Abiotic carbonate dissolution traps carbon in a semiarid desert

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It is generally considered that desert ecosystems release CO2 to the atmosphere, but recent studies in drylands have shown that the soil can absorb CO2 abiotically. However, the mechanisms and exact location of abiotic carbon absorption remain unclear. Here, we used soil sterilization, 13CO2 addition, and detection methods to trace 13C in the soil of the Mu Us Desert, northern China. After 13CO2 addition, a large amount of 13CO2 was absorbed by the sterilised soil, and 13C was found enriched both in the soil gaseous phase and dissolved inorganic carbon (DIC). Further analysis indicated that about 79.45% of the total 13C absorbed by the soil was trapped in DIC, while the amount of 13C in the soil gaseous phase accounted for only 0.22% of the total absorbed 13C. However, about 20.33% of the total absorbed 13C remained undetected. Our results suggest that carbonate dissolution might occur predominantly, and the soil liquid phase might trap the majority of abiotically absorbed carbon. It is possible that the trapped carbon in the soil liquid phase leaches into the groundwater; however, further studies are required to support this hypothesis.

Drylands (arid and semiarid desert ecosystems) cover about 41% of the global terrestrial surface1 and release a large amount of carbon via soil CO2 flux2,3. However, a net uptake of carbon is observed in some desert ecosystems, especially during nighttime, such as the Mojave Desert4,5, a playa of the Great Basin Desert6 and the Chihuahuan Desert7 in North America, the Gurbantunggut Desert8,9 and the Mu Us Desert10,11 in Asia and a dry valley in Antarctica12. Although the carbon absorptions in these cases are various, it may also play an important role in the terrestrial carbon cycle because of the great areal extent of drylands10,13. The anomalous and variable soil carbon accretion suggests that the soil carbon cycle in desert ecosystems is quite complicated14,15.

Although it is known that CO2 absorption by the soil is induced by abiotic processes and is relevant to some ambient factors (e.g. temperature, soil water content, and air pressure)8,10,12,16,17, previous studies failed to reveal the underlying mechanism of the abiotically absorbed CO2 by the soil, which is essential to better understand CO2 absorption by the soil in drylands18. Applying the 13C isotope tracer method to undisturbed soil, we previously found that the majority of absorbed carbon might conserve in the soil solid phase19; however, we could not demonstrate the mechanism of abiotic CO2 absorption and sequestration because of the influence of biotic processes. Hence, further studies on the abiotically absorbed carbon is required.

It is surmised that abiotic carbonate dynamic is a major contributor of CO2 absorption by the soil10,20 and the involved abiotic processes can be expressed as21:

\[
\text{CaSiO}_3 + 2\text{H}_2\text{O} + \text{CO}_2 \rightarrow \text{Ca(HCO}_3\text{)}_2 (\text{carbonate weathering}) \tag{2}
\]

Since Ca-silicate weathering is an extremely slow process, it can be ignored at the diel or annual scale22, while the process of carbonate weathering may help to better explain CO2 absorption by the soil. However, the reactions are reversible. Whether carbonate dissolution can predominately occur and induce the abiotic atmospheric CO2 absorption by soil in drylands, have not been proven directly by any previous study currently. Here, we hypothesized that carbonate dissolution can induce the abiotic atmospheric CO2 absorption by desert soils. To test the hypothesis, we used the sterilization, leaching method and 13CO2 isotope tracer technique to investigate

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the abiotic soil CO2 exchange and 13C abundance of soil liquid and vapour phase in the desert soil of the Mu Us Desert, northern China.

Results

The amount of abiotic 13CO2 absorption. After 12CO2 was replaced in the chamber, the initial 13CO2 amount in the atmosphere of the chamber was 446 μmol. After 12 h (throughout the nighttime), the amount of the final atmospheric 13CO2 in the chamber was 149 μmol. The amount of 13CO2 absorption by the sterilised soil reached to 297 μmol.

13C abundance (δ13C) in the leaching solution with and without 13CO2 addition. The 13C abundances and amounts in the leaching solution that removed by frequent leaching are illustrated in Fig. 1. The amount of the collected leaching solution of sterilised soil with 13CO2 addition did not differ remarkably from that without 13CO2 addition at each leaching operation, and both approximately equal to 1,000 ml since the second leaching operation. However, the leachate δ13C (δ13C of DIC in leachate) of sterilised soil with 13CO2 addition showed intense enrichment (although leachate δ13C decreased with the increasing number of leaching operations, all the values of leachate δ13C were positive, ranging from 564.48 ± 399.21% to 2.85 ± 1.31%), compared with the leachate δ13C of sterilised soil without 13CO2 addition (all the values of leachate δ13C were negative, ranging from −11.40 ± 3.64% to −21.21 ± 1.89%).

13C abundance (δ13C) in the soil gaseous phase with and without 13CO2 addition. The abundances of 13C of soil air with and without 13CO2 addition are showed in Fig. 2. The δ13C of soil air without 13CO2 addition was −9.3 ± 2.26% after soil-atmosphere CO2 exchange in the chamber. While the δ13C of soil air with 13CO2 addition was enriched after the CO2 exchange in the chamber (599.2 ± 192.87%).

Absorbed 13CO2 tracing. The total 13C in the soil leaching solution was calculated at about 235.98 μmol, and the 13C in the soil air was 0.66 μmol. Thus, the accumulated amount of absorbed 13C in the soil liquid phase accounted for 79.45% of the total absorbed 13C, while the amount in the soil gaseous phase accounted for only 0.22%. In addition, about 20.33% of the total absorbed 13C remained undetected.

Discussion

79.45% of the labelled carbon (13CO2) was found in dissolved inorganic carbon (H13CO3−), indicating that the majority of CO2 absorbed abiotically by the soil during nighttime is trapped in the soil liquid phase and converted into dissolved inorganic carbon (DIC) at this study site. A recent study also showed that the soil liquid phase in a saline/alkaline desert contains amounts of newly formed DIC, and the formation of DIC may be associated with atmospheric CO2.23. However, in our previous study, it was found that the majority of absorbed 13C was fixed in the solid phase of undisturbed soil19. The discrepancy between the two studies could be attributed to the influence of biotic processes. Verrecchia and Verrecchia24 reported that bacteria and fungi could accelerate the formation of needle fibre calcite (CaCO3). The impact of rhizosphere processes may also be important in the formation of carbonate. For instance, the roots absorb soil water and can cause bicarbonate decomposition producing carbonate25. In this study, the effect of biotic processes was excluded by soil sterilisation; therefore, the carbon absorbed by the soil could be only trapped abiotically. Moreover, soil water participation and carbon transportation may be another reason for the discrepancy. These may be overlooked in the previous study. Ignoring the interference of biotic processes, conserving in the soil solid phase may be a medium status for the carbon absorbed by soil from atmosphere according to the results in this study. Therefore, this work is an extension of the previous one.

Variations in the ambient temperature can affect in DIC formation and accumulation26–28. The high temperature results in DIC decomposition29, while the low temperature induces DIC accumulation30. In this study, the
ambient temperature was low during nighttime. As a result, the process of carbonate dissolution may predominately occur in the soil. In subsoil, the temperature may be persistently lower than topsoil (i.e. the sampled soil in this study)\(^3\). More DIC may be formed and accumulated without decomposition. Previous studies showed that the residence time of carbon in pedogenic DIC could be over three orders of magnitude higher than that of soil organic matter\(^31–34\). Therefore, the newly formed DIC may not easily turn into CaCO\(_3\) abiotically until the ambience is changed. The accumulation of DIC may induce a continuous consumption of soil CO\(_2\); inorganic consumption of soil CO\(_2\) through carbonate dissolution can create a pressure gradient of CO\(_2\) between the soil and the atmosphere, and then the pressure gradient can induce atmospheric CO\(_2\) to be pumped into the soil to replenish soil CO\(_2\) (as the ingredient in carbonate dissolution)\(^10\).

Although the results showed that the soil could absorb CO\(_2\) from the atmosphere and then trap large amounts of the atmospheric carbon by forming and accumulating DIC at this study site, we still cannot declare that this is an important soil carbon sink, because we have no direct evidence to support that DIC is sequestrated in the soil at a long-term scale. For the formation of soil carbonate induced by biotic processes, CO\(_2\), as a by-product, releases back to the atmosphere\(^34\) and overshadows the abiotic CO\(_2\) absorption\(^35\). As a result, carbonate in the soil solid phase may not be the main destination of absorbed atmospheric CO\(_2\) at the diel or annual scale. The hydrosphere beneath the soil may be a potential outlet of trapped carbon. Figure 1 shows that the vertical migration of soil water occurs easily, and the majority of the absorbed \(^{13}\)C in DIC can be washed out by four leaching operations. These results suggested that DIC may finally transport to the aquifer, regardless of being a long-term process. Similarly, Ma et al.\(^{18}\) and Li et al.\(^{23}\) reported that soil DIC could be leached into the aquifer by rainfall or snow glacier melt and irrigation water or river water in arid ecosystems. Moreover, Walvoord et al.\(^{36}\) found that the accumulated nitrate in the subsoil might also transport into the groundwater because of long-term leaching in xeric ecosystems. In the study site, groundwater is recharged mainly by rainfall\(^37\) and has almost no hydrological connection to rivers and oceans. If the trapped carbon transports deeply into the aquifer, the low and constant temperature and alkalinity of groundwater may convert the aquifer into a reservoir of carbon beneath the desert soil. However, further studies are required to test whether the newly formed DIC can be leached from the soil to the hydrosphere.

Although this is a case study in the Mu Us Desert, the results can provide the direct and effective support to the hypothesis that carbonate dissolution can induce the abiotic atmospheric CO\(_2\) absorption by desert soils. It is undeniable that the amount of carbon trapped in DIC may be overestimated, because the deionized water used for leaching can inevitably dissolve some of the solid fraction (soil carbonate) in theory. To diminish the overestimation, we multiply leached with much less deionized water each time instead of one time leaching with the large amount of deionized water (the duration of each leaching operation is much shorter and the amount of deionized water used for leaching is much less each time). Therefore, the overestimation may be small, and its influence should be slight.

After \(^{13}\)CO\(_2\) addition and exchange in the chambers for 12 h, \(^{13}\)C in the soil gaseous phase was enriched (Fig. 2); however, the amount of newly conserved \(^{13}\)C in the soil gaseous phase was much less than that fixed by the soil liquid phase (accounted for about 0.28% of the fixed \(^{13}\)C in the soil liquid phase). These results were in
agreement with those reported in our previous study\textsuperscript{19}, suggesting that the soil gaseous phase may only serve as the connecting medium between the atmosphere and the soil liquid phase. CO\textsubscript{2} in the soil gaseous phase may easily transport upwards to the atmosphere or downwards to the alkaline soil solution.

About 20.33\% of the total absorbed 13C remained undetected, probably because it was stored in the soil solid phase. Since the leaching operations started at 9:00 A.M., soil temperature might already begin to rise, and a part of DIC probably turned into CaCO\textsubscript{3} and released as CO\textsubscript{2}. The results from abiotic soil CO\textsubscript{2} flux measurement showed that soil could absorb atmospheric CO\textsubscript{2} during nighttime and release CO\textsubscript{2} during daytime, somewhat supporting our hypothesis. It is notable that even though DIC can turn into CaCO\textsubscript{3} during daytime, the amount of lapsed DIC may be small, because the abiotic net carbon exchange between the soil and the atmosphere is usually negative at the diel scale (negative net carbon exchange represents carbon absorption by soil)\textsuperscript{11}. Therefore, at the diel scale, most of the abiotically absorbed carbon may also be trapped in the soil liquid phase.

Methods

Site description. The study site is located on the southwestern fringe of the Mu Us Desert (37°42′ N, 107°13′ E; 1,509 m above sea level), northern China. It has a temperate continental monsoon climate with a mean annual temperature of 7.6 °C, mean annual solar radiation of 1.4 × 10\textsuperscript{5} J cm\textsuperscript{−2}, and mean annual wind speed of 3 m s\textsuperscript{−1} (prevailing northwest wind). The frost-free period lasts around 128 d. The mean annual precipitation is 275 mm (1954–2013), mainly occurring in August and September\textsuperscript{35}. The soil type is Aripsamment (derived from aeolian sand; soils with high CaCO\textsubscript{3} content tend to be salinised). The soil (0–20-cm depth) comprises 94.8\% sand, 4.5\% silt, and 0.7\% clay and has a pH of 8.6\textsuperscript{10}. Soil bulk density is 1.54 g cm\textsuperscript{−3} and soil porosity 42\%\textsuperscript{19}. The study site is sparsely vegetated by Artemisia ordosica, Astragalus mongolicum, Salix psammophila, and Tamarix chinensis (canopy coverage ≤ 30%).

\textsuperscript{13}CO\textsubscript{2} tracing. To test whether carbonate dissolution occurs and detect the location of the abiotically absorbed carbon in the soil, an improved leaching operation (adopted in Li et al.,\textsuperscript{23} for extraction DIC) and \textsuperscript{13}CO\textsubscript{2} tracing experiment (used in Liu et al.,\textsuperscript{19} for carbon tracing) were carried out in September and October 2014 using iron\textsuperscript{13}CO\textsubscript{2} exchange chambers (25 cm in length; 25 cm in width; 70 cm in height; 0.1 cm in thickness; Fig. 3). Three quadrate steel soil collars (25 cm in length; 25 cm in width; 20 cm in height; 0.2 cm in thickness) were placed randomly in the study site with a 2-cm wall to be exposed above the soil surface in order to allow the installation of the iron\textsuperscript{13}CO\textsubscript{2} exchange chambers. The soil within the collars was equilibrated with its surrounding for 24 h to minimise the disturbing effect. Subsequently, soil samples were collected and sterilised as described by Xie et al.\textsuperscript{8}. The sterilised soil was placed \textit{in situ} and equilibrated with its surrounding for 17 h to minimise the disturbing effect.

Each \textsuperscript{13}CO\textsubscript{2} exchange chamber (with backslash opened) was immersed in NaOH solution (5 mol L\textsuperscript{−1}) up to about 2 cm for 2 h to remove \textsuperscript{12}CO\textsubscript{2}. Then, two \textsuperscript{13}CO\textsubscript{2} exchange chambers (with backslash closed) were installed onto the collars (pushed 15 cm deep into the soil), while the third collar was used as a control. We injected 10 ml \textsuperscript{13}CO\textsubscript{2} (concentration > 99.99\%) into each chamber, opened the backslash, and allowed the soil to exchange \textsuperscript{13}CO\textsubscript{2} for 12 h.

The backslash of each chamber was closed after the \textsuperscript{14}CO\textsubscript{2} exchange, the gaseous samples (140 ml) in each chamber were collected with an aluminium foil gas-collecting bag, and the \textsuperscript{14}CO\textsubscript{2} exchange chambers were removed. In order to increase accuracy, a polyvinyl chloride (PVC) sheet (25 cm in length; 25 cm in width; 25 cm in height; 0.1 cm in thickness) was used to divide the sterilised soil into two parts; one for soil CO\textsubscript{2} extraction (25 cm in length; 5 cm in width; 18 cm in height) and the other for leaching (25 cm in length; 20 cm in width; 18 cm in height). An aluminium foil gas-collecting bag (200 ml) was used to reserve the extracted soil air (140 ml) from the one part of sterilised soil, and a PVC cylinder (15.3 cm outer diameter; 15 cm inner diameter; 25 cm in height) was inserted into the other part of sterilised soil to sample the soil column for leaching. The cylinder was sealed at the top and bottom with base plates and transported to the laboratory.

![Figure 3. Structure of the \textsuperscript{13}CO\textsubscript{2} exchange chamber. Neutral glass cement was used to seal the hole immediately after the backslash was opened or closed during the process of \textsuperscript{13}CO\textsubscript{2} addition to ensure the chamber sealability.](image-url)
A layer of gauze (100-mesh) was placed under the bottom plate to prevent mud from leaching. Then, the bottom plate was removed, and the sterilised soil cylinder was leached with 1,000 ml deionised water. The sterilised soil with 13CO2 addition was leached 10 times (about 10 times of leaching may leach out almost all the 13C according to the study of Ma et al.18), while the control (without 13CO2 addition) 4 times. We assumed that the abundance of 13C would not change considerably, because the natural 13C abundance of the soil was low38, and thus, the different number of leaching operations could not influence the comparison between the results from sterilised soil with and without 13CO2 addition. After each leaching operation, we weighed the total volume of leaching solution (Vleach) and collected leaching solution samples of 30 ml using brown glass bottles. During all these processes, the ambient temperature was below 10 °C.

The number of replications for the sterilised soil with and without 13CO2 addition was 18 and 9, respectively. The flow of 13CO2 tracing is shown in Fig. 4. The δ13C and CO2 concentration of all gaseous samples were measured by a carbon dioxide isotope analyser (CCIA-EP 912-0003; Los Gatos Research, Mountain View, CA, USA) and the δ13C of all liquid samples (δ13C of DIC in the soil liquid phase) by an isotope mass spectrum analyser (Finnigan MAT253 Gas Bench-IRMS; Thermo Fisher Scientific Inc., Waltham, MA, USA) as described by Liu et al.19.

Measurements of soil volumetric water content (VWC) and bicarbonate radical concentration (C_{DIC}). VWC was monitored from 20:00 to 7:00 the following day at a depth of 10 cm using the ECH2O system (LI-COR, Lincoln, NE, USA) with five Em50R sensors placed near the soil collars, and data were logged every 1 h. After collecting liquid samples (without 13CO2 addition), C_{DIC} was measured using the conventional method of acid base titration39.

Data processing and analysis. The absorbed 13CO2 within the soil collar was calculated as follows:

\[
N = \frac{1000 \times V_{\text{initial}}}{R_t} - \frac{C_{\text{final}} \times V_{\text{chamber}}}{R_t},
\]

where \(N\) is the total absorbed 13CO2 (μmol), \(V_{\text{initial}}\) is the initial volume of 13CO2 (10 ml), \(C_{\text{final}}\) is the final 13CO2 concentration (μmol mol\(^{-1}\)), \(V_{\text{chamber}}\) is the volume of the chamber exposed above the soil surface (L), and \(R_t\) is the molar volume of gas (L mol\(^{-1}\)).

The 13CO2 absorbed into the soil vapour phase was calculated as follows:

\[
N_{\text{vapour}} = 5 \times (N_{\text{added}} - N_{\text{control}})
\]

\[
N_{\text{added}} = C_{\text{CO2}} \times \frac{V_{\text{soil air}}}{R_t \times 1000} \times \frac{(1000 + \delta^{13}C_{\text{added}}) \times R_{st}}{1000 + (1000 + \delta^{13}C_{\text{added}}) \times R_{st}}
\]

\[
N_{\text{control}} = C_{\text{CO2}} \times \frac{V_{\text{soil air}}}{R_t \times 1000} \times \frac{(1000 + \delta^{13}C_{\text{control}}) \times R_{st}}{1000 + (1000 + \delta^{13}C_{\text{control}}) \times R_{st}}
\]

\[
V_{\text{soil air}} = \frac{V_{\text{soil}}}{1000} \times P_{\text{soil}} = \frac{V_{\text{soil}}}{1000} \times \text{VWC},
\]

where \(N_{\text{vapour}}\) is the amount of substance of absorbed 13C in the soil vapour phase (μmol), \(N_{\text{added}}\) and \(N_{\text{control}}\) are the amounts of substance of 13C in the soil vapour phase with and without 13CO2 addition, respectively, \(C_{\text{CO2}}\) is the concentration of CO2 in the soil vapour phase (μmol mol\(^{-1}\)), \(R_t\) is the molar volume of gas (L mol\(^{-1}\)), \(V_{\text{soil air}}\) is the volume of soil air (L), \(\delta^{13}C_{\text{added}}\) and \(\delta^{13}C_{\text{control}}\) are the δ13C values of the soil vapour phase with and without 13CO2 addition, respectively, \(R_{st}\) is the stable isotope ratio in the reference standard40, \(V_{\text{soil}}\) is the volume of soil sample (25 cm × 5 cm × 18 cm), \(P_{\text{soil}}\) is the soil porosity, and VWC is the mean value of soil volumetric water content during the measurement period.

The 13CO2 absorbed into the soil liquid phase was calculated as follows:

...
\[ N_{\text{liquid}} = \frac{5 \times (N'_{\text{added}} - N'_{\text{control}})}{4} \]  

\[ N'_{\text{added}} = \sum C_{\text{DIC}} \times \frac{V'_{\text{soil}} \times B}{1000} \times \frac{(1000 + 13C'_{\text{added}}) \times R_d}{1000 + (1000 + 13C'_{\text{control}}) \times R_d} \]  

\[ N'_{\text{control}} = \sum C_{\text{DIC}} \times \frac{V'_{\text{soil}} \times B}{1000} \times \frac{(1000 + 13C'_{\text{control}}) \times R_d}{1000 + (1000 + 13C'_{\text{control}}) \times R_d} \]

where \( N_{\text{liquid}} \) is the amount of substance of absorbed \(^{13}\text{C} \) in the soil liquid phase (μmol), \( N'_{\text{added}} \) and \( N'_{\text{control}} \) are the amounts of substance of \(^{13}\text{C} \) in the soil liquid phase with and without \(^{13}\text{CO}_2 \) addition, respectively. \( C_{\text{DIC}} \) is the soil bicarbonate radical concentration in the soil liquid phase (μmol kg\(^{-1}\)). \( V'_{\text{soil}} \) is the volume of soil sample (25 cm × 20 cm × 18 cm), \( B \) is bulk density (g cm\(^{-3}\)), and \( 13C'_{\text{added}} \) and \( 13C'_{\text{control}} \) are the \(^{13}\text{C} \) values of the soil liquid phase with and without \(^{13}\text{CO}_2 \) addition, respectively.

A one-way analysis of variance was used to test the differences in \(^{13}\text{C} \) and DIC. VWC and \( V_{\text{leachate}} \) between the chambers and samples. All statistical analyses were performed using MATLAB 7.12.0.635 (The Math Works, Natick, MA, USA).

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Author Contributions

All authors commented on manuscript at all stages. K.Y.F. and Y.Q.Z. developed the concept of the paper and supervised the study; K.Y.F. and Z. L. performed the in situ measurements of the soil volumetric water content and bicarbonate radical concentration and the \(^{13}\text{CO}_2\) tracing experiment; K.Y.F. analysed all the data and wrote the manuscript; Y.Q.Z., J.B.L. B.W., and S.G.Q. worked on each version of the manuscript and significantly contributed in its final structure and presentation.

Additional Information

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