



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

J. Ma<sup>1,2</sup>, R. Liu<sup>1,2</sup>, L.-S. Tang<sup>1,2</sup>, Z.-D. Lan<sup>1,2</sup>, and Y. Li<sup>1,2</sup>

<sup>1</sup>State Key Laboratory of Desert and Oasis Ecology, Xinjiang Institute of Ecology and Geography, Chinese Academy of Sciences, Urumqi, Xinjiang, China

<sup>2</sup>Fukang Station of Desert Ecology, Chinese Academy of Sciences, Fukang, Xinjiang, China

Correspondence to: Y. Li (liyan@ms.xjb.ac.cn)

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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 9 out of 10 years, and the average yearly NEE for the 10 years was  $-25.00 \pm 12.70 \text{ g C m}^{-2} \text{ year}^{-1}$ . 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 be leached downwards into the groundwater body in the process of groundwater fluctuation: rising groundwater absorbs soil dissolved inorganic carbon (DIC), and falling groundwater transports the DIC downward. Horizontal groundwater flow may send this DIC farther away and prevent it from being observed locally. This process has been called “passive leaching” of DIC, in comparison with the active DIC leaching that occurs during groundwater recharge. This passive leaching significantly expands the area where DIC leaching occurs and creates a literally “hidden” carbon sink process under the desert. This study tells us that when a downward CO<sub>2</sub> flux is observed, but seems to have nowhere to go, it does not necessarily mean that the flux measurement is unreliable. By looking deeper and farther away, a place and a process may be found “hidden” underground.

## 1 Introduction

Consistent enrichment of atmospheric CO<sub>2</sub> concentration from preindustrial times due to human activities (Stocker et al., 2013), together with global climate change, have focused much scientific and public attention on carbon cycling (Siegenthaler and Sarmiento, 1993; Chapin et al., 2006; Houghton, 2007). One of the most important issues is the imbalance between CO<sub>2</sub> released from anthropogenic activities and documented terrestrial or oceanic sinks, which is known as the “missing sink” or the “residual terrestrial sink” in the global carbon budget (Houghton, 2007, 2013; Friedlingstein et al., 2010). Substantial studies have been carried out to locate this residual sink, but the uncertainties remain large (Schimel et al., 2001; Houghton, 2007), especially in territorial ecosystems where the carbon flux and the source–sink processes vary greatly in magnitude and mechanism from region to region, such as from rainforest to desert (Schimel et al., 2001; Heimann and Reichstein, 2008).

With characteristics such as low productivity, low carbon exchange, and limited available data (Hastings et al., 2005), deserts remain one of the most underrepresented ecosystems in carbon cycle syntheses (Raich and Potter, 1995; Wohlfahrt et al., 2008; Cable et al., 2011), in spite of their huge area accounting for one-quarter of earth’s land surface (Reynolds, 2001). However, in recent years, a few studies have proposed that desert regions, which have long been considered negligible in global carbon budgeting, have strong downward fluxes into the ground and therefore might be a significant carbon sink (Jasoni et al., 2005; Stone, 2008; Wohlfahrt et al., 2008; Xie et al., 2009). The net carbon uptake in the Mojave Desert of the United States, based on flux-tower measurements, was reported to be around  $100 \text{ g C m}^{-2} \text{ year}^{-1}$

for a 3-year period (Jasoni et al., 2005; Wohlfahrt et al., 2008). Similar results were reported in the Gurbantunggut Desert in northwestern China based on closed-chamber measurements (Stone, 2008; Xie et al., 2009). These findings have understandably attracted scientific and public attention (Stone, 2008; Schlesinger et al., 2009; Serrano-Ortiz et al., 2010) but remain controversial (Schlesinger et al., 2009), in part because there is no agreement about the exact location of the fixed carbon (Schlesinger et al., 2009). Some researchers have argued that assimilation by cryptobiotic crust organisms (such as lichens, mosses, and cyanobacteria) or crassulacean acid metabolism (CAM) plants may account for a significant portion of C uptake (Hastings et al., 2005; Wohlfahrt et al., 2008), while others have argued that the fixed carbon is associated with precipitation or dissolution of carbonate (Xie et al., 2009; Shanahun et al., 2012). However, these interpretations have faced great challenges: cryptobiotic crust organisms and CAM plants are neither sufficiently active nor extensive to explain such an unusually high rate of uptake (Schlesinger et al., 2009; Serrano-Ortiz et al., 2010), not to mention that the negative flux (CO<sub>2</sub> down into the soil) occurred at night. Formation and accumulation of CaCO<sub>3</sub> is constrained by the sources of calcium derived from atmospheric deposition and rock weathering (Schlesinger, 1985; Monger and Gallegos, 2000). Hence, it is understandable that authors have been asked to reconfirm these flux measurements and to present a logical answer to the question of where the carbon goes (Schlesinger et al., 2009).

Recent publications have highlighted the need to include processes other than photosynthesis and respiration, such as losses of carbon as volatile compounds or dissolved carbon (Chapin et al., 2006; Heimann and Reichstein, 2008), in carbon budgeting. Leaching of dissolved inorganic carbon (DIC) through the hydrological cycle is an important component of regional or global carbon budgets (Kindler et al., 2011; Tobias and Bohlke, 2011). However, in (semi)arid regions, evaporative demand significantly exceeds rainfall on average (Noy-Meir, 1973), and surface runoff is constrained or limited, hampering direct groundwater recharge (de Vries and Simmers, 2002) and ultimately resulting in lack of conventional leaching. Instead, groundwater level fluctuations occur over various timescales (e.g., from daily to seasonal to decadal, 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 the rise and fall 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 the 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 Academy of Sciences, which is located at the southern periphery of the Gurbantunggut 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 5–7 °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

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

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

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

An uncertainty analysis, accounting for both random and systematic errors arising from random sampling error, the gap-filling procedure (Liu et al., 2012), and  $u^*$  filtering, was conducted to obtain confidence intervals for the annual carbon budgets. For random sampling error, the worst-case estimation method proposed by Morgenstern et al. (2004; Eq. 11) was used. As for the uncertainties in the gap-filling procedure, which resulted in a complete NEE time series, these were estimated by creating artificial gaps in the gap-filling series with added noise (Giasson et al., 2006); more details are 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 ( $u^* = 0.1 \text{ m s}^{-1}$ ) 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 are 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 nongrowing period (ngp) contained the remaining days of the year.

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

Generally, the chamber method is a direct and more accurate method for the estimation of ecosystem CO<sub>2</sub> flux, which provides a way to cross-check the reliability of the NEE observed from the eddy covariance system. Conceptually, NEE is representing the balance of ecosystem gross primary

**Table 1.** Errors introduced into estimates of annual net ecosystem CO<sub>2</sub> exchange (NEE) ( $\text{g C m}^{-2} \text{ yr}^{-1}$ ) from worst-case estimates of the effects of random errors, gap-filling, and  $u^*$  filtering, and the energy-balance analysis for the 10 years of eddy-covariance data.

Year	Error analysis ( $\text{g C m}^{-2} \text{ yr}^{-1}$ )			Energy balance		
	System error	Random error	Total error	Slope*	$r^2$	$n$
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

\* Slope means  $(LE + H)/(R_n - G)$ .

productivity (GPP) and ecosystem respiration ( $R_{\text{eco}}$ ). NEE integrates the carbon metabolism of all biota. Namely,

$$\begin{aligned} \text{NEE} &= \text{GPP} - R_{\text{eco}} = \text{GPP} - R_{\text{leaf}} - R_{\text{stem}} - R_{\text{soil}} \\ &= \text{NPP}_{\text{canopy}} - R_{\text{soil}}, \end{aligned}$$

where  $\text{GPP} - R_{\text{leaf}}$  represents net carbon assimilation or release by leaves, which can be estimated by scaling up the 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,  $\text{NPP}_{\text{canopy}}$  is equal to GPP minus  $R_{\text{leaf}}$ . Soil CO<sub>2</sub> flux ( $R_{\text{soil}}$ ) was scaled up by cumulating the relative contribution from different measurement sites, e.g., the site under canopy and in the bare area. Methods used to measure leaf net photosynthesis rates, LAI dynamic and soil CO<sub>2</sub> flux are briefly described below.

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

The 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 6 clear and sunny days evenly across 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 with photographs at intervals of 2 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 destructively sampled. On the basis of the accumulated leaf biomass and relative growth rate, the seasonal LAI dynamic was determined.

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

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

### 2.4 Calculating GPP and $R_{\text{eco}}$

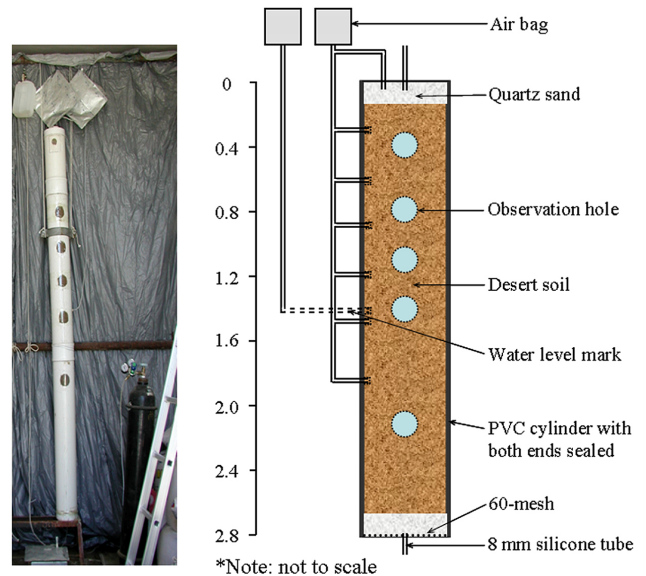
The  $R_{\text{eco}}$  at the study site and the GPP of the ecosystem were obtained through an algorithm (Reichstein et al., 2005) that first establishes a relationship between soil temperature and  $R_{\text{eco}}$  from nighttime data (e.g., the Lloyd-Taylor function, Lloyd and Taylor, 1994) and then uses this relationship to extrapolate  $R_{\text{eco}}$  from nighttime to daytime. GPP was then obtained by subtracting NEE from  $R_{\text{eco}}$ .

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

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

### 2.6 Investigation of biomass and soil carbon storage

A detailed survey of plants and soils at the study site, including living biomass and organic and inorganic carbon storage in the soil, was carried out in 1989 (the year that the Fukang Station of Desert Ecology was established) and 2009, respectively. Details of soil-sampling methods and quadrat investigations of plant biomass were described in Li et al. (2010). Soil total carbon and inorganic carbon were measured using a total organic carbon/total nitrogen analyzer (multi C/N 3100, Analytik Jena, Germany), and the difference between the two values was taken to represent the organic carbon content.



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

### 2.7 Laboratory soil column leaching experiment

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

A schematic representation of the soil columns is shown in Fig. 1. Briefly, soil columns (PVC cylinders, height 280 cm, inner diameter 20 cm) were packed with stratified (at intervals of 30 cm) soil samples at the corresponding depth according to the bulk densities in the field. The cylinders were sealed at the top and bottom with base plates furnished with an 8 mm drainpipe on each side. Above the bottom plate, a layer of gauze (60 mesh) was placed to prevent mud from leaching away (Fig. 1), and 20 cm thick quartz sand was placed on the surface of the gauze to accelerate water infiltration or drainage and to prevent disturbance to soil profiles during water injection. For the same purpose, another 20 cm thick layer of quartz sand was placed on the top of the soil surface to aid in even irrigation distribution. To equalize instantaneous pressure variations within the soil columns and let leachate flow out smoothly, six air vents were designed and all connected to a 10 L airbag (Fig. 1) with silicon tubes. The air vents were located at 30 cm intervals from 30 to 180 cm. To monitor water level in the soil columns, a water-level mark was included at the midpoint of the cylinder (at a position of 140 cm above the bottom). In addition,

five observation holes (diameter 8 cm) were drilled at heights of 100, 140, 170, 200, and 240 cm above the bottom and sealed by transparent resin plates, which enabled observation of color changes in the soil profile.

Once the soil columns were prepared, they were fixed on a metal shelf, which was a convenient method to collect the leachate from the bottom and to simulate groundwater-table fluctuations by manually moving the water tank up and down using a pulley fixed on the shelf. To eliminate the influence of dissolved carbon in the irrigation water on the carbon isotopic character of the leachate, air-free shallow groundwater (pH 8.16, EC = 3.7 dS m<sup>-1</sup>) was obtained from a 20 m deep well adjacent to the saline desert (groundwater level was about 4.5 m). The dissolved air in the water was removed by a vacuum degasser.

To simplify the isotope-marking process, <sup>13</sup>CO<sub>2</sub> (0.3 L, 99.3 % purity) was dissolved into air-free water (2 L) at room temperature in advance and then slowly poured into the soil column using the top tube. This way, dissolved <sup>13</sup>CO<sub>2</sub>, representing CO<sub>2</sub> derived from the atmosphere or from soil respiration, was added to the soil columns (infiltrated to a limited depth far from the bottom of the soil column). After this, the top tube was blocked with a small clip. The washing solution (air-free water) was applied to the bottom of the soil column by lifting up a water drum connected to the bottom tube until the water level reached the point where the labeled CO<sub>2</sub> (dissolved <sup>13</sup>CO<sub>2</sub>) could reach with infiltration. Then the bottom tube was switched to connect to a 10 L vacuum airbag, and the leachate was collected by gravity over 8 consecutive hours. The volume of leachate was recorded, and the δ<sup>13</sup>C of the leachate was measured by an isotope ratio mass spectrometer (IRMS) (Finnigan MAT 253, Thermo Finnigan, Bremen, Germany). This washing procedure was repeated until the δ<sup>13</sup>C of the leachate solution returned to its initial level (15 times overall). Leaching experiments were replicated with three columns.

## 2.8 Energy balance

To assess the reliability of the eddy-covariance measurements, an energy-balance analysis was carried out. The 30 min values of sensible heat flux ( $H$ ) plus latent heat flux (LE) were compared with net radiation ( $R_n$ ) minus soil heat flux ( $G$ ), with results listed in Table 1. These data showed an incomplete energy-balance closure: the slopes of the fitted lines between  $H + \text{LE}$  and  $R_n - G$  were all less than one, which means that the eddy-covariance measurements underestimated  $H + \text{LE}$ . This energy-closure deficit might be explained by the measurement error related to sensor separation, sampling frequency response, or interference from tower or instrument-mounting structures (Twine et al., 2000; Liu et al., 2012). In addition, energy stored under the plant canopy 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

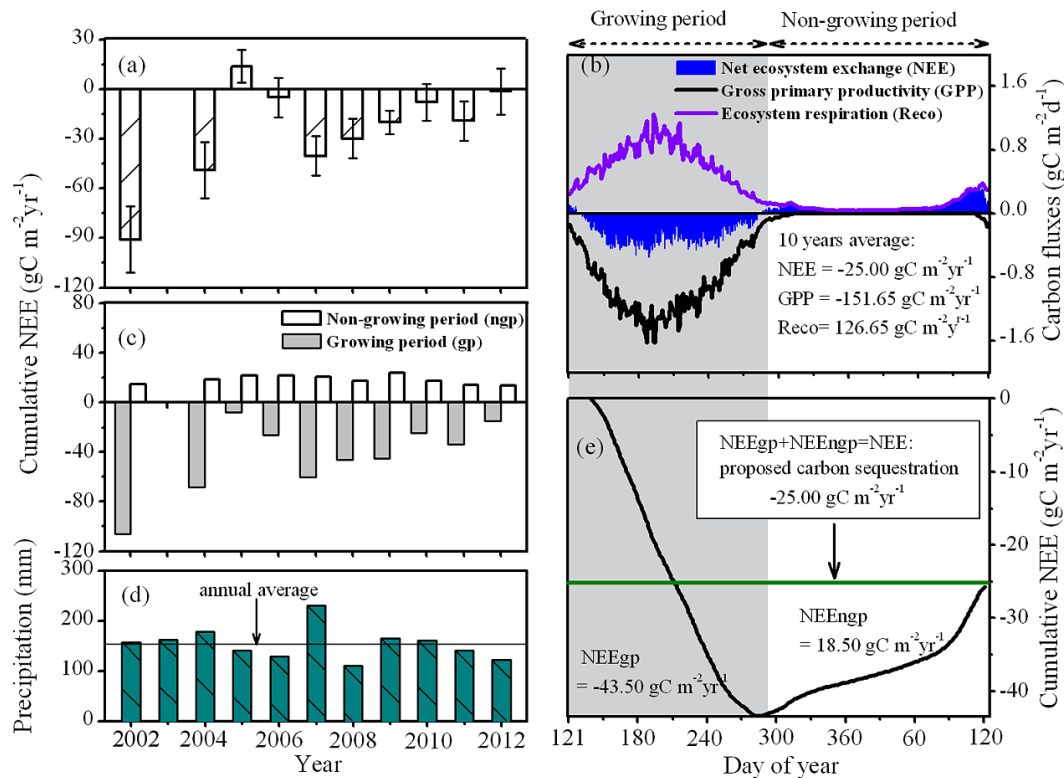
Figure 2 shows obvious annual variations in cumulative NEE ( $P < 0.01$ ), which ranged from  $-91$  to  $14 \text{ g C m}^{-2} \text{ year}^{-1}$ . For 9 out of the 10 years, NEE was negative (or downward into the desert) (Fig. 2a), and the average yearly NEE for the 10 years was  $-25.00 \pm 12.70 \text{ g C m}^{-2} \text{ year}^{-1}$ . By partitioning NEE into GPP and  $R_{eco}$  (Fig. 2b), it was found that  $151.65 \text{ g C m}^{-2} \text{ year}^{-1}$  was assimilated by green plants and  $126.65 \text{ g C m}^{-2} \text{ year}^{-1}$  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 10 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 nongrowing periods (Fig. 2c, 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 example, the accumulated NEE for the growing period varied over thirteenfold over the 10 years, ranging from a high of  $-8.13 \text{ g C m}^{-2} \text{ year}^{-1}$  in 2005 to a low of  $-106.51 \text{ g C m}^{-2} \text{ year}^{-1}$  in 2002, and had a significant positive relationship with total cumulated NEE (Pearson's correlation coefficient  $r = 0.99$ ,  $P < 0.001$ ). However, for the nongrowing period, the cumulated NEE (ecosystem carbon release), with a maximum of  $25.10 \text{ g C m}^{-2} \text{ year}^{-1}$  in 2009 and a minimum of  $13.6 \text{ g C m}^{-2} \text{ year}^{-1}$  in 2012, had no discernable correlation with total cumulated NEE (Pearson's correlation coefficient  $r = 0.29$ ,  $P = 0.43$ ). More importantly, in terms of values for the two periods, the absolute values of cumulative NEE during the growing period were always greater than the values during the nongrowing period, except in 2005, and the 10-year averages for the growing and nongrowing periods were  $-43.50$  and  $18.50 \text{ g C m}^{-2} \text{ year}^{-1}$ , respectively. In other words, the carbon sequestered during the growing period was not completely released during the nongrowing period, which strongly suggested a carbon sink.

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

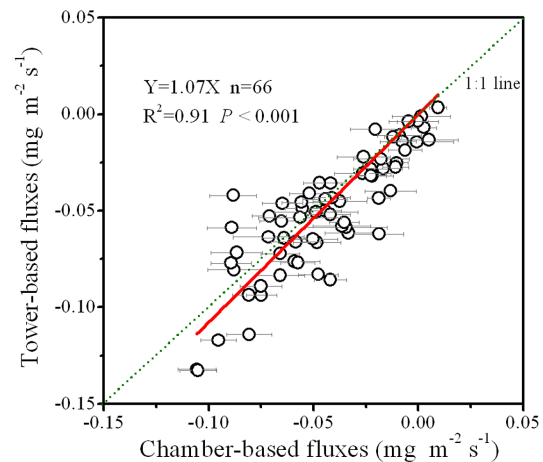
To cross-check the reliability of the observed downward CO<sub>2</sub> flux from eddy-covariance measurements, soil CO<sub>2</sub> flux and net primary canopy productivity were measured using the closed-chamber method (Figs. S1 and S2 in the



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

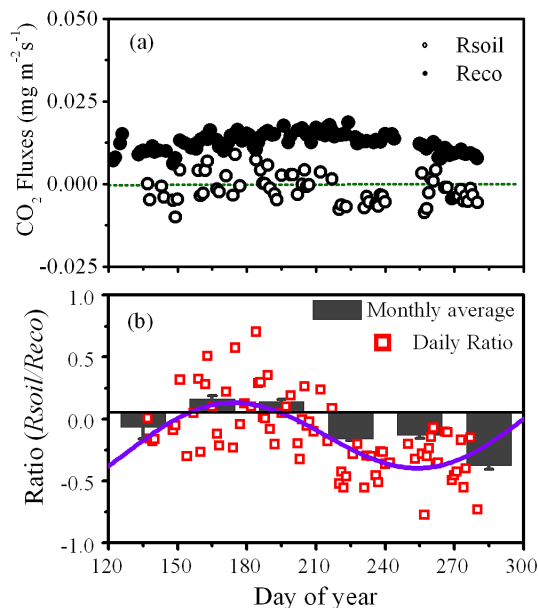
Supplement). Figure 3 illustrates that the tower-based measurements (NEE) agreed very well with the chamber-based CO<sub>2</sub> fluxes ( $R_{\text{soil}} + \text{NPP}_{\text{canopy}}$ ) for the daylight hours during the 6 days (Fig. 3). In other words, the reliability of daytime NEE values obtained from eddy-covariance measurements was verified by independent chamber-based measurements on soil and plants.

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



**Figure 3.** Agreement between chamber-based and tower-based CO<sub>2</sub> fluxes for 6 days during daylight hours in 2009. Original data for the 6 days are given in Fig. S1.



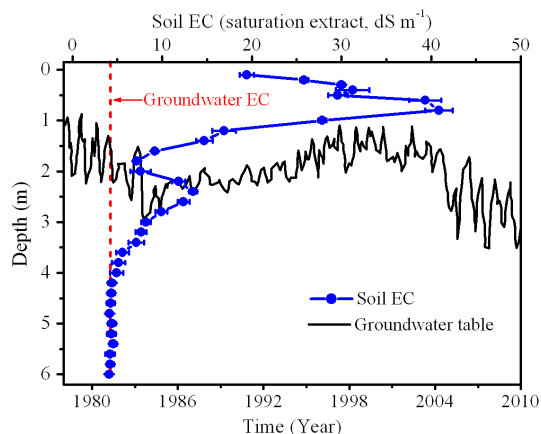


**Figure 4.** Seasonal variations in  $R_{\text{soil}}$  and  $R_{\text{eco}}$  (a) and the ratio between these two fluxes (b) in 2009. The seasonal variation in  $R_{\text{soil}}/R_{\text{eco}}$  was well fitted by a second-order Fourier function, where:  $R_{\text{soil}}/R_{\text{eco}} = -0.132 - 0.208 \sin(\text{DOY} \cdot -0.540) - 0.402 \sin(2 \times \text{DOY} \cdot -1.087)$ , and  $\text{DOY} \cdot (\text{in radians}) = \text{DOY} \times \frac{2\pi}{365}$ .  $N = 90$ ,  $R^2 = 0.474$ .

was passing through the soil–atmosphere interface into the soil.

### 3.3 Changes in biomass and soil carbon storage

The eddy-covariance data provided strong evidence for net CO<sub>2</sub> uptake at the study site. But where did the CO<sub>2</sub> uptake go? Fortunately, when the Fukang Station (see online Materials and Methods) was established in 1989, a detailed survey of plants and soils was conducted at the site, including biomass and organic and inorganic carbon in the soil. If the downward CO<sub>2</sub> flux detected in the eddy-covariance measurements were stored in plants or soil, it should be observable now after 20 years of accumulation. Over the 20 years, woody biomass (not including shrub foliage, annuals, or ephemeral plants) changed from  $0.78 \pm 0.06 \text{ kg m}^{-2}$  to  $0.74 \pm 0.05 \text{ kg m}^{-2}$ . Organic carbon storage in the 0–3 m soil layer was  $8.1 \pm 0.64 \text{ kg m}^{-2}$  in 1989 and  $8.4 \pm 0.67 \text{ kg m}^{-2}$  in 2009. Inorganic carbon storage at the same depth was  $32.7 \pm 2.1 \text{ kg m}^{-2}$  in 1989 and  $33.0 \pm 1.8 \text{ kg m}^{-2}$  in 2009. In other words, in the past 20 years, carbon storage in plants and soil at this site has not undergone significant change (independent-sample  $t$  tests,  $n = 8$ ,  $P = 0.05$  for all pairs). This test confirmed that it is unlikely that net CO<sub>2</sub> uptake at this desert site can be explained by changes in C stored in plant biomass or shallow soils.

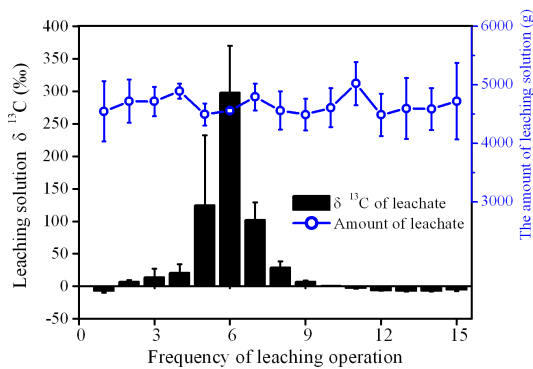


**Figure 5.** Profile distribution of soil EC and fluctuations in the groundwater table at the saline desert site. Generally, EC decreases with increasing depth, which means that passive leaching of salt is possible with fluctuations in the groundwater table. The “S” shape of the EC was created by rainfall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

### 3.4 Downward carbon leaching with groundwater fluctuations

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

The laboratory leaching experiment showed the isotopic characteristics and the amount of leachate removed with frequent leaching (Fig. 6). In total, the leaching operation was conducted 15 times. The amount of leaching solution, 4653.5 g on average, showed no significant difference among leaching operations ( $P < 0.01$ ). However, the



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

isotopic characteristics of the leachate,  $\delta^{13}\text{C}$ , showed strong enrichment following single-time labeling, changing from its initial value of  $-7.33 \pm 2.50\text{‰}$  to a peak value of around  $300\text{‰}$  at the sixth washing operation. After that, the  $\delta^{13}\text{C}$  of leachate diminished exponentially and returned to approximately background values. These data unambiguously demonstrate that, as expected, surface dissolved carbon (labeled  $^{13}\text{CO}_2$ ) can be leached out by fluctuations in groundwater.

#### 4 Discussion

Long-term trends in CO<sub>2</sub> exchange between the terrestrial ecosystem and the atmosphere have been well documented using the eddy-covariance method in different climatic zones and ecosystem types (Falge et al., 2002a; Law et al., 2002; Baldocchi et al., 2005), including at the present study site (Liu et al., 2012). With a 10-year average NEE of  $-25.00 \text{ g C m}^{-2} \text{ year}^{-1}$ , the saline desert was verified to be a net carbon sink, which was in agreement with the studies of Jasoni et al. (2005) and Wohlfahrt et al. (2008), exceeding the  $100 \text{ g C m}^{-2} \text{ year}^{-1}$  uptake by the Mojave Desert.

A comprehensive uncertainty analysis accounting for both random and systematic errors showed that the average confidence interval for annual NEE was  $\pm 12.70 \text{ g C m}^{-2} \text{ year}^{-1}$  (Table 1). This value was less than the 10-year average, which indicates that this sink was a fact and not caused by measurement errors. In addition, the comparison between NEE estimates from eddy-covariance and scaling-chamber measurements showed close agreement to NEE values (Fig. 3). Therefore, the net uptake derived from eddy-covariance measurements can be considered reliable, even though the possibilities of biological assimilation by soil crusts or CAM plants (Hastings et al., 2005) and changes in either soil or living biomass carbon storage (Stone, 2008) were insufficient to explain such an uptake (Hastings et al., 2005; Stone, 2008; Schlesinger et al., 2009).

It is worth noting that vegetation, in this study area, had no significant change over 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 shrub-dominated stable vegetation has long ago reached its maturity – the new fixed biomass offset dead biomass. A similar result is found in the repeated soil carbon content inventory: soil organic and inorganic carbon contents are also unchanged. Considering the characteristics of the study site, these results were very reasonable. In the study site, the shrubs are sparsely distributed (plant coverage is approximately 17%) and organic litter is mainly dispersed under the canopy (Li et al., 2007), where the microbial activity is strong. In addition, the desert shrubs have a strong canopy interception effect, which induces a higher soil water content under the canopy than in bare area (Wang et al., 2012), which also speeds up the litter decomposition rate to equalize the organic litter input rate. For the bare soil without almost any litter input, the carbon content hardly changes. Therefore, coupled with results of repeated plant biomass and soil carbon content inventories, it can be safely inferred that, in the long run, the CO<sub>2</sub> assimilated by plants is completely respired by autotrophic and heterotrophic respiration.

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

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



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

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

Leaching DIC into groundwater requires fluid input, which consists mainly of precipitation and surface-water (e.g., runoff) infiltration. However, at the present study site, rainfall alone may be insufficient to achieve this transport process (Amiaz et al., 2011). In addition, runoff from mountains was diverted to nearby agricultural oases decades ago. Therefore, due to these limitations, localized uptake of CO<sub>2</sub> was stored in the soil in the form of DIC (Gamnitzer et al., 2011; Ma et al., 2013). When fluctuations in the depth of the water table occur (Amiaz et al., 2011), a leaching process takes place: rising groundwater absorbs soil DIC, and falling groundwater transports the DIC downward (Fig. 6). These water-table fluctuations have been named as “passive leaching” of DIC, in comparison with the active DIC leaching that occurs during groundwater recharge. This passive leaching significantly expands the area where DIC leaching occurs and creates a “hidden carbon sink”. However, the passive leaching process may be constrained by the groundwater level and its fluctuation. For the vast desert, in which the groundwater can be from 10 to over 100 m deep, the passive leaching should be very limited. Instead, it probably play a greater role in carbon storage of groundwater, which is transported horizontally from the passive leaching active areas. We are fully aware that the passive leaching presented here may exist only within a limited geographic range. To clarify this, field measurements of DIC in soil and groundwater, containing DIC concentration and isotopic characters, should be ways to advance this line of research.

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

In conclusion, this study tells us that when a downward CO<sub>2</sub> flux is observed, but seems to have nowhere to go, it does not necessarily mean that the flux measurement is unreliable. By looking deeper and farther away, a territorial inorganic sink for anthropogenic CO<sub>2</sub> – the groundwater body under the saline arid land – was identified in this long-neglected region in global carbon budgeting. When a downward CO<sub>2</sub> flux has been confirmed, but seems to have nowhere to go, an unrecognized process may be hidden below.

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*Author contributions.* All authors commented on the manuscript at all stages. Y. Li developed the concept of the paper and oversaw the study; J. Ma did in situ measurements of soil CO<sub>2</sub> flux; R. Liu analyzed the eddy-covariance data; Z.-D. Lan did the soil column leaching experiment; J. Ma and Y. Li wrote the paper; and L.-S. Tang contributed significantly to the structuring and presentation of the paper.

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