24 h at 37°C, the solution was centrifuged (4,000 g, 10 min). Supernatant (1 ml) was mixed with sodium phenoxide solution (4 ml, 2.7 M) and sodium hypochlorite solution (3 ml, 0.9%) in a 50-ml volumetric flask. After 20 min, the reaction solution was diluted to 50 ml and the absorbance at 460 nm measured. Urease activity was expressed according to the number of milligrams of NH₃−N in 1 g of soil.

For SOC determination, dry soil (2 g) was added to HCl solution (40 ml, 5%), and the mixture blended about 10 min until gas is no longer generated. The tubes were then spun at 8,000 g for 5 min. After this, the samples were rinsed three times with ultrapure water. After drying at 105°C, the residual solids were weighed and the carbon content tested using the elemental analyzer (Elementar Vario MACRO, Germany).

The microbial biomass in the soil was measured using the method outlined by Vance, Brookes, and Jenkinson (1987) involving chloroform fumigation and extraction. Moist soil was fumigated in a sealed desiccator using ethanol-free chloroform for 24 h at 25°C. Water (20 ml) and the same amount of NaOH (1 M) were placed in the bottom to trap any evolved CO₂. Non-fumigated soil was used as a control. Fumigated and non-fumigated soils were subjected to extraction using 0.5 M K₂SO₄ solution for 30 min using an “end-over-end” shaker at 350 rpm and centrifuged. The supernatant was filtered through a 0.45-μm membrane. The filtrate was tested using a total organic carbon analyzer (Shimadzu TOC-VCSN, Japan). The soil microbial biomass carbon (B) was estimated using $B_{EC} = \frac{E_{ec}}{k_{EC}}$, where $E_{ec} = [\text{organic carbon extracted by } K_2SO_4 \text{ from fumigated soil} - \text{carbon extracted by } K_2SO_4 \text{ from non-fumigated soil}]$ (Wu, Joergensen, Pommerening, Chaussod, & Brookes, 1990). The value of the $k_{EC}$ parameter (the proportion of the extracted microbial biomass carbon evolved as organic carbon) was taken to be 0.45, following the work of Wu et al. (1990).

The available potassium content of the soil was measured. Briefly, a portion (0.5 g) of crushed dry sample (50–80 mesh) and ammonium acetate solution (50 ml, 1 M) were mixed in an extraction bottle (flask) following the method of Zhu, Lian, Yang, Liu, and Zhu (2013). Then, the bottle was stoppered and oscillated for 30 min. The mixture was filtered using a filter paper, and the filtrate collected for testing using a full-spectrum, direct-reading plasma emission spectrometer (Thermo Fisher Scientific, UK).

### 2.3.3. Determination of SIW parameters

The pH and temperature of the SIW measured using a pH meter (S20 SevenEasy, Mettler-Toledo). The concentrations of certain cations (K⁺, Na⁺, Ca²⁺, and Mg²⁺) were determined using an atomic
absorption analyzer (AA900F, PerkinElmer, US). Anion concentrations ($\text{Cl}^-$ and $\text{SO}_4^{2-}$) were determined using ion chromatography (DIONEX ICS-90, US). An acid–base titration method was used to measure the content of the bicarbonate in the aqueous solution according to the published literature with a little modification (Verma, 2004; Zangen, 1962). The SIW was filtered using a 0.45-μm microporous membrane and 20 ml titrated with a standardized HCl solution. The above parameters (pH, water temperature, ion concentrations) were imported into appropriate software (MINTEQ) to calculate the saturation index of the calcite, and the disordered-dolomite and ordered-dolomite.

2.4. Statistical analysis
StatSoft’s STATISTICA 6.0 software was used to analyze the data. The significance of the differences between the treatments was tested separately using one-way ANOVA tests followed by Fisher LSD tests for mean comparisons. All analyses were performed in triplicate. The data shown correspond to the means (along with the standard deviation) of at least three independent experiments.

3. Results

3.1. Amaranth carbon content
For the average carbon content, dolomite or/and K-feldspar is conducive to plant growth (see Figure 1). The minerals thus improve organic carbon fixation. From a statistical point of view, only adding dolomite (d-s) or increasing the K-feldspar content (p-d-s and p-s) were beneficial to the formation of organic carbon sinks. However, there was no difference between the all-s and d-p-s treatments.

3.2. Soil parameters
Soil moisture content was not significantly different (statistically) among the five different kinds of treatment (ranging from 11.6% to 16.8%). Dolomite significantly increased the soil pH, but K-feldspar did not (Figure 2(a)). The amount of dolomite added and the elevation of the soil pH were positively correlated. As can be seen from Figure 2(b) (compared with all-s treatment), the increase in SOC per gram of moist soil was significantly boosted after adding a large amount of dolomite (p-s and d-p-s). However, adding a large amount of K-feldspar did not have this effect (p-d-s and p-s). This may be due to the dissolution of the dolomite, which can consume gaseous or liquid CO2. In contrast, K-feldspar does not have this ability over a relatively short period of time.
Figure 2. The effect of adding mixed minerals (dolomite and feldspar) to the soil on several soil parameters: (a) soil pH, (b) increased SOC, (c) microbial biomass, (d) polyphenol oxidase, (e) urease, and (f) available potassium.
To our surprise, the mineral (dolomite or feldspar) caused the soil microbial biomass to decline a certain amount over the experimental period. The effect of the added minerals on microbial survival in the microenvironment may be key here. Compare Figure 2(b) and (c). The microbial biomass and increase in organic carbon show some negative correlation. However, different minerals had no significant effect on soil microbial biomass. From Figure 2(a)–(c), the increase in SOC may be mainly pH dependent and little to do with the total number of micro-organisms. The minerals made the activity of the polyphenol oxidase (except for all-p) and urease (except for p-d-s) increase (Figure 2(d) and (e)). There was no significant difference between dolomite and K-feldspar with respect to polyphenol oxidase activity. Nevertheless, dolomite alone induced an increase in soil urease activity. As was expected, the amount of available potassium rose to a certain extent after K-feldspar was added (see Figure 2(f)). However, excessive addition did not significantly increase the available potassium content.

3.3. SIW parameters

3.3.1. pH
Both dolomite and K-feldspar promoted an increase in the pH of the SIW (Figure 3(a)). The impact of these two minerals on the pH of the SIW was little different. In the initial stages of the trial, the average pH in the sample with dolomite added was the highest. As the experiment progressed, a mixture of the minerals was most propitious to enhancing the SIW’s pH.

3.3.2. The concentration of $\text{HCO}_3^-$
The addition of minerals had a great effect on $\text{HCO}_3^-$ concentration (Figure 3(b)). No matter which mineral (dolomite or feldspar) was added, there was a significant difference at each of the eight sampling times (compared with the all-s treatment). Overall, the change in $\text{HCO}_3^-$ concentration showed a sudden increase, followed by a rapid decrease, and then it gently changed. Dolomite alone was more influential with respect to $\text{HCO}_3^-$ concentration compared to K-feldspar. However, adding
more dolomite did not cause more $\text{HCO}_3^-$ to be produced. If standard deviation is ignored, then the $\text{HCO}_3^-$ produced was the most in the d-s treatment at the first two sampling times, yet mixed minerals are more conducive to generating $\text{HCO}_3^-$ in the long term.

3.3.3. $K^+$ concentration

Overall, the $K^+$ concentration gradually decreased (see Figure 3(c)). Only the average $K^+$ concentration in the p-s sample always exceeded that in all-s. Using mixed minerals or only K-feldspar did not significantly increase the concentration of $K^+$ in soil in the beginning of this experiment. The ability of K-feldspar to release $K^+$ can be seen in the latter part of the experiment (22 June and 28 June). Thus, it is feasible that, for long-term farming, fertilizers containing K-feldspar could be used to continue to provide $K^+$.

3.3.4. Saturation index calculation

MINTEQ software was used to calculate the saturation index of the carbonate using the following parameters: water temperature, pH, and the concentrations of $K^+$, $Na^+$, $Ca^{2+}$, $Mg^{2+}$, $Cl^-$, $\text{SO}_4^{2-}$, and $\text{HCO}_3^-$ ions. Three kinds of carbonate, calcite, ordered-dolomite, and disordered-dolomite, were selected. Interestingly, the saturation indices of these three carbonates were almost the same (Figures 3(d) and S2). In the case of the calcite saturation index (Figure 3 (d)), no matter which minerals were added, the calcite saturation index increased significantly. Comparing the effects of the different minerals on the saturation index, it seems mixed minerals (dolomite and K-feldspar) are more conducive to increasing the saturation index.

4. Discussion

The atmospheric $\text{CO}_2$ concentration continues to rise; thus researchers are constantly looking at a variety of solutions. Some of the research has shown that only a minor increase in the natural uptake is required to compensate for the extra anthropogenic $\text{CO}_2$ emission (Oelkers et al., 2008; Salek, Kleerebezem, Jonkers, Witkamp, & van Loosdrecht, 2013). Up until the last decade, mineral weathering has been a subject of some concern. However, this is the most important way in which nature keeps the $\text{CO}_2$ levels stable (Schuiling & Krijgsman, 2006). This oversight may be because people have not found a practical way to accelerate weathering (Schuiling & Krijgsman, 2006) or they have ignored the natural regulation effect. In this study, we have investigated the impact on $\text{CO}_2$ fixation of adding mixed dolomite–K-feldspar power to the soil. The results suggest that artificially adding suitable amount of dolomite and K-feldspar to the soil is likely to increase the amount of SOC. This is similar to the finding of previous reports investigating the potential of artificial soils (i.e. made by adding demolition waste or basic slag to soil). These were used to capture some of the transferred carbon as geologically stable CaCO$_3$ (Renforth et al., 2009). Artificial soils were also prepared by blending compost with dolerite and basalt quarry fines to be used for the purpose of $\text{CO}_2$ capture (Manning, Renforth, Lopez-Capel, Robertson, & Ghazireh, 2013).

The significantly increased pH value of the soil implies that adding dolomite to the soil is possible to mitigate the effects of acid rain (Allen & Brent, 2010; Teir, Eloneva, Fogelholm, & Zevenhoven, 2006) and/or alleviates soil acidification caused by agricultural fertilizer. Although the addition of the minerals made the total microbial biomass decrease, the increased polyphenol oxidase and urease activity suggests that adding minerals may promote the reproduction and activity of the microbes in the soil which are associated with the carbon and nitrogen cycles.

Manning and Renforth (2013) studied sequestration of atmospheric $\text{CO}_2$ through coupled plant–mineral reactions in urban soils. They showed that the rate-limiting factor seems to be the availability of Ca, not carbon (Manning & Renforth, 2013). Our results showed that the increase in SOC was doubled by dolomite addition, while K-feldspar addition did not do this. Both dolomite (d-s) and K-feldspar (p-f-s and p-s) improved the total carbon content of the amaranth. This means that the mineral may act as a regulatory component of the soil. After adding K-feldspar, the availability of K was significantly increased. Potassium is the most abundant cation in plants, comprising up to 10% of a plant’s dry weight (Leigh & Wynjones, 1984). In China, soluble potassium is a scarce
resource—there is only a 35% self-supply, so most is sourced via imports (Sun et al., 2013). Adding K-bearing minerals to soil is an attemptable way to improve the potassium content of the soil.

The following reaction occurs when dolomite is added to the soil:

$$\text{CaMg(CO}_3\text{)}_2 + 2\text{CO}_2 + 2\text{H}_2\text{O} \rightarrow \text{Ca}^{2+} + \text{Mg}^{2+} + 4\text{HCO}_3^-$$

The experimental results showed that the amount of $\text{HCO}_3^-$ generated was in some way related to the amount of dolomite added. In other words, the more dolomite added meant that more inorganic carbon produced (to some extent). However, the amount of $\text{Ca}^{2+}$ in the d-s sample was lower than in the d-p-s-treated sample in the latter part of the experiment. Precipitation of carbonate mineral is controlled by the saturation state of the soil solution which itself depends on the activities of the dissolved species (cation and bicarbonate). A higher saturation index is beneficial to the formation of carbonate precipitation. From the saturation index of calcite or dolomite (Figures 3(d) and S2), we see that the saturation index of the minerals added is always higher than the all-s sample. This indicated that the addition of minerals can accelerate the fixation of gaseous carbon into the form of relatively stable inorganic carbon, e.g. $\text{HCO}_3^-$ and $\text{CO}_3^{2-}$ as well. Moreover, others minerals, such as anorthite, can be tested as well. Further researches are still needed to explore if the long-term minerals addition can cause soil desertification. Anyway, these results show some prospects for mineral application in the farming.

5. Conclusions

The addition of minerals was likely to accelerate the fixation of organic and inorganic carbon in the soil. Moreover, the available K content in the soil was increased when K-feldspar was added. Taken together, adding moderate amounts of carbonate and silicate minerals into soil is an attemptable way to accelerate CO$_2$ fixation and improve the soil’s K content. However, further research is needed to improve the application of mixed minerals so as to increase soil quality and carbon sequestration.

Supplementary material
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