SCIENTIFIC REPORTS

Received: 08 December 2015 Accepted: 09 March 2016 Published: 29 March 2016

OPEN 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 predominately, 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 flux^{2,3}. However, a net uptake of carbon is observed in some desert ecosystems, especially during nighttime, such as the Mojave Desert^{4,5}, a playa of the Great Basin Desert⁶ and the Chihuahuan Desert⁷ in North America, the Gurbantunggut Desert^{8,9} and the Mu Us Desert^{10,11} 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 drylands^{10,13}. The 'anomalous' and variable soil carbon accretion suggests that the soil carbon cycle in desert ecosystems is quite complicated^{14,15}.

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 CO_2 by the soil, which is essential to better understand CO_2 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 soil^{10,20} and the involved abiotic processes can be expressed as²¹:

$$CO_2 + 2H_2O + CaAl_2Si_2O_8 \rightarrow Al_2Si_2O_5(OH)_4 + CaCO_3$$
 (Ca-silicate weathering) (1)

$$CO_2 + H_2O + CaCO_3 \xrightarrow[carbonate precipitation]{} Ca(HCO_3)_2$$
 (carbonate weathering) (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 CO_2 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 CO_2 absorption by desert soils. To test the hypothesis, we used the sterilization, leaching method and ${}^{13}CO_2$ isotope tracer technique to investigate

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the abiotic soil CO_2 exchange and ^{13}C abundance of soil liquid and vapour phase in the desert soil of the Mu Us Desert, northern China.

Results

The amount of abiotic ${}^{13}CO_2$ **absorption.** After ${}^{12}CO_2$ was replaced in the chamber, the initial ${}^{13}CO_2$ amount in the atmosphere of the chamber was 446 µmol. After 12 h (throughout the nighttime), the amount of the final atmospheric ${}^{13}CO_2$ in the chamber was 149 µmol. The amount of ${}^{13}CO_2$ absorption by the sterilised soil reached to 297 µmol.

¹³C abundance (δ^{13} C) 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 δ^{13} C (δ^{13} C of DIC in leachate) of sterilised soil with ¹³CO₂ addition showed intense enrichment (although leachate δ^{13} C decreased with the increasing number of leaching operations, all the values of leachate δ^{13} C were positive, ranging from 564.48 ± 399.21% to 2.85 ± 1.31%), compared with the leachate δ^{13} C of sterilised soil without ¹³CO₂ addition (all the values of leachate δ^{13} C were negative, ranging from $-11.40 \pm 3.64\%$ to $-21.21 \pm 1.89\%$).

¹³C abundance (δ^{13} C) 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 δ^{13} C of soil air without ¹³CO₂ addition was $-9.3 \pm 2.26\%$ after soil-atmosphere CO₂ exchange in the chamber. While the δ^{13} C of soil air with ¹³CO₂ addition was enriched after the CO₂ exchange in the chamber (599.2 ± 192.87%).

Absorbed ¹³**CO**₂ **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 ($^{13}CO_2$) was found in dissolved inorganic carbon ($H^{13}CO_3^{-}$), 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 CO_2^{23} . However, in our previous study, it was found that the majority of absorbed ^{13}C 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 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 accumulation^{26–28}. The high temperature results in DIC decomposition²⁹, while the low temperature induces DIC accumulation¹². 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)³⁰. 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 CaCO₃ abiotically until the ambience is changed. The accumulation of DIC may induce a continuous consumption of soil CO₂: inorganic consumption of soil CO₂ through carbonate dissolution can create a pressure gradient of CO₂ between the soil and the atmosphere, and then the pressure gradient can induce atmospheric CO₂ to be pumped into the soil to replenish soil CO₂ (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, CO₂, as a by-product, releases back to the atmosphere³⁴ and overshadows the abiotic CO_2 absorption³⁵. 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 ¹³C 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 deserts. 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 rainfall³⁷ 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 CO2 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 rather than 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 study¹⁹, suggesting that the soil gaseous phase may only serve as the connecting medium between the atmosphere and the soil liquid phase. CO_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 CaCO₃ and released as CO₂. The results from abiotic soil CO₂ flux measurement showed that soil could absorb atmospheric CO₂ during nighttime and release CO₂ during daytime, somewhat supporting our hypothesis. It is notable that even though DIC can turn into CaCO₃ 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)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 1.4×10^5 J cm⁻², and mean annual wind speed of 3 m s⁻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 September³⁵. The soil type is Aripsamment (derived from aeolian sand; soils with high CaCO₃ 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 8.6¹⁰. Soil bulk density is 1.54 g cm⁻³ and soil porosity 42%¹⁹. The study site is sparsely vegetated by *Artemisia ordosica, Astragalus mongolicum, Salix psammophila*, and *Tamarix chinensis* (canopy coverage \leq 30%).

¹³**CO**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.*²³; for extraction DIC) and ¹³CO₂ tracing experiment (used in Liu *et al.*¹⁹; for carbon tracing) were carried out in September and October 2014 using iron¹³CO₂ 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 ¹³CO₂ 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.*⁸. The sterilised soil was placed *in situ* and equilibrated with its surrounding for 17 h to minimise the disturbing effect.

Each ${}^{\bar{13}}CO_2$ exchange chamber (with backsplash opened) was immersed in NaOH solution (5 mol L⁻¹) up to about 2 cm for 2 h to remove ${}^{12}CO_2$. Then, two ${}^{13}CO_2$ exchange chambers (with backlash 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 ${}^{13}CO_2$ (concentration > 99.99%) into each chamber, opened the backsplash, and allowed the soil to exchange ${}^{13}CO_2$ for 12 h.

The backsplash of each chamber was closed after the ${}^{13}CO_2$ exchange, the gaseous samples (140 ml) in each chamber were collected with an aluminium foil gas-collecting bag, and the ${}^{13}CO_2$ 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 CO₂ 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 4. Flow diagram of ¹³CO₂ tracing. Red line indicates CO₂ exchange in the chamber.

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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 ${}^{13}CO_2$ addition was leached 10 times (about 10 times of leaching may leach out almost all the ${}^{13}C$ according to the study of Ma *et al.*¹⁸), while the control (without ${}^{13}CO_2$ addition) 4 times. We assumed that the abundance of ${}^{13}C$ would not change considerably, because the natural ${}^{13}C$ 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 ${}^{13}CO_2$ addition. After each leaching operation, we weighed the total volume of leaching solution ($V_{leachate}$) 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 ${}^{13}CO_2$ addition was 18 and 9, respectively. The flow of ${}^{13}CO_2$ tracing is shown in Fig. 4. The $\delta {}^{13}C$ 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 $\delta {}^{13}C$ of all liquid samples ($\delta {}^{13}C$ 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 13 CO₂ within the soil collar was calculated as follows:

$$N = \frac{1000 \times V_{\text{initial}}}{R_{\text{t}}} - \frac{C_{\text{final}} \times V_{\text{chamber}}}{R_{\text{t}}},\tag{3}$$

where N is the total absorbed ¹³CO₂ (µmol), V_{initial} is the initial volume of ¹³CO₂ (10 ml), C_{final} is the final ¹³CO₂ concentration (µmol mol⁻¹), V_{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:

$$N_{\rm vapour} = 5 \times (N_{\rm added} - N_{\rm control}) \tag{4}$$

$$N_{\rm added} = C_{\rm CO_2} \times \frac{V_{\rm soil\,air}}{R_{\rm t} \times 1000} \times \frac{(1000 + \delta^{13} C_{\rm added}) \times R_{\rm st}}{1000 + (1000 + \delta^{13} C_{\rm added}) \times R_{\rm st}}$$
(5)

$$N_{\rm control} = C_{\rm CO_2} \times \frac{V_{\rm soil\,air}}{R_{\rm t} \times 1000} \times \frac{(1000 + \delta^{13} C_{\rm control}) \times R_{\rm st}}{1000 + (1000 + \delta^{13} C_{\rm control}) \times R_{\rm st}}$$
(6)

$$V_{soil\ air} = \frac{V_{soil}}{1000} \times P_{soil} - \frac{V_{soil}}{1000} \times VWC,\tag{7}$$

where N_{vapor} is the amount of substance of absorbed ¹³C in the soil vapour phase (µmol), N_{added} and N_{control} are the amounts of substance of ¹³C in the soil vapour phase with and without ¹³CO₂ addition, respectively, C_{CO_2} 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 δ^{13} C values of the soil vapour phase with and without ¹³CO₂ addition, respectively, R_{st} is the stable isotope ratio in the reference standard⁴⁰, V_{soil} is the volume of soil sample (25 cm × 5 cm × 18 cm), P_{soil} is the soil porosity, and *VWC* is the mean value of soil volumetric water content during the measurement period.

The¹³CO₂ absorbed into the soil liquid phase was calculated as follows:

$$N_{\text{liquid}} = \frac{5 \times (N'_{\text{added}} - N'_{\text{control}})}{4}$$
(8)

$$N'_{\rm added} = \sum C_{\rm DIC} \times \frac{V'_{\rm soil} \times B}{1000} \times \frac{(1000 + \delta^{13}C'_{\rm added}) \times R_{\rm st}}{1000 + (1000 + \delta^{13}C'_{\rm added}) \times R_{\rm st}}$$
(9)

$$N_{\rm control}' = \sum C_{\rm DIC} \times \frac{V_{\rm soil}' \times B}{1000} \times \frac{(1000 + \delta^{13} C_{\rm control}') \times R_{\rm st}}{1000 + (1000 + \delta^{13} C_{\rm control}') \times R_{\rm st}}$$
(10)

where N_{liquid} is the amount of substance of absorbed ¹³C in the soil liquid phase (µmol), N'_{added} and N'_{control} are the amounts of substance of ¹³C in the soil liquid phase with and without ¹³CO₂ addition, respectively, C_{DIC} is the soil bicarbonate radical concentration in the soil liquid phase (µmol kg⁻¹), V'_{soil} is the volume of soil sample (25 cm × 20 cm × 18 cm), *B* is bulk density (g cm⁻³), and $\delta^{13}C'_{\text{added}}$ and $\delta^{13}C'_{\text{control}}$ are the δ^{13} C values of the soil liquid phase with and without ¹³CO₂ addition, respectively.

A one-way analysis of variance was used to test the differences in δ^{13} C, C_{DIC} , VWC and V_{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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Acknowledgements

The study was supported by the National Natural Science Foundation of China (No. 31170666) (NFSC). We would like to thank the staff of the Yanchi Research Station for their assistance with field and laboratory work. We are grateful to the anonymous reviewers for their valuable comments that helped us improve this paper.

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 CO₂ 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

Competing financial interests: The authors declare no competing financial interests.

How to cite this article: Fa, K. *et al.* Abiotic carbonate dissolution traps carbon in a semiarid desert. *Sci. Rep.* **6**, 23570; doi: 10.1038/srep23570 (2016).

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