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Environmental feasibility of soil amendment with flue gas desulfurization gypsum (FGDG) for terrestrial carbon sequestration

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Abstract

Technologies for increasing carbon storage in soils are gathering attention as a means for mitigating atmospheric CO₂ emissions. Carbon sequestration can be achieved by controlling the organic carbon stock in soil and by accelerating mineral carbonation. In this study, carbon sequestration capacity was measured in soil columns treated with flue gas desulfurization gypsum (FGDG), a by-product of electric power generation. The feasibility of using FGDG as an environmentally benign alternative to gypsum or anhydrite was examined using a toxicity characteristic leaching procedure and Microtox bioassay. While no toxic leachate was generated from the FGDG treatment, some toxic elements in the soil were removed through absorption reactions. Test results for carbon sequestration based on unsaturated soil column experiments suggest that the application of FGDG for soil treatment holds promise of less microbial CO₂ emission from soil. The net benefits of carbon sequestration from the FGDG treatment were calculated as 87 and 621 g C/m²/m of infiltrated water, for the 1 % calcite-added column and 3 % calcite-added columns, respectively. The presented test results show that the FGDG treatment for soil carbon sequestration holds a promise when it is applied to slightly alkaline soils.

Keywords: Carbon sequestration, Mineral carbonation, Organic carbon storage, Soil amendment, Terrestrial soil carbon, Flue gas desulfurization gypsum (FGDG) recycling

Introduction

Carbon sequestration to mitigate rising atmospheric CO₂ levels has been recognized as an emerging focus area for technological development. In recent decades, numerous studies have investigated CO₂ capture at point sources (combustion gases of electric power generation plants that burn fossil fuels) and its geologic sequestration in deep reservoirs. Technologies that boost mineral carbonation and soil organic carbon (SOC) retention within the earth's surface have also been considered to be potentially important. According to the report by the Intergovernmental Panel on Climate Change (IPCC 2014), removing CO₂ from the atmosphere will be required to achieve the goal to keep temperature rise to a maximum 2 °C

above preindustrial times. The IPCC suggested that this may be partly possible by managing the CO₂ cycle of soils as well as planting trees. As the role of terrestrial soil in carbon sequestration has been receiving more attention, there has been an increase in studies examining the feasibility of cost-effective soil management using industrial by-products or waste materials (Bobicki et al. 2012; Cardenas-Escudero et al. 2011; Litynski et al. 2006; Palumbo et al. 2004). Mitigating atmospheric CO₂ by soil management using alkaline materials is distinguished from other studies aiming to reduce concentrated industrial CO₂ (Iizuka et al. 2004; Lee et al. 2012) by sequestration of nonpoint source CO₂ and not require any energy input.

Flue gas desulfurization gypsum (FGDG) is a by-product of the SO₂ scrubbing process used in coal-fired power plants. It is produced from wet scrubbers when SO₂ gas is sequentially reacted with calcite (CaCO₃) and wet limestone, yielding gypsum (CaSO₄·2H₂O). When produced in this way, FGDG is inexpensive relative to the price of commercial gypsum (DeSutter and Cihacek 2009) and is therefore recognized as a good alternative to the latter. Flue gas desulfurization gypsum has been primarily used as material for construction (ACAA 2008; Tzouvalas et al. 2004), but its use in agriculture is also being considered (DeSutter et al. 2011; Dick et al. 2000). Gypsum treatments in agriculture are beneficial as they are a source of sulfur (DeSutter et al. 2011) and serve as soil amendment agents to improve soil structure, drainage, and aeration (Chen and Dick 2011). More recently, the benefits of using FGDG (a calcium source) for carbon sequestration have been the subject of increasing attention (Lee et al. 2012; Noack et al. 2014).

The addition of Ca-rich compounds (such as FGDG) to soils can aid in CO₂ fixation through calcite precipitation, especially under alkaline conditions (Renforth et al. 2012; Washbourne et al. 2012). Soil inorganic carbon (SIC) accumulation in urban soils containing soluble Ca minerals has been estimated to be substantial, with a carbonation potential of 700–1200 million tons of CO₂ annually, or an enhanced weathering potential of 1190–2040 million tons of CO₂ (Manning and Renforth 2012). The carbon sequestration rate for soil in an urban center of Newcastle (UK) was estimated to be 12.5 kg CO₂/Mg soil/year (Washbourne et al. 2012). More recently, benefits in carbon (C) sequestration of CaSO₄ minerals (anhydrite and gypsum) treatment were investigated (Han and Tokunaga 2014). As a result, they reported net carbon benefit of the treatment as around 130–260 g C/m²/m infiltrated water. This benefit was acquired through SOC retention and suppressing microbial activity due to the increased ionic strength by mineral treatment (Han and Tokunaga 2014). Such accumulation reactions of carbonate minerals would also occur in natural alkaline soils with the addition of Ca-rich industrial by-product, such as FGDG.

The study presented here investigated the feasibility of adding FGDG as a Ca source, substituting commercial gypsum or anhydrite, to increase the amount of carbon storage in slightly alkaline soils. To achieve high enough

soil pH, calcite content of the tested soil was amended by adding chemical calcite as one of the experimental parameters in this study. It was well known that as small as 1 % calcite may control the overall soil pH through the buffering effect of calcite. The conceptual diagram of CO₂ sequestration mechanism expected from FGDG treatment is illustrated in SM Figure 1 in supplementary materials (SM). In this study, we investigated the environmental acceptability of FGDG as a soil amendment agent by conducting a chemical toxicity test in accordance with TCLP procedure and a bioassay using Microtox[®] system. Also, we quantified the changes in SOC and SIC in FGDG-treated soil with varying carbonate contents. The results of this study will contribute to determining the feasibility of recycling FGDG for soil carbon sequestration in the region with alkaline soils.

Materials and methods

Soils and FGDG

The soil used for the laboratory column experiments is an alkaline soil collected from the surface layer (0–40 cm) of San Joaquin Valley (Merced County, California, 37°6′21″N, 120°45′43″W), and the sampling area is mapped as the Alros soil series (Typic Epiaqualf) (Nazar 1990). FGDG used in this study, which comprised of a yellowish-white powder in the form of a wet cake (approximately 24.5 % moisture content), was obtained from South Carolina Electric & Gas Co. (Cayce, SC). The details of physicochemical properties of soil and FGDG are presented in supplementary material (SM).

TCLP test

The environmental characteristics of FGDG were determined by the toxicity characteristic leaching procedure (TCLP) (US EPA 1992). For the leaching test, pH 2.88 acetic acid solution (5.7 mL of glacial acetic acid/L) was added to a mixture of 2 g FGDG and/or column soil in 1:20 solid-to-solution ratio in each 6 replicated batch. The prepared soil/solution mixture was mixed for 18 h using an end-over-end rotator, and the supernatant was filtered using a 0.1- μ m nylon filter and acidified with HNO₃ by diluting the sample using 1 % HNO₃ solution and stored in a refrigerator until an inductively coupled plasma mass spectrometer (ICP-MS, Perkin-Elmer) measurement. From the leaching solution, seven heavy metal (As, Cd, Cr, Hg, Ba, Pb, and Se) concentrations were measured using ICP-MS.

Microtox[®] bioassay

The Microtox[®] bioluminescent assay of FGDG was carried out in accordance with solid-phase test (SPT). The test sample was prepared with 7 g of FGDG and 35 mL of 2 % sterilized NaCl in a 50-mL plastic tube and was mixed for 1 h using an end-over-end rotator.

In SPT, solid suspension directly contacts the microbes for 20 min, and right before the light reading, solid particles are filtered using the SPT filter (Evergreen Scientific, USA) to separate coarse particles from the solid

suspension mixed with bacteria. The toxicity of sample was calculated as a parameter of EC_{50} using the readings of light reduction 15 min after the sample's contact with *A. fischeri* reagent. The calculated EC_{50} refers to the sample concentration when 50 % of light emission is diminished. The detail of SPT can be found in the Microtox user manual (Azur Environmental 1998).

Column design and experimental conditions

An 85-cm-long acrylic tube with a 7.5 cm internal diameter (1.27 cm wall thickness) was used for the column tests. The columns were designed specifically to collect gaseous, aqueous, and solid phases of carbon in the headspace at the top, the effluent collecting port, and the sidewall solid sampling port, respectively. A schematic figure of column apparatus is provided in Fig. 1.

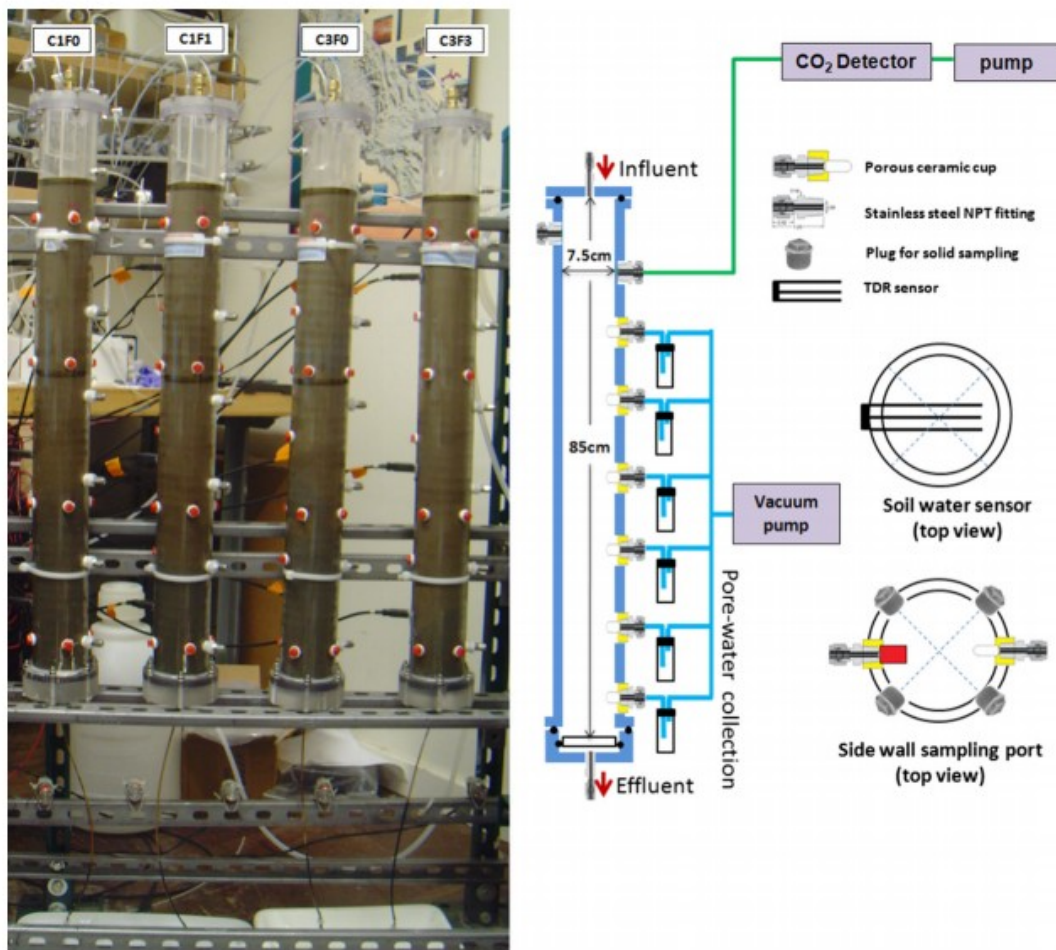


Fig. 1 Photograph and schematic diagram of soil columns

Four columns were prepared and packed with different soil mixtures using calcite and FGDG. Each column was assigned a reference code based on the mass percentage of calcite (C) and FGDG (F) addition. The first control and test columns containing 0 and 1 % FGDG within soil mixed with 1 % calcite

are referred to as C1F0 and C1F1, respectively. Two other columns prepared with 3 % calcite addition are called C3F0 (no initial FGDG addition) and C3F3 (3 % FGDG addition), respectively. C1F0 and C3F0 are control columns with no FGDG additions. SM Table 1 provides details of physicochemical and experimental column running conditions.

Sampling of liquid, gaseous and solid column samples

Column effluents at the bottom of each column were collected in a gastight Tedlar bag. In samples, pH and electrical conductivity (EC) were measured immediately after collection, with remaining samples stored in septum-sealed glass vials until measurement of other chemical properties. The collected aqueous samples were used for measurement of pH, EC, cation concentrations for Ca, Mg, Na, and K, anion concentrations for SO_4^{2-} , total dissolved inorganic carbon (DIC), and total dissolved organic carbon (DOC). Additionally, heavy metal concentrations (As, Cd, Cr, Hg, Ba, Pb, and Se) were periodically measured until 54 weeks (the whole column operation period was 80 weeks) after the start of effluent collection.

Headspace gas was retrieved with a peristaltic pump. The collected gas was continuously passed through an air tube connected to a CO_2 gas analyzer (Li-840A, Licor, NE, USA), and the concentration of CO_2 was measured using an infrared gas analyzer (IRGA). The detail of headspace gas collecting method and the calculation method of measured CO_2 can be found in Han and Tokunaga (2014). Note that the each cumulative CO_2 emission point was calculated with continuously monitored values by integrating the measured CO_2 levels for certain period of time; thus, each calculated point displays the averaged values of many measured points (each point represents the averaged values of every 3 min readings for several days) in spite of no statistical remark. Solid samples were collected periodically from the sidewall sampling ports placed at four vertical locations (22-cm intervals) along the length of each column and prepared for total carbon (TC), total organic carbon (TOC), and X-ray diffraction (XRD) measurement. The detail of solid-phase sample measurement can be found in SI.

Results and discussion

TCLP test result

Results indicate that heavy metal leaching was not negatively affected by the FGDG treatment. The leaching concentration results for As, Cd, Cr, Hg, Ba, Pb, and Se (Table 1) indicate a small concern with respect to Se concentrations, which are close to the regulatory level for hazardous waste. Otherwise, results for the soil/FGDG mixture indicate that no detrimental effect would likely occur from the addition of FGDG. These observations are similar to those obtained in other toxicity investigations of the flue gas desulfurization (FGD) by-product (Desutter et al. 2011; Dick et al. 2000). Relatively high Se concentrations in FGDG by-products have also been reported in these studies. The high Se concentration is likely to be

attributable to the volatility of Se causing accumulation during the coal processing.

Table 1 Heavy metal concentrations leached during TCLP test in mg heavy metal/L tested solution (error: \pm SD, $n = 6$)

Unit	Regulatory level for hazardous waste ^a µg/L	FGDG µg/L	Soil µg/L	Soil + 1 % FGDG µg/L	Soil + 3 % FGDG µg/L
As	5000	2.03 (\pm 0.05)	10.92 (\pm 0.10)	14.22 (\pm 0.16)	11.71 (\pm 0.74)
Cd	1000	0.08 (\pm 0.03)	1.71 (\pm 0.09)	1.84 (\pm 0.05)	1.73 (\pm 0.09)
Cr	5000	25.47 (\pm 0.59)	20.33 (\pm 0.25)	21.02 (\pm 0.21)	21.93 (\pm 0.08)
Hg	200	1.79 (\pm 0.13)	0.99 (\pm 0.43)	2.94 (\pm 2.45)	0.56 (\pm 0.14)
Ba	100,000	17.04 (\pm 0.38)	396.09 (\pm 5.12)	185.27 (\pm 4.60)	177.35 (\pm 7.20)
Pb	5000	20.04 (\pm 0.38)	0.37 (\pm 0.11)	0.34 (\pm 0.04)	0.21 (\pm 0.01)
Se	1000	193.48 (\pm 14.12)	1.92 (\pm 0.77)	2.05 (\pm 0.54)	3.61 (\pm 0.70)

^a 40CFR §261.24, Table 1, EPA (<https://www.gpo.gov/fdsys/pkg/CFR-2012-title40-vol27/xml/CFR-2012-title40-vol27-sec261-24.xml>)

Heavy metal concentrations were also measured in the effluent collected from columns. The concentrations of heavy metal leaching measured from the column effluent can be found in SM Table 2. The column effluents were alkaline (around pH 8) contrasting to the acidic TCLP test reagent, and the applied solid/solution ratio was higher in the measured soil column, as porous systems mostly show a solid/solution ratio of 3:1–5:1. The test results of As, Hg, Ba, and Se concentration exceeded the maximum contaminant levels (MCLs) set by the Environmental Protection Agency (EPA) for primary drinking water and by the Resource Conservation and Recovery Act (RCRA) for groundwater. Although these heavy metal concentrations exceeded the MCL, the measured high concentrations are attributable to the chemical properties of the selected soil and not to the FGDG treatment. Based on the results of the heavy metal leaching column test, and as reported by other studies (Sun et al. 2012), some beneficial effects can be noted in FGDG-treated soil columns. The Hg and Se concentrations were lower in the column effluent of FGDG-treated columns than in the effluent of the untreated C1F0 column; this remediation effect was more noticeable with higher FGDG content. Because Hg and Se concentrations decreased with the eluted amount of pore water, it can be inferred that soil concentrations of these two elements were somehow originally elevated. Conversely, the continuous leaching of As and Ba implies that the soil may contain some As and/or Ba minerals.

Microtox bioassay results

The Microtox bioassay test result of FGDG is presented in Fig. 2. As it displayed in the graph, the series of diluted FGDG suspension did not present any significant reduction in bacteria's luminescence and all light intensities were higher than 50 % of the initial intensity. The EC₅₀ value could not be calculated due to low toxicity of FGDG. Based on this result, it could be concluded that the eco-toxicity effect of FGDG may be negligible.

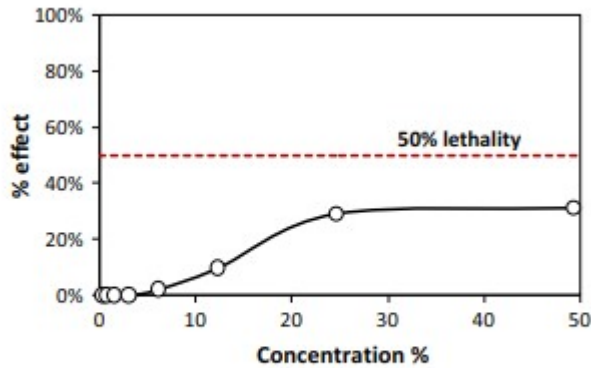


Fig. 2 Microtox toxicity test of FGDC showing no toxicity

Results of liquid-, gas-, and solid-phase sample measurement of column samples

The amount of carbon contained and discharged from the column soil was measured in all of three phases: effluent samples as liquid, emitted CO₂ gas from the column headspace, and soil itself as solid samples.

Results of liquid-phase measurement

The results of measured effluent chemistry are presented in Fig. 3. The pH values and DOC/DIC concentrations were for the most part similar in all columns, regardless of the FGDC treatment and applied soil type. This is because the pore water pH of columns is mostly controlled by calcite worked as a pH buffer in soils. Generally, when carbonate minerals are present in silicate or aluminosilicate rocks or soils at concentrations of 1 % or more, the minerals tend to dominate the chemistry of the soil or groundwater (Langmuir 1997). The DOC concentrations measured for four columns are also presented similar results regardless of the treatment or soil composition primarily due to the similar pH conditions in all columns (Fig. 3b). However, the amount of DOC leached during the test period was less than 1 % of initial SOC, implying that the controlling factor of SOC loss is not chemical dissolution. The DIC concentration measured in effluent was well above the injected 120 ppm C (as NaHCO₃), demonstrating that active microbial respiration was occurring in all columns (Fig. 3c). In other words, this elevated DIC was caused by the dissolution of gaseous CO₂ generated by respiration of microbes in soil column. The source of carbon turned over to CO₂ by microbial activity is considered to be SOC existing in initial soils.

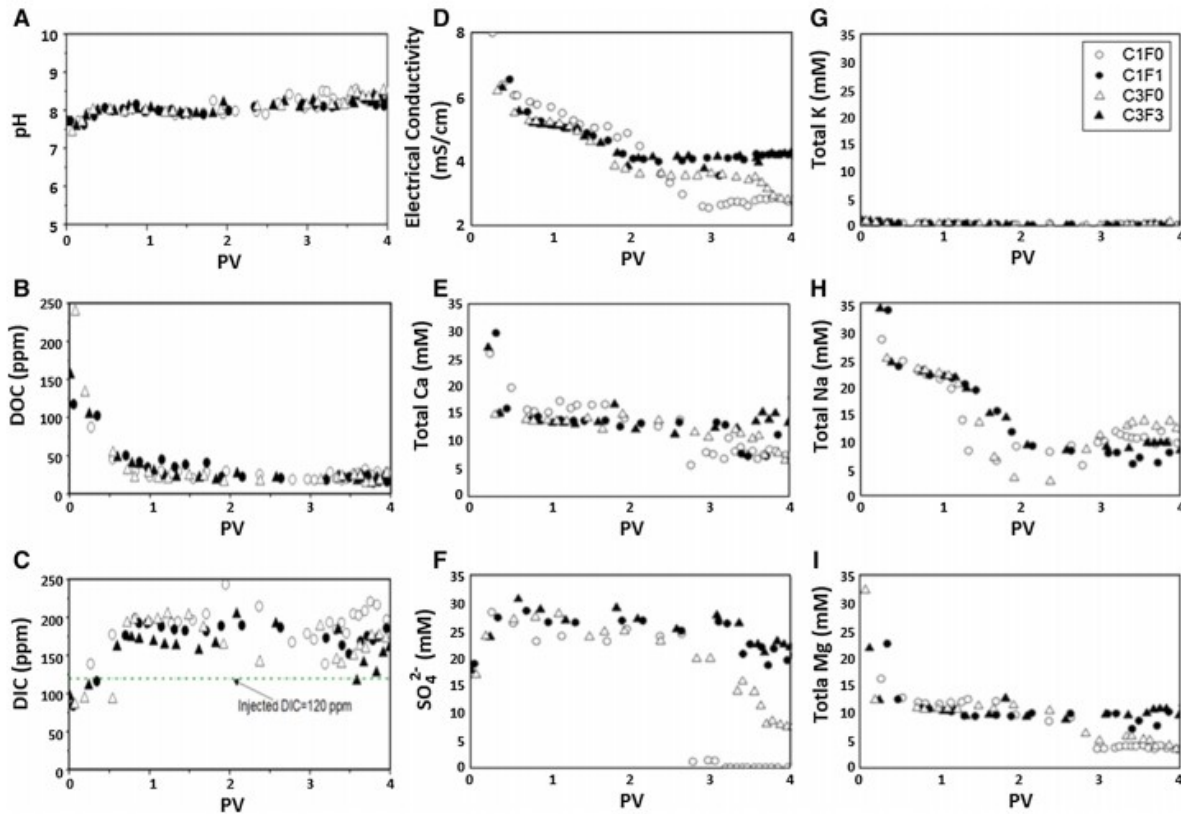


Fig. 3 The chemistry of column effluent. **a** pH, **b** total organic carbon (TOC), **c** total inorganic carbon, **d** electrical conductivity, **e** total Ca, **f** SO_4^{2-} , **g** total K, **h** total Na, and **i** total Mg concentration

Figure 3 also shows the cation and anion concentrations of column effluent. The dissolution of FGDG directly affected temporal trends with respect to EC, total Ca, and SO_4^{2-} concentrations in the column drainage waters. The EC trends reflect the influence of displacement of original soluble salts, different soil treatments, and different influent solutions. The continuous release of Na and Mg (Fig. 3h, i) resulted from cation exchange from the soil and/or added NaHCO_3 with the retention of the injected 2.5 mM of K by cation exchange (Fig. 3g). The overall chemistry of anion and cation concentrations in effluent was, however, generally similar in all columns regardless of the calcite contents in soil until the gypsum or added FGDG was exhausted. This is because the calcite was a dominant solid species under the tested pH and partial CO_2 concentration and gypsum was a dissolving species, as it was simulated using a thermodynamic chemical model under similar chemical conditions in Han and Tokunaga (2014). The continuous release of SO_4^{2-} is an evidence of gypsum dissolution along with XRD result.

Results of gas-phase measurement

The main desired effect of applying FGDG treatment is the reduction in CO_2 emissions from the soil surface. Lower microbial respiration can be explained by suppressed microbial activity due to the higher osmotic potential resulting from the addition of FGDG. Figure 4 shows the cumulative

amounts of CO₂ emissions in each column over 5 months, indicating the effect of the FGDG treatment on different soil types. However, the resultant microbial respiration was almost identical in treated and untreated columns of C1F1 and C1F0. This may be because the measured values of EC of treated and untreated 1 % calcite-containing columns (C1F0 and C1F1) were very similar in the initial phase, as the soil used in the test naturally contained a fair amount of gypsum. The beneficial effect of carbon sequestration by the added FGDG is expected after exhausting this naturally contained gypsum.

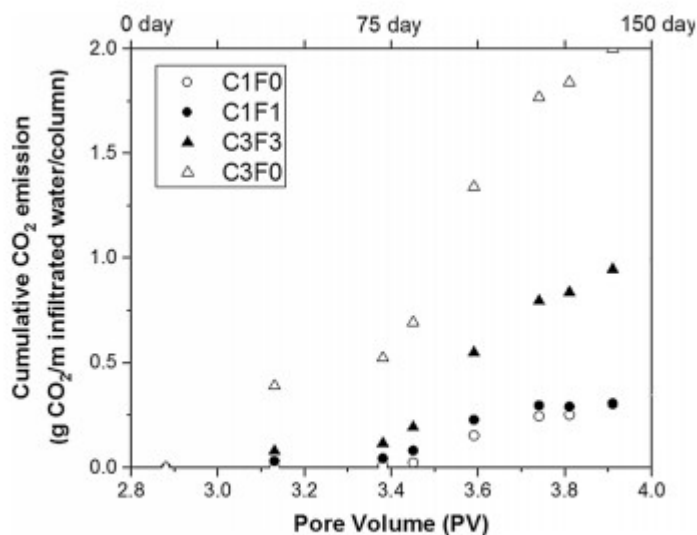


Fig. 4 Cumulative CO₂ gas emissions from soil columns

When compared with the generally low and similar microbial respiration values obtained for C1F0 and C1F1, the 3 % calcite-containing C3F0 and C3F3 columns displayed higher and significantly different CO₂ emission values. In soil with 3 % added calcite, the amount of CO₂ gas released from the FGDG-treated C3F3 column was less than half of the CO₂ emitted from the non-treated C3F0. Higher CO₂ emissions may result from higher calcite content which does not affect the dissolution of gypsum but stimulates the microbial activity. Higher microbial respiration with a high calcite ratio has also been reported in other studies (Bolan et al. 2011; Ingvar Nilsson et al. 2001). Lundström et al. (2003) reported that soil respiration increased from 10 to 36 % after the liming process, demonstrating the influence of higher calcite content in promoting microbial activity. In the present study, the 3 % calcite-containing soil produced higher CO₂ emissions in both FGDG-treated and untreated columns, compared to the 1 % calcite-containing soil columns. This result implies that FGDG treatment is distinctly suppressing the microbial activity within the soil column.

Results of solid-phase measurement

According to XRD analysis, the spectrum of the samples included the peaks of an initial soil sample comprised mainly of quartz, plagioclase, K-feldspar, and amphibole with traces of chlorite and mica, as well as the spiked FG DG (detected as gypsum) and calcite (Fig. 5). The XRD spectrum of gypsum has the characteristic peaks, not overlapped by major peaks of other minerals, at around $2\theta = 11.6^\circ$ and 29.1° which are assigned to crystalline structure at the (020) and (041) planes, respectively. For the gypsum after the reaction, the diffraction peaks which are typical fingerprints of gypsum were disappeared for the soil collected from C1F1 and steeply reduced for the soil collected from C3F3, indicating the active dissolution reaction of FG DG. The characteristic peak of calcite is indexed as (104) plane at around $2\theta = 29.5^\circ$, which showed the difference in the peak intensity with different initial amounts of the spiked calcite, but no prominent evidence of secondary precipitation of calcite was observed from the XRD pattern. The analytical result for measurement of TIC may become more direct evidence of secondary precipitation of calcite. The amount of solid-phase TIC change is given in section “Discussion about carbon sequestration efficiency of FG DG treatment” with mass balance of other two phases of C.

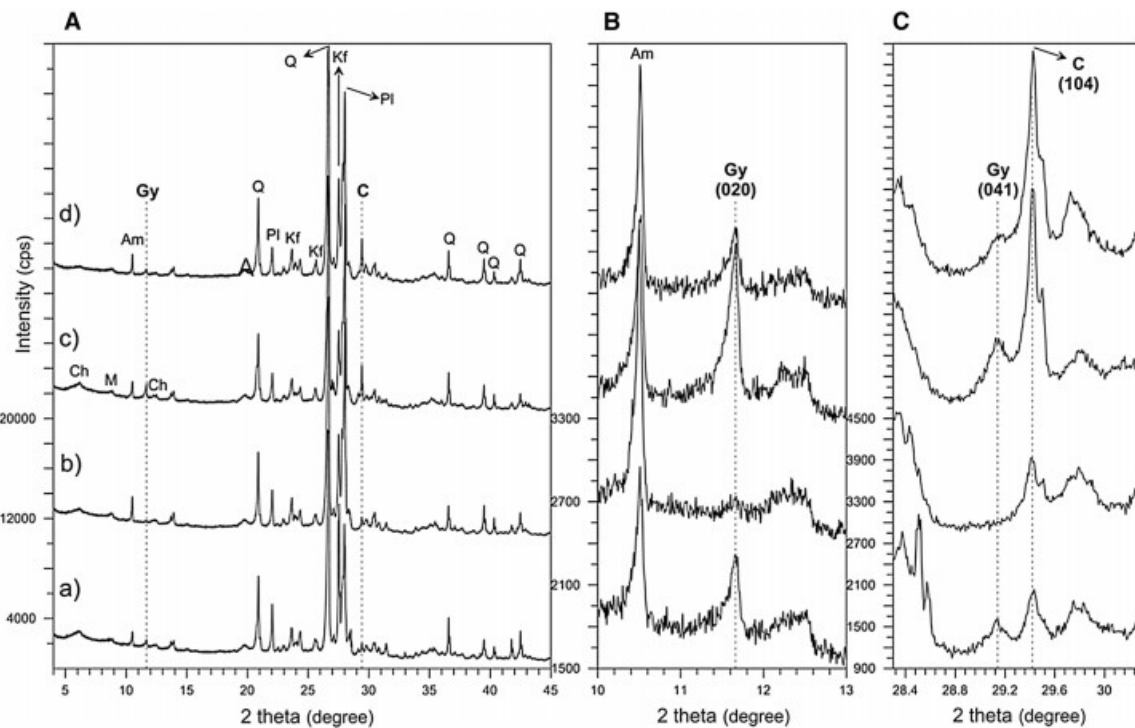


Fig. 5 X-ray diffraction patterns for a) initial soil of C1F1, b) 12-month tested soil of C1F1, c) initial soil of C3F3, and d) 12-month tested soil of C3F3. Graph a presents the overall XRD spectrum and graph b, c are enlarged view of the diffractions peaks of the 2θ ranges

of 10° – 13° (for gypsum) and 28.4° – 30.2° (for calcite). *Q* quartz, *Pl* plagioclase, *Kf* K-feldspar, *Gy* gypsum, *C* calcite, *Am* amphibole, *Ch* chlorite, *M* mica

Discussion about carbon sequestration efficiency of FG DG treatment

The carbon mass balance was calculated using carbon measurement of gaseous- and aqueous- phase samples using the method provided in Han

and Tokunaga (2014). Figure 6 shows the amount of SOC loss and precipitated SIC, along with the overall change in the amount of carbon. The loss in SOC is higher in 3 % calcite-containing columns (C3F0 and C3F3), mostly due to high microbial respiration. The role of calcite in increasing soil respiration has also been observed in other studies (Shukla et al. 2005). The SOC changes in 1 % calcite-containing columns were similar, regardless of FGDG treatment, based on DIC and DOC in effluents and CO₂ respiration rate. However, SOC loss was actually slightly higher in the treated column (C1F1). Nevertheless, this result does not mean that FGDG treatment had no carbon sequestration benefit for this soil condition. As shown in SM Table 3, the net carbon loss in C1F0 is still higher compared with that in C1F1, considering the portion of carbon precipitated to form calcite.

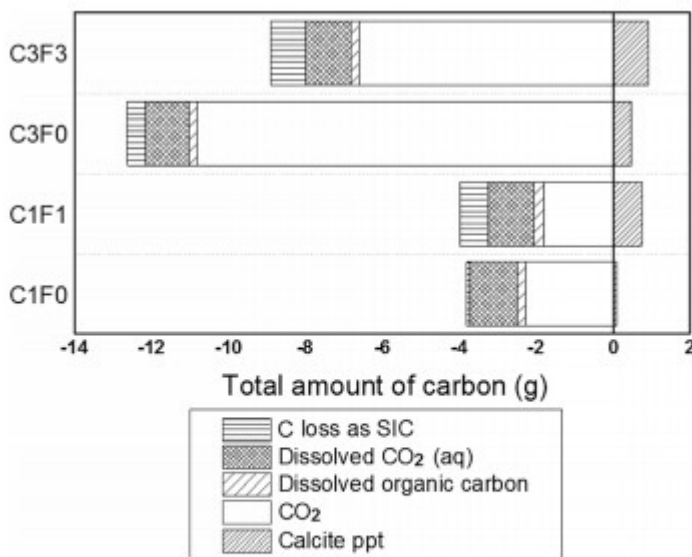
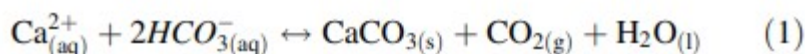


Fig. 6 Amount of carbon change and distribution of carbon in columns. The left side of the plot (negative number) indicates carbon loss, and the right side of the plot indicates carbon gain as calcite precipitation

With respect to SIC, results indicated that the amount of calcite precipitated was higher in FGDG-treated columns compared to their controls. Under pH around 8 or higher, the solubility of calcite is low, but that of gypsum is relatively high; therefore, the calcite precipitation reaction is favorable over the gypsum dissolution. The calcite precipitation reaction occurring in the column can be expressed as follows:



As the pH conditions of the columns were slightly alkaline (close to pH 8), bicarbonate was the controlling species among carbonate species. Under the soil column conditions, the limiting reactant was Ca²⁺ because the bicarbonate was injected into the influent, and more bicarbonate was

partitioned into the aqueous phase by microbial respiration, as shown in Fig. 5. The concentration of bicarbonate in column effluent was always 120–150 mM after some of the bicarbonate was utilized in the calcite precipitation reaction. The FGDG treatment provided calcium ions, and the calcite precipitation reaction was thus stimulated.

In this study, two aspects of the carbon sequestration benefit of FGDG treatment were demonstrated. The first aspect is the reduced SOC loss by suppressed microbial activity, while the second aspect is increased calcite precipitation resulting from added calcium ions. Overall, during the column test period, the SOC loss was much higher than SIC gain. However, the FGDG treatment helped to reduce the SOC loss. The net benefits of carbon sequestration from the FGDG treatment were 87 and 621 g C/m²/m of infiltrated water, for the C1F1 and C3F3 columns, respectively. Contrast to other previous studies focused more on the CO₂ sequestration effect by entering bicarbonate downward to the groundwater (Ma et al. 2014), this study emphasizes more on the CO₂ sequestration effect from the reduction in SOC loss.

In a review article, Sanderman (2012) noted that depending on the source of Ca and HCO₃, the calcite precipitation reaction represents a net carbon source, is carbon neutral, or is a carbon sink. From Sanderman's analysis, the source of HCO₃ is the critical factor to determine whether the calcite precipitation induced by Ca supply from FGDG treatment is a carbon sink or carbon source of atmospheric CO₂. The source of HCO₃ driving precipitation in these soils is from microbial respiration rather than the influent water, as evidenced by the excess concentration of DIC in the effluent relative to the injected HCO₃. Thus, the carbonate precipitation measured in our experiment may constitute a net sink of atmospheric CO₂ (Schlesinger 1982). Monger et al. (2015) also pointed out that the secondary precipitation of pedogenic CO₂ without extra addition of calcium does not sequester atmospheric carbon. This reaction of carbon sequestration in SIC change is slow comparing to the faster SOC change, but since this soil amendment intends to the natural and slow reaction of soil without any intensive input of energy or materials, the SIC accumulation by FGDG treatment may help the sequestration of atmospheric CO₂ in the long term.

Conclusions

The results of this study demonstrate that utilization of FGDG as a soil amendment agent in alkaline soils can help reduce atmospheric CO₂ emissions by limiting SOC losses and promoting calcite precipitation. Based on the toxicity tests, it can be concluded that the use of FGDG may not deteriorate the soil leaching solution or nearby ecological environments. The CO₂ sequestration benefit was confirmed by the tested results from unsaturated soil columns treated with FGDG. The obtained results also demonstrated that the FGDG treatment in high calcite-containing alkaline soil would create a good synergy effect of CO₂ sequestration by suppressing

microbial activity. This study was conducted with a limited number of soil columns, and therefore, further experiments with extended replications and a wider variety of soil conditions are needed. Nevertheless, the relatively large size of the soil columns and relatively long experiment duration indicate that proposed reactions resulted in quantifiable moderation of soil CO₂ emissions upon FGDG treatment in natural alkaline soils.

Acknowledgments

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