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Hidden carbon sink beneath desert

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Key Points:
- An active carbon sink results from irrigation of saline/alkaline soils in arid zones
- A huge inorganic carbon pool has accumulated in the groundwater beneath deserts

Supporting Information:
- Text S1, Tables S1–S6, and Figure S1

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Abstract For decades, global carbon budget accounting has identified a “missing” or “residual” terrestrial sink; i.e., carbon dioxide (CO_2) released by anthropogenic activities does not match changes observed in the atmosphere and ocean. We discovered a potentially large carbon sink in the most unlikely place on earth, irrigated saline/alkaline arid land. When cultivating and irrigating arid/saline lands in arid zones, salts are leached downward. Simultaneously, dissolved inorganic carbon is washed down into the huge saline aquifers underneath vast deserts, forming a large carbon sink or pool. This finding points to a direct, rapid link between the biological and geochemical carbon cycles in arid lands which may alter the overall spatial pattern of the global carbon budget.

1. Introduction

The global carbon balance includes a large terrestrial carbon sink, but that sink has not been fully identified nor its mechanisms explained [*Houghton*, 2007; *Ballantye et al.*, 2012; *Evans et al.*, 2014]. Recent findings that desert regions remove carbon dioxide (CO_2) from the atmosphere at a magnitude of ~100 g C m^{-2} yr^{-1} suggest that these systems may explain at least a portion of that terrestrial carbon sink [*Jasoni et al.*, 2005; *Stone*, 2008; *Wohlfahrt et al.*, 2008; *Xie et al.*, 2009; *Ma et al.*, 2014]. The findings remain controversial, however, because the carbon apparently removed from the atmosphere has not been observed in terrestrial ecosystems [*Schlesinger et al.*, 2009]. The two known active carbon pools on land, living biomass and soil organic carbon, are unlikely to accumulate carbon at such high rates because deserts are sparsely vegetated with humus-poor soil [*Schlesinger et al.*, 2009]. It is also impossible for carbonate accumulation to reach such high rates since it is limited by air deposition of calcium [*Schlesinger et al.*, 2009]. Here we demonstrate a carbon sink that is not observable in plant or soil, with dissolved inorganic carbon (DIC) leached from irrigated arid land and deposited in the saline/alkaline aquifers under bare deserts. It starts with soil-respired CO_2 dissolving into the solution of saline/alkaline soils [*Rengasamy*, 2006], which is then transported downward into the saline/alkaline groundwater aquifers [*Ma et al.*, 2014]. The DIC sink formed during groundwater recharge has been reported before. It has been proposed that its average input rate is 1.34 g C m^{-2} yr^{-1} globally and lower in the arid zone [*Kessler and Harvey*, 2001]. Here we show that DIC sequestration rates in irrigated arid zone soils can be more than 20 g C m^{-2} yr^{-1} on average, 1–2 orders of magnitude greater than previously thought. More importantly, the DIC goes into an almost untouched DIC pool in saline/alkaline aquifers hidden beneath deserts, which is estimated to be up to 1000 Pg globally, large enough to be recognized as the third largest active carbon pool on land.

2. Materials and Methods

2.1. ^{14}C Dating of Inorganic Carbon

Conventional radiometric analysis was used for ^{14}C dating of soil organic/inorganic carbon (SOC/SIC, respectively; Figure 1) at the ^{14}C Laboratory of Lanzhou University, China (lab code Lug), where the organic matter or carbonate salts in the soils were extracted by acidification to produce gaseous CO_2, and the CO_2 was purified and converted into C_6H_6 for radioactive counting by a liquid scintillation spectrometer. The ^{14}C dating of the DIC (Figures 1 and 2) in the groundwater was sent to Beta Analytic at Miami, FL, USA, where the samples were treated and dated with an Accelerator Mass Spectrometry. Pretreatment procedures of the water samples included: precipitation of DIC by first converting it to ammonium carbonate prior to carbon dating. Then strontium chloride was added to the ammonium carbonate solution, and strontium carbonate was precipitated for radiocarbon analysis. The half-life of $T_{1/2}$ = 5730 years was applied as commonly used in hydrology [*Mook*, 2000]. The $^{13}C/^{12}C$ ratios ($\delta^{13}C$) were

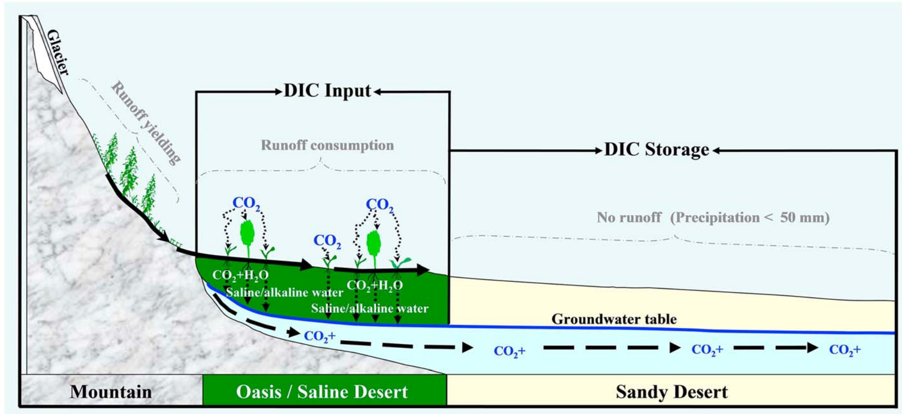

Figure 1. Schematic diagram of DIC (dissolved inorganic carbon) leaching and transport in a closed arid basin: Tarim Basin as an example.

also provided by Beta Analytic and used for correction against dissolution of old carbon in the Tarim Basin. The ^{13}C correction model was assumed to be applicable for both open and closed systems for the evolution of DIC in groundwater, with the following equations [*Salem et al.*, 1980; *Gonfiantini*, 1988]:

$$t = 8267\ln(C_0/C) \quad (1)$$

$$C_0 = [100(\delta_{\mathrm{DIC}} - \delta_R)(1 + 2\varepsilon_{13}/1000)]/(\delta_S - \delta_R + \varepsilon_{13}) \quad (2)$$

where t is the age of groundwater; C is the ^{14}C concentration measured in the sample; C_0 is the "initial" one, corrected for changes due to interaction with the aquifer matrix; δ_{DIC} is the ^{13}C content of the carbonate species dissolved in the sample; δ_R is the ^{13}C content of the aquifer carbonate; δ_S is the ^{13}C content of soil CO_2; and ε_{13} is the equilibrium enrichment factor between bicarbonate and CO_2.

All ^{14}C dates in groundwater were corrected against old carbon dissolution based on equations (1) and (2). In equation (2), the value of 0 ± 1‰ was adopted for δ_R for coated carbonate in sand, and the measured value of δ_S = 20.3‰ (mean value) was adopted for calculating initial ^{14}C activity in equation (2) [*Reardon et al.*, 1979; *Hesterberg and Siegenthaler*, 1991]. In addition, the value of ε_{13} was set to 8‰, a value usually adopted in this model [*Salem et al.*, 1980; *Gonfiantini*, 1988].

2.2. DIC Storage in the Tarim Basin

DIC storage in the saline aquifers of the Tarim Basin was calculated as $\sum(\Delta H \times P \times C \times A)$, where ΔH is the thickness of the aquifer; P the porosity of the aquifer; C the DIC content in the groundwater, which is taken as the sum of carbon in the form of carbonate and bicarbonate; and A is the surface area.

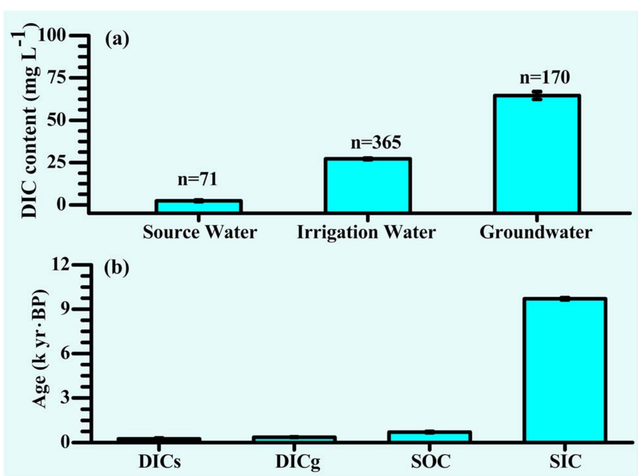

Figure 2. DIC content and the age of DIC in water and soil in the Tarim Basin. (a) DIC concentrations in source water, irrigation water, and groundwater in the Tarim Basin (mean ± standard error). Source water was sampled (n = 71) from direct rainfall or snow glacier in the mountain region; irrigation water was sampled from irrigation channels or rivers in the plain region; groundwater was sampled as shown in Figure 3a. (b) The carbon ages (by ^{14}C dating) for DICs (DIC in the soil of 0–2 m layer), DICg (DIC in the groundwater), SOC (soil organic carbon in the 0–2 m layer), and SIC (soil inorganic carbon in the 0–2 m layer) at the edge of the Tarim Basin.

There were 170 groundwater samples taken from the saline aquifers of the Tarim Basin during two sampling campaigns: 12 in 2011 and the others in 2012. Sampling was by digging, drilling, and pumping, with locations recorded by GPS. Source water was sampled ($n = 71$) from direct rainfall or snow glaciers in the mountain region; irrigation water was sampled from irrigation channels or rivers in the plain region ($n = 365$). DIC content was determined for the sampled water [*Sun et al.*, 1997].

Ordinary kriging interpolation was used to quantify the spatial distribution of DIC age in the Tarim Basin. An ordinary kriging module in ArcGIS's geostatistical analysis (Environmental Systems Research Institute Inc., Redlands, CA, USA) was used. Carbon storage in each age group of DIC was obtained by combining the digital maps of DIC with the isolines of saline aquifer thickness.

The Kolmogorov-Smirnov (K-S) test for DIC age suggested a normal distribution (Table S1 in the supporting information, $p < 0.05$), indicating that the data and size of the samples were appropriate for geostatistical analysis with a semivariogram [*Li and Reynolds*, 1995]. The optimal theoretical model of DIC age was exponential. The value of the coefficient of determination (R^2) was > 0.5; the residual sum of square was small, and the F test for R^2 was significant (Table S2, $p < 0.01$). These parameters indicate that the exponential model was representative of the spatial structure of the DIC age measurements. For the spatial prediction of DIC age, the standardized mean error (ME) of prediction was 0.007 (i.e., approximating 0), and the root-mean-square standardized error (RMSSE) was in the range of 0.908–1.084, indicating that the spatial map of DIC age obtained by ordinary kriging interpolation was reliable/acceptable [*Kitanidis*, 1997].

The parameters for carbon storage of the saline aquifers of the Tarim Basin for each age group are listed in Table S3. DIC content in different age groups within the Tarim Basin (mg L^{-1}) are listed in Table S4.

3. Results

3.1. DIC Leaching and Transport in a Closed Arid Basin

Figure 1 illustrates the water cycle in an interior basin in an arid region. Runoff from precipitation in adjacent mountains is intercepted at the edge of the basin or along rivers, creating oases. Evaporation is greater than precipitation, and thus, the entire oasis/desert landscape is saline. To combat soil salinity and maintain sustainable agriculture, the generally accepted practice is overirrigation (above that required for crop production) to leach the salt away, i.e., application of a leaching fraction [*Abrol et al.*, 1988; *Amiaz et al.*, 2011]. The continuous deposition from this additional irrigation washes salt out of the soil and down to the saline/alkaline aquifers beneath sandy deserts. CO_2 solubility in saline/alkaline water is much higher than in pure or acidic water: the solubility increases linearly with electric conductivity and exponentially with alkalinity (pH) of the soil solution [*Lindsay*, 1979]. As a result, the saline/alkaline water leached into the saline/alkaline aquifer contains large amounts of dissolved CO_2 (i.e., DIC). Thus, the leaching of salt during irrigation in arid regions [*Abrol et al.*, 1988] acts to create a DIC "pump" from surface layers to the saline/alkaline aquifers hidden beneath deserts.

3.2. A Modern Carbon Sink

This DIC pump was characterized in the Tarim Basin (Figure 2). Irrigation water or river water was not carbon free but contained less than half of the DIC content in groundwater (Figure 2a); however, the source water (rainfall or snow glacier melt) was nearly carbon free (Figure 2a). This means that rain or snow glacier meltwater dissolved considerable amounts of CO_2 as it flowed and converged into rivers of the mountain region, which was absorbed from soil air or directly from the atmosphere. When deposited into groundwater, the DIC content was more than doubled (Figure 2a) after flowing through the largely saline/alkaline soils in the plain region. The ^{14}C dating of SOC and SIC of the oasis soil and DIC in the soil and groundwater (DICs and DICg, respectively) showed that DIC was much younger than SIC and even younger than SOC (Figure 2b). These observations indicate that the DIC was not from SIC but rather from dissolved CO_2 produced by soil/root respiration [*Karberg et al.*, 2005]. Thus, whether dissolved in the mountain or plain region, the DIC content of recharge water constituted a modern carbon sink.

3.3. The Active Carbon Pool

Quantification of sink strength and size is difficult. First, the sink strength depends on the recharge rate of groundwater, which remains a challenge for hydrological studies [*Scanlon et al.*, 2006]. Second, DIC

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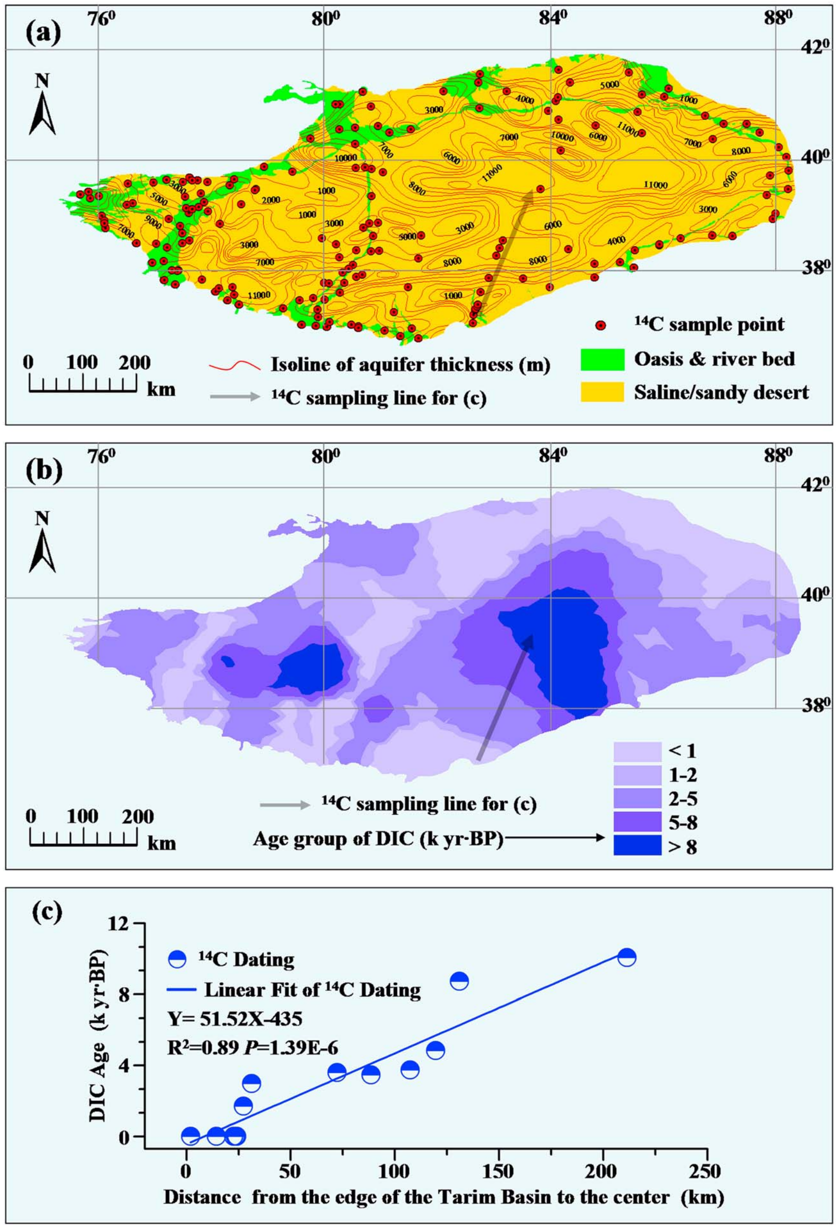

Figure 3. Distribution of samples, thickness of the saline aquifer, and DIC age in the Tarim Basin. (a) Isoline of saline aquifer thickness based on a local geohydrological survey. (b) Isoline for DIC age from ^{14}C dating. (c) DIC ages for groundwater from the edge to the center of the Tarim Basin.

contained in recharge water is even more difficult to quantify because it depends not only on the salinity/alkalinity of the soil [*Karberg et al.*, 2005] but also on the rate of root respiration, which alters the CO_2 partial pressure in the soil air [*Richter and Markewitz*, 1995]. Furthermore, CO_2 solubility is temperature dependent [*Karberg et al.*, 2005] and thus varies seasonally and diurnally. Because of the large spatial and temporal variability, direct measurement of the total amount of DIC leaching in the basin was not feasible. Instead, we used an inversion approach to quantify the DIC input rate in the Tarim Basin: the amount of DIC stored in the saline/alkaline groundwater aquifers divided by its age.

Figure 3a illustrates the sampling points for ^{14}C dating and DIC content analysis, as well as isograms of the thickness of the saline/alkaline aquifer in the Tarim Basin [*Xinjiang Bureau of Geology and Mineral Resources*, 1993]. Sampling points were not evenly distributed because much of the desert is inaccessible. Roads at the edge

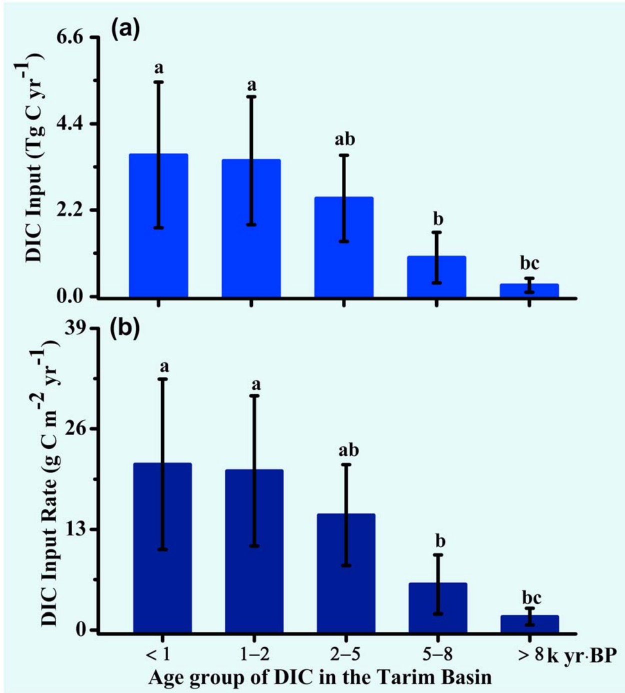

Figure 4. DIC input rate for different aged pools within the Tarim Basin: (a) for the entire basin and (b) per unit area, assuming zero input for the bare sandy desert region and equal input for the rest of the plain area of the basin. Different letters above the columns indicate significant differences at $p < 0.05$.

and two cross-desert highways [*Sun et al.*, 2010] provided access points for data collection used for spatial interpolation, which yielded the contour map of DIC age after ^{14}C dating and correction (see section 2) against old carbon dissolution (Figure 3b). DIC ages in groundwater increased linearly from modern carbon at the edge of the Tarim Basin to more than 10,000 years old at the center (Figure 3c). Figure 3b demonstrates in two dimensions what Figure 3c shows in one dimension: that the DIC ages were youngest near the river beds or oases, where DIC deposition occurred. We conclude that the DIC leached at the edge of arid basins is of modern atmospheric origin and that the process of leaching and transport in groundwater represents a mechanism of CO_2 removal from the atmosphere. With annual precipitation of 30–50 mm in the Tarim Basin, there is no other source of groundwater and no other source of DIC except through recharge from the oasis region that extends from the rivers, mostly at the edge of the basin (Figures 1 and 3).

The distribution of aquifer thickness enabled us to determine the volume and total DIC for each subaquifer within the Tarim Basin. DIC storage was grouped into subaquifers according to ages (Figure 3b). We then calculated the total DIC for different age groups (0–1000, 1000–2000 years, etc.). Dividing the total DIC of each age group by its mean age, we obtained the mean deposition rate for DIC in different age groups (Figure 4a). Dividing the mean deposition rate by the surface area of each age group, we obtained the unit area deposition rate (the potential area (1.682×10^5 km^2) was determined as the plain area of the Tarim Basin (5.058×10^5 km^2) minus the Taklamagan Desert (3.376×10^5 km^2)). Over human history, the DIC deposition rate increased from 1.7 ± 1.2 g C m^{-2} yr^{-1}, more than 8000 years ago, to 21.4 ± 11 g C m^{-2} yr^{-1} for the last 1000 years (Figure 4b). Before agricultural development, deposition occurred mainly in river beds or terminal lakes with much less saline/alkaline water, and thus, less DIC was transported into aquifers. As agriculture developed in this region, irrigation of saline land increased the salinity/alkalinity of the recharge water [*Rengasamy*, 2006], thus delivering more DIC into aquifers. As a result, agricultural development over human history has enhanced the carbon sink (Figure 4). For instance, the highest DIC deposition started 2000 years ago (Figure 4) when the Silk Road opened (and anthropogenic activity intensified) along the edge of the Tarim Basin.

A crude assessment of the global magnitude of the DIC sink in saline/alkaline aquifers may be obtained by assuming that all 24×10^6 km^2 of saline aquifers worldwide [*Weert et al.*, 2009] deposit DIC at the same rate as the Tarim Basin, for which we found 3.6 ± 1.9 Tg C yr^{-1}. The global DIC sink calculated this way is ~0.2 Pg C yr^{-1}. Assuming the same DIC storage as in the Tarim Basin, the global DIC pool in 24×10^6 km^2 of saline aquifers [*Weert et al.*, 2009] is nearly 1000 Pg C. We consider the extrapolations reasonable since the processes shown in Figure 1 apply to all active saline aquifers [*Rengasamy*, 2006; *Weert et al.*, 2009]. However, we are also aware that these extrapolations necessarily remain simplistic and subject to large uncertainty, as arid basins vary in magnitude in their amount of runoff, recharge, and DIC storage in aquifers (Tables S5 and S6: DIC input and storage for arid basins in China and worldwide). Notably lower in DIC recharge and storage than Tarim are arid basins in Northern Africa (Table S6), where water inputs from snow melted are lacking and thus DIC input and storage are limited in corresponding aquifers. Therefore, the global extrapolations presented may constitute a reasonable upper bound. More reliable estimation is hindered by a lack of detailed information on saline water recharge and saline aquifers, as saline waters are generally not usable and thus are largely neglected in hydrological studies.

4. Discussion

Often the downward flux of CO_2 is not equal to carbon accumulation in plants or soils [*Jasoni et al.*, 2005; *Luo et al.*, 2007; *Wohlfahrt et al.*, 2008]. For instance, in European grasslands DIC leaching out of the ecosystem can account for a large portion (22% or 24 g C m^{-2} yr^{-1}) of the net ecosystem exchange of carbon (NEE) [*Kindler et al.*, 2011]. DIC leaching in saline/alkaline desert/oasis regions should be much higher because CO_2 solubility in water increases exponentially with alkalinity and linearly with salinity [*Lindsay*, 1979]. In a saline/alkaline desert, DIC leaching might account for 100% of the observed NEE [*Ma et al.*, 2014]. Thus, a downward CO_2 flux that is not matched by an equivalent increase in plant or soil carbon does not necessarily mean the flux measurement is wrong [*Schlesinger et al.*, 2009]. If we look deeper and further away, the carbon can be found in the saline/alkaline aquifer (Figure 1). The fact that such a huge carbon pool and active sink has been unstudied for so long may simply be because it is remote and hidden under deserts: out of sight, out of mind.

The CO_2 sink reported here is not in the saline/alkaline soils themselves [*Rengasamy*, 2006; *Schlesinger et al.*, 2009] but in the groundwater beneath (Figures 1 and 4b). We found that the direct flux of CO_2 into saline/alkaline soils, as proposed by *Xie et al.* [2009], is only a transient flux and thus is not related to the carbon sink [*Stone*, 2008]. The strong CO_2 absorption found in sterilized saline/alkaline soils [*Xie et al.*, 2009] is simply a recovery of CO_2, as all dissolved CO_2 in the soil solution is driven off [*Lindsay*, 1979; *Xie et al.*, 2009] during sterilization by steam vapor at $>$100°C. In fact, in unvegetated sandy desert like the Taklamagan, diurnal variation of temperature itself may create a night-in/day-out CO_2 flux due to dissolution and diffusion, resulting in zero daily mean flux and no net sink or source [*Ma et al.*, 2013] (see also Figure S1).

Leaching into groundwater does not necessarily create a carbon sink, as the carbon may be released back into the atmosphere after being discharged into streams, rivers, lakes [*Kling et al.*, 1991; *Worrall and Lancaster*, 2005], and the ocean [*Cai et al.*, 2003]. However, rivers in closed (terminal) arid basins have no outlet and no hydrological connection to the oceans, creating aquifers isolated from the river-ocean system (Figure 1). Saline aquifers function as the "ocean" for these inland river-aquifer hydrological systems. The only difference is that this ocean is covered by a thick layer of sandy soil (Figure 1). As CO_2 partial pressures in the soil air can be an order of magnitude higher than in the atmosphere [*Richter and Markewitz*, 1995], the CO_2 solubility or DIC content in the saline aquifer (see Table S4) is significantly higher, on a volume basis, than in seawater (by more than 200%), despite the groundwater being less saline/alkaline than seawater [*Lindsay*, 1979].

It could be argued that pumping of groundwater for irrigation will release DIC back into atmosphere. However, the use of saline/alkaline water for irrigation is still in the experimental stage and has never been and will most likely never be (in the foreseeable future) practiced on a large scale [*Beltrán*, 1999], as saline/alkaline water is not usable by crop plants. Diffusion of CO_2 back into the atmosphere through the coarse-textured sandy layer is possible but has been shown to be negligible, as surface CO_2 flux has a zero mean [*Ma et al.*, 2013] (see Figure S1). Freshwater aquifers in arid regions are rare and are usually old geological waters buried long ago [*Leaney et al.*, 2003]. Even if fresh groundwater were used for irrigation, it would not release DIC into the atmosphere as its DIC content is low [*Lindsay*, 1979].

Through irrigation, however, water is made saline/alkaline by contact with the soil [*Abrol et al.*, 1988; *Leaney et al.*, 2003], thereby leaching carbon into saline aquifers (Figure 1). Overall, we demonstrated that this DIC sink in groundwater increased over human history due to expansion of irrigated land. Of course, this expansion has a limit as water resources in arid zones are limiting.

Although this is a case study on a closed arid basin of Tarim, the process of the DIC sink proposed here is not limited to our case. Any places with soil pH $>$ 7 (roughly half of the global soil cover) can have significant DIC deposition into groundwater, provided that there is groundwater recharge per se.

The long-term fate of DIC in saline/alkaline aquifers is worthy of further study. The fact that we overestimated the direct CO_2 absorption by saline/alkaline soils in our previous study [*Xie et al.*, 2009] does not mean that direct absorption is not possible. For instance, the CO_2 sequestration process proposed in the current study, must have been, and will be intensified as atmospheric CO_2 concentration increases [*Hamerlynck et al.*, 2013], in which CO_2 is pushed into saline/alkaline soil and thus the process itself must constitute a carbon sink. The temperature-driven abiotic (or inorganic, geochemical) CO_2 fluxes observed in desert [*Yates et al.*, 2013] and karst regions [*Roland et al.*, 2013] or Antarctic dry valleys [*Shanhun et al.*, 2012] should also contribute significantly to diurnal, seasonal, or even interannual variations in the global carbon cycle. Overall, we believe that nonbiological processes in the modern global carbon cycle may have been greatly underestimated and further study of these processes is highly warranted.

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