CHEMICAL WEATHERING PROCESSES AND ATMOSPHERIC CO₂ CONSUMPTION OF HUANGHE RIVER AND CHANGJIANG RIVER BASINS

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ABSTRACT: Rock weathering plays an important role in studying the long-term carbon cycles and global climatic change. According to the statistics analysis, the Huanghe (Yellow) River water chemistry was mainly controlled by evaporite and carbonate weathering, which were responsible for over 90% of total dissolved ions. As compared with the Huanghe River basin, dissolved load of the Changjiang (Yangtze) River was mainly originated from the carbonate dissolution. The chemical weathering rates were estimated to be $39.29t/(km^2 \cdot a)$ and $61.58t/(km^2 \cdot a)$ by deducting the HCO_3^- derived from atmosphere in the Huanghe River and Changjiang River watersheds, respectively. The CO₂ consumption rates by rock weathering were calculated to be $120.84 \times 10^3 mol/km^2$ and $452.46 \times 10^3 mol/km^2$ annually in the two basins, respectively. The total CO₂ consumption of the two basins amounted to $918.51 \times 10^9 mol/a$, accounting for 3.83% of the world gross. In contrast to other world watersheds, the stronger evaporite reaction and infirm silicate weathering can explain such feature that CO₂ consumption rates were lower than a global average, suggesting that the sequential weathering may be go on in the two Chinese drainage basins.
KEY WORDS: Huanghe River basin; Changjiang River basin; chemical weathering rate; CO₂ consumption

1 INTRODUCTION

The atmospheric CO₂ consumed by rock weathering has long been recognized as providing a major loss of carbon and decreasing the atmospheric CO₂ (STALLARD and EDMOND, 1981; MEYBECK, 1983). The $0.7 \times 10^9 - 0.8 \times 10^9 t$ of carbon derived from atmospheric CO₂ is transferred from atmosphere and biosphere to the ocean by surface weathering processes (SUCHET and PROBST, 1993, 1995).

In order to monitor the quality of river water, it is also important to understand the natural flux of dissolved ions. So far, most studies on water chemistry in China had focused on water quality and dissolved fluxes (HU *et al.*, 1982; QU *et al.*, 1993; CHEN *et al.*, 1985, 2000; XIA *et al.*, 2000; ZHANG *et al.*, 1990, 1995), scarce literatures could be used to understand the sources of solute loads and CO₂ consumption in China. However, many scholars had attempted to fill in the gap in our knowledge of atmospheric CO₂ consumption by rock weathering and tried to link the water chemistry and weathering reactions in the major world watersheds (GAILLARDET *et al.*, 1997, 1999a, 1999b; ROY *et al.*, 1999; GALY and FANCE-LA-NORD, 1999; DATTA and SUBRAMANIAN, 1997; BOEGLIN and PROBST, 1998).

The main objective of this study will provide the different contribution proportions to the major dissolved ions and estimate the average chemical weathering rates by deducting the HCO_3^- derived from atmosphere in the Huanghe (Yellow) River and Changjiang (Yangtze) River drainage basins where previous information was indeed limited. The paper focused on the consumption of atmospheric CO₂ by rock weathering and the weathering reactions that were attributable for dissolved loads in the two drainage basins.

Hundreds of water samples from the upper reaches to lower reaches of the two rivers were collected and

stored in acid-washed polypropylene bottles from 1999 to 2001, after collection, all samples were filtered in time through 0.45μm acetate fibrous filters. The analytical procedure for water analysis included conventional flame atomic absorption spectrophotometry for Ca, Na, Mg and K (analytical precision higher than 10%), and hydrochloric acid titration for HCO_3^-. The analysis of Cl^- and SO_4^{2-} were performed by $AgNO_3$ titration and $BaSO_4$ nephelometery. Dissolved silica was measured by spectrophotometric measurement of the Mo blue complex. Analytical errors in concentration measurements were estimated to be less than 2%. The mass balance was tested within 1±10% between anion and cation equivalent concentrations.

2 WEATHERING PROCESSES DEDUCED FROM WATER CHEMISTRY

2.1 Water Chemistry and Weathering Reactions of the Huanghe River Basin

The Huanghe (Yellow) River water has very higher ion concentration of 300–900mg/L, with an average of 600mg/L, in comparison with a world medial river (100mg/L). The content order on an equivalent basis was: $Na^+>Ca^{2+}>Mg^{2+}>K^+$ among all cations. The Ca^{2+} and Na^+ account for 75% of total dissolvedions (in μeq/L). HCO_3^- is the most abundant anion; it constitutes 30%–50% of total dissolved ions (in μeq/L). The weighted soluble silica concentration was around 120μmol/L.

The ratios of major ions in the river water are relatively constant and usually imply the main weathering processes. Atmospheric input proportion of Cl^- is less than 0.1% in the Huanghe River water, additionally Cl^- behaves conservatively in the surface water, it can initially assumed that Cl^- is entirely derived from the dissolution of evaporite minerals.

A distinct positive correlation between Cl^- and Na^+ was found for all the Huanghe River water samples, which suggests that Cl^- and Na^+ were of evaporite origin in the Huanghe River basin. The Na^+ concentration was also significantly correlated with SO_4^{2-}, reflecting that a large proportion of dissolved Na^+ and SO_4^{2-} and Cl^- could be mainly originated from the evaporite weathering (correlation coefficients r=0.90, 0.93, p<0.0001).

The equivalent concentration ratios Cl^-/Na^+ of halite reaction and SO_4^{2-}/Na^+ of Glauber′s salt dissolution are 1.00 and 2.00, respectively. The better positive correlation between Na^+ and ($SO_4^{2-}+Cl^-$) (r=0.95, p<0.0001) and the ratio ($SO_4^{2-}+Cl^-$)/Na^+ =1.62 showed that only 38% of Na^+ come from halite dissolution, the Glauber′s

salt weathering reaction would offer the remainder 62% Na^+ if SO_4^{2-} and Cl^- were only derived from dissolution of the two minerals. As a matter of fact, SO_4^{2-} may come from other mineral origins (e.g., gypsum) because a good relationship was also found between Ca^{2+} and SO_4^{2-} even if most of SO_4^{2-} might be from Glauber′s salt dissolution yet.

High alkalinity was closely related to divalent cations ($Mg^{2+}+Ca^{2+}$) (r=0.70, p<0.001), more than to Mg^{2+} or Ca^{2+}, suggesting that dolomite $CaMg(CO_3)_2$ dissolution may contribute more solutes than calcites. The average ratio 1.46 of ($Ca^{2+}+Mg^{2+}$)/(Na^++K^+) was 34% lower than the world average value of 2.20, which indicates that the effect of evaporite weathering on the dissolved ions might overstep carbonate dissolution, despite the contribution of dissolved Mg^{2+} and Ca^{2+} from carbonate weathering would still dominate.

Silicate weathering consumed atmospheric CO_2, which was transferred into river in the form of HCO_3^-, and only this weathering process produced Si. The positive correlations were found among dissolved Si and Na^+, HCO_3^-, which implied that sodium silicates (for instance, albite dissolution) maybe had undergone weathering and erosion. However, the low Si concentration and slight relationship between Si and Na^+ showed that silicate weathering developed weakly.

For the major ions, component loadings of the Huanghe River water chemistry were calculated by the principal component analysis (Varimax with Kaiser Normalization) and 3 major factors were obtained by statistics software SPSS10.0. The relative contribution proportions for the three factors to total coefficient of variation were 36.5%, 33.3% and 15.4%, respectively. The factor 1 exhibited a significant correlation with SO_4^{2-} and Cl^- and Na^+, likely representing the evaporite weathering; the factor 2 with HCO_3^- and Ca^{2+} and Mg^{2+}, denoting the carbonate dissolution; and the factor 3 with Si and K^+, implying the silicate weathering. The similar contribution proportions for the factor 1 and the factor 2 were far higher than the factor 3, suggesting the intensive carbonate and evaporite weathering and slight silicate dissolution in the Huanghe River basin. The results were almost consistent with that the high ($HCO_3^-+Ca^{2+}+Mg^{2+}$) and ($Na^++SO_4^{2-}+Cl^-$) concentrations accounted for 47% and 52% of total ion equivalent concentrations as above analysis, respectively.

The contributions to the dissolved load of the different end-members varied greatly, the relative contribution proportions of carbonates, evaporites and silicates occupied 32.8%, 54.7% and 3.23% (Table 1) of total dissolved loads in the Huanghe River watershed, respec-

Table 1 Contribution proportions of rock and atmospheric CO_2 in the Huanghe River and Changjiang River watersheds of China versus the global average (%)

Source	River	Mg^{2+}	Ca^{2+}	Na^+	K^+	Si	HCO_3^-	SO_4^{2-}	Cl^-	Contribution proportion
Carbonate	Huanghe	72.0	96.4	0	0	0	45.0	0	0	32.80
	Changjiang	14.8	93.0	0	0	0	45.7	0	0	46.90
	World	42.0	67.0	0	0	0	33.0	0	0	35.00
Evaporite	Huanghe	24.1	3.2	97.6	1.1	0	0	93.2	100	54.70
	Changjiang	7.5	0.2	100.0	42.3	0	0	70.0	100	18.00
	World	10.0	7.0	54.0	5.0	0	0	42.0	100	11.00
Silicate	Huanghe	3.9	0.4	2.40	98.9	100	0	6.8	0	3.23
	Changjiang	77.7	6.8	0	57.7	100	0	0	0	12.90
	World	48.0	26.0	46.0	95.0	100	0	58.0	0	15.00
Atmosphere	Huanghe	0	0	0	0	0	55.0	0	0	9.32
	Changjiang	0	0	0	0	0	54.3	0	0	19.60
	World	0	0	0	0	0	67.0	0	0	37.00
Others	Changjiang	0	0	0	0	0	0	30.0	0	2.65
	World	–	–	–	–	–	–	–	–	2.00

tively, based on the component analysis loadings. Namely, 90% of solutes came from carbonate and evaporite weathering, only 9.32% was related with atmospheric CO_2 input. Silicate weathering merely produced 3.23% of total solutes in the Huanghe River.

The result was consistent with those from the bedrock underlain in the Huanghe River drainage basin. Loess terrain covered 40% of drainage area, but contributed 90% of sediment load to the Huanghe River. The loess enriched the highest evaporite minerals (5%–10 %) and $CaCO_3$ minerals (WU *et al.*, 1996). In terms of water/rock interaction, over 90% of the input of ions to the Huanghe River water was largely controlled by the dissolution of carbonate and evaporite minerals. Undoubtedly, the total component loadings did not amount to 100% and could not absolutely correspond to three lithologies. Whereas to some extent, the numerals only reflect the relative intensity of weathering reactions and the contribution percentage could not express accurately as numerals itself.

In most rivers of the world, the dissolved ions input was controlled by silicate and carbonate weathering throughout the hydrological cycle. The carbonate and silicate weathering processes usually contribute over 50% of river solutes on a global scale, while evaporite weathering only accounts for 11% of dissolved ions averagely (GAILLARDET *et al.*, 1999b). However, the relative contribution percentage of evaporites was up to approximately 55% in the Huanghe River watershed. So an outstanding characteristic was its dominant contribution of evaporites and nearly neglectable effect of silicates on the water chemistry in the Huanghe River drainage basin.

2.2 Water Chemistry and Weathering Reactions of the Changjiang River Basin

The total ion charge of the Changjiang River water ranged between 140mg/L and 180mg/L with an annual average of 160mg/L. The cation abundance sequence was: $Ca^{2+}>Na^+\geqslant Mg^{2+}>K^+$. ($Ca^{2+}+Mg^{2+}$) accounted for over 80% of the cations and anions, while the Na^+, SO_4^{2-} and Cl^- were responsible for about 20% of the total equivalent concentrations. On average, weighted soluble silica concentration was about $145\mu mol/L$.

The relationship between HCO_3^- and ($Ca^{2+}+Mg^{2+}$) concentration showed significantly positive correlations, and was close to 1:1 ratio (0.82) as dolomite dissolution in the Changjiang River. In addition, the highest HCO_3^- and ($Ca^{2+}+Mg^{2+}$) concentrations suggested that dissolution of carbonate minerals, especially dolomite and calcite, added significant amounts of Ca^{2+} and Mg^{2+} to the river. Nevertheless, the sum of divalent cations ($Ca^{2+}+Mg^{2+}$) was higher than HCO_3^-, indicating a majority of Ca^{2+} and Mg^{2+} were from carbonate weathering and a small part from other origins.

($Ca^{2+}+Mg^{2+}$) was well balanced by ($HCO_3^-+SO_4^{2-}$) and their equivalent concentration ratio (0.84, <1.00) illustrated that ($Ca^{2+}+Mg^{2+}$) might be partly combined with SO_4^{2-}, namely the gypsum dissolution furthest probably took place because SO_4^{2-} was found to clearly correlate with Ca^{2+} ($r=0.82$) than other cations. Certainly, in our present study, the carbonate weathering processes mainly controlled the HCO_3^- and ($Ca^{2+}+Mg^{2+}$) concentrations in the Changjiang River basin.

The Na^+ concentration was only less than Ca^{2+} concentration, and the obvious relationships were gained

between Na^+ and Cl^- ($r=0.83$, $p<0.001$) and between Na^+ and SO_4^{2-} ($r=0.67$, $p<0.001$) for the Changjiang River water, implying the evaporites (such as halite and Glauber′s salt) dissolution was a major source of Na^+, Cl^- and SO_4^{2-} to the river water.

It could be assumed that all Cl^- were derived from dissolution of evaporite minerals as described above, then Cl^- should balance Na^+. If Na^+ was only derived from dissolution of evaporite minerals, the equivalent concentration ratio of $(Na^+-Cl^-)/SO_4^{2-}=0.28$ indicated that only 56% of SO_4^{2-} was from the dissolution of Glauber′s salt ($Na^+/SO_4^{2-}=0.50$). Other potential sources of SO_4^{2-} might be the weathering of gypsum as above discussed and anthropogenic input.

Silica of stream water is mostly completely derived from silicate weathering, producing HCO_3^- by using atmospheric CO_2 into the stream water. No evident relationship between Si and HCO_3^- was found in the Changjiang River samples, which implied that silicates contributed only a few solutes to the Changjiang River, as compared to the remarkable carbonate weathering. The Si correlated only with Ca^{2+} among all cations, reflecting that anorthite weathering maybe be primary silicate weathering.

Component loadings of the Changjiang River water chemistry were calculated by the principal component analysis and 3 major factors were obtained. The relative contribution proportions for the factor 1, factor 2 and factor 3 to total coefficient of variations were 75.8%, 12.7% and 8.41%, respectively. The factor 1 exhibited a close correlation with Ca^{2+}, Mg^{2+} and HCO_3^-, indicating that there are mainly carbonate weathering; the factor 2 with Ca^{2+} and Si, and the factor 3 with Na^+, Cl^- and SO_4^{2-}, denoting the silicate and evaporite dissolutions, respectively. The similar contribution proportions for the factor 2 and the factor 3 were far less than that of the factor 1, suggesting carbonates weathered intensively and silicates and evaporites dissolved slightly in the Changjiang River basin. The results were almost consistent with the above correlation analysis.

The component analysis loadings displayed that about one half of dissolved elements with a carbonate origin were exported, 18.0% with an evaporite origin, 12.9% with a silicate origin, and 19.6% from atmospheric CO_2 in the Changjiang River drainage basin (Table 1). The evaporites and silicates supported the similar dissolved loads and totally contributed 31% of the river solutes.

By this token, about 80% of the total annual dissolved flow was produced by rock weathering, the remainder were mainly imported by atmospheric CO_2 in the form of the HCO_3^- into the Changjiang River, while the an-

thropogenic SO_4^{2-} may contribute to 2%–3% of total dissolved fluxes.

So the water chemistry and weathering reactions were controlled by the dolomite and calcite hydrolyzation in the Changjiang River basin. The complex rock compositions, including abundant sedimentary bedrock, widely-distributed carbonate minerals, and igneous rocks, developed widely in the Changjiang River watershed. Especially carbonate minerals plentifully outcroped in the Changjiang headstream and its important tributaries (e.g., the Wujiang River and the Minjiang River), and rapid weathering rate of carbonate minerals might bring on the above carbonates-controlled weathering characteristic.

In summary, the major ion chemistry of the Huanghe River and the Changjiang River basins were dominated by dissolution of evaporite and carbonate minerals, respectively. The silicate weathering was of minor importance on the water chemistry for the both basins, but in contrast to the Huanghe River basin, the silicate weathering degree of the Changjiang River basin might be stronger because the dissolved Si concentration and relative contribution percentage of silicates of the Changjiang River basin were always higher than that of the Huanghe River basin.

3 CHEMICAL WEATHERING RATES

The river chemistry discussed above only provided information on the weathering processes, not on chemical weathering rates and mass transfer from the basins. By combining chemical data with discharge and area information, the chemical weathering rate could be calculated by normalizing the weighted average annual fluxes from rock weathering over the basin area. However, the atmospheric CO_2 participated in the weathering processes and occurred in the form of HCO_3^- of the stream water. The correcting atmospheric CO_2 loading was hardly engaged scholars′ attention in the past, in fact, for example, the HCO_3^- derived from atmospheric CO_2 usually dominates total solutes in most rivers. Accordingly, it is very necessary to firstly correct the measured water chemistry for the non-weathering components as atmospheric CO_2.

During carbonate weathering process, only one half the HCO_3^- in the river water is from rock weathering, the reminder from atmospheric CO_2. Furthermore, all HCO_3^- for the silicate weathering come from atmospheric CO_2, while the evaporite weathering hardly consumed atmospheric CO_2.

For the Huanghe River, the ion concentration by

rock weathering was 490.20mg/L averagely, deducting the HCO_3^- from atmospheric CO_2. Due to our lack of knowledge of the carbonate and silicate outcrop areas, the weathering rate was simply calculated using the total basin area. So the average chemical weathering rate was estimated to be 39.29t/(km²·a) according to the relative contribution proportions from different origins as Table 1 showed, which was close to the world mean value of 36t/(km²·a).

For the Changjiang River basin, the dominated carbonate weathering supplied 45.7% of HCO_3^-, the remainder 54.3% of HCO_3^- were derived from atmospheric CO_2. Thus, 122.5mg/L of ion concentration of the stream water was wholly from rock weathering. So on average, the chemical weathering rate was estimated to be 61.58t/(km²·a), 1.6 times higher than the Huanghe River basin. The results reflected that the eroded flux by chemical weathering in unit area in the Changjiang River basin was obviously higher than that in the Huanghe River basin. The warmer climate and more abundant precipitation might correspond with the higher weathering rate in the Changjiang River basin.

4 ATMOSPHERIC CO_2 CONSUMPTION BY ROCK WEATHERING

The dissolution of carbonates produces two mole of HCO_3^- for one mole of $(Ca^{2+}+Mg^{2+})$, but only one is derived from the atmosphere. By contrast all the HCO_3^- derived from silicates were originated from the atmosphere. The inorganic consumption of CO_2 by rock weathering can be calculated by using the proportions of HCO_3^- derived from silicates and carbonates calculated above.

For the Huanghe River basin, 55% of HCO_3^- was derived from atmospheric CO_2 by rock weathering, while 45% of HCO_3^- was supported by the carbonates. Therefore, the average HCO_3^- concentration from atmospheric CO_2 was 1.56mol/m³, and the atmospheric CO_2 consumption rates resulting from silicate and carbonate weathering were up to 22.00×10^3mol/km² and 98.84×10^3mol/km² in the Huanghe River drainage basin, respectively (Table 2). The results indicated that CO_2 consumption rate by silicate weathering was only 1/4 lower than that of carbonate weathering in the Huanghe

Table 2 Weathering rate and CO_2 consumption for the Huanghe River and the Changjiang River basins compared with the world watersheds

Basin	Runoff ($\times 10^9$m³)	Chemical weathering rate (t/(km²·a))	CO_2 consumption rate by silicate weathering ($\times 10^3$mol/(km²·a))	CO_2 consumption rate by carbonate weathering ($\times 10^3$mol/(km²·a))	CO_2 consumption ($\times 10^9$mol/a)
Huanghe	58.02	39.29	22.0	98.84	90.51
Changjiang	920.00	61.58	380.8	71.66	828.00
World*	37400.00	36.00	100.9	145.10	24000.00

 * After GAILLARDET *et al*., 1996b

River basin. The estimations of total CO_2 consumption of 90.51×10^9mol/a were similar to those from GAILLARDET *et al*. (1999b) based on the lithologic model.

Similarly, on average, 0.9mol/m³ concentration of HCO_3^- in the Changjiang River water was from atmospheric CO_2. Hence, the CO_2 consumption rate of the Changjiang River basin was calculated to be 452.46×10^3mol/(km²·a). The total annual CO_2 consumption amounted to 828.00×10^9mol/a, and 84.16% from carbonate weathering, 15.84% from silicate weathering. Compared with the world average atmospheric CO_2 consumption rate of 246×10^3mol/(km²·a), the Changjiang River basin was 1.84 times higher than the world average. The annual CO_2 consumption fluxes of the Huanghe River and Changjiang River basins were close to 0.38% and 3.45% of the total world watersheds, respectively. In comparison, the atmospheric CO_2 consumed by weathering for the Changjiang River basin was 10 times higher than that for the Huanghe River basin.

As usual, carbonate weathering is more productive for the dissolved loads than silicate weathering. We estimated a total carbon flux of consumed CO_2 of 248.5×10^3t C in the Changjiang River and the Huanghe River watersheds where carbonate weathering equally dominated the atmospheric CO_2 sink. In the Huanghe River and the Changjiang River basins, the results tended to ascribe a minor importance to silicate weathering compared to the world average for the uptake of CO_2 from the atmosphere, especially in the Huanghe River basin without obvious dissolution of silicate minerals, which maybe imply that the rock would be continuously undergone erosion in the future.

5 CONCLUSIONS

The major components were used to evaluate the weathering reactions in two large drainage basins of China. The dissimilar weathering processes determined the dis-

tinct differences from water chemistry and CO$_2$ consumption rates in the Huanghe River and the Changjiang River basins.

The contribution percentage of evaporite weathering on the water chemistry would approach to one half; the effect of carbonate weathering on the solutes took second place in the Huanghe River basin. No pronounced effect of silicate weathering on the Huanghe river chemistry was found.

For the Changjiang River basin, carbonate dissolution controlled the major dissolved ions such as HCO$_3^-$ and (Ca^{2+}+Mg^{2+}). The silicate and evaporite weathering processes of the Changjiang River basin were similarly weak without prominent contribution to the river, even if its silicate weathering might be stronger than the Huanghe River basin.

The chemical weathering rates of the Huanghe River and the Changjiang River basins were about 39.29 t/(km^2·a) and 61.58t/(km^2·a), respectively, higher than the global average. The average CO$_2$ consumption rates of the Huanghe River and the Changjiang River basins were about 120.8 ×10^{3}mol/km^2 and 452.5 ×10^3 mol/km^2, respectively. The rate of CO$_2$ consumption by rock weathering in the Huanghe River basin is about a quarter of the Changjiang River basin, but in the same order of magnitude. The CO$_2$ consumption in the Changjiang River basin was estimated to be 828× 10^{9}mol/a，10 times approximately higher than the Huanghe River basin. The total CO$_2$ consumption amounted to 918.5×10^{9}mol/a for the two major basins. The difference was mostly attributable for stronger silicate weathering and plenty runoff in the Changjiang River basin.

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