



# A new direction in effective accounting for the atmospheric CO<sub>2</sub> budget: Considering the combined action of carbonate dissolution, the global water cycle and photosynthetic uptake of DIC by aquatic organisms

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## ABSTRACT

The magnitudes, variations, locations and mechanisms responsible for the global atmospheric CO<sub>2</sub> sink are uncertain and under continuing debate. Previous studies have focused mainly on the sinks in the oceans, and soil and vegetation on the continents. Here, we show, based on theoretical calculations and field monitoring evidence, that there is an important but previously underestimated sink for atmospheric CO<sub>2</sub> as DIC-dissolved inorganic carbon that results from the combined action of carbonate dissolution, the global water cycle and the photosynthetic uptake of DIC by aquatic organisms in ocean and land. The sink constitutes up to 0.8242 Pg C/a, amounting to 29.4% of the terrestrial CO<sub>2</sub> sink, or 10.4% of the total anthropogenic CO<sub>2</sub> emission. 0.244 Pg C/a are transferred to the sea via continental rivers and 0.2278 Pg C/a by meteoric precipitation over the seas. 0.119 Pg C/a is released back to the atmosphere again, and 0.2334 Pg C/a is stored in the continental aquatic ecosystem. Therefore, the net sink is estimated as 0.7052 Pg C/a. This sink may increase with an intensification of the global water cycle as a consequence of global warming, rising anthropogenic emissions of CO<sub>2</sub> and carbonate dust in atmosphere, and afforestation, which increases the soil pCO<sub>2</sub> and thus the carbonate dissolution. Fertilization with the elements N, P, C, Fe, Zn, and Si increases the organic matter storage/burial by aquatic organisms and thus decreases the CO<sub>2</sub> return to the atmosphere. Based on the ensemble mean projection of global warming for the year 2100 by IPCC, it is estimated that the atmospheric CO<sub>2</sub> sink will increase by 21%, or about 0.18 Pg C/a. However, the uncertainty in the estimation of this sink needs further exploration.

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## Contents

1. Introduction . . . . .	163
2. Methods and data . . . . .	163
3. Solubility of CO <sub>2</sub> in the systems of CO <sub>2</sub> –H <sub>2</sub> O and CaCO <sub>3</sub> –CO <sub>2</sub> –H <sub>2</sub> O and calculation of equilibrium DIC concentration . . . . .	163
4. DIC of rainwaters and runoffs . . . . .	165
4.1. DIC of world rainwaters . . . . .	165
4.2. DIC of runoffs . . . . .	166
5. Calculation of CO <sub>2</sub> sinks by the global water cycle . . . . .	166
6. Photosynthetic uptake of DIC by aquatic organisms . . . . .	167
7. The future of the atmospheric CO <sub>2</sub> sink in the global water cycle . . . . .	169
8. Conclusions and perspective . . . . .	170
Acknowledgements . . . . .	170
References . . . . .	170

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## 1. Introduction

One of the most important challenges in the science of global change is effective accounting of the global budget for atmospheric CO<sub>2</sub> (Broecker et al., 1979; Tans et al., 1990; Sundquist, 1993; Joos, 1994; Schindler, 1999; Melnikov and O'Neill, 2006). Anthropogenic activities have clearly altered the global carbon cycle and significant gaps exist in our understanding of this cycle. Roughly half of the CO<sub>2</sub> emitted by burning fossil fuels remains in the atmosphere, and the other half is absorbed by the oceans and the terrestrial biosphere (Tans et al., 1990; Siegenthaler and Sarmiento, 1993; Raven and Falkowski, 1999; Sabine et al., 2004; Reay et al., 2007). The partitioning between these two sinks is the subject of considerable debate (Tans et al., 1990; Sabine et al., 2004). Without robust accounting for the fate of CO<sub>2</sub> leaving the atmosphere predictions of future CO<sub>2</sub> concentrations will remain uncertain (Siegenthaler and Oeschger, 1978; Jones et al., 2003).

As defined by Melnikov and O'Neill (2006), increase in atmospheric CO<sub>2</sub> concentration ( $N_a$ ) is a function of emissions from fossil fuels ( $E_{ff}$ ) together with net emissions from changes in land use ( $E_{lu}$ ) minus the uptake in oceans ( $F_{oc}$ ) and the residual (terrestrial) sink ( $F_{res}$ ).

$$N_a(t) = E_{ff}(t) + E_{lu}(t) - F_{oc}(t) - F_{res} \quad (1)$$

Current estimates suggest that  $E_{ff} = 6.3$  Pg C/a,  $E_{lu} = 1.6$  Pg C/a,  $F_{oc} = 1.9$  Pg C/a, and  $N_a = 3.2$  Pg C/a. Therefore, the terrestrial sink is  $F_{res} = 2.8$  Pg C/a.

Previous studies have focused mainly on sinks in the oceans and soil/vegetation on the continent (Detwiler and Hall, 1988; Sarmiento and Sundquist, 1992; Hudson et al., 1994; Davidson et al., 1995; Friedlingstein et al., 1995; Khashgi et al., 1996; Nabuurs et al., 1997; Fan et al., 1998; Schulze et al., 1999; Lal, 2004; Woodbury et al., 2007). Here, we show, based on theoretical calculations and evidence from field monitoring results, that there is an important but previously underestimated sink (Meybeck, 1993; Liu and Zhao, 2000; Gombert, 2002) for atmospheric CO<sub>2</sub> by the combined action of carbonate dissolution (Dreybrodt, 1988), the global water cycle (Shiklomanov, 1993), and the photosynthetic uptake of DIC (dissolved inorganic carbon) by aquatic organisms (Yang et al., 1996; Einsele et al., 2001; Lerman and Mackenzie, 2005; Iglesias-Rodriguez et al., 2008). This sink is much larger than previous estimations where only riverine transport of atmospheric carbon was considered (Meybeck, 1993). It is also larger than the sink that arises from dissolution of carbonate rocks in the world's karst areas (Liu and Zhao, 2000; Gombert, 2002). The sink is caused by CO<sub>2</sub> uptake in rainwater and soil water. Dissolution of carbonates by these waters consumes their enhanced CO<sub>2</sub> concentrations worldwide and not only in the karst areas.

## 2. Methods and data

The atmospheric CO<sub>2</sub> sink in rainfall is given by:

$$\text{Rainwater flux} \times \text{observed mean DIC concentration in rainwater.} \quad (2)$$

The CO<sub>2</sub> sink in the continental runoff is computed as:

$$\text{Runoff flux} \times \text{calculated equilibrium DIC concentration with respect to the world's mean soil CO}_2 \text{ and calcite.} \quad (3)$$

The rainwater flux and runoff flux were taken from the work of Shiklomanov (1993). To estimate the mean DIC of rainwater, 16 references (Munger, 1982; Panettiere et al., 2000; Lara et al., 2001; Zhang et al., 2003; Al-Khashman, 2005a; Kulshrestha et al., 2005; Liu et al., 2005; Rastogi and Sarin, 2005; Jawad Al Obaidy and Joshi, 2006;

Arsene et al., 2007; Báez et al., 2007; Singh et al., 2007; Özsoy et al., 2008; Anatolaki and Tsitouridou, 2009; Celle-jeanton et al., 2009; Ladouche et al., 2009) were used, which report representative DIC or HCO<sub>3</sub><sup>-</sup> data of the rainwater from various locations around the globe (Fig. 1). The carbon flux calculated from these data is considered only for rainwater falling into the oceans. The CO<sub>2</sub> from rainfall on land contributes to the soil pCO<sub>2</sub> and must therefore be excluded from this balance; otherwise it would be counted twice. To obtain the sink of CO<sub>2</sub> due to continental runoff we assume that the groundwater is in equilibrium with the soil pCO<sub>2</sub> and also with calcite in carbonate terrains. The corresponding equilibrium values of DIC are listed in Table 1. The runoff fluxes were calculated separately for carbonate and non-carbonate regions, the former having a much higher DIC for the same pCO<sub>2</sub> because of the presence of dissolved carbonate.

Global mean soil pCO<sub>2</sub> was calculated by use of the equation by Brook et al. (1983) as:

$$\log(\text{pCO}_2) = -3.47 + 2.09 \left(1 - e^{-0.00172 \text{ AET}}\right), \quad (4)$$

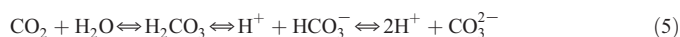
where AET is the annual evapotranspiration in mm.

The equilibrium DIC concentration in calculating the CO<sub>2</sub> sink via continental runoff is based on both theoretical and field observational evidences (Dreybrodt, 1988; Ford and Williams, 1989, 2007; White, 1997; Liu et al., 2007), which are given in the Sections 3 and 4.

The net CO<sub>2</sub> sink is calculated as: (the total CO<sub>2</sub> sink by dissolution of carbonate and CO<sub>2</sub>) – (CO<sub>2</sub> release flux from runoff waters). The latter depends on the difference in pCO<sub>2</sub> between water and the atmosphere, which depends again on the balance of carbonate deposition and organic matter storage/burial in the water (Einsele et al., 2001; Lerman and Mackenzie, 2005).

## 3. Solubility of CO<sub>2</sub> in the systems of CO<sub>2</sub>–H<sub>2</sub>O and CaCO<sub>3</sub>–CO<sub>2</sub>–H<sub>2</sub>O and calculation of equilibrium DIC concentration

CO<sub>2</sub> is readily soluble in water. The reaction:



establishes an equilibrium mixture of carbonic acid, bicarbonate and carbonate ions, which make up the “dissolved inorganic carbon” (DIC) fraction. The proportion of each species depends on pH. At high pH the reaction shifts to the right hand side of Eq. (5). At pH between 7 and 9, about 95% of the carbon in the water is in the form of bicarbonate (HCO<sub>3</sub><sup>-</sup>). At high pH > 10.5, carbonate predominates (Dreybrodt, 1988).

At a global mean annual surface temperature of 15 °C and global mean atmospheric CO<sub>2</sub> partial pressure (pCO<sub>2</sub>) of 380 ppmv, the equilibrium value of the DIC for the CO<sub>2</sub>–H<sub>2</sub>O system is calculated to be 20 μmol/l according to Dreybrodt (1988). However, in the CaCO<sub>3</sub>–CO<sub>2</sub>–H<sub>2</sub>O system, uptake of CO<sub>2</sub> by water is enhanced remarkably by carbonate dissolution, which can be presented as:



For each Ca<sup>2+</sup> dissolved one CO<sub>2</sub> molecule is converted to HCO<sub>3</sub><sup>-</sup>. For example, at 15 °C and a pCO<sub>2</sub> of 380 ppmv, the equilibrium concentration of the DIC for the CaCO<sub>3</sub>–CO<sub>2</sub>–H<sub>2</sub>O system is 1231 μmol/l, or about 66 times larger than that in the CO<sub>2</sub>–H<sub>2</sub>O system (Table 1). If carbonate dissolution proceeds in soil water or in groundwater recharged by soil water with higher pCO<sub>2</sub>, the amount of the CO<sub>2</sub>-uptake by water is even larger (Table 1). For example, according to Brook et al.'s (1983) Eq. (4) using global mean annual evapotranspiration of 548 mm in the continents (Shiklomanov, 1993), world soil pCO<sub>2</sub> may reach up to ~6400 ppmv. In this case,

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