

# **A downward CO<sub>2</sub> flux seems to have nowhere to go**

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## **Abstract.**

Recent studies have suggested that deserts, which are a long-neglected region in global carbon budgeting, have strong downward CO<sub>2</sub> fluxes and might be a significant carbon sink. This finding, however, has been strongly challenged because neither the reliability of the flux measurements nor the exact location of the fixed carbon has been determined. This paper shows, with a full chain of evidence, that there is indeed strong carbon flux into saline/alkaline land in arid regions. Based on continuous measurement of net ecosystem CO<sub>2</sub> exchange (NEE) from 2002 to 2012 (except for 2003), the saline desert in western China was a carbon sink for nine out of the ten years, and average yearly NEE for the ten years was  $-25.00 \pm 12.70 \text{ g Cm}^{-2} \text{ y}^{-1}$ . Supporting evidence for the validity of these NEE estimates comes from the close agreement of NEE values obtained from the chamber and eddy-covariance methods. After ruling out the possibility of changes in C stored in plant biomass or soils, the C uptake was found to

1 be leached downwards into the groundwater body in the process of groundwater  
2 fluctuation: rising groundwater absorbs soil dissolved inorganic carbon (DIC), and  
3 falling groundwater transports the DIC downward. Horizontal groundwater flow may  
4 send this DIC farther away and prevent it from being observed locally. This process has  
5 been called “passive leaching” of DIC, in comparison with the active DIC leaching that  
6 occurs during groundwater recharge. This passive leaching significantly expands the  
7 area where DIC leaching occurs and creates a literally “hidden” carbon sink process  
8 under the desert. This study tells us that when a downward CO<sub>2</sub> flux is observed, but  
9 seems to have nowhere to go, it **does not necessarily mean** that the flux measurement is  
10 unreliable. By looking deeper and farther away, a place and a process may be found  
11 that are “hidden” underground.

12

## 13 **1 Introduction**

14 Consistent enrichment of atmospheric CO<sub>2</sub> concentration from pre-industrial times  
15 due to human activities (Stocker *et al.*, 2013), together with global climate change, have  
16 focused much scientific and public attention on carbon cycling (Siegenthaler and  
17 Sarmiento, 1993; Chapin *et al.*, 2006; Houghton, 2007). One of the most important  
18 issues is the imbalance between CO<sub>2</sub> released from anthropogenic activities and  
19 documented terrestrial or oceanic sinks, which is known as the “missing sink” or the  
20 “residual terrestrial sink” in the global carbon budget (Houghton, 2007, 2013;  
21 Friedlingstein *et al.*, 2010). Substantial studies have been carried out to locate this  
22 residual sink, but the uncertainties remain large (Schimel *et al.*, 2001; Houghton, 2007),

1 especially in territorial ecosystems where the carbon flux and the source-sink processes  
2 vary greatly in magnitude and mechanism from region to region, such as from  
3 rainforest to desert (Schimel et al., 2001; Heimann and Reichstein, 2008).

4 With characteristics such as low productivity, low carbon exchange, and limited  
5 available data (Hastings et al., 2005), deserts remain one of the most under-represented  
6 ecosystems in carbon cycle syntheses (Raich and Potter, 1995; Wohlfahrt et al., 2008;  
7 Cable et al., 2011), in spite of their huge area that accounts for one-quarter of the earth's  
8 land surface (Reynolds, 2001). However, in recent years, a few studies have proposed  
9 that desert regions, which have long been considered negligible in global carbon  
10 budgeting, have strong downward fluxes into the ground and therefore might be a  
11 significant carbon sink (Jasoni et al., 2005; Stone, 2008; Wohlfahrt et al., 2008; Xie et  
12 al., 2008). The net carbon uptake in the Mojave Desert of the United States, based on  
13 flux-tower measurements, was reported to be around  $100 \text{ g C m}^{-2} \text{ y}^{-1}$  for a three-year  
14 period (Jasoni et al., 2005; Wohlfahrt et al., 2008). Similar results were reported in the  
15 Gurbantonggut Desert in northwestern China based on closed-chamber measurements  
16 (Stone, 2008; Xie et al., 2008). These findings have understandably attracted scientific  
17 and public attentions (Stone, 2008; Schlesinger et al., 2009; Serrano-Ortiz et al., 2010)  
18 but remain controversial (Schlesinger et al., 2009), in part because there is no  
19 agreement about the exact location of the fixed carbon (Schlesinger et al., 2009). Some  
20 researchers have argued that assimilation by cryptobiotic crust organisms (such as  
21 lichens, mosses, and cyanobacteria) or crassulacean acid metabolism (CAM) plants  
22 may account for a significant portion of C uptake (Hastings et al., 2005; Wohlfahrt et al.,

1 2008), while others have argued that the fixed carbon is associated with precipitation or  
2 dissolution of carbonate (Xie et al., 2008; Shanhun et al., 2012). However, these  
3 interpretations have faced great challenges: cryptobiotic crust organisms and CAM  
4 plants are neither sufficiently active nor extensive to explain such an unusually high  
5 rate of uptake ( Schlesinger et al., 2009; Serrano-Ortiz et al., 2010), not to mention that  
6 the negative flux ( $\text{CO}_2$  down into the soil) occurred at night. Formation and  
7 accumulation of  $\text{CaCO}_3$  is constrained by the sources of calcium derived from  
8 atmospheric deposition and rock weathering (Schlesinger, 1985; Monger and Gallegos,  
9 2000). Hence, it is understandable that authors have been asked to reconfirm these flux  
10 measurements and to present a logical answer to the question of where the carbon goes  
11 (Schlesinger et al., 2009).

12 Recent publications have highlighted the need to include other processes than  
13 photosynthesis and respiration, such as losses of carbon as volatile compounds or  
14 dissolved carbon (Chapin et al., 2006; Heimann and Reichstein, 2008), in carbon  
15 budgeting. Leaching of dissolved inorganic carbon (DIC) through the hydrological  
16 cycle is an important component of regional or global carbon budgets (Kindler et al.,  
17 2011; Tobias and Bohlke, 2011). However, in (semi-)arid regions, evaporative demand  
18 significantly exceeds rainfall on average (Noy-Meir, 1973), and surface runoff is  
19 constrained or limited, hampering direct groundwater recharge (de Vries and Simmers,  
20 2002) and ultimately resulting in lack of conventional leaching. Instead, groundwater  
21 level fluctuations occur over various time scales (e.g. from daily to seasonal to decadal,  
22 Healy and Cook, 2002; Amiaz et al., 2011), especially for desert-oasis ecotones which

are usually located at the foot of mountains or along rivers. Here, we hypothesized that rises and falls in the groundwater table may carry DIC downward into the groundwater body, forming a carbon sink process that is literally hidden under the desert. If proven to be true, this would mean the discovery of a piece of the “hidden loop in the carbon cycle” (Stone, 2008) and might be able to explain the seemingly unexplainable CO<sub>2</sub> absorption into desert (Hastings et al., 2005; Wohlfahrt et al., 2008; Xie et al., 2009).

## 2 Methods

### 2.1 Site description

The carbon flux (soil CO<sub>2</sub> flux and tower flux) measurements were conducted at the Fukang Station of Desert Ecology, Chinese Academic of Sciences, which is located at the southern periphery of the Gurbantonggut Desert, in the hinterland of the Eurasian continent (87°56'E, 44°17'N and 475 m a.s.l.). The climate is temperate continental arid, with hot summers and cold winters. Mean annual temperature is 5°C–7°C, and mean annual precipitation is 164 mm. Soils are silty clay-loam textured with high salinity and alkalinity [electrical conductivity (EC) > 4 dS m<sup>-1</sup>, pH > 8.2 for a soil solution at a soil/water ratio of 1:5] and are classified as Solonchaks in the FAO/UNESCO soil-classification system. Average plant cover is approximately 17% and is dominated by the phreatophytic desert shrub *Tamarix ramosissima*, but also includes sporadic *Reaumuria songarica*, *Nitraria tangutorum*, and various annual herbaceous plants (primarily *Salsola nitraria* Pall.).

### 2.2 Eddy-covariance measurement and data processing

1       An eddy-covariance system was used to measure the fluxes of CO<sub>2</sub>, water vapor,  
2       and sensible heat over the saline desert continuously from 2002 to 2012, except for  
3       2003 due to instrument malfunction. The system consisted of a three-dimensional  
4       ultrasonic anemometer-thermometer (STA-5055, Kaijo Corporation, Tokyo, Japan) and  
5       an open infrared gas (CO<sub>2</sub>/H<sub>2</sub>O) analyzer (LI-7500, LI-COR, USA). The instruments  
6       were placed on a tower at a height of 3 m, which is about 1–1.5 m above the plant  
7       canopy, with data recorded at a frequency of 10 Hz. Two soil heat-flux plates  
8       (HFP01SC, Hukseflux, The Netherlands) were placed at 2 cm below the soil surface,  
9       and additional meteorological instruments were installed to measure soil and air  
10      temperature, humidity, photosynthetically active photon flux density (PPFD), and  
11      incoming and outgoing total radiation (CM21F, Kipp & Zonen, The Netherlands). The  
12      flux-measurement system and calculation methods are described in more detail in Liu et  
13      al. (2012). Following the sign convention in flux research, positive values represents  
14      net carbon gain by the atmosphere and loss from the ecosystem; conversely, negative  
15      values means loss of CO<sub>2</sub> from the atmosphere and gain by the ecosystem. This  
16      convention is equally applicable to research on **soil CO<sub>2</sub> flux**.

17      Half-hourly flux values were excluded from further analysis if the data were  
18      anomalous or showed spurious spikes (Liu et al., 2012) which might have been derived  
19      from sensor maintenance, rain or snow events, power failure, or instrument malfunction.  
20      Details of the procedure were as follows: first, reject values collected during rain and  
21      snow events, belonging to incomplete 30-minute data sets, or taken at times of  
22      instrument malfunction; second, remove the net ecosystem exchange (NEE) data where

1 the H<sub>2</sub>O concentrations differed by more than 30% from those estimated from relative  
2 humidity data (Rogiers et al., 2005); third, filter the steady-state data (both in daytime  
3 and nighttime), using a threshold - the friction velocity ( $u^*$ ) lower than 0.1 m·s<sup>-1</sup> (Xu  
4 and Baldocchi, 2004; Reichstein et al., 2005) - to determine whether the atmosphere  
5 provided sufficient turbulent mixing (Aubinet et al., 2001; Baldocchi, 2003). Over the  
6 2002–2012 period (except for 2003), roughly 30%–40% of the data were discarded  
7 according to these exclusion principles and therefore required gap-filling. For short  
8 time periods (e.g., one hour or less), missing fluxes were estimated by linear  
9 interpolation; for large gaps (e.g., days) during the daytime, the relationship between  
10 PPFD and good CO<sub>2</sub> flux was used to fill the gaps using a Michaelis-Menten  
11 rectangular hyperbolic equation (Ruimy et al., 1995; Falge et al., 2001). Gap-filling  
12 during night and winter, when the CO<sub>2</sub> flux is respiration only, was accomplished using  
13 Lloyd-Taylor function between respiration and soil temperature at 5 cm depth (Lloyd and  
14 Taylor, 1994).

15 An uncertainty analysis, accounting for both random and systematic errors  
16 arising from random sampling error, the gap-filling procedure (Liu et al., 2012), and  
17  $u^*$ -filtering, was conducted to obtain confidence intervals for the annual carbon budgets.  
18 For random sampling error, the worst-case estimation method proposed by Morgenstern  
19 et al. (2004, Equation 11) was used. As for the uncertainties in the gap-filling procedure,  
20 which resulted in a complete NEE time series, these were estimated by creating  
21 artificial gaps in the gap-filling series with added noise (Giasson et al., 2006); more  
22 details were described in Liu et al. (2012). Systematic error associated with the choice

of the  $u^*$  threshold was estimated by comparison of the annual NEE calculated using the standard method ( $u^*=0.1 \text{ m}\cdot\text{s}^{-1}$ ) with that derived using a different  $u^*$  threshold (Morgenstern et al., 2004). For different  $u^*$  thresholds during the night, the relationship between soil temperature at the 5-cm depth and nighttime NEE ( $NEE_{\text{nighttime}}$ ) was determined, and all nighttime data below the chosen  $u^*$  threshold were modeled accordingly, providing an annual cumulative estimate of NEE. The errors were as shown in Table 1.

The growing period (gp) was defined as extending from the day assimilation (photosynthesis) first occurred to the day when ecosystem carbon assimilation stopped, from about DOY (day of year) 121 to DOY 290. Correspondingly, the non-growing period (ngp) contained the remaining days of the year.

### 2.3 Cross-check the reliability of the observed NEE

Generally, chamber method is a direct and more accurate method in estimation of ecosystem  $\text{CO}_2$  flux, which provides a way to cross-check the reliability of the NEE observed from eddy covariance system. Conceptually, NEE is representing the balance of ecosystem gross primary productivity (GPP) and ecosystem respiration ( $R_{\text{eco}}$ ). NEE integrates the carbon metabolism of all biota. Namely:

$$NEE = GPP - R_{\text{eco}} = GPP - R_{\text{leaf}} - R_{\text{stem}} - R_{\text{soil}} = NPP_{\text{canopy}} - R_{\text{soil}}$$

where ( $GPP - R_{\text{leaf}}$ ) represented net carbon assimilation or release by leaves, which can be estimated by scaling up leaf net photosynthesis rate with synchronous leaf area index (LAI). For the stem respiration ( $R_{\text{stem}}$ ), results of preliminary experiments showed that



the respiration rate is so low in terms of contribution to the total ecosystem respiration (no more than 2%, unpublished data) that it can be reasonably ignored. Therefore, in this context,  $NPP_{canopy}$  is equal to GPP minus  $R_{leaf}$ . Soil  $CO_2$  flux ( $R_{soil}$ ) was scaled up by cumulating the relative contribution from different measurement site, e.g., the site under canopy and in the bare area. Methods used to measure leaf net photosynthesis rates, LAI dynamic and soil  $CO_2$  flux are briefly described below.

### 2.3.1 Measurements of leaf net photosynthesis rate and LAI

Diurnal course of leaf net photosynthesis rate was measured according the protocols described in Xu and Li (2006) in 2009. Each diurnal measurement was carried out hourly from sunrise to sunset over six clear and sunny days evenly cross the growing season (DOY 141, 163, 189, 216, 236 and 260). Five plants were selected and two sets of leaves were measured from each plant. LAI was derived from combined leaf (assimilating branch) relative growth rate with accumulated leaf biomass as described in Zou et al. (2010). Totally, 15 branches, in the footprint area of the eddy-covariance instrument, were labeled. Branch areas were monitored by photograph at interval of two weeks and were converted to branch biomass by defining the relationship between surface area and dry mass. After that, relative growth rate was calculated between every two sequential measurements. At the end of the growing season, all the leaves in a 50 m by 3 m transect, which was located in the center of footprint area, were destructive sampled. On the basis of the accumulated leaf biomass and relative growth rate, seasonal LAI dynamic was determined.

### 2.3.2 Chamber measurements of $R_{soil}$

1        Soil CO<sub>2</sub> flux ( $R_{\text{soil}}$ ) was measured independently in 2009 (from DOY 125 to 283)  
2        by an LI-8150 system (Lincoln, Nebraska, USA) with six long-term monitoring  
3        chambers. To address the question of whether chamber measurements adequately  
4        represent soil CO<sub>2</sub> flux for comparison with eddy-covariance measurements of total  
5        ecosystem respiration at a large scale, soil collars were distributed along a transect from  
6        a site near a plant stem to the interplant space to capture individual-based variation.  
7        Measurements were made on each of the chambers every half-hour; details of the  
8        system design are given by Ma et al. (2012).

#### 9        **2.4 Calculating gross primary productivity (GPP) and ecosystem respiration ( $R_{\text{eco}}$ )**

10        Ecosystem respiration ( $R_{\text{eco}}$ ) at the study site as well as the gross primary  
11        production (GPP) of the ecosystem were obtained through an algorithm (Reichstein et  
12        al., 2005) that first establishes a relationship between soil temperature and  $R_{\text{eco}}$  from  
13        nighttime data (e.g., the Lloyd-Taylor function, Lloyd and Taylor, 1994) and then uses  
14        this relationship to extrapolate  $R_{\text{eco}}$  from night-time to day-time. GPP was then obtained  
15        by subtracting NEE from  $R_{\text{eco}}$ .

#### 16        **2.5 Calculating the $R_{\text{soil}}/R_{\text{eco}}$ ratio**

17        For each effective measurement of CO<sub>2</sub> flux data, the average  $R_{\text{soil}}/R_{\text{eco}}$  ratio was  
18        calculated. Half-hourly nighttime ( $\text{PPFD} < 5 \mu\text{mol m}^{-2} \text{s}^{-1}$ ) eddy-covariance estimates of  
19        the NEE were selected, binned by day and averaged. These estimates of mean daily  
20        nighttime  $R_{\text{eco}}$  were paired with the corresponding  $R_{\text{soil}}$  to calculate a ratio.

#### 21        **2.6 Investigation of biomass and soil carbon storage**

22        A detailed survey of plants and soils at the study site, including living biomass and

1 organic and inorganic carbon storage in the soil, was carried out in 1989 (the year that  
2 the Fukang Station of Desert Ecology was established) and 2009 respectively. Details of  
3 soil-sampling methods and quadrat investigations of plant biomass were described in Li  
4 et al.(2010). Soil total carbon and inorganic carbon were measured using a Total  
5 Organic Carbon/Total Nitrogen analyzer (Multi C/N 3100, Analytik Jena, Germany),  
6 and the difference between the two values was taken to represent the organic carbon  
7 content.

## 8 **2.7 Laboratory soil column leaching experiment**

9 To test whether leaching processes could occur due to fluctuations in the  
10 groundwater table, a laboratory soil column leaching experiment was carried out. Soil  
11 samples were collected from three random locations near the flux-tower. For each  
12 location, soil cores were collected at intervals of 30 cm from the surface to the depth of  
13 2.1 m. Soil core samples were air-dried and samples from same depth were mixed  
14 before transferring to columns.

15 A schematic representation of the soil columns is shown in Fig. 1. Briefly, soil  
16 columns (PVC cylinders, height 280 cm, inner diameter 20 cm) were packed with  
17 stratified (at intervals of 30 cm) soil samples at the corresponding depth according to  
18 the bulk densities in the field. The cylinders were sealed at the top and bottom with  
19 base plates furnished with an 8-mm drainpipe on each side. Above the bottom plate, a  
20 layer of gauze (60-mesh) was placed to prevent mud from leaching away (Fig. 1), and  
21 20-cm-thick quartz sand was placed on the surface of the gauze to accelerate water  
22 infiltration or drainage and to prevent disturbance to soil profiles during water injection.

1 For the same purpose, another 20-cm-thick layer of quartz sand placed on the top of the  
2 soil surface to aid in even irrigation distribution. To equalize instantaneous pressure  
3 variations within the soil columns and let leachate flow out smoothly, six air vents were  
4 designed and all connected to a 10L air bag (Fig. 1) with silicon tubes. The air vents  
5 were located at 30-cm intervals from 30 cm to 180 cm. To monitor water level in the  
6 soil columns, a water-level mark was included at the midpoint of the cylinder (at a  
7 position of 140 cm above the bottom). In addition, five observation holes (diameter 8  
8 cm) were drilled at heights of 100, 140, 170, 200, and 240 cm above the bottom and  
9 sealed by transparent resin plates, which enabled observation of color changes in the  
10 soil profile.

11 Once the soil columns were prepared, they were fixed on a metal shelf, which was  
12 a convenient method to collect the leachate from the bottom and to simulate  
13 groundwater-table fluctuations by manually moving the water tank up and down using a  
14 pulley fixed on the shelf. To eliminate the influence of dissolved carbon in the irrigation  
15 water on the carbon isotopic character of the leachate, air-free shallow groundwater (pH  
16 8.16,  $EC=3.7\text{ dS}\cdot\text{m}^{-1}$ ) was obtained from a 20-m-deep well adjacent to the saline desert  
17 (groundwater level was about 4.5 m). The dissolved air in the water was removed by a  
18 vacuum degasser.

19 To simplify the isotope-marking process,  $^{13}\text{CO}_2$  (0.3L, 99.3% purity) was  
20 dissolved into air-free water (2L) at room temperature in advance and then slowly  
21 poured into the soil column using the top tube. This way, dissolved  $^{13}\text{CO}_2$ , representing  
22  $\text{CO}_2$  derived from the atmosphere or from soil respiration, was added to the soil

1 columns (infiltrated to limited depth far from the bottom of the soil column). After this,  
2 the top tube was blocked with a small clip. The washing solution (air-free water) was  
3 applied to the bottom of the soil column by lifting up a water drum connected to the  
4 bottom tube until water level reached the point where the labeled CO<sub>2</sub> (dissolved <sup>13</sup>CO<sub>2</sub>)  
5 could reach with infiltration. Then the bottom tube was switched to connect to a 10L  
6 vacuum airbag, and the leachate was collected by gravity over eight consecutive hours.  
7 The volume of leachate was recorded, and the δ<sup>13</sup>C of the leachate was measured by an  
8 Isotope Ratio Mass Spectrometer (IRMS) (FinniganMAT 253, Thermo Finnigan,  
9 Bremen, Germany). This washing procedure was repeated until the δ<sup>13</sup>C of the leachate  
10 solution returned to its initial level (15 times overall). Leaching experiments were  
11 replicated with three columns.

## 12 **2.8 Energy balance**

13 To assess the reliability of the eddy-covariance measurements, an energy-balance  
14 analysis was carried out. 30-min values of sensible heat flux ( $H$ ) plus latent heat flux  
15 ( $LE$ ) were compared with net radiation ( $R_n$ ) minus soil heat flux ( $G$ ), with results listed  
16 in Table 1. These data showed an incomplete energy-balance closure: the slopes of the  
17 fitted lines between  $H+LE$  and  $R_n-G$  were all less than one, which means that the  
18 eddy-covariance measurements underestimated  $H+LE$ . This energy-closure deficit  
19 might be explained by measurement error related to sensor separation, sampling  
20 frequency response, or interference from tower or instrument-mounting structures  
21 (Twine et al., 2000; Liu et al., 2012). In addition, energy stored under the plant canopy  
22 and in the layer of soil between the surface and the soil heat flux plates was not

considered (Twine et al., 2000; Wilson et al., 2002).

### 3 Results

#### 3.1 Net ecosystem exchange of carbon in the saline desert

Figure 2 shows obvious annual variations in cumulative NEE ( $P < 0.01$ ), which ranged from  $-91 \text{ g C m}^{-2} \text{ y}^{-1}$  to  $14 \text{ g C m}^{-2} \text{ y}^{-1}$ . For nine out of the ten years, NEE was negative (or downward into the desert) (Fig. 2a), and the average yearly NEE for the ten years was  $-25.00 \pm 12.70 \text{ g C m}^{-2} \text{ y}^{-1}$ . By partitioning NEE into GPP and  $R_{\text{eco}}$  (Fig. 2b), it was found that  $151.65 \text{ g C m}^{-2} \text{ y}^{-1}$  was assimilated by green plants and  $126.65 \text{ g C m}^{-2} \text{ y}^{-1}$  was released in the form of ecosystem respiration, including autotrophic and heterotrophic respiration. In other words, the saline desert is a significant net carbon sink which sequestered as much as 16.7% of GPP.

Further analysis revealed that there was no significant relationship between annual NEE and precipitation (Pearson's correlation coefficient  $r=0.37$ ,  $P=0.29$ , Fig. 2d). The average precipitation for the ten years was 153.5 mm, which is very close to the long-term average of 164 mm (Fig. 2d), indicating that this sink was not a result of unusually heavy precipitation, but rather a long-term average.

To gain more insight into the process of carbon sequestration, the cumulative NEE for the growing and non-growing periods (Figs. 2c and e), were analyzed separately. The results showed that the accumulated NEE demonstrated significant annual variation for both periods, but that these variations were not synchronized with each other. For

1 example, the accumulated NEE for the growing period varied over thirteen-fold over  
 2 the ten years, ranging from a high of  $-8.13 \text{ g C m}^{-2} \text{ y}^{-1}$  in 2005 to a low of  $-106.51 \text{ g C}$   
 3  $\text{m}^{-2} \text{ y}^{-1}$  in 2002, and had a significant positive relationship with total cumulated NEE  
 4 (Pearson's correlation coefficient  $r=0.99$ ,  $P<0.001$ ). However, for the non-growing  
 5 period, the cumulated NEE (ecosystem carbon release), with a maximum of  $25.10 \text{ g}$   
 6  $\text{Cm}^{-2} \text{ y}^{-1}$  in 2009 and a minimum of  $13.6 \text{ g C m}^{-2} \text{ y}^{-1}$  in 2012, had no discernable  
 7 correlation with total cumulated NEE (Pearson's correlation coefficient  $r=0.29$ ,  $P=0.43$ ).  
 8 More importantly, in terms of values for the two periods, the absolute values of  
 9 cumulative NEE during the growing period were always greater than the values during  
 10 the non-growing period except in 2005, and the ten-year averages for the growing and  
 11 non-growing periods were  $-43.50 \text{ g C m}^{-2} \text{ y}^{-1}$  and  $18.50 \text{ g C m}^{-2} \text{ y}^{-1}$  respectively. In  
 12 other words, the carbon sequestered during the growing period was not completely  
 13 released during the non-growing period, which strongly suggested a carbon sink.

### 14 **3.2 Cross-check of the CO<sub>2</sub> flux**

15 To cross-check the reliability of the observed downward CO<sub>2</sub> flux from  
 16 eddy-covariance measurements, soil CO<sub>2</sub> flux, and net primary canopy productivity  
 17 were measured using closed-chamber method respectively (Supplementary Figs. S1 and  
 18 S2). Figure 3 illustrates that the tower-based measurements (NEE) agreed very well  
 19 with the chamber-based CO<sub>2</sub> fluxes ( $R_{\text{soil}} + \text{NPP}_{\text{canopy}}$ ) for the daylight hours during the  
 20 six days (Fig. 3). In other words, the reliability of daytime NEE values obtained from  
 21 eddy-covariance measurements was verified by independent chamber-based  
 22 measurements on soil and plants.

1 During the nighttime, CO<sub>2</sub> exchange becomes simple: respiration at the ecosystem  
2 level should equal soil CO<sub>2</sub> flux plus plant-canopy respiration. As the chamber used to  
3 measure CO<sub>2</sub> flux from the plant canopy was not automated, nighttime respiration was  
4 even more difficult to quantify. Therefore, only limited amounts of eddy-covariance and  
5 soil CO<sub>2</sub> flux data are available for the nighttime. The R<sub>soil</sub> to R<sub>eco</sub> ratio was calculated,  
6 which could provide useful information for partitioning relative contributions to R<sub>eco</sub>.  
7 Figure 4 shows that the ratio of R<sub>soil</sub> to R<sub>eco</sub> was very small and even negative during  
8 most of the year. The maximum monthly average ratio was only 0.11 (± 0.03). The  
9 direct reason for this was that soil CO<sub>2</sub> flux was so low that atmospheric CO<sub>2</sub> was  
10 sucked into the soil (Fig. 4a), which contradicts common sense (see discussion for  
11 details). However, these observations further confirmed that atmospheric CO<sub>2</sub> was  
12 passing through the soil-atmosphere interface into the soil.

### 13 **3.3 Changes in biomass and soil carbon storage**

14 The eddy-covariance data provided strong evidence for net CO<sub>2</sub> uptake at the  
15 study site. But where did the CO<sub>2</sub> uptake go? Fortunately, when Fukang Station (see  
16 online Materials and Methods) was established in 1989, a detailed survey of plants and  
17 soils was conducted at the site, including biomass and organic and inorganic carbon in  
18 the soil. If the downward CO<sub>2</sub> flux detected in the eddy-covariance measurements were  
19 stored in plants or soil, it should be observable now after 20 years of accumulation.  
20 Over the 20 years, woody biomass (not including shrub foliage, annuals, or ephemeral  
21 plants) changed from 0.78 ± 0.06 kg m<sup>-2</sup> to 0.74 ± 0.05 kg m<sup>-2</sup>. Organic carbon storage  
22 in the 0–3 m soil layer was 8.1 ± 0.64 kg m<sup>-2</sup> in 1989 and 8.4 ± 0.67 kg m<sup>-2</sup> in 2009.



1 Inorganic carbon storage at the same depth was  $32.7 \pm 2.1 \text{ kg m}^{-2}$  in 1989 and  $33.0 \pm$   
2  $1.8 \text{ kg m}^{-2}$  in 2009. In other words, in the past 20 years, carbon storage in plants and  
3 soil at this site has not undergone significant change (independent-samples *t*-test,  $n = 8$ ,  
4  $P = 0.05$  for all pairs). This test confirmed that it is unlikely that net CO<sub>2</sub> uptake at this  
5 desert site can be explained by changes in C stored in plant biomass or shallow soils.

#### 6 **3.4 Downward carbon leaching with groundwater fluctuations**

7 Since the downward CO<sub>2</sub> flux had not been stored in plants or soil, the next  
8 possibility was the groundwater body, because saline/alkaline soils are by their nature  
9 connected to groundwater (Rengasamy, 2006). The first piece of evidence was the  
10 vertical distribution of soil EC (Fig. 5). Generally, soil EC decreases with increasing  
11 soil depth, which means that passive leaching of salt is possible with fluctuations in the  
12 groundwater table. Restricted by the limited rainfall at the study site (annual average  
13 precipitation  $\sim 164 \text{ mm}$ ), salt could be washed down to a limited depth and thereby  
14 create the “S” shape of the EC (Fig. 5), but not to the depth of groundwater. The  
15 maximum value of EC was  $41.05 \pm 1.52 \text{ dS m}^{-1}$  at a depth of 1 m. High values reached  
16 as far as 2 m depth, probably from very rare events of heavy rainfall or snowmelt in this  
17 cold desert. The ground water is saline/alkaline with EC of  $4.27 \pm 0.04 \text{ dS m}^{-1}$ , which  
18 have high solubility to CO<sub>2</sub> (Lindsay, 1979). Furthermore, the last 33 years data (from  
19 1978 to 2010) showed that the groundwater table fluctuated greatly (Fig. 5) between 1  
20  $\sim 3$  meters. Therefore, it is reasonable that the CO<sub>2</sub> respired can be absorbed by the  
21 saline/alkaline soil solution (Lindsay, 1979; see discussion for details) and be washed  
22 downward into the groundwater body by rises and falls in the groundwater table.

1       The laboratory leaching experiment showed that the isotopic characteristics and  
2       the amount of leachate removed with frequent leaching (Fig. 6). In total, the leaching  
3       operation was conducted 15 times. The amount of leaching solution, 4653.5 g on  
4       average, showed no significant difference among leaching operations ( $P<0.01$ ).  
5       However, the isotopic characteristics of the leachate,  $\delta^{13}\text{C}$ , showed strong enrichment  
6       following single-time labeling, changing from its initial value of  $-7.33 \pm 2.50 \text{ ‰}$  to a  
7       peak value of around 300‰ at the sixth washing operation. After that, the  $\delta^{13}\text{C}$  of  
8       leachate diminished exponentially and returned to approximately background values.  
9       These data unambiguously demonstrate that, as expected, surface dissolved carbon  
10      (labeled  $^{13}\text{CO}_2$ ) can be leached out by fluctuations in groundwater.

11

#### 12   **4 Discussion**

13      Long-term trends in  $\text{CO}_2$  exchange between the terrestrial ecosystem and the  
14      atmosphere have been well documented using the eddy-covariance method in different  
15      climatic zones and ecosystem types (Falge et al., 2002a; Law et al., 2002; Baldocchi et  
16      al., 2005), including at the present study site (Liu et al., 2012). With a ten-year average  
17      NEE of  $-25.00 \text{ g C m}^{-2} \text{ y}^{-1}$ , the saline desert was verified to be a net carbon sink, which  
18      was in agreement with the studies of Jasoni et al. (2005) and Wohlfahrt et al. (2008),  
19      exceeding the  $100 \text{ g C m}^{-2} \text{ y}^{-1}$  uptake by the Mojave Desert.

20      A comprehensive uncertainty analysis accounting for both random and systematic  
21      errors showed that the average confidence interval for annual NEE was  $\pm 12.70 \text{ g C m}^{-2}$   
22       $\text{y}^{-1}$  (Table 1). This value was less than the 10-year average, indicated that this sink was

1 not caused by measurement errors but rather a fact. In addition, comparison between  
2 NEE estimates from eddy-covariance and scaling-chamber measurements yielded close  
3 agreement of NEE values (Fig. 3). Therefore, the net uptake derived from  
4 eddy-covariance measurements can be considered reliable, even though the possibilities  
5 of biological assimilation by soil crusts or CAM plants (Hastings et al., 2005) and  
6 changes in either soil or living biomass carbon storage (Stone, 2008) were insufficient  
7 to explain such an uptake (Hastings et al., 2005; Stone, 2008; Schlesinger et al., 2009).

8 It is worth to note that vegetation, in this study area, has no significant change over  
9 the 20 years ( $0.78 \pm 0.06 \text{ kg m}^{-2}$  in 1989 and  $0.74 \pm 0.05 \text{ kg m}^{-2}$  in 2009). Namely, this  
10 shrub-dominated stable vegetation has long reaching its maturity - the new fixed  
11 biomass offset dead biomass. Similar result found in the repeated soil carbon content  
12 inventory, soil organic and inorganic carbon contents also unchanged. Considering the  
13 characteristic of the study site, these results were very reasonable. In the study site, the  
14 shrubs are sparsely distributed (plant coverage is approximately 17%) and organic litter  
15 mainly dispersed under the canopy (Li et al., 2007), where the microbial activity is  
16 strong. In addition, the desert shrubs have strong canopy interception effect, which  
17 induces higher soil water content under canopy than in bare area (Wang et al., 2012),  
18 which also speeds up the litter decomposition rate to equalize the organic litter input  
19 rate. For the bare soil without almost any litter input, the carbon content hardly changes.  
20 Therefore, coupled with results of repeated plant biomass and soil carbon content  
21 inventories, it can be safely inferred that, in the long run,  $\text{CO}_2$  assimilated by plants is  
22 all respired by autotrophic and heterotrophic respiration.

1       The magnitude of the NEE estimates for this desert ecosystem is comparable to net  
2       ecosystem production for many grassland and forest ecosystems (Falge et al., 2002b;  
3       Bergeron et al., 2007; Luyssaert et al., 2007) which have much higher GPP than desert.  
4       Because NEE is the residual of GPP and  $R_{eco}$ , this suggested that  $R_{eco}$ , especially soil  
5       CO<sub>2</sub> flux, was very low in desert ecosystems (Wohlfahrt et al., 2008). This inference is  
6       supported by results of the very tiny or even negative contribution of  $R_{soil}$  to  $R_{eco}$  (Fig. 4,  
7       maximum value of  $0.11 \pm 0.03$ ). Although the magnitude of negative values of  $R_{soil}/R_{eco}$   
8       was meaningless, it did, however, confirm findings of negative soil CO<sub>2</sub> flux based  
9       solely on soil-chamber measurements in some extreme environments (Ma et al., 2012,  
10      2013; Shanhun et al., 2012). This observation contradicts common sense - soil  
11      respiration by definition should not be negative - and similar studies, where soil  
12      respiration was commonly proposed to be the most important portion of ecosystem  
13      respiration, accounting for 50%–100% of total respiration (Lavigne et al., 1997; Bolstad  
14      et al., 2004; Curtis et al., 2005; Davidson et al., 2006). **Therefore, the term “soil CO<sub>2</sub>**  
15      **flux” is more appropriate than “soil respiration” within the context of the current study.**

16      Therefore, these results provide a basis for a plausible explanation: the soil did  
17      not respire as much as it should have (Figs. 2 and 4). Although most of the CO<sub>2</sub>  
18      produced in soil escaped to the atmosphere as gas efflux, a fraction of CO<sub>2</sub> was  
19      dissolved in the soil solution and formed a significant DIC reservoir, which was then  
20      leached downwards into groundwater (Serrano-Ortiz et al., 2010; Shanhun et al., 2012;  
21      Ma et al., 2013). **It may be argued that the leached carbon would also contain dissolved**  
22      **organic carbon (Kindler et al., 2011). While we can not rule out the possibility of**

1   dissoluble organic carbon leaching from the soil, the organic matter content in the study  
2   area is very low (less than 1%) and dissolvable organic carbon must be even lower.  
3   More important, soil organic carbon mainly concentrates at the topsoil and decreases  
4   with soil depth (Li et al., 2010), where the dissoluble organic carbon are hardly leached  
5   by limited rainfall in “passive leaching” pattern (Fig. 5). Therefore, within this context,  
6   we assume the leaching carbon is in the dissolved inorganic carbon form. Namely, it is  
7   proposed here that leaching of DIC is responsible for carbon uptake in deserts (Fig. 1;  
8   Chapin et al., 2006).

9       Substantial evidence has suggested that the amount of DIC leaching may be large  
10   (Ciais et al., 2008; Kindler et al., 2011). In European grasslands, which are much less  
11   saline/alkaline than desert regions, the DIC leaching rate was  $24 \text{ g C m}^{-2} \text{ y}^{-1}$  and  
12   accounted for 22% of NEE (Kindler et al., 2011). A recent study of soil dissolved  
13   inorganic carbon (SDIC) at the present study site showed that SDIC storage was  
14   approximately  $24.2 \text{ kg} \cdot \text{m}^{-2}$  at 6 m depth, most of which was in leachable form (Wang et  
15   al., 2013).

16       Leaching DIC into groundwater requires fluid input, which consists mainly of  
17   precipitation and surface-water (e.g., runoff) infiltration. However, at the present study  
18   site, rainfall alone may be insufficient to achieve this transport process (Amiaz et al.,  
19   2011). In addition, runoff from mountains was diverted to nearby agricultural oases  
20   decades ago. Therefore, due to these limitations, localized uptake of  $\text{CO}_2$  was stored in  
21   the soil in the form of DIC (Gamnitzer et al., 2011; Ma et al., 2013). When fluctuations  
22   in the depth of the water table occur (Amiaz et al., 2011), a leaching process takes place:

1 rising groundwater absorbs soil DIC, and falling groundwater transports the DIC  
2 downward (Fig. 6). These water-table fluctuations have been named as “passive  
3 leaching” of DIC, in comparison with the active DIC leaching that occurs during  
4 groundwater recharge. This passive leaching significantly expands the area where DIC  
5 leaching occurs and creates a “hidden carbon sink”. However, the passive leaching  
6 process may be constrained by groundwater level and its fluctuation. For the vast desert,  
7 of which the groundwater can be 10 to over 100 meters deep, the passive leaching  
8 should be very limited. Instead, they probably play a greater role in carbon storage of  
9 groundwater, which is transported horizontally from the passive leaching active areas.  
10 We fully aware that the passive leaching presented here may only within a limited  
11 geographic range. To clarify this, field measurements of DIC in soil and groundwater,  
12 containing DIC concentration and isotopic characters, should be ways to advance this  
13 line of research.

14 The leaching process in this alkaline deserts leads to a third carbon pool on land,  
15 which differs completely from the carbon pools in living biomass and soils (Houghton,  
16 2007). One important reason is that the mean residence time for carbon in groundwater  
17 may be much longer than the residence time in soils (Kessler and Harvey, 2001),  
18 although the leaching flux into groundwater is small compared to the rapid diffusion of  
19 CO<sub>2</sub> out of soils.

20 In conclusion, this study tells us that when a downward CO<sub>2</sub> flux is observed,  
21 but seems to have nowhere to go, it is not necessarily mean that the flux measurement  
22 is unreliable. By looking deeper and farther away, a territorial inorganic sink for

1 anthropogenic CO<sub>2</sub>—the groundwater body under the saline arid land—was identified  
2 in this long-neglected region in global carbon budgeting. When a downward CO<sub>2</sub> flux  
3 has been confirmed, but seems to have nowhere to go, an unrecognized process may be  
4 hidden below.

#### 6 **Author Contributions**

7 All authors commented on manuscript at all stages. Y. L. developed the concept of the  
8 paper and oversaw the study; J. M. did *in situ* measurements of soil CO<sub>2</sub> flux; R. L.  
9 analyzed the eddy-covariance data; Z. -D. L. did the soil column leaching experiment; J.  
10 M. and Y. L. wrote the paper; L. -S. T. contributed significantly to the structuring and  
11 presentation of the paper.

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1    **Supporting material legend**

2    **Supplement Figure**

3    **Fig. S1.** Diurnal variations of leaf net photosynthesis rate of *Tamarix ramosissima* (a)  
4    and leaf area index (LAI) dynamic (b) around the growing season in 2009.

5    **Fig. S2.** Diurnal CO<sub>2</sub> flux measured at the soil surface, the plant canopy and the tower  
6    for six days in 2009.

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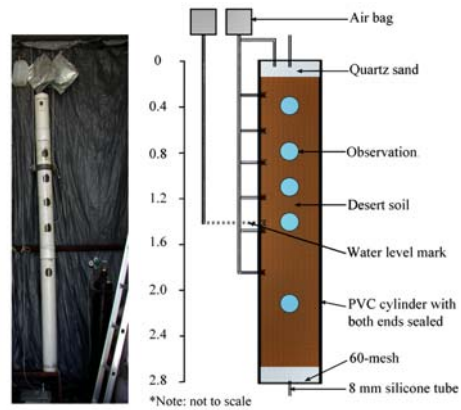
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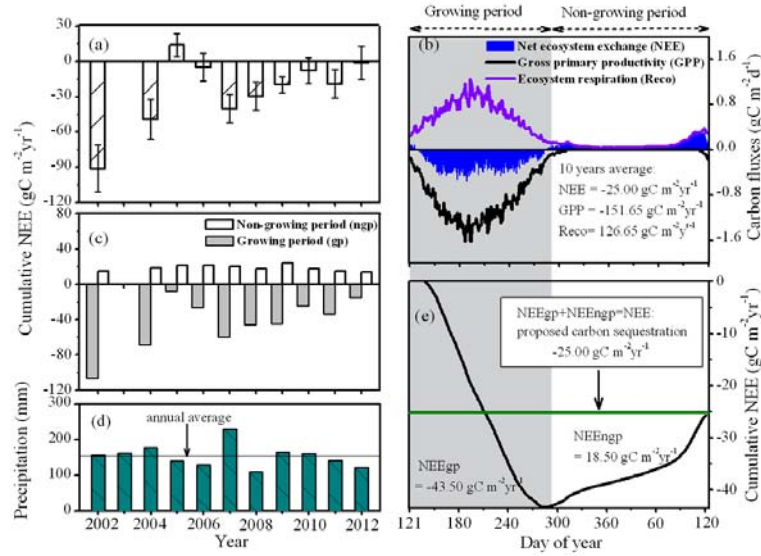
**Table 1.** Errors introduced into estimates of annual net ecosystem CO<sub>2</sub> exchange (NEE) (g C m<sup>-2</sup> y<sup>-1</sup>) from worst-case estimates of the effects of random errors, gap-filling, and u<sup>\*</sup>-filtering and energy-balance analysis for the ten years of eddy-covariance data.

| Year    | Error analysis (g C m <sup>-2</sup> y <sup>-1</sup> ) |              |             | Energy balance     |                |      |
|---------|---|--------------|-------------|--------------------|----------------|------|
|         | System error  | Random error | Total error | Slope <sup>*</sup> | r <sup>2</sup> | n    |
| 2002    | 11  | 9            | 20          | 0.69               | 0.75           | 7011 |
| 2004    | 11  | 6            | 17          | 0.62               | 0.88           | 6608 |
| 2005    | 5   | 5            | 10          | 0.83               | 0.70           | 7972 |
| 2006    | 8   | 4            | 12          | 0.82               | 0.81           | 7250 |
| 2007    | 7   | 5            | 12          | 0.75               | 0.77           | 7740 |
| 2008    | 5   | 7            | 12          | 0.78               | 0.73           | 8015 |
| 2009    | 3   | 4            | 7           | 0.79               | 0.82           | 7150 |
| 2010    | 6   | 5            | 11          | 0.66               | 0.71           | 5325 |
| 2011    | 5   | 7            | 12          | 0.80               | 0.73           | 4752 |
| 2012    | 4   | 10           | 14          | 0.74               | 0.82           | 6127 |
| Average | 6.5   | 6.2          | 12.7        | 0.74               | 0.77           | 7341 |

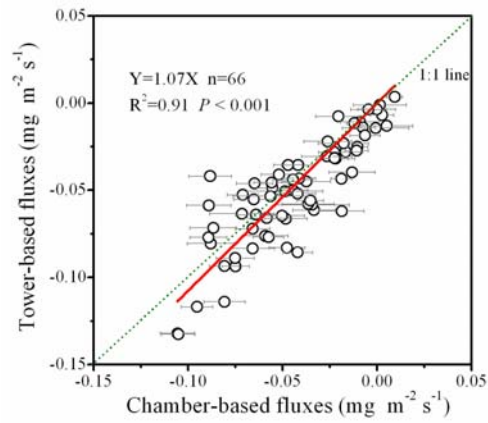
Note: Slope means  $(LE+H)/(Rn-G)$ .



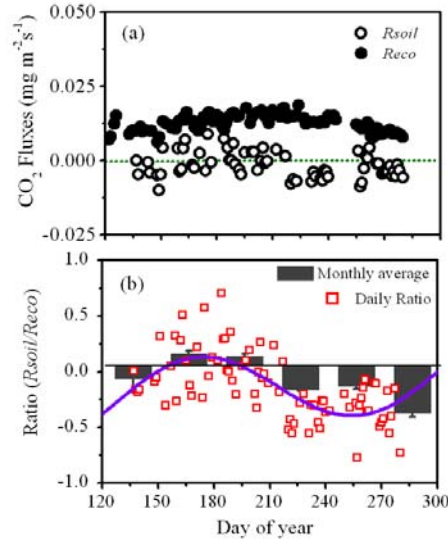
**Fig. 1.** Section of the soil column in the PVC cylinder in which the leaching experiments were performed to model the groundwater fluctuation process.



**Fig. 2.** Carbon fluxes and proposed carbon sequestration at the saline desert site: (a) ten years' net ecosystem exchange (NEE) of carbon; (b) ten-year averages of daily NEE, GPP, and  $R_{eco}$ ; (c) accumulated NEE for the growing and non-growing periods over the 10 years; (d) yearly precipitation from 2002 to 2012; (e) ten-year average of accumulated NEE for the growing and non-growing periods; the difference between these two periods was proposed as carbon sequestration by the saline desert.



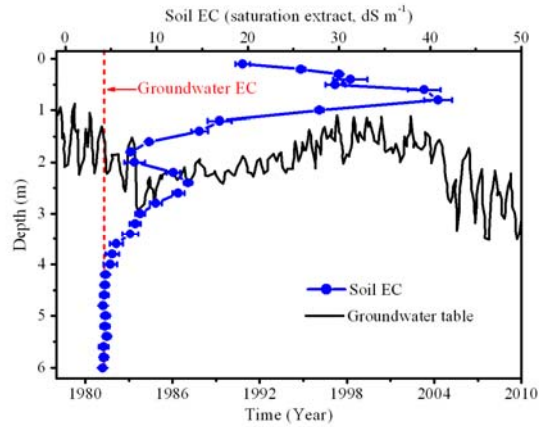
**Fig. 3.** Agreement between chamber-based and tower-based CO<sub>2</sub> fluxes for six days during daylight hours in 2009. Original data for the six days are given in Supplementary Fig. S1.



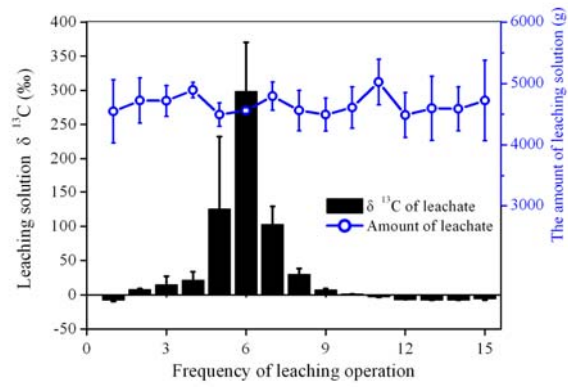
**Fig. 4.** Seasonal variations in  $R_{soil}$  and  $R_{eco}$  (a) and the ratio between these two fluxes (b) in 2009. The seasonal variation in  $R_{soil}/R_{eco}$  was well fitted by a second-order Fourier function, where:

$$R_{soil} / R_{eco} = -0.132 - 0.208 \sin(DOY * -0.540) - 0.402 \sin(2 \times DOY * -1.087), \text{ and}$$

$$DOY * (\text{in radians}) = DOY \times \frac{2\pi}{365}. \text{ N}=90, \text{ R}^2=0.474.$$



**Fig. 5.** Profile distribution of soil EC and fluctuations in the groundwater table at the saline desert site. Generally, EC decreases with increasing depth, which means that passive leaching of salt is possible with fluctuations in the groundwater table. The “S” shape of the EC was created by rainfall infiltration, which reached no more than 2 m depth, probably from very rare events of heavy rainfall or snowmelt in this cold desert.



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2 **Fig. 6.**  $^{13}\text{C}$  isotopic characteristics and amount of leaching solution removed with  
 3 frequent leaching operations.

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