

Large invisible carbon sink potential of global clay minerals to mitigate climate change

Huipeng Xi

Institute of Geochemistry, Chinese Academy of Sciences

Xiaoyong Bai (✉ baixiaoyong@vip.skleg.cn)

Institute of Geochemistry, Chinese Academy of Sciences

Luhua Wu

Institute of Geochemistry, Chinese Academy of Sciences

Chaojun Li

Institute of Geochemistry, Chinese Academy of Sciences

Huan Chen

State Key Laboratory of Environmental Geochemistry, Institute of Geochemistry, Chinese Academy of Sciences

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1 **Large invisible carbon sink potential of global clay minerals to**
2 **mitigate climate change**

3 **Abstract:** The chemical weathering of clay minerals widely distributed in the soil
4 have great potential of carbon sink (CS), but the magnitude and influence mechanisms
5 of this CS are unclear. Here we analyse recent changes in five major clay minerals
6 (chlorite, smectite, mica, illite, and vermiculite) carbon sink and its driving factors,
7 using a process-based model (PROFILE) and satellite data assimilation. We show that
8 magnitude of CS in five major clay minerals is about $0.11 \text{ Pg C yr}^{-1}$ from 0 to 2m
9 depth of soil, which is one third of CS in rocks and also may be mainly responsible
10 for the world's missing carbon sink. According to our simulations, the linear trend of
11 CS during 1970-2018 showing that CS in 56% of the world increasing significantly,
12 although the intensification of CS cannot be explained by soil moisture (SM) or soil
13 temperature (STMP) alone, they are the dominant cause of the intensification of CS in
14 the high latitude area and the decrease of CS in parts of the tropics, while in areas
15 where SM is drier, STMP may weaken the former's negative effect on CS. Besides,
16 simulation results based on medium emission scenarios indicating that CS may
17 increase by about 36% by the end of this century. These results highlight that a more
18 comprehensive understanding of the magnitude and driving mechanism of the soil
19 minerals' CS is the key to realizing their potential as a nature-based climate solution.

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23 **Key words:** Chemical weathering; Carbon sink; Soil minerals; Climate change;
24 carbon budget

25

26 **Introduction**

27 There is still a carbon imbalance of about 0.1-0.6 Pg C yr⁻¹ in the current carbon
28 cycle system([Stocker et al., 2013, Fig. 1](#)), previous studies have regarded the carbon
29 sink (CS) produced by chemical weathering of rocks as the key to solving the carbon
30 imbalance problem, and have estimated the magnitude and pattern of the CS of
31 various rocks based on this ([Hartmann, 2009; Li et al., 2018; Romero-Mujalli et al.,](#)
32 [2018; Xi et al., 2021](#)). However, due to the shielding effect of the soil layer, the
33 chemical weathering flux budget of the underlying bedrocks often loses more than
34 40% under natural conditions ([Hartmann et al., 2014](#)), and the main reason is the
35 incomplete weathering of rocks, resulting in the formation of smaller primary and
36 secondary minerals distributed in the soil, such as feldspar, pyroxene, chlorite,
37 smectite and mica, etc ([Zolkos et al., 2018](#)). These minerals can not only store about
38 600Pg of organic carbon per year ([Ferdush and Paul, 2021](#)), but also absorbing a
39 certain scale of inorganic carbon through chemical reaction with CO₂ in the
40 atmosphere/soil ([Doetterl et al., 2018](#)), especially for most clay minerals with silicate
41 as the main component, they are not only widely distributed, but also having a large
42 specific surface area for reaction, resulting in a high degree of stability of the carbon
43 sink effect on the geological time scale ([Bibi et al., 2016; Whitfield et al., 2018](#)).
44 Therefore, estimating the magnitude and pattern of the weathering CS of clay
45 minerals can not only provide new ideas for solving the problem of carbon budget
46 imbalance, but also will hopefully update the understanding of soil carbon pools.

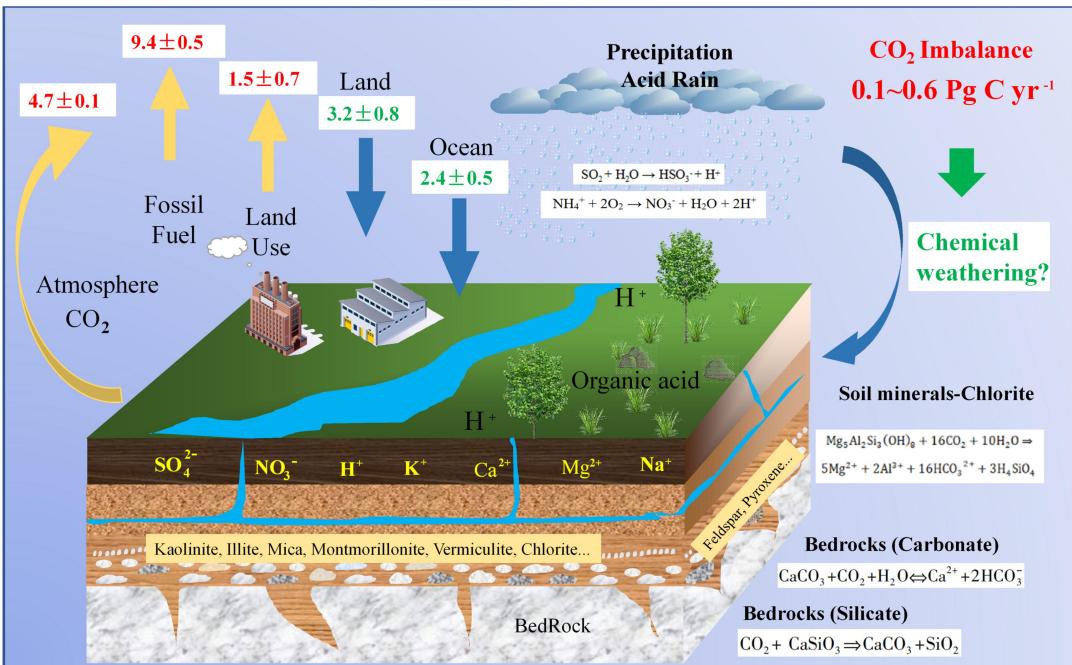
47 The weathering of minerals in the soil can not only generate CS, but also can
48 release lots of basic cations, which play a key role in maintaining the development of
49 terrestrial ecosystems and mitigating the harm of acid deposition. Therefore, scholars
50 have done a lot of modeling and inversion for the chemical reaction kinetics of

51 various minerals in the soil (Goddéris et al., 2006; Olsson et al., 1993; Sverdrup and
52 Warfvinge, 1993; Sverdrup and Belyazid, 2014), and then estimated the ability of the
53 minerals to release cations (that is, the weathering rate of the soil). For example,
54 Erlandsson et al estimated the soil weathering rate at the site scale based on long-term
55 measured results and the PROFILE model (Erlandsson et al., 2016), similarly, Roy et
56 al used the same model and the ISRIC-Wise database to estimate the ratio of basic
57 cation to Al concentration on a global pixel scale (Roy et al., 2012). At the watershed
58 scale, Goddéris et al (2006) considered the migration of chemical elements between
59 different layers of soil, and using the WITCH model to estimate the weathering rate of
60 the soil on the granite bedrock, on this basis, Roelandt et al (2010) and Beaulieu et al
61 (2011) estimated the distribution characteristics of weathering rate and CS in pixels
62 based on the spatial distribution map of minerals in the soil, respectively. Therefore, it
63 can be seen from previous studies that based on a suitable kinetic model and soil
64 mineral distribution map, it is expected to estimate the spatial distribution pattern of
65 CS produced by chemical weathering of minerals in the soil. It can be seen from
66 previous studies that based on a suitable kinetic model and the soil minerals
67 distribution map, it is expected to estimate the spatial distribution pattern of CS of soil
68 minerals. However, due to the lack of data on the spatial distribution of soil physical
69 properties and mineral composition at the global scale, only a few studies have
70 discussed the CS of minerals weathering in soil at the site and small watershed scales
71 (Beaulieu et al., 2011; Goddéris et al., 2013), studies on the magnitude and temporal
72 and spatial distribution of weathering CS of soil minerals, especially clay minerals,
73 have not yet been discussed on the global pixel scale.

74 In addition, the weathering of minerals in the soil is very sensitive to the
75 disturbance of climate change. For example, the experimental results of Akselsson et

76 al (2016) in Sweden showed that warming temperature can increase the weathering
77 rate of minerals in the soil by more than 20%, and the study by Yu et al (2020) in the
78 subtropical regions of China found that regional warming reduced the basal cation
79 concentration of soil weathering. Previous studies have shown that soil water is also
80 an important factor affecting soil weathering (Kronnäs et al., 2019), but as soil
81 moisture (SM) in most regions of the world decreases (Deng et al., 2020), Not only
82 the chemical weathering process of rocks may be weakened (Xi et al., 2021), the
83 weathering of soil minerals is also affected (Yu et al., 2020), especially in arid and
84 semi-arid areas, SM is not only an important factor in controlling the soil carbon pool
85 (Zhao et al., 2019), but the drought can cause a large amount of soil carbon pool loss,
86 especially the bottom soil carbon pool (Canarini et al., 2017; Soong et al., 2021; Su et
87 al., 2020). Therefore, in the context of global warming and soil drying (Deng et al.,
88 2020), evaluating their respective effects on CS of soil minerals is of great
89 significance for correcting soil weathering models and re-understanding soil inorganic
90 carbon pools.

91 Hence, based on the widely used steady-state soil geochemical profile model
92 (PROFILE model), and using the newly released spatial distribution map of the global
93 clay mineral content (Ito and Wagai, 2017), we estimated the monthly-scale CS of
94 clay-grade minerals in the global soil from 1970 to 2018, aiming to discuss the
95 following questions: 1) What is the magnitude, pattern and evolution trend of the CS
96 of the chemical weathering of clay minerals in the soil? 2) What are the spatial
97 characteristics of the effects of global warming and soil drying on CS of soil minerals?
98 3) How does the CS of soil minerals compare with other CS?



99

100 Fig. 1 Schematic diagram of the global carbon budget from 2008 to 2019

101 **Results**102 ***Spatial differentiation of carbon sink***

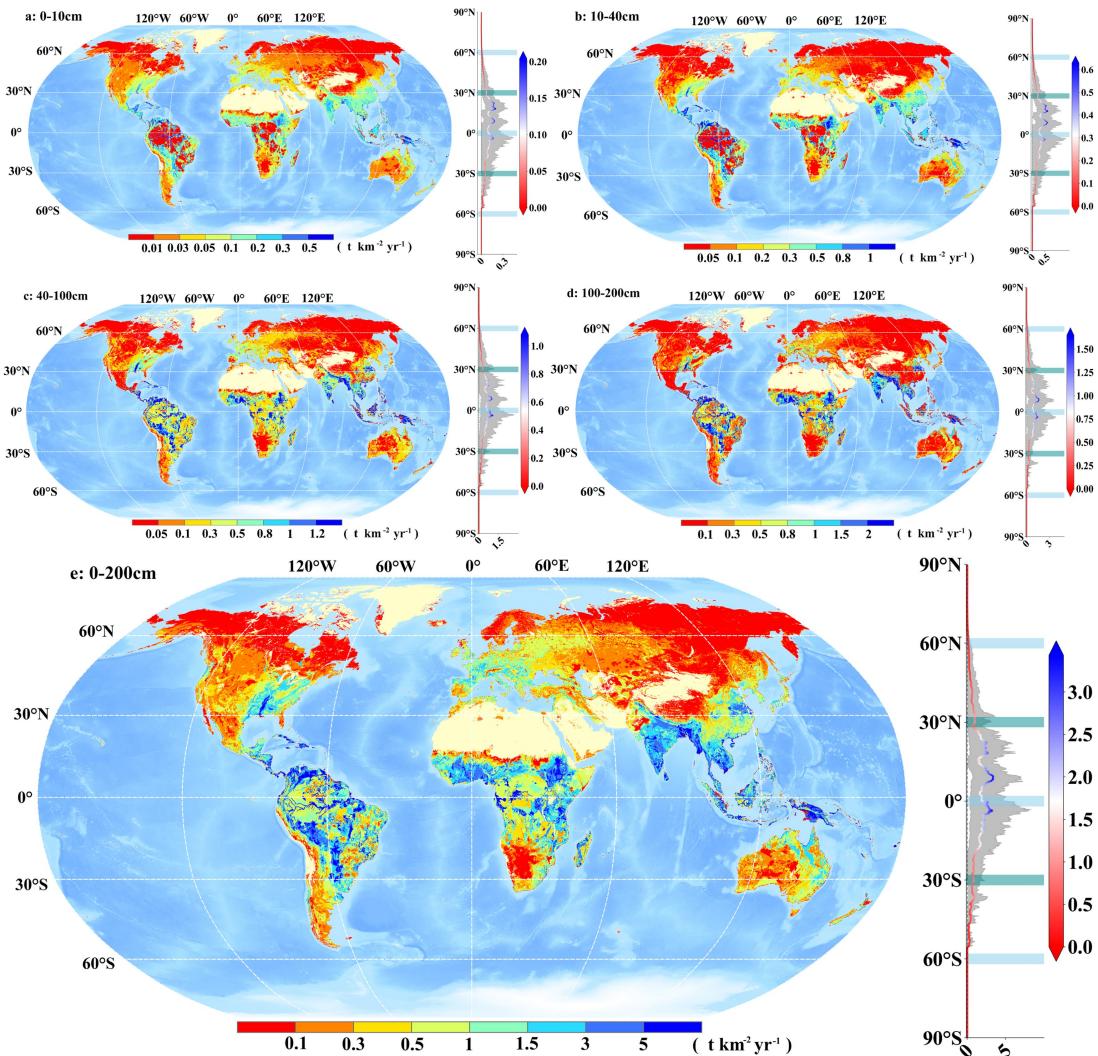
103 In terms of magnitude, combined with the chemical formula of typical mineral
 104 weathering (Goddéris et al., 2006), we have summarized the CS of clay minerals in
 105 soils at depths of 0-200 cm around the world. It was also found that the annual
 106 average carbon sink flux (CSF) of five types of clay minerals (including chlorite,
 107 smectite, illite, mica, and vermiculite) was about 0.50 t C km⁻², and the total CS was
 108 about 110.86 Tg C yr⁻¹, which accounts for about 34% to 42% of the CS of rocks
 109 (Hartmann., 2009) and about 18% of the missing carbon sink (Xi et al., 2021), in
 110 addition, the magnitude of this CS is basically equivalent to the CS of global silicates
 111 (Zhang et al., 2021). Based on the statistics of climate zones, it is found that the CSF
 112 (2.43 t C km⁻²) and carbon storage (59.69 Tg C yr⁻¹) in tropical are much higher than
 113 other climate regions, followed by temperate (20.83 Tg C yr⁻¹), arid (18.17 Tg C yr⁻¹),
 114 cold zone (11.60 Tg C yr⁻¹) and polar zone(0.58 Tg C yr⁻¹) (Table S1). In addition,
 115 affected by the mineral content, the capacity of CS clay minerals in the soil is

116 positively correlated with soil depth, the average CS at depths of 0-10cm, 10-40cm,
117 40-100cm, and 100-200cm are about 6.77 Tg C, 21.22 Tg C, 33.10 Tg C, 49.77 Tg C,
118 etc. ([Fig. 3](#)), the CSF increases with depth, and the average values of CSF are 0.03 t C
119 km^{-2} , 0.10 t C km^{-2} , 0.15 t C km^{-2} , and 0.23 t C km^{-2} , respectively.

120 In terms of spatial distribution, the high-value areas of CSF are mainly
121 concentrated in the range of 30° north-south latitude, and the low-value areas are
122 mainly distributed in areas above 30°N in the northern hemisphere, such as Russia,
123 Canada, and Alaska. Affected by soil thickness, there are differences in CS at different
124 depths in the same area. For example, in the karst area of southwest China, despite
125 abundant hydrothermal conditions, most of the soil thickness in this area is less than 1
126 meter ([Li et al., 2020](#)), so the high value of CSF is mostly only within the depth of 0
127 to 100cm, while the CS potential of minerals at the depth of 100-200cm is greatly
128 reduced ([Fig. 2d](#)). However, in the Amazon River Basin in South America, due to the
129 large runoff and serious surface soil erosion ([Flores et al., 2019](#)), the mineral content
130 in the soil is low, so the CSF of the minerals in the surface soil is also very low ([Fig.](#)
131 [2a](#)), but the CS of soil below 40cm starts to increase ([Fig. 2c](#)). In addition, near the
132 Congo Basin in Africa, high temperature and drought have reduced the water content
133 of the topsoil ([Zhu et al., 2021](#)), which will also limit the CS of potential of the topsoil,
134 while in the subsoil, with the increase of soil moisture Increase, this restriction has
135 been alleviated.

136 At different latitudes, the CSF presents an obvious three-peak distribution.
137 Taking the latitude profile of CSF at a depth of 0 to 200 cm as an example ([Fig. 2e](#)),
138 the CSF of world's major clay minerals first increase and then decrease from north to
139 south, among them, the average CSF in the 30°N-60°N latitude zone is mostly lower
140 than 1 t C km^{-2} . From 30°N to the south, the CSF gradually increases, in the range of

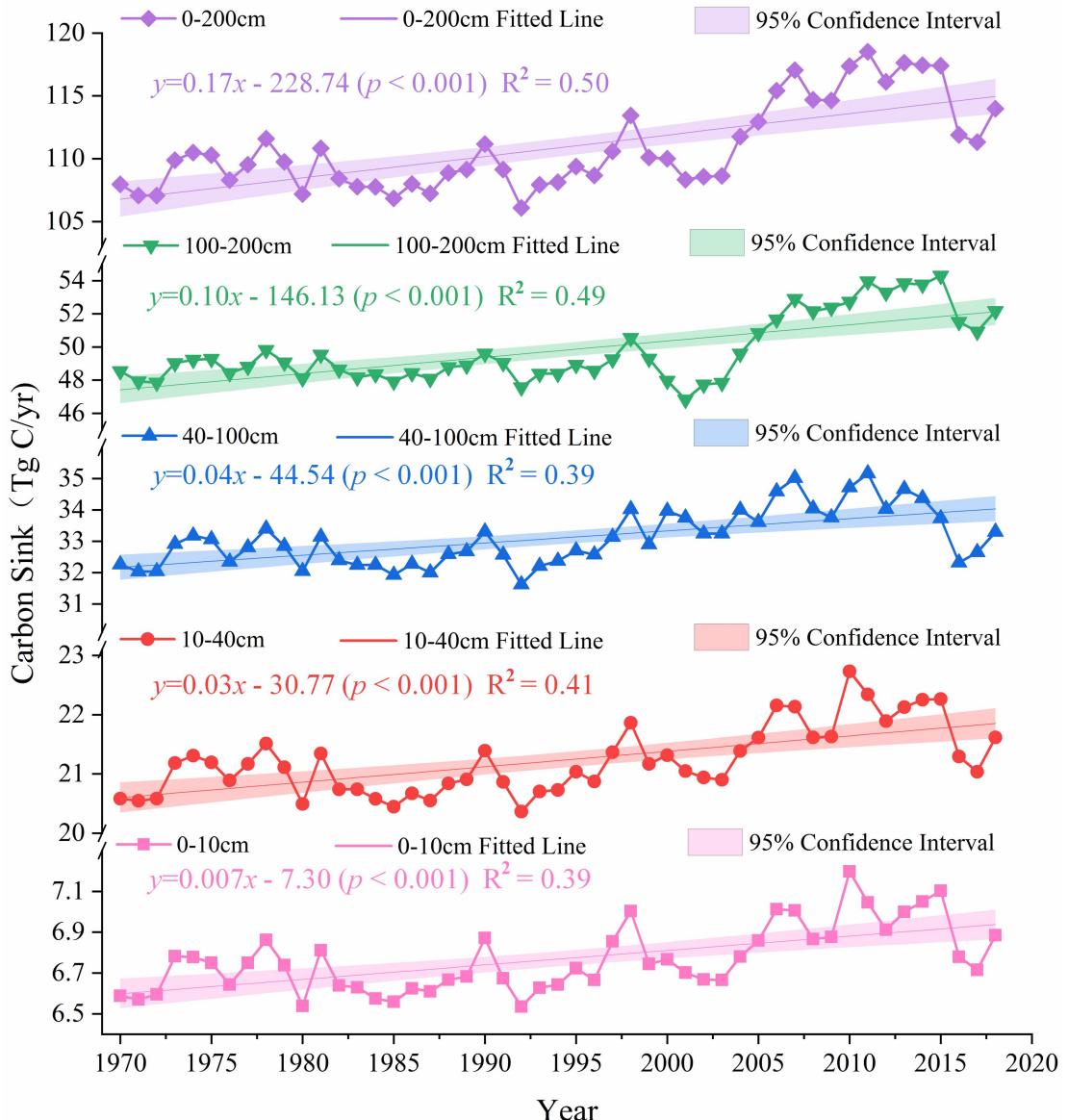
141 17°N to 24°N, the average CSF is higher than 2.0 t C km⁻², and in this latitude zone,
 142 the CS of it accounts for about 7.4% of the global. Moreover, the CSF reached its first
 143 peak (2.80 t C km⁻²) near 18°N. Thereafter, to the south, the CSF first decreased and
 144 then increased, and reached the second peak (3.47 t C km⁻²) near 8°N. The average
 145 CSF in the range of 7°N to 11°N is higher than 3 t C km⁻², and the CS in this latitude
 146 zone accounts for a higher proportion (9.7%). The third peak appeared near 3°S (3.31
 147 t C km⁻²), and the average CSF in the latitude zone between 0°S and 5°S reached 2.75
 148 t C km⁻², and the CS accounted for about 10.64%.



149
 150 Fig. 2 Spatial and latitude distribution map of global weathering carbon sink of major clay
 151 minerals

152 *Spatial differentiation of carbon sink trends*

153 Clarifying the temporal and spatial evolution characteristics of CS of soil
154 minerals is of great significance for understanding the fluctuations of regional or
155 global soil inorganic carbon storage. To this end, we analyzed the magnitude and
156 spatial evolution trends of CS of five major types of clay minerals in the world from
157 1970 to 2018. The results show that the change trend of CS of clay minerals at
158 different depths basically shows a fluctuating upward trend, and the change trend is
159 significant ([Fig. 3](#)). The growth rate increases with soil depth, from $0.007 \text{ Tg C yr}^{-1}$ to
160 0.1 Tg C yr^{-1} in 0 to 200 cm deep soil, and the average growth rate of global CS is
161 $0.17 \text{ Tg C yr}^{-1}$. This rate is higher than the change trend of CS of major rocks in recent
162 years (0.1 Tg C yr^{-1}), but lower than that of carbonate rocks ($0.31 \text{ Tg C yr}^{-1}$) ([Xi et al.,](#)
163 [2021](#)).



164

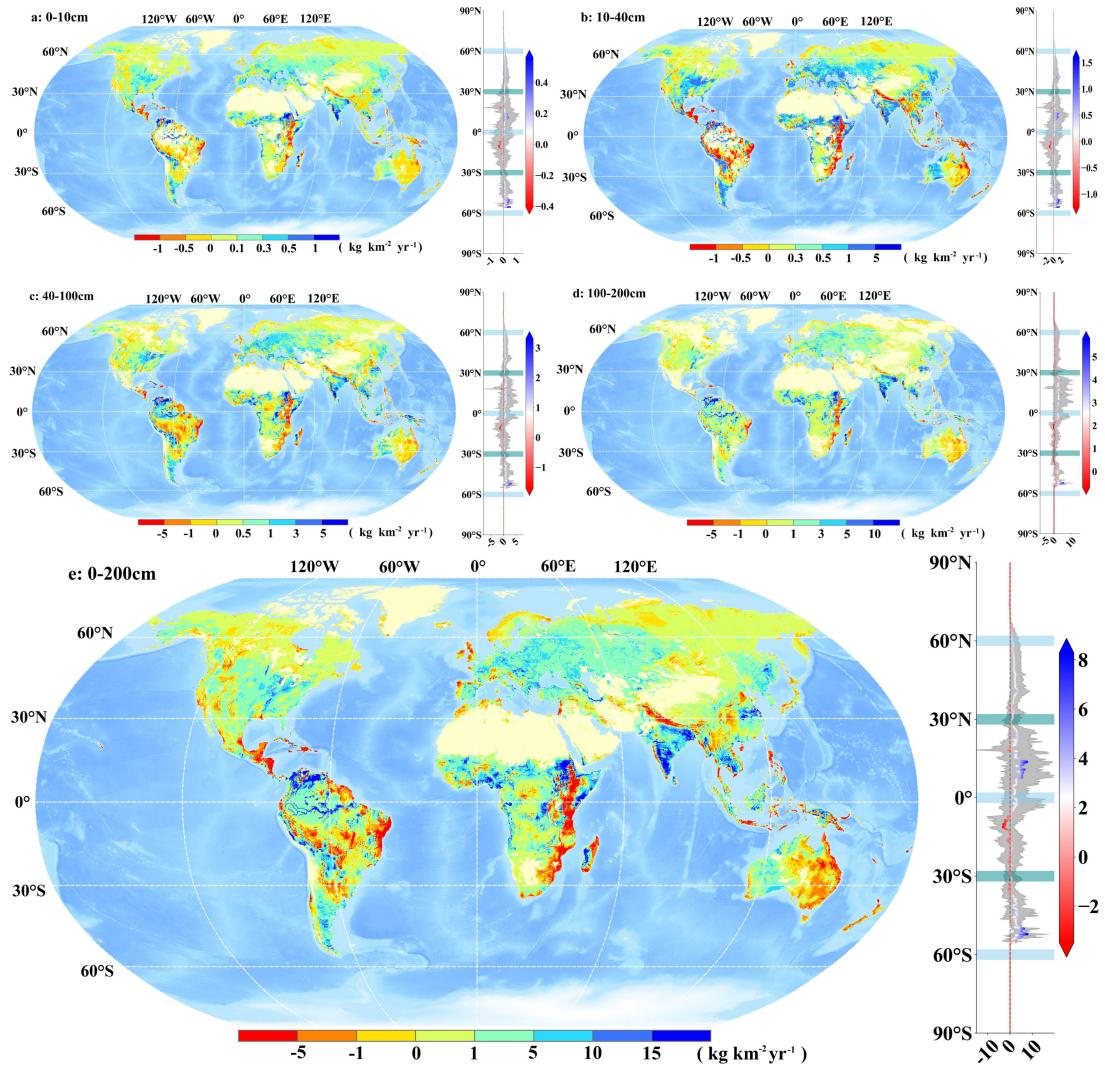
165 Fig. 3 Change trend of carbon sink of clay minerals at a depth of 0 to 2m from 1970 to 2018

166 Spatially, we estimated the changing trend of CS in soil at different depths on a
 167 pixel scale (Fig. 4), and found that the trend of CS in clay minerals is opposite to
 168 that of silicate rocks ($-0.027 \text{ t C km}^{-2} \text{ yr}^{-1}$) (Zhang et al., 2021), in the 0-200cm depth
 169 of soil, the CS of clay minerals in about 56.47% of the world has shown a significant
 170 increase. Since 1970-2018, about 1.4 kg of more carbon can be sequestered per
 171 kilometer per year on average. In addition, the growth rate of CS in the bottom soil
 172 (100-200cm) is about 8 times that of the surface soil (0-40cm), which indicating that
 173 the carbon storage of minerals in the bottom soil has been increasing in the past half

174 century, and the role of balancing the carbon cycle of the minerals in the bottom soil
175 is growing. Most of the CS in the northern hemisphere above 30°N are
176 growth-oriented, and the high-value areas of growth are mainly concentrated in the
177 Indian Peninsula, the Nile River Basin, the Amazon River Plain, South America near
178 the Caribbean, and the middle and lower reaches of the Yangtze River in China.

179 Combined with the climate zoning map, we found that in tropical regions, the
180 significant increase or decrease of surface soil (0-40cm) is basically the same, and the
181 rate of CS is negative, indicating that the carbon sequestration capacity of surface soil
182 in tropical regions is weakening. However, the growth rate of CS in the bottom soil
183 can offset the deceleration of CS in the surface soil, eventually, the CS in the tropics
184 are still mainly increasing. In addition, although the CS in the temperate zone is
185 higher than that in the arid zone, the growth rate of the CS in this region is smaller,
186 which indicating that the potential CS of minerals in the soil is gradually increasing in
187 the arid zone. In the cold and polar zone, the CS of 71.89% and 55.17% of the regions
188 are increasing significantly, respectively.

189 In the latitude zone, the overall trend of CS increasing firstly and then decreasing
190 from north to south. Taking the profile of the CS trend at a depth of 0-200 cm as an
191 example ([Fig. 4e](#)), in the range of 20°N to 60°N, the trend of CS is mostly positive. In
192 the range of 5°N to 15°N, the rate of CS is mostly greater than 4.5 kg C km⁻² yr⁻¹, and
193 a peak value (8.05 kg C km⁻² yr⁻¹) appears near 14°N, and then, the change trend of
194 CS turned negative as the latitude decreased, and the lowest value (-3.57 kg C km⁻²
195 yr⁻¹) appeared near 11°S. The decreasing trend in the southern hemisphere did not
196 gradually turn positive until around 35°S, and reached a peak again around 52°S (8.65
197 kg C km⁻² yr⁻¹).



198

199 Fig. 4 Spatial and latitude differentiation of the change trend of clay mineral weathering carbon
200 sink

201 ***The influence of soil moisture on carbon sink of clay minerals***

202 Evaluating the impact of changes in soil hydrothermal properties on the CS of
203 clay minerals at different soil depths is of great significance for understanding and
204 improving the response mechanism of soil inorganic carbon storage to climate change.
205 Based on the method mentioned in 2.2.3, we separated the relative contribution of SM
206 and STMP to the CS of clay mineral weathering. The relative contribution of SM at a
207 depth of 0-200cm is shown in Fig. 5. Although the area of the positive contribution
208 area (52.91%) is slightly larger than the negative contribution area (47.08%), the
209 average pixel value is -2.4%, which indicating that the SM has a negative effect on

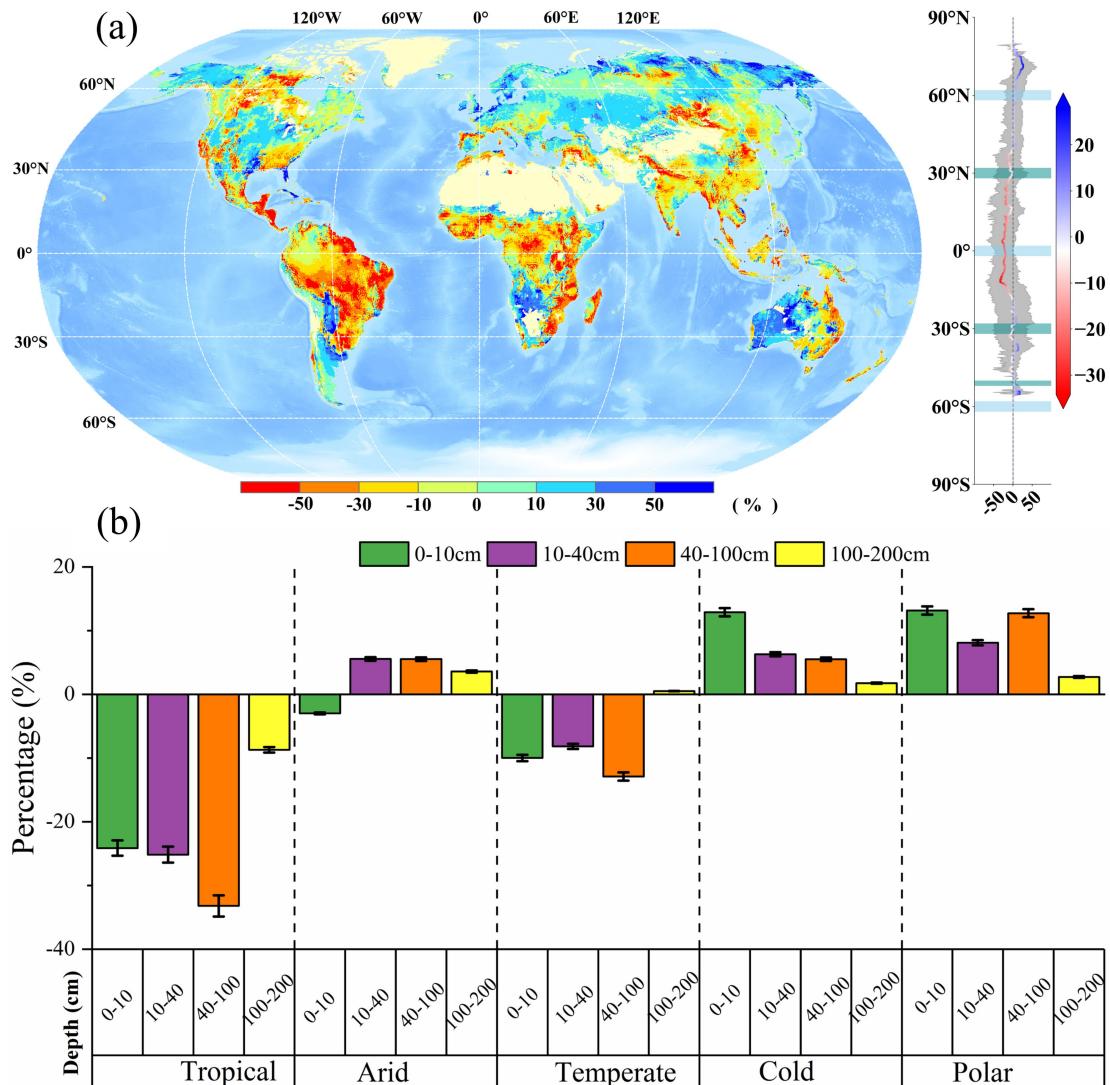
210 the weathering of clay minerals as a whole.

211 Spatially, the area where soil moisture is negatively affected are mainly
212 concentrated in the southern hemisphere. Except for Western Australia, Western
213 Africa, and Western South America, the influence of SM in other areas is mostly
214 negative, while in areas above 30°N north latitude, the area where the soil moisture
215 has a positive contribution is larger. In terms of latitude, the negative influence area of
216 SM is mainly concentrated in the latitude zone of 30°N to 25°S. The highest value of
217 the relative contribution of SM appears near 70°N (29%), and the lowest value
218 appears near 12°S (-34%).

219 In climatic zones, the contribution of SM varies greatly in space. In tropical and
220 temperate zones, the SM is mostly shows a negative contribution, especially in the
221 soil with a depth of 0 to 100cm, the negative contribution of SM in the tropics is the
222 highest (-24.28%). In the arid zone, the contribution of surface soil moisture is
223 negative, but as the soil depth increases, the contribution of SM becomes positive,
224 which further illustrating the potential of CS of minerals in the deep soil of the arid
225 zone. In cold and polar zones, SM has a positive contribution, which may be related to
226 the moistening of the soil in the high latitudes of the northern hemisphere in recent
227 years ([Deng et al., 2020](#)).

228 Combining with the change trend of soil moisture ([Fig. S1](#)), we found that in the
229 0 to 100cm depth of soil, the average relative contribution of the area with reduced
230 SM is -23.59%, and the average contribution of about 78.4% of the regional soil
231 moisture is negative. The average contribution of 78.4% of the SM in this area is
232 negative, which indicating that the surface soil moisture does weaken the weathering
233 CS of clay minerals when there is a drying trend. However, in the bottom soil, this
234 weakening effect is somewhat reduced, with a relative contribution of about -11.48%

235 at a depth of 100-200cm, and an average of about 88.72% of the area shows a
 236 negative contribution area.



237

238 Fig. 5 Spatial differentiation of the relative contribution of soil moisture

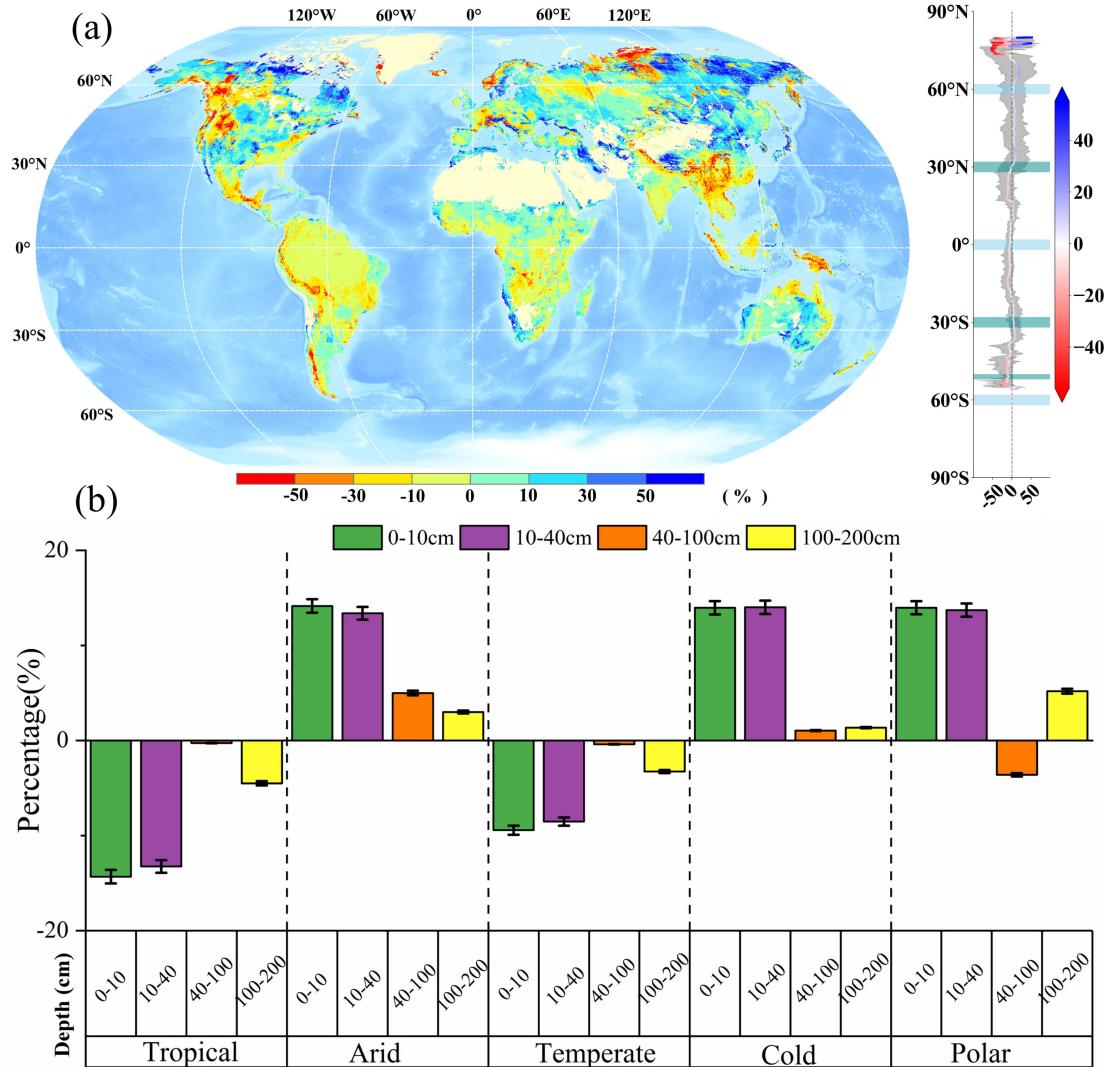
239 *The influence of soil temperature on carbon sink of clay minerals*

240 The relative influence of STMP is shown in the figure below (Fig. 6). The results
 241 show that in the 0-200cm deep soil, the average value of the STMP is 3.29%, and
 242 more than half of the area shows a positive contribution (55.90%), which indicating
 243 that STMP accelerates the chemical weathering of minerals in soil, which is consistent
 244 with previous research conclusions (Akselsson et al., 2016).

245 In terms of spatial distribution, areas where STMP shows a negative contribution

246 are mostly consistent with SM, but the contribution of STMP is less than that of SM.
247 The positive contribution of the high value of STMP is mainly concentrated in the
248 middle and high latitudes of the northern hemisphere, and the overall characteristics
249 are similar to the relative contribution of SM. In terms of latitude differentiation, the
250 overall characteristics of STMP are less significant than that of SM. The average
251 contribution of STMP in most latitude zones is around 0, while the average value in
252 areas above 60°N is mostly shows positive. However, the variability in areas above
253 75°N is extremely large, which may be disturbed by the data itself, but the CSF in
254 these areas is low, so it has little impact on the overall situation.

255 In climatic zones, the contribution of STMP is quite different ([Fig. 6b](#)). It can be
256 seen that the contribution of STMP in tropical and temperate zones is all negative,
257 while the contribution in the arid, cold and polar zones are basically positive, and the
258 average contribution in the arid zone is the highest (8.23%), which indicating that in
259 arid regions, SM and STMP both promote the carbon sequestration of soil minerals.
260 Combining with the spatial map of the change trend of STMP ([Fig. S2](#)), we further
261 found that in areas where STMP is rising, the average relative contribution of STMP
262 is 10.51% (0-40cm), and there are about 70.55% of the area showed a positive
263 contribution on the whole. Although the average relative contribution in mid to deep
264 soil is only 0.5% (40-200cm), in areas where the soil is warmed in the middle and
265 deep layers, there is still 61% of the area where the contribution of STMP is positive.
266 It further shows that when the STMP is warming, it does promote the CS of clay
267 minerals, and this effect mainly occurs in the surface soil.



269 Fig. 6 Spatial differentiation of the relative contribution of soil temperature

270 **Discussion**

271 ***Evaluation of our results***

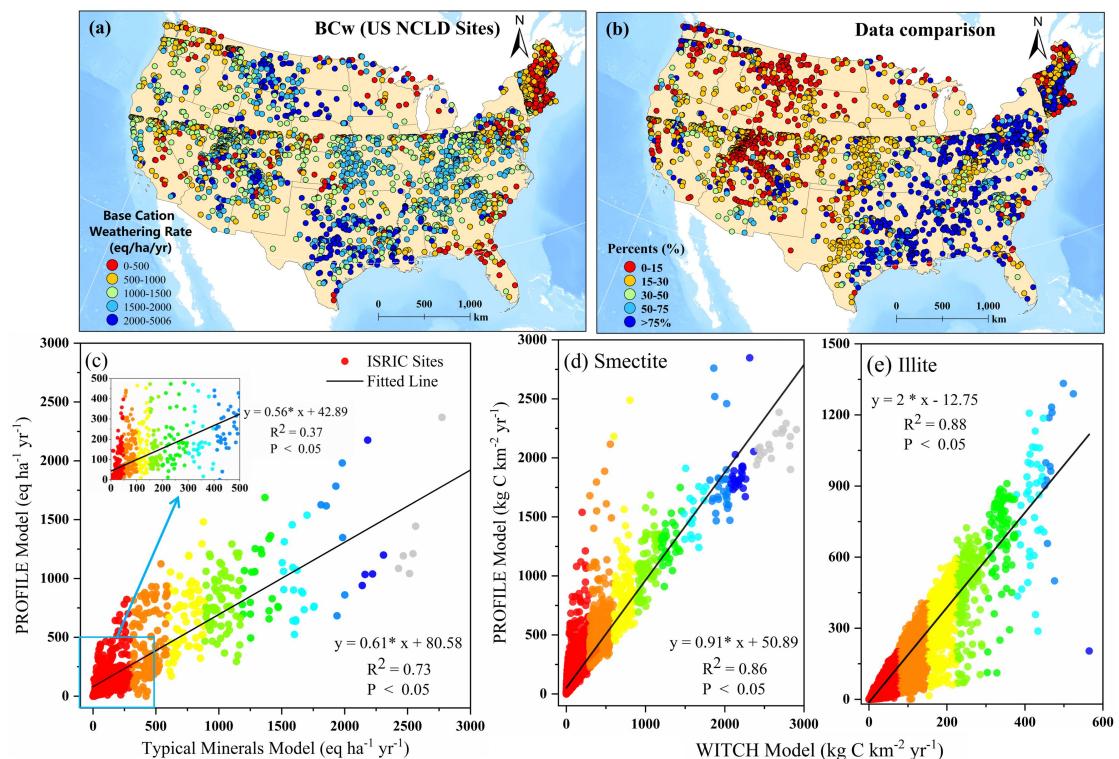
272 In order to verify our estimated results, we carried out comparisons and
 273 discussions from site monitoring results, simulation results of other models, and
 274 research results of other scholars, respectively. Since there is no research on the global
 275 CS of clay mineral weathering, we first compared the soil weathering cation site data
 276 published in the National Nitrogen and Sulfur Deposition Database ([Fig. 7a](#)). This
 277 data set consists of the estimation results of multiple models and a summary of the
 278 measured data. The average value of cationic weathering at these sites is 1046.23 eq

279 ha⁻¹, and our result is 174.92 eq ha⁻¹, with an average proportion of 20.65% and a
280 median of 9.70% ([Fig. 7b](#)), and it can be seen that the ratio is lower in the western
281 United States, while higher in the eastern United States, which may be caused by
282 higher soil temperature and soil moisture in the eastern United States than in the
283 western United States. In general, the results of this paper are lower than the data of
284 the site, but since only five types of minerals are considered, and the more easily
285 weathered feldspar, pyroxene and other minerals are not considered, the estimated
286 result is definitely lower than the overall weathering rate. However, because the
287 values are in the same magnitude, the results have a certain degree of reliability as a
288 whole.

289 In order to further verify the reliability of the model and the results, we
290 compared the calculation results of two different models. First, we estimated the
291 weathering rate of chlorite, mica and vermiculite with reference to a simple mineral
292 model ([Sverdrup and Warfvinge, 2018](#)) with some site data in the ISRIC database ([Fig.](#)
293 [7c](#)), it turns out that our result is slightly smaller than the estimation result of the
294 simple mineral model, and the average is about 0.8 times that of the latter, but most of
295 the sample points are at the same magnitude. This further implies that our estimation
296 results are more reliable.

297 In addition, we also used another set of more general model to characterize soil
298 weathering in watersheds which named WITCH ([Goddéris et al., 2006](#)). This model
299 assumes that each layer of soil is an independent box shape, and the weathering
300 module of it also considering the weathering kinetics of silicate minerals, and this
301 model can be used to simulate the soil weathering rate at the pixel scale in a small
302 watershed ([Roelandt et al., 2010](#)). Therefore, we used this model to estimate the CS of
303 smectite and illite ([Fig. 7d, e](#)). The results show that the CSF of smectite estimated

304 using the WITCH model is smaller than that of the PROFILE model, but the CSF of
 305 illite is higher than the latter. Due to the complexity of soil minerals, the parameter
 306 settings between different models are quite different. Previous studies have also found
 307 that the estimation results between different models can differ by several orders of
 308 magnitude (Erlandsson et al., 2016), but our research show that the results between
 309 different models are almost on an order of magnitude, which once again shows that
 310 our results are highly reliable.



311 Fig. 7 Comparison of results. The Fig. 7a shows the distribution of the US nitrogen and sulfur
 312 deposition database, the Fig. 7b shows the ratio of the estimated results to the site data, the Fi. 7c
 313 shows the comparison of the weathering rate estimated by the PROFILE model and the typical
 314 mineral model, and the Fi. 7d and Fi. 7e show the comparison of the carbon sink flux estimated by
 315 the PROFILE model and the WITCH model.

317 In addition, we have also compared the research results of other scholars (Table
 318 1). In terms of CS, our results are basically consistent with the deduction results of
 319 other studies, and are equivalent to CS of silicate, but smaller than the CS of rocks. In

320 terms of CSF, we compared with the CS results of mineral weathering in loess and
 321 founding that our results are within its range, however, since carbonate minerals that
 322 are more susceptible to weathering were not considered, the value was low. In
 323 addition, although the CS is equivalent to that of silicate rocks, due to the limited
 324 distribution of silicate rocks, its CSF is higher than our results. In terms of weathering
 325 rate, we compared the weathering rate on site and regional scales. Although the
 326 weathering rate is lower than the results of other studies, the ratio is in the range of
 327 13% to 32%. Therefore, this result is more consistent with the comparison result of
 328 the US National Nitrogen and Sulfur Deposition Database.

329 Table1 Comparison with other research

Compared Target	Object	Region	This Study	Others'	References
	Soil			0.12	Andrews and Schlesinger, 2001
Carbon Sink (Pg C)	Silicate	Global	0.11	0.13	Zhang et al., 2021
	Rocks			0.32	Xi et al., 2021
Carbon Sink Flux (t C/km ²)	Loess	America	0.7	0.6~2.4	Goddéris et al., 2013
	Silicate	Global	0.5	1.67	Zhang et al., 2021
Weathering Rate (Eq/ha)		Sweden	44	224~332	Akselsson et al., 2016
	Forest	Västra Torup	169	530~575	Kronnäs et al., 2019
	Soil	Hissmossa	47	152~180	

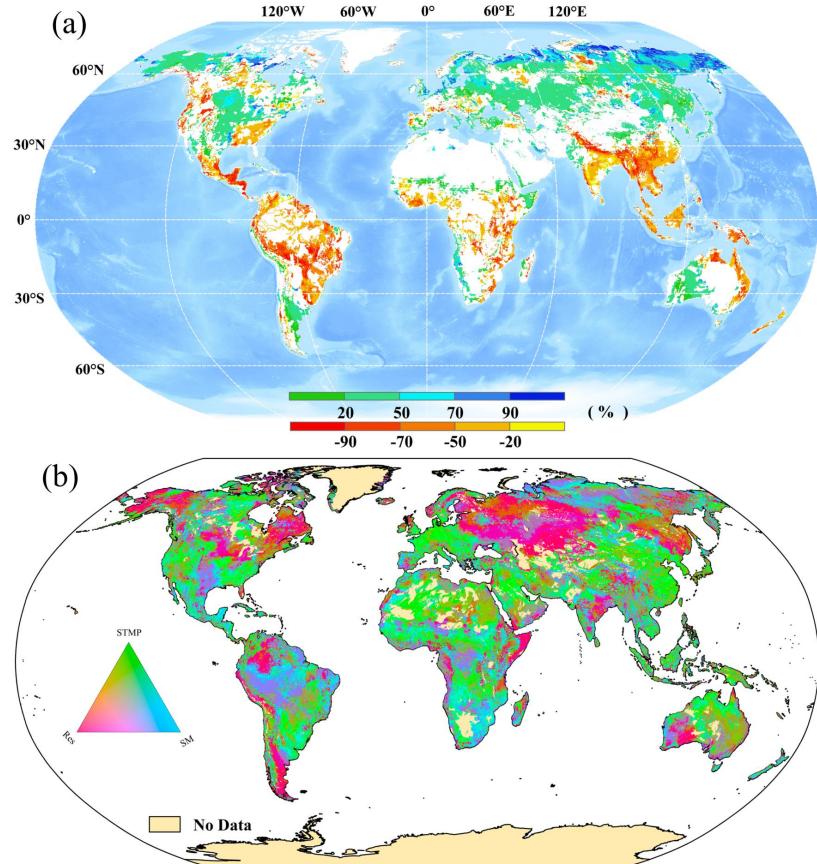
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331

332 *Synergistic and Trade-off effects of SM and STMP on CS*

333 Under natural conditions, soil moisture and soil temperature do not individually
334 affect the weathering of minerals, and they interact to promote or limit the weathering
335 process of minerals. For this reason, we further discussed the synergistic and offset
336 effects of SM and STMP on the CS of clay mineral weathering. By superimposing the
337 distribution map of the relative contribution of SM and STMP, we found ([Fig. 8](#)) that
338 the land area driven by the two together accounted for about 43.17% of the total land
339 area. It can be seen that the area of the main control area of STMP is higher than that
340 of SM ([Fig. 8b](#)). Among them, the areas where both soil moisture and soil temperature
341 contribute positively accounted for about 28.7%, with an average relative contribution
342 of 44.9%. They are mainly distributed in the northern hemisphere, especially in high
343 latitudes. This phenomenon is consistent with the results of other scholars ([Zolkos et](#)
344 [al., 2018](#)), which may be related to the significant warming in the high latitudes of the
345 northern hemisphere in recent years. The melting of frozen soil in high latitudes not
346 only exposes minerals, but also makes the soil environment warm and humid, which
347 is very conducive to weathering reactions ([Cuozzo et al., 2020](#)).

348 The area where SM and STMP are both negatively contributing account for
349 about 14.47%, and the average contribution on the pixel is -51.04%. That is to say,
350 both SM and STMP have led to a reduction of more than half of the CS of clay
351 minerals in this area, which is mainly distributed in most parts of the southern
352 hemisphere, especially in climatic regions such as tropical and temperate zones, the
353 reason may be that the soil moisture in these areas is greatly reduced ([Deng et al.,](#)
354 [2020](#)).



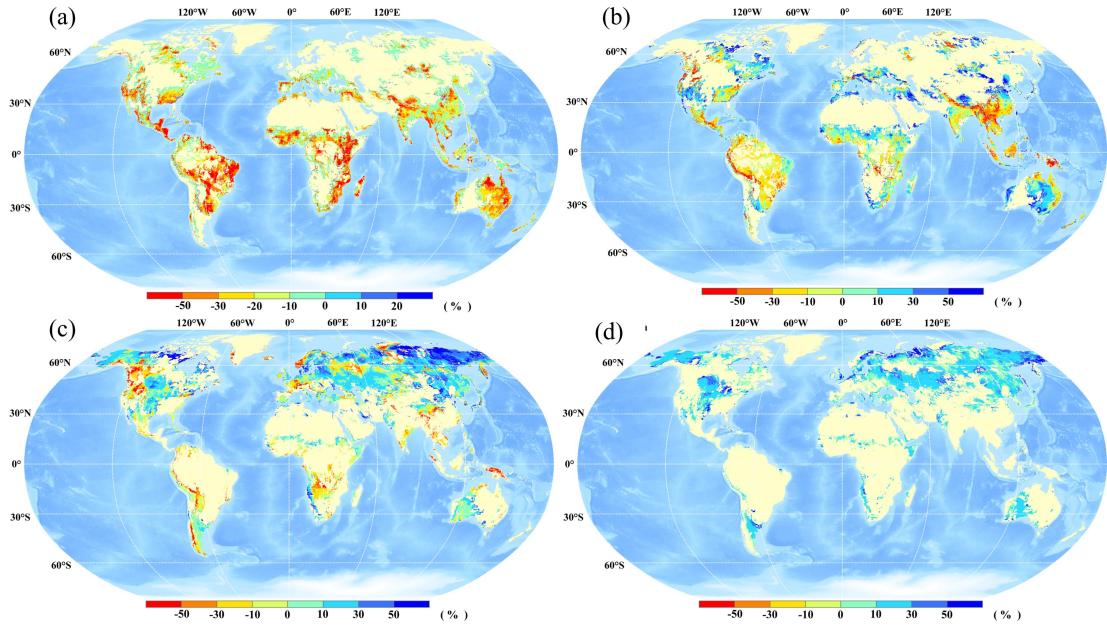
355

356 Fig. 8 The synergistic effect of soil moisture and soil temperature. Fig. 8a is the area where soil
 357 moisture and soil temperature are both promoted or weakened, and Fig. 8b is the main control area
 358 of different factors

359 Combined with the change trend of SM, we further found that in areas where SM
 360 dries out and SM is negatively affected, the average relative contribution of STMP is
 361 1.60%, and in 55.06% of this area, the influence of STMP is positive ([Fig. 9a, b](#)),
 362 indicating that in more than half of the area, STMP may offset part of the negative
 363 impact of soil moisture drying on the CS of clay minerals.

364 However, combined with the change trend of soil temperature, we found that in
 365 areas where soil warming and the contribution of STMP is positive, the average
 366 relative contribution of SM is 20.21%, and these areas are mainly concentrated in the
 367 high latitudes of the northern hemisphere ([Fig. 9c, d](#)), the reason further validates the
 368 findings in the previous section that warming can lead to soil warming and soil

369 wetness in high latitudes, which to some extent accelerates the chemical weathering
370 process of minerals in the soil.



371
372 Fig. 9 The offset effect of soil moisture and soil temperature. The Fig. 9a shows the relative
373 contribution of soil moisture in the soil desiccation area, the Fig. 9b shows the relative
374 contribution of soil temperature in areas where the soil is drying and soil moisture is showing a
375 negative contribution, the Fig. 9c shows the relative contribution of soil temperature in the soil
376 warming area, the Fig. 9d shows the relative contribution of soil moisture when the soil is warming
377 up and the soil temperature is positively contributing to the area.

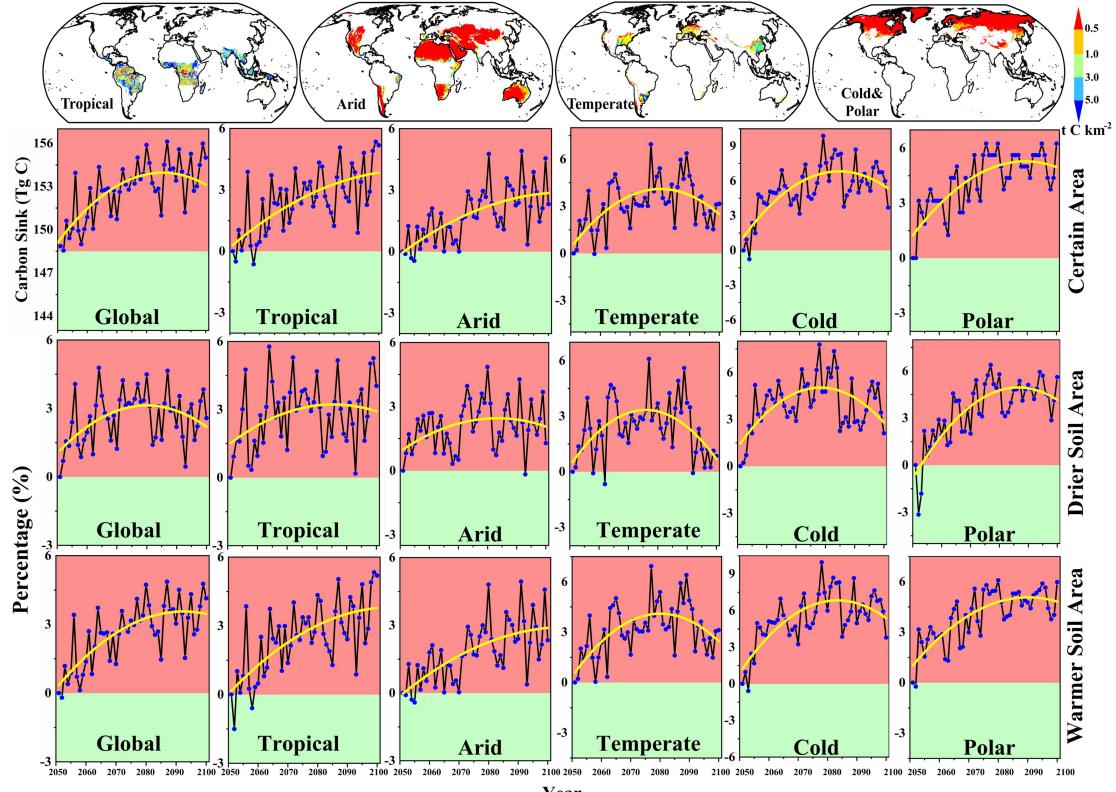
378 ***Trends of CS in future scenarios***

379 Referring to the SM and STMP data based on the medium emission scenario
380 RCP4.5, we used the PROFILE model to extend the global CS of clay minerals from
381 2051 to 2100 ([Fig. 10](#)). Under the RCP4.5 scenario, the average weathering CS of the
382 world's major clay minerals is about $0.15 \text{ Pg C yr}^{-1}$, which means that after
383 government intervention, the high concentration of carbon dioxide emitted in the
384 future can still increase the capacity of CS in clay minerals by about 36%. In general,
385 CS of clay minerals shows a trend of rapid increase first, and then a slow decrease,
386 however, there are slightly differences in different climate zones. For example, in

387 tropical with the largest amount of CS, the trend is basically a continuous increase,
388 but in temperate areas, CS shows a trend of first increasing and then decreasing.

389 In addition, combined with the future trends of SM and STM, we extracted the
390 percentage changes of CS in drier and warmer soil regions. The results show that the
391 trend of CS in these regions is basically the same as that of CS that have not been
392 partitioned, which shows that under the future medium emission scenario, the CS will
393 be affected by soil desiccation and soil warming slightly less than the current impact.
394 The reason for this phenomenon may be related to the high concentration of carbon
395 dioxide emitted. For example, in the study of Roelandt et al (2005), it was found that
396 high concentration of CO₂ can increase the cations released by minerals after
397 weathering by about 20%. Furthermore, the positive impact of soil warming on
398 weathering may also offset the negative impact caused by the reduction of SM, for
399 example, based on future climate simulation experiments, Akselsson found that by
400 2050 the temperature rising can increase the weathering rates of soil minerals by up to
401 30% (2016).

402 High-concentration carbon emissions not only mean an increase in climate
403 warming, but also accelerating the growth rate of vegetation (Chen et al., 2021),
404 which in turn will indirectly participating in the weathering reaction of minerals by
405 affecting the hydrological environment of soil. The changes in these environmental
406 factors in the future will have a certain degree of impact on the weathering process of
407 minerals. However, due to the limitation of data resolution, we did not conduct a
408 detailed analysis and only estimating the change trend of CS in clay minerals in the
409 future, and the above results suffice to illustrate the potential of clay minerals in the
410 soil as a stable carbon-sequestered method in the context of possible intensified
411 climate warming in the future.



412

413 Fig. 10 Time series of CS and its trends under the medium emissions scenario RCP4.5 in the
 414 different areas. The top four maps represent the distribution of global clay minerals' carbon sink
 415 flux in different climatic regions. The line chart shows the changing trends of carbon sink in the
 416 world and in different climatic regions. Among them, except that the unit of the ordinate of the
 417 first line chart is carbon sink, the rest are expressed as the percentage of changes in carbon sink in
 418 other years compared with the carbon sink in 2050.

419 ***The uncertainty of this study***

420 Given that the chemical formula of the mineral itself is extremely complex, our
 421 study only refers to the typical chemical formula of the mineral determined in the
 422 laboratory by the predecessors ([Goddéris et al., 2006](#)). However, the actual situation is
 423 very complicated and the weathering evolution stage between different minerals is not
 424 considered, for example, the mica may be weathered first to form illite and then
 425 subjected to secondary weathering. So our thinking is consistent with estimating the
 426 CS response of the weathering of the bedrock, that is, the estimated CS after the
 427 minerals are completely reacted ([Hartmann et al., 2009](#)). Therefore, the results

428 estimated in this paper are theoretically potential CS.

429 Although the content distribution map of clay-grade minerals has been widely
430 cited ([Ito and Wagai, 2017](#)), under natural conditions, this set of data still has certain
431 uncertainties. By summarizing the uncertainty of the mineral content on the surface
432 and bottom grids, we know that the average value on the pixel is about 36.52%.
433 Therefore, the estimated CS based on this data set should also have a considerable
434 degree of uncertainty.

435 The PROFILE model itself has a high degree of uncertainty. Even under the
436 premise of ensuring the quality of the input data, the error of the model can reach
437 about 40% ([Jönsson et al., 1999](#)), when compared with the soil weathering results of
438 other models, the error can reach 33%~300% ([Futter et al., 2012](#)), which is mainly
439 affected by the input data. Although some remote sensing and hydrological data are
440 used in this paper, there are still uncertainties. For example, the error of
441 evapotranspiration data in the land surface model assimilation data product (GLDAS)
442 in East Asia can be as high as 40% ([Khan et al. , 2018](#)), and the SM is lower than the
443 observed value ([Deng et al., 2020](#)). In addition, due to the low resolution of some data,
444 downscaling methods such as resampling have to be used, and it is found that the
445 average error in our data preprocessing process is about 6.22% through calculations,,
446 and these errors are all errors generated in the data production process.

447 In addition, we only estimated the relative contribution of soil moisture and soil
448 temperature, but we know from [Fig. 8b](#) that the main controlling factors in most areas
449 have not yet been determined. Factors such as vegetation, CO₂ concentration, acid
450 deposition, human activities and other factors will affect the process of soil
451 weathering. For example, vegetation can control soil weathering by controlling soil
452 hydrology, and the high CO₂ concentration of deep soil can increase the weathering

453 rate by at least 15% (Roelandt et al., 2010), moreover, nitrogen deposition can cause
454 global soil acidification and destroy the weathering reaction of carbonate minerals in
455 the soil, causing the soil to release a large amount of CO₂ (L. Deng et al., 2020; Raza
456 et al., 2020), therefore, all of the above factors will play a key role in the weathering
457 of minerals in the soil.

458 In summary, due to the constraints of the original model, input data, influencing
459 factors, etc., there are still a lot of uncertainties in our research results. However, after
460 comparing with sites, other models, and other research results, we found that the
461 results of CS in this paper have a certain degree of credibility. The greatest value of
462 this paper is that we have produced a dataset of clay mierals in soil with high temporal
463 and spatial resolution, and clarifying the contribution of the CS produced by the
464 weathering of clay minerals to the carbon imbalance. These data and research results
465 will provide more novel and unique ideas for re-understanding the soil inorganic
466 carbon pool and improving the carbon cycle framework.

467 **Materials**

468 ***Clay minerals***

469 This article refers to the latest global spatial distribution map of clay-grade
470 minerals released by Ito and Wagai (2017), this data set compiles data including the
471 content of the main clay minerals in the soil and their soil bulk density, etc. According
472 to the classification of soil genesis, the percentages of minerals including kaolinite,
473 smectite, vermiculite, chlorite, illite, mica and quartz were estimated, respectively.
474 This data set is divided into two sets of surface and deep data, with spatial resolution
475 ranging from 2' to 2°, and has been widely cited in the field of geochemical simulation
476 (Charbonnier et al., 2020; Dabat et al., 2019).

477 ***Soil attribute dataset***

478 The soil moisture (SM), soil temperature (STMP) and other data used to simulate
479 the CS of clay minerals come from the GLAS Noah 2.0 and GLAS Noah 2.1 data sets
480 released by Global Land Data Assimilation System (LDAS-NASA) of NASA. Among
481 them, the GLAS Noah 2.0 data set was used from 1970 to 1999, and the GLAS Noah
482 2.1 data set (<https://ldas.gsfc.nasa.gov/gldas/>) was used after 2000. This set of data is
483 divided into 0-10cm, 10-40cm, 40-100cm, 100-200cm and other different soil layers
484 according to the soil depth from top to bottom, with a spatial resolution of 0.25°, it
485 mainly combines satellite and ground observation data, and uses advanced data
486 assimilation and ground simulation technologies, which has been widely cited in the
487 field of chemical weathering simulation research on rocks (Li et al., 2018; Gong et al.,
488 2021).

489 In addition, since the weathering reaction of clay minerals is limited by soil
490 temperature, in frozen soil areas, too low temperature will inhibit the weathering
491 process. Therefore, this paper estimates the percentage of frozen soil at the pixel scale
492 from 1970 to 2018 based on soil temperature. The calculation formula is as follows:

$$493 \quad r = \frac{\sum_{i=1}^{12} con(T_{soil} > 0,1,0)}{12} \quad (1)$$

494 Where, r is the annual percentage of frozen soil with %, and Tsoil is the soil
495 temperature with K.

496 The soil bulk density data of the surface and bottom layers comes from the
497 global clay mineral and its attribute database compiled by Ito and Wagai (2017).
498 Compared with a single soil bulk density data, this data can finely describe the
499 difference in soil bulk density between the surface and bottom soils. Among them, the
500 average bulk density of the top soil is 1.32 g/cm³, and the average bulk density of the
501 bottom soil is 1.38 g/cm³.

502 The soil covered by rock weathering often results in uneven soil thickness due to
503 differences in lithology and local hydrothermal conditions. If the CS of global clay
504 minerals is estimated at a thickness of 2m, the estimation result will inevitably be too
505 high. To this end, this paper uses the absolute soil thickness data released by Oak
506 Ridge National Laboratory ([Pelletier et al., 2016](#)) to correct the rate of mineral
507 weathering in areas with different soil thicknesses.

508 ***Climate attribute dataset***

509 As an important indicator of CO₂ concentration in soil, this paper uses
510 evapotranspiration data to estimate the CO₂ partial pressure characteristics in the
511 atmosphere/soil month by month ([Li et al., 2019; Zeng et al., 2019](#)). The formula is as
512 follows (2):

$$513 \quad \log(pCO_2) = -3.7 + 2.09 \times (1 - e^{-0.00172 \times ET}) \quad (2)$$

514 Here, ET is evapotranspiration with mm. The monthly scale evapotranspiration
515 data from 2001 to 2014 comes from the MOD16/ET data set released by the
516 University of Montana. Based on the 8-day resolution, monthly scale data with a
517 global resolution of 0.05°×0.05° was produced. The evapotranspiration data for other
518 years comes from the GLDAS Noah 2.0 and Noah 2.1 data sets provided by NASA.

519 In addition, in order to calculate the spatial differentiation of CS of clay minerals,
520 we also used the latest Köppen climate classification map ([Beck et al., 2018](#)), this
521 climate zoning data represents the current (1980-2016) climate background, with a
522 spatial resolution ranging from 0.0083° to 0.5°.

523 ***Site data***

524 This study refers to the soil weathering rate in the Critical Load Database of
525 Nitrogen and Sulfur Deposition (NCLD) in the United States to compare with our
526 estimated results of the weathering rate of clay-grade minerals, this database was

527 developed by the United States Scientific Committee on Atmospheric Deposition
528 Critical Load ([Lynch, J.A et al., 2020](#)), based on a set of databases shared by the
529 United States Atmospheric Deposition Program (NADP). Among them, the key
530 element used to evaluate the critical load is the weathering rate of minerals in the soil.
531 In this database, a number of measured data including soil weathering rate, nitrogen
532 and sulfur deposition are recorded in detail. The details of the site are shown in [Figure](#)
533 [7a.](#)

534 In order to verify our estimation results, we also used the WITCH model to
535 recalculate, and used the site data in the ISRIC database ([Batjes et al., 2020](#)), this data
536 set includes key parameters such as soil texture, soil temperature, and soil moisture on
537 the site.

538 ***Methods***

539 ***POFILE Model***

540 We used the steady-state soil geochemical profile model (PROFILE) to estimate
541 the weathering rate and CS of global clay minerals in soil. The PROFILE model is a
542 steady-state mechanical soil biogeochemical model ([Sverdrup and Warfvinge, 1993](#)),
543 it can be used not only to estimate the rate of soil weathering, but also to determine
544 the critical load of acid deposition in soil and surface water ([Erlandsson Lampa et al.,](#)
545 [2020](#)). This model couples independent reactions between minerals and different
546 media, making it not only able to simulate the reaction process of minerals with a
547 single medium, but also combining environmental data such as soil temperature and
548 soil moisture, making the scale expansion is possible ([Whitfield et al., 2018](#)). In
549 addition, the model also considers the influence of time on the weathering process,
550 and can simulate the weathering rate under a long time sequence. The core formula is
551 as follows:

552

$$R_w CO_2 = \sum_{i=1}^{\text{minerals}} R_i \times A_w \times x_i \times \phi \times Z \quad (3)$$

553 Where, $R_w CO_2$ is the total weathering rate of the chemical reaction of clay
 554 minerals in which carbonic acid participates, and R_i is the weathering reaction rate of
 555 the i -th type of clay minerals (Equations 4, 5), A_w is the reaction contact area of the
 556 mineral (Equation 6), x_i is the content of the i -th clay mineral in the soil, ϕ is the soil
 557 moisture saturation (Equation 7), and Z is the soil thickness with m. The weathering
 558 kinetics module of the PROFILE model takes into account the reaction of soil
 559 minerals with water, CO_2 , exogenous acid H^+ and organic acids. The formula for
 560 estimating the reaction rate of minerals with CO_2 at $8^\circ C$ is as follows:

561

$$R_{i-8} = K_{CO_2} \times \left(\frac{P_{CO_2}}{1 + f_{CO_2} \times (P_{CO_2} + P_{CO_2\text{Limit}})} \right)^{nCO_2} \quad (4)$$

562 Where, R_{i-8} is the mineral reaction rate measured at $8^\circ C$ in the laboratory, and
 563 K_{CO_2} is the mineral dissolution kinetic constant, and its value is often related to the
 564 temperature (Equation 8), P_{CO_2} is the partial pressure of carbon dioxide in the
 565 atmosphere/soil (Equation 2), $P_{CO_2\text{limit}}$ is the limit of carbon dioxide absorption,
 566 nCO_2 is the reaction order, and f_{CO_2} is the suppression constant 1. After temperature
 567 correction, the following formula can be used to estimate the mineral reaction rate at
 568 room temperature:

569

$$R_i = e^{R_{i-8} \times A_w \times \left(\frac{1}{T_{lab}} - \frac{1}{T_{soil}} \right)} \quad (5)$$

570 Here, A is the Arrhenius index factor, with a fixed value of 3600K, T_{lab} is the
 571 laboratory temperature of $8^\circ C$ (281.15 k), and T_{soil} is the soil temperature in K. The
 572 mineral reaction contact area A_w is mainly related to the content of clay, sand, and silt
 573 in the soil ([Erlandsson et al., 2016](#)). The estimation formula is as follows:

574 $A_w = 8 \times X_{clay} + 2.2 \times X_{slit} + 0.3 \times X_{sand}$ (6)

575 The moisture saturation ϕ in the soil is related to the soil water content, soil
576 density, etc., and can be estimated in combination with the following formula:

577
$$\phi = \frac{\theta \times \rho_{soil}}{\rho_{soil} - \rho_i + \theta \times \rho_{water}}$$
 (7)

578 Among them, θ is soil moisture, ρ_{soil} is soil density, ρ_{water} is water density, and ρ_i
579 is soil bulk density.

580 **Trend analysis**

581 We use time as the independent variable, and the year-by-year carbon sink flux at
582 the pixel as the dependent variable, and combined the unary linear regression and
583 F-test to estimate the change slope of the weathering CS of the world's major clay
584 minerals from 1970 to 2018 ([Li et al. al., 2019; Zhang et al., 2021](#)). Based on this
585 result, we discuss the change trend of the weathering CS of clay minerals in the past
586 half century. The core calculation formula of the unary linear regression is as follows:

587
$$\theta = \frac{n \times \sum_{i=1}^n (i \times \gamma_i) - \sum_{i=1}^n i \sum_{i=1}^n \gamma_i}{n \times \sum_{i=1}^n (i^2) - (\sum_{i=1}^n i)^2}$$
 (8)

588 Where, θ is the evolution slope of the CS γ on the pixel, and $\theta > 0$ indicates that
589 the CS γ has an overall increasing trend within the current research time limit. i is the
590 current year, n is the overall period of the study, and γ_i is the value of the CS γ in the
591 i -th year at a certain pixel.

592 **Separation the effect of SM and STMP**

593 In order to quantify and distinguish the effects of the two key driving factors, soil
594 moisture and soil temperature, on the CS of the main clay mineral weathering, this
595 paper sets up two independent experiments to simulate the impact of soil moisture

596 changes and soil temperature changes on CS (Liu et al., 2019), in the simulation
597 driven by changes in soil moisture, by limiting the soil temperature, CO₂
598 concentration and the percentage of frozen soil in 1970, and combined with the soil
599 moisture data from 1970 to 2018 year by year, the carbon sink CS_(SM) based on soil
600 moisture is estimated, that is, the simulated CS_(SM) from 1970 to 2018 is driven by soil
601 moisture. In the same way, the CS_(STMP) driven by the soil temperature is simulated.

602 In order to further analyze the relative contribution of soil moisture/soil
603 temperature/residual factors to the CS of clay minerals, we uses the time as the
604 dependent variable, and use actual CS/the CS driven by SM/the CS driven by STMP
605 as independent variables to do a linear regression analysis to estimate changes in CS
606 under different scenarios. Taking the CS driven by soil moisture (CS_(SM)) as an
607 example, the calculation formula is as follows:

$$608 \quad y = \alpha_0 + \alpha_1 t + \tau \quad (9)$$

609 Among them, y is the CS based on SM simulation with t C km⁻² (CS_(SM)); t is the
610 corresponding year, α_0 is the intercept, α_1 represents the change trend of CS driven by
611 SM ($\Delta CS_{(SM)}$), τ is the residual error.

612 In the same way, the actual weathering CS of clay minerals and the change trend
613 of CS driven by soil temperature can be obtained, which are represented by ΔCS and
614 $\Delta CS_{(STMP)}$, respectively. In addition, the change trend of clay mineral weathering CS
615 (ΔRes), which is not driven by the remaining factors explained by soil moisture and
616 soil temperature, is calculated by subtracting the sum of $\Delta CS_{(STMP)}$ and $\Delta CS_{(SM)}$ from
617 the change trend of actual CS ΔCS . Finally, the relative contributions of soil moisture,
618 soil temperature, and remaining factors are calculated by the following formulas:

$$619 \quad Contr.S.Moi = \frac{|\Delta CS_{SM}|}{|\Delta CS_{SM}| + |\Delta CS_{ST}| + |\Delta CS_{RES}|} \times 100 \% \quad (10)$$

620

$$Contr.S.Tem = \frac{|\Delta CS_{ST}|}{|\Delta CS_{SM}| + |\Delta CS_{ST}| + |\Delta CS_{RES}|} \times 100\% \quad (11)$$

621

$$Contr.S.Res = \frac{|\Delta CS_{RES}|}{|\Delta CS_{SM}| + |\Delta CS_{ST}| + |\Delta CS_{RES}|} \times 100\% \quad (12)$$

622 **Data availability.** All relevant data are available upon request from the authors.

623

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