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Wollastonite addition stimulates soil organic carbon mineralization: Evidences from 12 land-use types in subtropical China

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19 **Abstract**

20 Enhanced rock weathering through adding silicate rock powder to soil has been increasingly
21 considered as an effective means of removing CO₂ from the atmosphere. However, the potential
22 impact of silicate rock powder addition on the stability of soil organic carbon remains largely
23 unknown, which adds an uncertainty to its effectiveness in mitigating atmospheric CO₂. In this study,
24 the response of soil CO₂ efflux to wollastonite addition was evaluated for 12 land-use types in a
25 subtropical region of China through an incubation experiment. Results showed that wollastonite
26 addition significantly ($P < 0.05$) increased soil CO₂ efflux over the control by an average of $330.97 \pm$
27 29.25% during the incubation period. Approximately 90% of the variance in soil CO₂ efflux was
28 explained by soil properties in this study. Increased soil pH, dissolved organic carbon and available
29 silicon induced by wollastonite addition were the main reasons responsible for the increase in soil
30 CO₂ emission. Soils having lower pH values responded to wollastonite addition with a higher increase
31 in CO₂ emissions. Based on numerous land-use types, our study showed that wollastonite application
32 will cause substantial increase in the mineralization of soil organic carbon in acidic soil, which may
33 weaken the effectiveness of enhanced rock weathering strategies as a tool for CO₂ sequestration.

34

35 **Key words:** Soil organic carbon, Soil CO₂ emissions, Soil properties, Enhanced rock weathering,
36 Silicate rock powder

37 1 Introduction

38 Climate change is one of the most serious environmental and societal issues currently faced by
39 humanity (IPCC, 2018). The 2015 climate summit in Paris has agreed to take steps towards limiting
40 the global mean annual surface air temperature increase to well below 2 °C above pre-industrial levels,
41 and to pursue efforts towards a target of 1.5 °C (UNFCCC, 2015). Researchers generally believe that
42 reaching this target requires a combination of decreasing CO₂ emission and increasing CO₂ removal
43 from the atmosphere (IPCC, 2018; Strefler et al., 2018; Bellamy et al., 2019). Recently, enhanced
44 rock weathering (ERW) by adding crushed, fast-reacting silicate rocks (e.g., wollastonite, basalt and
45 olivine) to arable or forest soils has been demonstrated to be an effective practice to remove
46 atmospheric CO₂ through immobilizing it to soil inorganic carbon (SIC) pool (Andrews and Taylor;
47 2019; Beerling et al., 2020; Goll et al., 2021).

48 Whereas before ERW can be recognized as an effective means of removing atmospheric CO₂,
49 evaluations must be carried out on the potential influence of ERW on soil organic matter (SOC)
50 mineralization, since a small change of which has substantial effect on atmospheric CO₂ concentration
51 (Kirschbaum, 2004; Qiao et al., 2014). ERW likely contributes to SOC accumulation as it may
52 increase the cationic species (e.g., Ca²⁺ ions) and silicates in soil, which can reduce the decomposition
53 of organic matter by facilitating its adsorption to clay or aggregation (Song et al., 2014; Rowley et
54 al., 2018). There are, however, also evidences that soils respond to silicate addition with an increase
55 in CO₂ emission (Groffman et al., 2006; Borges et al., 2019; Taylor et al., 2021). In a potting
56 experiment with leguminous beans (*Phaseolus vulgaris* L.) and nonleguminous corn (*Zea mays* L.),
57 Haque et al. (2019) even found a decrease in SOC concentration following silicate addition. These
58 unexpected results are mostly attributed to the silicate-induced increase in soil pH values, whereby
59 the microbial decomposition raised. Therefore, the direction and magnitude to which SOC
60 mineralization responds to silicate addition depends on the balance among its effects on diverse
61 physicochemical and biological processes in soil. To date, information is still lacking with regard to
62 the relative importance of these processes in determining the effect of ERW on SOC mineralization,

which will limit the use of ERW as a valid practice to mitigate atmospheric CO₂.

The differences in soil traits might also be important factors affecting the response of SOC mineralization to ERW (Hartmann et al., 2014; Bothe, 2015; Vicca et al., 2022). For instance, compared to higher pH soils, the addition of silicate rock powder in lower pH soils has more significant effects on soil chemistry or biology (Ramezani et al., 2013; Ramezani et al., 2015), which is considered an important factor affecting SOC mineralization (Stockmann et al., 2013). Moreover, most previous site studies on this topic regarded only a few soil types (Dietzen et al., 2018; Borges et al., 2019; Haque et al., 2020a), which inevitably would increase the uncertainty of the conclusion. Theoretically, this limitation can be in part overcome by meta-analysis study that involves a large number of soil types. While there is not enough data to conduct a meta-analysis on this topic at present, and not all of the properties of soil were provided in the original studies. Therefore, site studies that include various soil types with contrasting traits are highly urgent, since such studies have rarely been performed.

To fill this knowledge gap, we explored the effects of silicate rock (in the form of wollastonite) addition on the mineralization of SOC for 12 different land use types in a subtropical region of China using a laboratory incubation. Meanwhile, the effects of wollastonite addition on soil physicochemical properties mainly including soil pH and nutrients (available silicon, available nitrogen and available phosphorus, etc.), were also evaluated. This study aimed to clarify how SOC mineralization responds to wollastonite addition and the underlying mechanisms. We hypothesized that wollastonite addition will slow SOC mineralization, considering that the input of cationic species and silicates mediated by wollastonite may improve the SOC stability (Song et al., 2014; Rowley et al., 2018). Furthermore, we hypothesized that the wollastonite effect on soil organic carbon mineralization would depend on the soil properties change caused by wollastonite.

2 Materials and methods

2.1 Location

89 This study was conducted in the Huitong Experimental Station of Forest Ecology, Chinese
90 Academy of Sciences (26°50' N, 109°36' E). This region, which belongs to sub-tropical monsoon
91 humid climate, has a mean annual temperature of 16.5 °C, and a mean annual precipitation of
92 approximately 1,200 mm. The soil developed from shale is classified as Plinthudults according to the
93 U.S. Soil Taxonomy system (Wang et al., 2013). Paddy fields and plantations are the primary land
94 uses in the study area.

95

96 **2.2 Soil sampling**

97 We found 12 co-existing cover all land-use types at our study sites, namely *Oryza sativa* L., *Glycine*
98 *max* (Linn.) Merr. and *Anorphophallus konjac* K. Koch fields; *Camellia sinensis* (L.) O. Ktze.,
99 *Camellia oleifera* Abel., *Citrus reticulata* Blanco, *Myrica rubra* (Lour.) S. et Zucc., *Cunninghamia*
100 *lanceolata* (Lamb.) Hook., *Phyllostachys edulis* (Carriere) J. Houzeau, *Michelia macclurei* Dandy,
101 *Schima superba* Gardn. et Champ. and *Pinus massoniana* Lamb. plantations. At each site, a
102 rectangular area of approximately 20 m × 20 m was demarcated and 10-12 soil samples were randomly
103 collected along an S-shaped route. The samples were mixed, and pooled to a composite sample. These
104 soil samples were thoroughly homogenized and passed through a 2-mm-mesh sieve. The visible plant
105 debris and stones in the soils were carefully removed by hand. Subsequently, these soil samples were
106 divided into two parts. One of the two parts was stored at 4 °C was used for incubation and microbial
107 measurement and the other part was air-dried for soil physical and chemical proprieties determination.
108 Relevant information of soil physical-chemical proprieties for all sites is listed in Table1.

109

110 **2.3 Incubation study**

111 The moist-weight equivalent of 30 g of dry soil was placed into each 120 ml incubation bottles
112 after being sieved to 2 mm. These incubation bottles had pierced lids that enabled gas exchange, but
113 minimized evaporation and soil drying. Prior to the beginning of the treatments, all soils were pre-
114 incubated at 25 °C for 7 days to minimize potential disturbances (Zhao et al., 2022). Our experiment

involved two treatments: wollastonite mineral ($0.1 \text{ g g soil}^{-1}$) addition and no wollastonite mineral added as control. There were three replicates for each treatment and each land-use type, resulting in 72 incubation bottles in total. Wollastonite was chosen because of its simple chemistry and high dissolution rate (Haque et al., 2019). The wollastonite mineral in this study was sourced from Diaobing Mountain, Liaoning, China in the form of finely ground rock ($\leq 50 \mu\text{m}$). The major chemical compositions obtained from X-ray fluorescence analyses were CaO (42.51%), SiO₂ (53.14%), CaCO₃ (5%), Fe₂O₃ (0.25%), MgO (0.20%), Na₂O (0.11%), and K₂O (0.02%). The soils to be incubated were adjusted to a 60% water holding capacity (WHC) and incubated at 25 °C in the dark. Throughout the experiment, soil moisture was maintained at 60% WHC by adding de-ionized water at regular intervals.

Soil CO₂ efflux were determined based on the headspace of the closed chamber with an infrared gas analyzer (America, Li-Cor-820) connected via a closed loop. Measurements were taken on days 3, 5, 7, 9, 12, 16, 20, 25, 30, 38, 46, 54, 62, and 90 after the incubation began.

2.4 Soil physical-chemical properties analysis

The pH value was detected using the water extraction method (water: soil = 2.5: 1). The contents of dissolved organic carbon (DOC) were measured with a TOC analyzer (Multi N/C 3000, Germany). Soil microbial biomass carbon (MBC) was determined using the chloroform fumigation-extraction method (Brookes et al., 1985) and measured using the TOC analyzer. Soil available nitrogen (AN) and available phosphorus (AP) were measured in accordance with the methods described by Lu (2000) using a continuous flow analyzer (AA3, Seal Analytical, Germany). The soil exchangeable Ca cation contents were measured according to Lu (2000) using an atomic absorption spectrometer. The soil available silicon (ASi) concentration was determined using the acetate buffer method (pH 4.0) (Huang et al. 2020). Soil bulk density samples were obtained using a standard container with a fixed volume size of 100 cm³ and oven-dried at 105 °C to obtain their masses. The oven-dried soil mass and container volume were then used to quantify the bulk density. The soil C and N contents were

141 analyzed with an elemental analyzer (Elementer CARIO Macro, Germany), and the C: N ratios are
142 presented as mass ratios.

143

144 2.5 Calculations and statistical analyses

145 Soil CO₂ efflux (R , mg C kg⁻¹ soil day⁻¹) was calculated as follows:

$$146 \quad R = \frac{C \times V \times M \times 273.15}{22.4 \times (273.15 + T) \times W \times t}$$

147 Where C is the measured CO₂ concentration (ppm); V is the effective volume of an incubation flask
148 (L); M is the molar mass of C (12 g/mol); 22.4 (L) is the molar volume of an ideal gas at 1 atm and
149 273.15 K; W is the gram dry weight of the soil; t is the time of CO₂ accumulation (days); and T is the
150 incubation temperature (25°C).

151 Cumulative soil CO₂ efflux (T , mg C kg⁻¹ soil) was calculated as follows:

$$152 \quad T = \sum_{i=1}^n \frac{R_i + R_{i+1}}{2} \times (t_{i+1} - t_i)$$

153 Where R_i and R_{i+1} are soil CO₂ efflux at i th and $(i + 1)$ th incubation time (mg C kg⁻¹ day⁻¹),
154 respectively; $t_{i+1} - t_i$ is the interval between the i th and $(i + 1)$ th incubation time (day); and n is the
155 number of incubation times.

156 The intensity of treatment effects (Δ , %) was calculated by comparing the difference in T between
157 wollastonite addition treatment and the unamended control.

$$158 \quad \Delta = \left(\frac{\text{treatment} - \text{control}}{\text{control}} \right) \times 100\%$$

159 Shapiro-Wilk and Levene's test were used to check the normality and homogeneity of variances
160 prior to the statistical analysis. One-way ANOVA and the least significant difference (LSD) test were
161 adopted to determine the effects of wollastonite treatments among different land use types. Two
162 sample t-tests were conducted to assess the differences in each measured variable between the
163 treatment and control soils. The relationships between Δ soil CO₂ efflux and Δ soil properties were
164 determined via linear regression analyses. Pearson correlation coefficients were utilized to analyze

relationships between Δ soil CO₂ efflux and initial soil properties. To calculate the relative influence of each initial soil factor, we also employed the hierarchical partitioning method to distinguish a single variable's contribution via the "rdacca.hp" package in R (Lai et al., 2022). All statistical analyses above were performed using R (version 4.1.1).

3 Results

3.1 Soil properties

We observed a strong wollastonite effect on the soil properties (Fig. 1). The addition of wollastonite markedly increased soil pH (+ 2.78 pH units on average) ($P < 0.001$). Wollastonite addition is conducive to improve of soil nutrients, particularly, the Ca, ASi, AN, DOC and MBC were significantly increased 17.54, 14.16, 1.63, 2.04 and 1.75 folds respectively ($P < 0.05$). In addition, AP was not significantly affected by wollastonite (Fig. 1).

3.2 Soil CO₂ efflux

The soil CO₂ efflux at 12 sites drastically dropped until day 38, and then remained steady until the end of incubation (Fig. S1). The soil CO₂ efflux rate at all sites increased after wollastonite addition compared to the control (Fig. S1). Wollastonite addition significantly increased the cumulative soil CO₂ efflux ($P < 0.05$) (Fig. 2). Among all sampling sites, the overall percentage change in soil CO₂ efflux was $330.97 \pm 29.25\%$. There are significant differences in CO₂ efflux among soils ($P < 0.05$), the lowest percentage change in soil CO₂ efflux with a value of 170.21% occurred at the *Amorphophallus konjac* field, and the highest value was 465.63% at the *Michelia macclurei* plantation ($P < 0.05$, Fig. 2).

3.3 Relationships between soil properties and CO₂ efflux

We performed regression analyses with Δ soil CO₂ efflux and Δ soil properties. The results showed that Δ soil CO₂ efflux was significantly positively correlated with soil Δ ASi, Δ Ca, Δ pH, Δ AP and

191 ΔDOC (R^2 values were 0.55, 0.33, 0.57, 0.17 and 0.26, respectively; $P < 0.05$, Fig. 3). The Δsoil
192 properties explained 90.70% of the variations in $\Delta\text{soil CO}_2$ efflux induced by wollastonite addition
193 (Fig. 4), and ΔDOC , ΔASi and ΔpH were the main controlling factors for the $\Delta\text{soil CO}_2$ efflux
194 accounting for 22.59%, 22.42%, and 22.30% of the variation, respectively. In addition, $\Delta\text{soil CO}_2$
195 efflux was negatively correlated with initial soil Ca, ASi and pH (R^2 values were 0.29, 0.33, 0.55,
196 respectively; $P < 0.05$, Fig. 5).

197

198 **4 Discussion**

199 In the present study, the response of SOC mineralization to wollastonite addition was evaluated for
200 12 land-use types that are common in southern China. Wollastonite addition was found to promote
201 SOC mineralization as indicated by the increased soil CO_2 emission during the entire experimental
202 period (Fig. 2), thus no support was obtained for our first hypothesis. Similar trends have also been
203 reported in many previous studies (Groffman et al., 2006; Dietzen et al., 2018; Borges et al., 2019).
204 Based on a 11-year experiment in northeastern United States, Johnson et al. (2014) found a more than
205 40% decrease in the stock of SOC in the Oa horizon following wollastonite addition. As such, the
206 direction of the response of SOC mineralization to wollastonite addition appears to be irrespective of
207 the time the treatment has lasted. We admitted that our experiment might have induced a bias to the
208 estimation of SOC mineralization, given the wollastonite used contains 5% CaCO_3 , which may be
209 one of the sources of CO_2 . However, the CO_2 related to the interaction of CaCO_3 originating from
210 wollastonite and H^+ from soil is, at the most, equivalent to 1.01%-1.82% of the total CO_2 emission.
211 Therefore, we proposed that abiotic CO_2 emission could be ignored in the present study and that the
212 major CO_2 emission resulted from SOC mineralization.

213 We observed that approximately 90% of the variance in $\Delta\text{soil CO}_2$ efflux was explained by Δsoil
214 property (Fig. 4), which supported our second hypothesis that wollastonite addition affects soil CO_2
215 emission by changing soil properties. For instance, the positive effects of wollastonite addition on
216 soil CO_2 efflux could be a consequence of the increased soil pH following wollastonite addition, as a

217 significant positive correlation was observed between the soil ΔpH and accumulated $\Delta\text{soil CO}_2$ efflux
218 (Fig. 3). Increased soil pH value with silicate application has also been reported in many previous
219 studies due to the OH^- release caused by silicate hydrolysis (Groffman et al., 2006; Anda et al., 2015;
220 Xiao et al., 2016). There are two possible pathways for the decreased soil acidity to mediate the
221 influence of wollastonite addition on soil CO_2 emission. First, the acidic stress on the soil microbes
222 was in part loosened. Microbial growth and relevant SOC utilization are often limited in highly acidic
223 soils (Khalil et al., 2005; Kemmitt et al., 2006; Zhang et al., 2020), thus an increase in soil pH in
224 response to wollastonite addition may favor microbial activity and further the SOC mineralization
225 (Haynes and Naidu, 1998; Borges et al., 2019; Ribeiro et al., 2020). Many other studies involving
226 lime application also have published similar results as indicated by the increased soil MBC and
227 respiration as the soil pH increases (Fuentes et al., 2006; Wu et al., 2021). Second, more recalcitrant
228 C has been translated to labile form, an easily utilized fraction of soil C for microbes, as a response
229 to the soil pH increase. The release of DOC from organic soil layers has been positively related to
230 soil pH before (Andersson et al., 2001; Jokinen et al., 2006). The theories behind this phenomenon
231 included the enhanced deprotonation of organic substances in response to soil acidity decrease and,
232 as a result, the bonding between organic compounds and soil particles decreases, leading to an
233 increased amount of dissolved organic matter in the soil (Curtin et al., 1998; You et al., 1999).
234 Moreover, higher microbial activity generally enhanced the production rate of DOC (Gödde et al.,
235 1996; Andersson et al., 2000). DOC leaching and consumption in soil may further prompt microbial
236 decomposition of organic matter to obtain C, resulting in a lower SOC content.

237 Soil nutrients, in addition to pH, also played a vital role in relating wollastonite addition to SOC
238 mineralization (Nowinski et al., 2008). First of all, the silicon contained in wollastonite should have
239 accounted for a significant portion of the variance in SOC decomposition as Si can desorb organic C
240 from mineral binding sites (Reithmaier et al., 2017; Schaller et al., 2019). Recently, Ma et al. (2021)
241 also found that the amount of soil organic matter decreased after Si application to paddy soil. This
242 explanation was supported in part by the fact that ΔASi was positively correlated with $\Delta\text{soil CO}_2$

243 efflux (Fig. 3). Furthermore, the higher Si availability also may stimulate microbial decomposition
244 by improving the availability of some potential limiting nutrients, such as P, in the soil. Although
245 wollastonite addition did not significantly increase the AP concentration in our study (Fig.1), this
246 phenomenon is unsurprising given Si often strongly interfere with Fe mineralogy or Al-oxides to
247 compete with P for binding sites, thereby increasing P mobilization and availability (Koski-Vähälä et
248 al., 2001; Borges et al., 2019; Schaller et al., 2019). In addition, we also found a significant increase
249 in the availability of Ca^{2+} , which is thought can stabilize SOC by enhancing its resistance to microbial
250 decomposition (Rowley et al., 2021; Wan et al., 2021). Altogether, it seems that the increments in soil
251 pH, DOC, and ASi have exerted a predominant effect over Ca^{2+} increase on microbial decomposition,
252 resulting in a faster SOC mineralization following wollastonite addition. In our study, Δ soil properties
253 explained relatively low variation of Δ soil CO_2 efflux (17%-57%), suggesting that there may be other
254 influencing factors that contribute to the variation of soil CO_2 efflux. This point towards the need to
255 pay further attention to the mechanisms behind the effect of wollastonite addition on SOC
256 mineralization in the future.

257 In the present study, land use type was found to be a factor regulating the magnitude of the response
258 of SOC mineralization to wollastonite addition (Fig.2). This phenomenon indicated that soils with
259 various traits responded differentially to wollastonite addition in the SOC mineralization. For
260 example, our results showed that the extent of wollastonite-induced increase in CO_2 efflux was related
261 to soil pH, with soils having lower pH values received stronger wollastonite effect (Fig. 5). This
262 finding was supported by that of Zhang et al. (2012), who even detected a 5%-12% decrease in soil
263 CO_2 efflux after adding calcium silicate to a neutral soil (pH 6.5). In addition, larger treatment effect
264 also took place in the soils with lower ASi and Ca cation (Fig.5). However, it should be noted that the
265 initial soil physicochemical properties are still relatively low in explaining Δ soil CO_2 efflux
266 variation (29%-55%) (Fig.5), which indicated that the understanding of the regulation of wollastonite
267 effect on SOC is limited in our study, and further research is needed for evaluating the wollastonite
268 effect on SOC mineralization from the aspects of wollastonite addition concentration and more soil

269 types, and exploring its main regulation factors. Nevertheless, our study suggests to some extent that
270 the increased SOC mineralization induced by the wollastonite addition may offset the effectiveness
271 of ERW, especially in soils with lower pH.

272 Wollastonite application is likely of very limited values for these subtropical areas, which generally
273 dominated by acidic soil, to remove CO₂ from the atmosphere, since the decomposition of organic C
274 in acidic soils is highly sensitive to wollastonite addition. However, this speculation seems not that
275 accurate, considering the potential influence of wollastonite addition on the plant-derived C input was
276 not quantized and, whether wollastonite addition will negatively affect SOC accumulation is therefore
277 unclear at this stage. Potentially, there are two pathways of plant production can be promoted
278 following wollastonite addition, one being the loosened acidic stress on plants due to the soil pH
279 increase, just as it did for microbes (Anda et al., 2013; Haque et al.2020b; Taylor et al., 2021); and
280 the other is the increased availability of mineral nutrients, such as Ca, Si, P, and N, which stem from
281 silicate rock dissolution (Basak et al., 2017; Kelland et al., 2020). We might therefore expect more
282 root exudates and litter inputed to the soil after wollastonite addition, offsetting at least a part of the
283 C loss associated with the enhancement in microbial decomposition (Kantola et al., 2017). To
284 determine the net effect of wollastonite addition on SOC pool, future studies regarding the responses
285 of soil and plants within a system are needed.

286

287 **5 Conclusion**

288 Before wollastonite addition can be considered an effective practice of removing atmospheric CO₂,
289 what role it will play in determining SOC mineralization should be assessed first. Our results revealed
290 that wollastonite addition did facilitate SOC mineralization for all 12 land use types included in this
291 study, as indicated by the enhanced CO₂ emission. Wollastonite addition achieved its effect on SOC
292 mineralization via increasing the soil pH value, Si and P availability, which partially loosened the
293 limitations for microbial metabolism caused by soil acidity and nutrients shortage, resulting in a faster
294 C utilization by microbes. Overall, our findings did not underpin that wollastonite addition can be

295 used as a C sequestration tool in the acidic soils at this stage. Taken together, our study contributes to
296 a better awareness to the necessity of wollastonite addition as a practice of removing CO₂ from the
297 atmosphere. We also acknowledge that our study lasted for a relatively short period, and whether the
298 findings obtained can be applied to chronic situations still awaits further experimental evidences.

299

300 **Conflict of interest statement**

301 All authors have approved the submission and none declare any conflict of interest in the work
302 performed or in the submission of the manuscript.

303

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309

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