Contribution of carbonate rock weathering to the atmospheric CO_2 sink

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Abstract To accurately predict future CO_2 levels in the atmosphere, which is crucial in predicting global climate change, the sources and sinks of the atmospheric CO_2 and their change over time must be determined. In this paper, some typical cases are examined using published and unpublished data. Firstly, the sensitivity of carbonate rock weathering (including the effects by both dissolution and reprecipitation of carbonate) to the change of soil CO_2 and runoff will be discussed, and then the net amount of CO_2 removed from the atmosphere in the carbonate rock areas of mainland China and the world will be determined by the hydrochem-discharge and carbonate-rock-tablet methods, to obtain an estimate of the contribution of carbonate rock weathering to the atmospheric CO_2 sink. These contributions are about 0.018 billion metric tons of carbon/a and 0.11 billion metric tons of carbon/a for China and the world, respectively. Further, by the DBL (Diffusion Boundary Layer)-model calculation, the potential CO_2 sink by carbonate rock dissolution is estimated to be 0.41 billion metric tons of carbon/a for the world. Therefore, the potential CO_2 source by carbonate reprecipitation is 0.3 billion metric tons of carbon/a.

Key words Carbonate rock weathering · Soil CO_2 · Atmospheric CO_2 sink

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Introduction

It is known (Quay 1992; Watson and others 1990) that the combustion of fossil fuels releases about 5.4 billion tons of carbon a year as CO_2 into the atmosphere. In addition, deforestation practices contribute about 1.6 billion tons of carbon a year to atmospheric CO_2. Therefore, the total input of CO_2 from human activities is about 7.0 billion tons of carbon annually. However, only about 3.4 billion tons of carbon a year accumulates in the atmosphere. That means there is an atmospheric CO_2 sink of about 3.6 billion tons of carbon a year.

To accurately predict future CO_2-levels in the atmosphere, which is crucial in predicting climate change, the CO_2 sinks and their change with time must be determined. Although extensive efforts (Berner 1997; Degens and others 1991; Hesshaimer and others 1994; Quay and others 1992; Ritschard 1992; Sarmiento and Sundquist 1992; Siegenthaler and Sarmiento 1993; Tans and others 1990; Yager and others 1995; Yoshimura and Inokura 1997; Yuan 1997) have been made to trace the missing carbon, the explanation is still unclear.

As the world's biggest carbon reservoir, carbonate rocks contain about 6.1×10^7 billion tons of carbon, which is 1694 times and 1.1×10^5 times larger than those of oceans and world vegetation respectively (Houghton and Woodwell 1989). Carbonate rocks occupy an area of about 22 million km^2 in the world (Yuan 1997).

The basic reactions for carbonate rocks weathering can be expressed by:

$$CaCO_3 + CO_2 + H_2O \Leftrightarrow Ca^{2+} + 2\,HCO_3^- \quad (1)$$

for limestone,

$$CaMg(CO_3)_2 + 2\,CO_2 + 2\,H_2O \Leftrightarrow Ca^{2+} + Mg^{2+} + 4\,HCO_3^- \quad (2)$$

for dolomite, here CO_2 may come from the atmosphere directly in bare carbonate rock areas, or from soil in overlying and/or buried carbonate rock regions.

It can be easily visualized from the above reactions that carbonate rock weathering contributes to the atmospheric CO_2 sink. (Note that the consumption of CO_2 in soil by the weathering decreases the release of soil CO_2 into the atmosphere, and thus also contributes to the atmospheric CO_2 sink). For limestone weathering, the removal of 1 mol $CaCO_3$ needs 1 mol of CO_2 from the atmosphere;

and for dolomite weathering, the removal of 1 mol $CaMg(CO_3)_2$ needs 2 mol of CO_2 from atmosphere. It is also clear that only half of the carbon in solution is from atmospheric CO_2 [Eqs. (1) and (2)]. On the other hand, the backward reactions of Eqs. (1) and (2), i.e, reprecipitation of carbonate (e.g. the formation of tufas), are related to the release of CO_2 into the atmosphere. It is very difficult to estimate the net CO_2 flux at individual cases. For example, the corrosion rate of limestone tablets in soil indicates only the CO_2 exhausted in the soil, where the deposition of calcite may occur. In addition, the net amount of CO_2 removed from the atmosphere in a given catchment area is equivalent to the total amount of limestone dissolved and transported outside the area via groundwater flow and/or rivers. Therefore, the net amount can be estimated by the limestone corrosion and the discharge of groundwater and/or rivers.

In this paper, some typical cases are examined using published (Drogue and Yuan 1987; Gong and Huang 1984; Liu 1992; Liu and others 1997; Ogden 1982; Yoshimura and Inokura 1997; Yuan 1997; Zhou and others 1988) and unpublished data. Firstly, the sensitivity of carbonate rock weathering (including the effects by both dissolution and reprecipitation of carbonate) to the change of soil CO_2 and runoff will be discussed, and then the net amount of CO_2 removed from the atmosphere in the carbonate rock areas of mainland China and the world will be determined by the hydrochem-discharge method and carbonate-rock-tablet method, to obtain an estimate of the contribution of carbonate rock weathering to the atmospheric CO_2 sink. It will be shown that these contributions amount to about 0.018 billion metric tons of carbon/a and 0.11 billion metric tons of carbon/a for China and the world, respectively.

Methods

1 CO_2 partial pressure (P_{CO_2}) at a depth of 50 cm in soil was measured in situ monthly with a CO_2-GASTEC meter to monitor the variation in soil CO_2 with time (Liu 1992; Yoshimura and Inokura 1997).
2 Temperature, pH, $[Ca^{2+}]$ and $[HCO_3^-]$ of water were measured monthly in situ with portable pH-meter and alkalinity meter (Liu 1992). The CO_2 partial pressure in water was calculated with the WATSPEC computer program (Wigley 1972), by using the field observation data. These data were used to examine the sensitivity of carbonate rock weathering to the change in soil CO_2 (Liu and others 1997).
3 By using the hydrochemical and discharge data by Li (1992), the contribution of carbonate rock weathering to the atmospheric CO_2 sink was estimated (Liu 1992; Yoshimura and Inokura 1997; Yuan 1997; hydrochem-discharge method).
4 To compare the results by the hydrochem-discharge method, corrosion tests with limestone tablets in the atmosphere and soil (carbonate-rock-tablet-test method) were used (Yuan 1997), and finally the maximum potential contribution by carbonate rock dissolution to the atmospheric CO_2 sink was given by using the DBL Model (Dreybrodt and Buhmann 1991).

Sensitivity of carbonate rock weathering to environmental change

Soil CO_2 change

As examples, two cases will be shown in the following: Figure 1(a) shows seasonal change in $[Ca^{2+}]$, $[HCO_3^-]$ and CO_2 partial pressure (P_{CO_2}) in water, and soil CO_2 partial pressure at the observation site of Yudong (Fishcave) Underground Stream, which is located at Zhen'an County of Shanxi Province, in a climatically transitional zone between North and South China. The mean annual air temperature here is about 11 °C, and mean annual precipitation is 850 mm. The karstified rock is predominately Carboniferous-Permian limestone. Due to the sinkholes in the recharge area, the Yudong Underground Stream is connected to the peak-cluster depressions, where terra rossa and loess formed. The length of the stream is about 30 km, with a catchment area of 85 km^2 and flood peak discharge of about 10 m^3/s.

It can be seen that soil CO_2 partial pressure changes remarkably during a year, with the maximum in the summer growing season, and minimum in cold winter. Related to this, the $[Ca^{2+}]$, $[HCO_3^-]$ and P_{CO_2} in water also show remarkable coincident change. That means that carbonate rock weathering is very sensitive to the soil CO_2 change [refer to Eq. (1)].

The sensitivity of carbonate rock weathering to soil CO_2 change was also found at the Guilin Karst Experimental Site, which is situated in the southeast of Guilin, about 8 km away from Guilin City, and near Yaji village (Fig. 1b). The site is at the boundary of a peak-cluster depression and peak-forest plain. The catchment area of the site is about 1.1 km^2. The strata of the experimental site is mainly pure limestone of Upper Devonian, with thin soil cover in the depressions. The major types of vegetation are bushes and grasses. The annual mean air temperature and the annual mean precipitation are 19 °C and 1900 mm, respectively. Precipitation is the sole recharge to the groundwater in the site (Liu 1992).

In addition to the seasonal change, Figure 1b also shows the increase in soil P_{CO_2} in a multi-year scale. The latter is related to the reforestation at the site since 1993, and the increase in the atmospheric CO_2 content (Harrison and others 1993). The increase in soil P_{CO_2} drives the dissolution of carbonate rock, resulting in the increase in $[Ca^{2+}]$, $[HCO_3^-]$ of karst water (Fig. 1b). This is also proven by the fact that the corrosion flux of limestone tablets in the Guilin experimental site increased from 1993 to 1995 (Table 1).

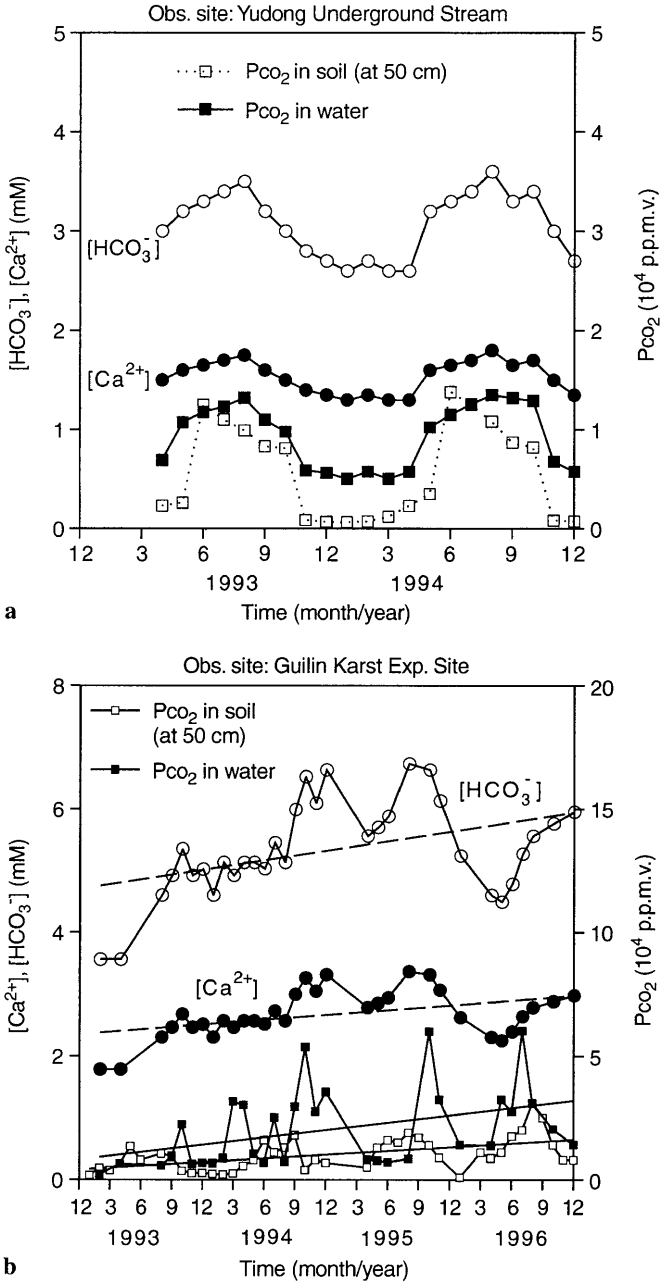

Fig. 1
Seasonal and multi-year change of hydrochemistry and its sensitivity to the change in soil CO_2 partial pressure

Precipitation-evapotranspiration or runoff change

Sensitivity of carbonate rock weathering to the precipitation-evapotranspiration (P-E) or runoff change is reflected in the relationship between the denudation rate of carbonate rocks and runoff. Karst denudation rate is defined as the annual removal of carbonate rock from a carbonate area and is measured in $m^3\ km^{-2}\ a^{-1}$. This unit corresponds to an average lowering of the area by 1 mm in a thousand years (1 mm/ka).

Figure 2 gives some reported denudation rates as a function of (P-E; White 1984; Yoshimura and Inokura 1997).

Table 1
Change in corrosion flux of limestone tablet in the Guilin Experimental site from 1993 to 1995 (unit: $mg \cdot cm^{-2}\ a^{-1}$), – sample lost

Sample location	1993	1994	1995
In the air	3.88	–	4.69
On ground surface	4.29	5.04	5.11
20 cm below the surface	3.79	7.69	10.22
50 cm below the surface	4.71	9.19	11.45

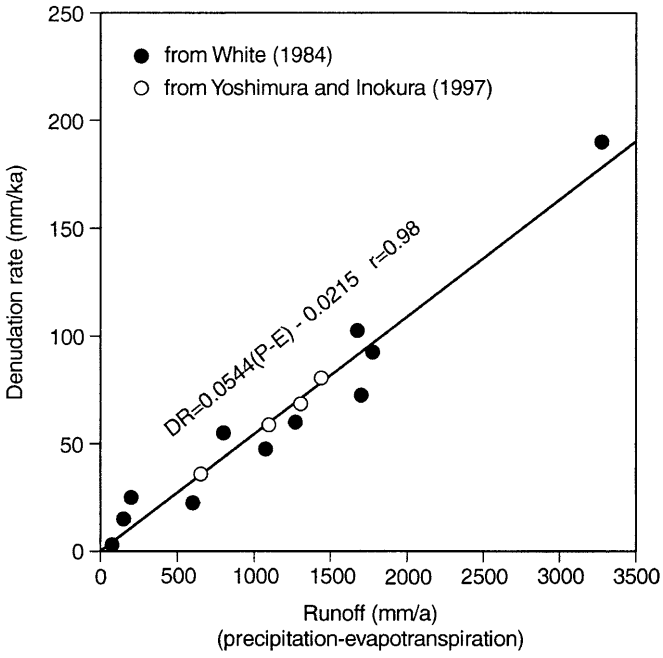

Fig. 2
Relationship between denudation rate of carbonate rocks and runoff

The data points can be fitted by a linear relation $DR = 0.0544(P–E) – 0.0215$ with a correlation coefficient $r = 0.98$. That means the carbonate rock weathering is very sensitive to the runoff, i.e. the larger the runoff, the more intensive the carbonate rock weathering. This may be the main reason for the great difference in karstification between south and north China. It also explains why the contribution of carbonate rock weathering to the atmospheric CO_2 is much larger in south China, where there is a strong runoff, than that in north China.

Estimate of contribution of carbonate rock weathering to the atmospheric CO_2 sink

Hydrochem-discharge method

By using hydrochemical and discharge data, the flux of atmospheric CO_2 consumed in carbonate rock weathering can be estimated by:

Table 2
The relevant parameters to calculate CO_2 sink during carbonate rock weathering in south and north China

Distribution of carbonate rocks	Bare area ($\times 10^4$ km^2)	HCO_3^- content in karst water ($g \cdot l^{-1}$)	Runoff module of karst water ($l \cdot s^{-1}$ km^{-2})	Atmospheric CO_2 sink ($g \cdot a^{-1}$)
South China	44.6	0.236	11.67	1.397×10^{13}
North China	46.1	0.245	2.62	3.366×10^{12}

$$F = 1/2 \times [HCO_3^-] \times Q \times M_{CO_2}/M_{HCO_3} \quad (3)$$

where $[HCO_3^-]$ is HCO_3^- concentration in water (g/l); 1/2 means that only half of the carbon in solution is from atmospheric CO_2 [Eqs. (1) and (2)]; Q is discharge of water in a studied area(l/s), equal to the product of the area and the runoff module; M_{CO_2} and M_{HCO_3} are molecular weights of CO_2 and HCO_3^- respectively.
Table 2 gives the area of the bare carbonate rocks, HCO_3^- concentration in karst water, runoff module of karst water in China (Li 1992), and then the amount of atmospheric CO_2 consumed during carbonate rock weathering in both south and north China could be calculated by the Eq. (3) (Table 2). It is seen that the contribution of carbonate rock weathering to the atmospheric CO_2 sink is 1.397×10^{13} g/a in the bare karst areas of south China, and 0.337×10^{13} g/a in the bare karst areas of north China. The total is 1.734×10^{13} g/a. It would be 6.577×10^{13} g/a (or 0.018 billion metric tons of carbon/a) and 4.206×10^{14} g/a (or 0.11 billion metric tons of carbon/a) if the rate is applied to the whole China karst areas (3.44 million km^2) and the whole world karst area (22 million km^2), respectively.

Carbonate-rock-tablet-test method

Seven monitoring stations which represent the major typical karst areas in China were built (Fig. 3), where corrosion tests of the standard limestone tablet (with a surface area of 28.91 cm^2 and insoluble matter content of 0.97 %) was carried out in the implementation of IGCP 299 *Geology, Climate, Hydrology and Karst Formation* and IGCP 379 *Karst Processes and the Carbon Cycle*(Yuan 1997). Table 3 gives the corrosion test results from six of these seven monitoring stations (one failed due to the loss of the samples), then the amount (A) of atmospheric CO_2 consumed in carbonate rock weathering can be estimated with the following formula:

$$A = F \times S \times C \times M_{CO_2}/M_{CaCO_3} \quad (4)$$

where F is corrosion flux of limestone tablet (g/cm$^2 \cdot$ a), S the area of studied carbonate rock (cm^2), C the $CaCO_3$ content of the limestone tablet, M_{CO_2} and M_{CaCO_3} – the molar weights of CO_2 and $CaCO_3$, respectively.
The calculation results of the amount of atmospheric CO_2 consumed in carbonate rock weathering in various karst types in China are shown in Table 4. The total amount of atmospheric CO_2 consumed in carbonate rock weather-

Fig. 3
Distribution of bare karst areas and monitoring stations of karst system in China, *1* bare carbonate rocks, *2* reef, *3* monitoring station

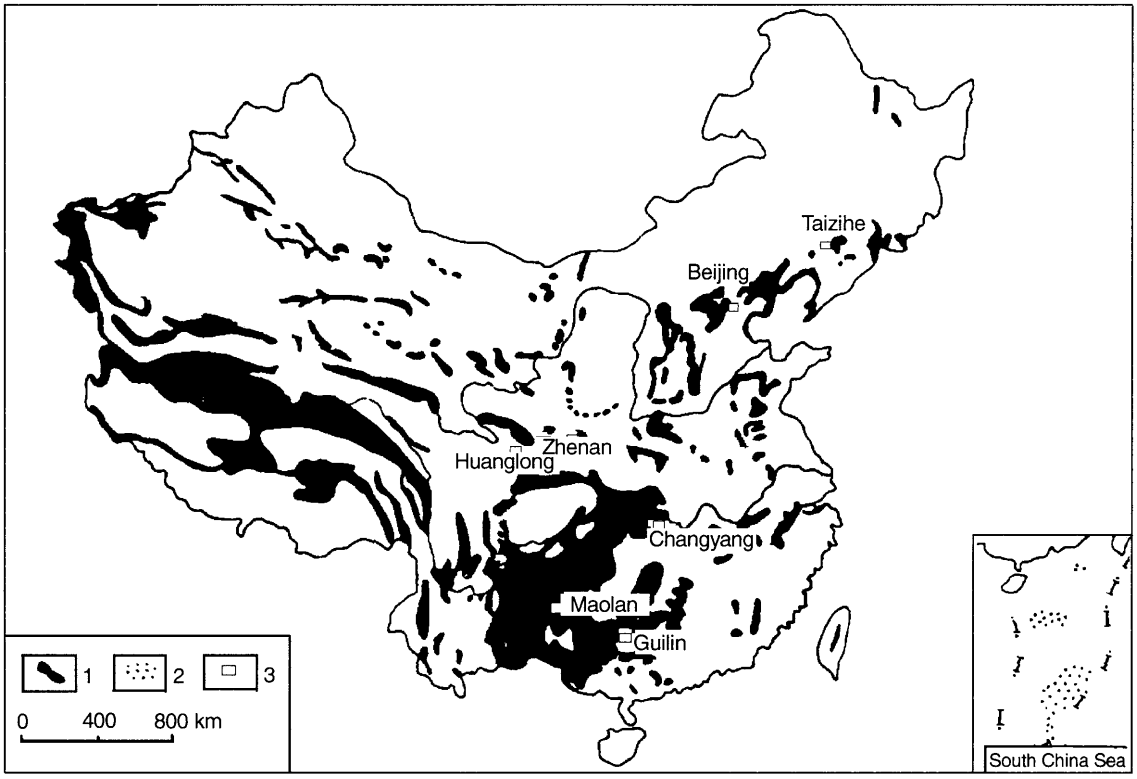

Table 3
Corrosion of the standard limestone tablets in some monitoring stations in 1994 (unit: $g \cdot ^{-1}$), – sample lost

Sample location	Guilin	Maolan	Huanglong	Zhen'an	Beijing	Taizihe
in the air	0.1356[a]	0.0290	0.0151	0.0068	–	0.0136
Ground surface	0.1456	0.0380	0.0170	0.0013	0.0336	0.0048
Average flux (10^{-4} g·cm^{-2}·a^{-1})	48.634	11.588	5.569	1.557	11.622	3.182
20 cm below soil surface	0.2222	0.1670	0.1690	–	0.0021	0.0014
50 cm below soil surface	0.2657	0.0520	0.1920	–	0.0018	0.0014
average flux (10^{-4} g·cm^{-2}·a^{-1})	84.399	37.876	62.435	–	0.675	0.484

[a] the value in 1995

Table 4.
Distribution of the main karst types and the atmospheric CO_2 sink during carbonate rock weathering in China

Karst type	Distribution	Bare area (10^4 km^2)	Mean annual rainfall (mm)	Representative station	CO_2 sink (g·a^{-1})
Tropical and sub-tropical karst	Guangdong, Guangxi, Taiwan, Zhejiang, Yunnan, Guizhou, Hunan, Jiangxi, west Hubei etc.	44.6	1000 – 1850	Guilin	1.305×10^{13}
High mountain and plateau karst	West Sichuan, Tibet, Guilun Mountains	22.1	300 – 800	Huanglong	3.306×10^{12}
Semiarid karst	Shanxi, Hebei, Henan, north Shangxi, Shandong etc.	21.4	400 – 600	Beijing	5.789×10^{11}
Humid temperate karst	Taizihe River basin Huai River basin	2.6	800 – 1000	Taizihe	2.097×10^{10}

ing is about 1.696×10^{13} g/a in China bare karst areas. It would be 6.432×10^{13} g/a and 4.114×10^{14} g/a if this value is applied to the whole China karst areas and the whole world karst areas, respectively, which are very close to those obtained by the hydrochem-discharge method.

DBL-model calculation

According to Liu and Dreybrodt (1997), the dissolution rate of calcite in CO_2-H_2O solutions with turbulent motion can be approximated by a linear rate law $R = \alpha(C_{eq}\text{-}C)$, where c_{eq} is the equilibrium concentration with respect to calcite and α a rate constant, dependent on temperature T, CO_2 partial pressure P_{CO_2}, DBL (diffusion boundary layer) thickness ε and the thickness of the water sheet flowing on the mineral δ. If we take $T = 10\,^{\circ}C$, $P_{CO_2} = 5 \times 10^{-3}$ atm, $\delta = 1$ cm and $\varepsilon = 5 \times 10^{-3}$ cm, which are the reasonable mean values in nature (Dreybrodt 1988), then $\alpha = 2.03 \times 10^{-5}$ cm/s and $C_{eq} = 1.62 \times 10^{-3}$ mmol/cm^3 . Taking the average value 2×10^{-4} mmol/cm^3 of [Ca^{2+}] in rainfall, $R = 2.88 \times 10^{-8}$ mmol cm^{-2} s^{-1} were obtained. This corresponds to 288 mm/ka if water runs down continuously. Assuming rainfall to occur only during 20 % of the time (Dreybrodt 1988), an annual retreat of bedrock of about 57.6 mm/ka will result. That means potential atmospheric CO_2 sink by carbonate rock dissolution is estimated to be 2.354×10^{14} g/a (or 0.0642 billion metric tons of carbon/a) and 1.505×10^{15} g/a (or 0.41 billion metric tons of carbon/a) in all of China's carbonate rock area and the whole world's carbonate rock area, respectively. These values are about 3.6 times those obtained by the hydrochem-discharge method or by carbonate-rock-tablet-test method (average 0.018 billion metric tons of carbon/a and 0.11 billion metric tons of carbon/a for China and the world, respectively). The latter may represent the net effect of carbonate dissolution and reprecipitation. So, carbonate reprecipitation releases 0.3 (0.41 – 0.11) billion metric tons of carbon/a to the atmosphere for the world.

Conclusion

The net contribution of carbonate rock weathering to the atmospheric CO_2 sink is about 0.11 billion metric tons of carbon/a by both the hydrochem-discharge method and carbonate-rock-tablet-test method. The potential value by carbonate dissolution, according to the DBL-model calculation, is about 0.41 billion metric tons of carbon/a, while the release of CO_2 by carbonate reprecipitation is 0.3 billion metric tons of carbon/a.

Moreover, according to the data from the Guilin monitoring station during 1993 – 1995 (Fig. 1), the consumption of atmospheric CO_2 during carbonate rock weathering increased from 6.129×10^9 g c/a in 1993 to 11.582×10^9 g c/a in 1995 due to the increase of soil CO_2, which was related to reforestation and global increase in atmospheric CO_2. This means that the contribution of carbonate rock weathering to the atmospheric CO_2 sink increases with the lifting of the atmospheric CO_2 content. Therefore, the carbonate rock functions as an adjustor to the atmospheric CO_2.

It is worthwhile to note that there is quite a difference between the estimates of contribution of carbonate rock weathering to the atmospheric CO_2 sink by different researches, though ours is very close to that (0.1×10^9 tons C/a) of Ichikuni (1976). Yoshimura and Inokura

(1997) and Kitano (1984) obtained the value of about 0.2 billion metric tons of carbon/a; Yuan (1997) reported the value of 0.6 billion metric tons of carbon/a; and others (Sarmiento and Sundquist 1992) obtained 0.4 billion metric tons of carbon/a. Therefore, further work is needed for more accurate calculations, especially under the condition of lifting atmospheric CO_2 content.

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