Hydrogeochemistry of Wujiang River Water in Guizhou Province, China*

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Abstract: The chemical composition of Wujiang River water represents that of river water from the typical carbonate areas. Its hydrogeochemical characteristics are different from those of global major rivers. The Wujiang River and its tributaries have high total dissolved solid concentrations, with Ca^{2+} and HCO_3^- being dominant, Mg^{2+} and SO_4^{2-} coming next. Both $Na^+ + K^+$ and $Cl^- + Si$ account for 5% - 10% of the total cations and anions, respectively. These general features show the chemical composition of river water is largely controlled by carbonate weathering, with the impact of silicate and evaporate weathering being of less importance. Production activity, mining practice and industrial pollution also have some influence on the chemical composition of river water.

Key words: **carbonate; chemical weathering; water chemistry; river water; karst**

The geochemical study of river water permits us to develop much important imformation on the chemical weathering of catchment, climate, the chemical and isotopic compositions of the upper continental crust (UCC) and the cycling of elements in the continent-river-ocean system (Gibbs, 1972; Reeder et al., 1972; Hu et al., 1980; Stallard and Edmond, 1983, 1987; Goldstein and Jacobsen, 1987; Elderfield et al., 1990; Huh et al., 1998; Zhang, 1995). Because the product of carbonate weathering controls the geochemical composition of river water in some degree, systematic studies of the geochemical composition of river water in carbonate areas will help us to know the relation between chemical weathering in carbonate areas and hydrogeochemical characteristics of river water and to understand the factors controlling the geochemical composition of river water.

This paper focuses on the hydrogeochemistry of Wujiang River water in Guizhou karst areas of southweatern China, in an attempt to know the general geology of the drainage areas and understand the chemical weathering of rock/soil in karst areas and assess the impact of human activities on the environment.

General Geology of the Drainage Areas

Guizhou Province is located in the center of the Southeast Asia Karst Region where karstification is most developed, karst types are most diverse and the distribution area is largest in the world. The carbonate rocks of Guizhou Province cover an area of approximately 130×10^3 km^2,

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which accounts for 73.6% of the whole province (Wan Guojiang et al., 1995). The Wujiang River originates in the Wumeng Ranges on the Yunnan-Guizhou Plateau and winds its way through four provinces including Yunnan, Guizhou, Sichuan and Hubei. The Wujiang River is the largest tributary in the upper reaches of the Changjiang River and also is the largest river in Guizhou Province with a total length of 874.2 km and a mean water discharge of 534×10^8 $m^3 \cdot a^{-1}$(Zhang Licheng et al., 1996).

The Wujiang River drainage system is located in the karst areas of Guizhou Province. The catchment represents many different and complicated lithologies. The strata exposed in the Wujiang River catchment consist mostly of the Pre-Jurassic system, and carbonate rocks are widely spread. The upper reaches of the Wujiang River Valley are located in the west of Guizhou Plateau and are dominated by Permian and Triassic carbonate rocks, coal-bearing formations and basalts. In the middle reaches are widely distributed the Permian and Triassic limestones, dolomitic limestones and dolomites. While in the lower reaches are distributed carbonate rocks, as well as shales, sandy shales and siltstones. A large number of tributaries were sampled. In consideration of their complicated geographic names, all the samples are marked by numbers as shown in Fig. 1.

Sampling and Analytical Methods

This work is focussed on the variations of the concentrations of the dissolved major elements Ca, Mg, K, Na, HCO_3^-, SO_4^{2-}, Cl^- and NO_3^-. Thirty-seven river water samples were collected during the period from Jan. 7 to 27, 1999. Water temperature, dissolved oxygen, pH and conductivity were measured at the sample localities with a portable pH and SC (salt conductivity) meter. HCO_3^- was titrated by HCl on the spot. The river water samples were collected by way of placing an acid-cleaned linear polyethylene bottle in a plastic holder. Immediately after collection, all the samples were filtered through 0.22 μm membrane filters (Millipore) and a small portion of these samples was stored for measuring anions, while another portion was acidified with ultra-pure hydrochloric acid ($pH<2$) for measuring cations. All the samples were stored in darkness. Major cations (K, Na, Ca, Mg) were determined by AAS. Anions (Cl^-, SO_4^{2-}, NO_3^-) were measured by anion chromatography. The PO_4^- concentrations of all the samples are at or beyond the detection limit ($\sim 0.01 \times 10^{-6}$). Uncertainties involved in major ion analyses were estimated to be less than ±5%. Silica was measured by colorimetry.

During the sampling period, the flow of the Wujiang River was characterized by minimum discharge in winter. The concentrations of dissolved major elements are listed in Table 1.

Results and Discussion

Physical and chemical parameters and the total concentrations of dissolved ions

River water samples present the pH values ranging from 7.9 to 8.9, with a mean value of 8.4. These relatively high pH values revealed the important effects of the dissolution of limestones and dolomites in the watershed on the chemistry of river water. The river water samples vary in conductivity from 0.046 to 1.269 mS/cm with a mean value of 0.407 mS/cm. The total amount of dissolved cations in the river water is linearly correlated with conductivity.

Sample No. 14 had been polluted by waste water discharged from factories, and its ion concentrations are several times higher than those of the other samples. Except sample No. 14, the

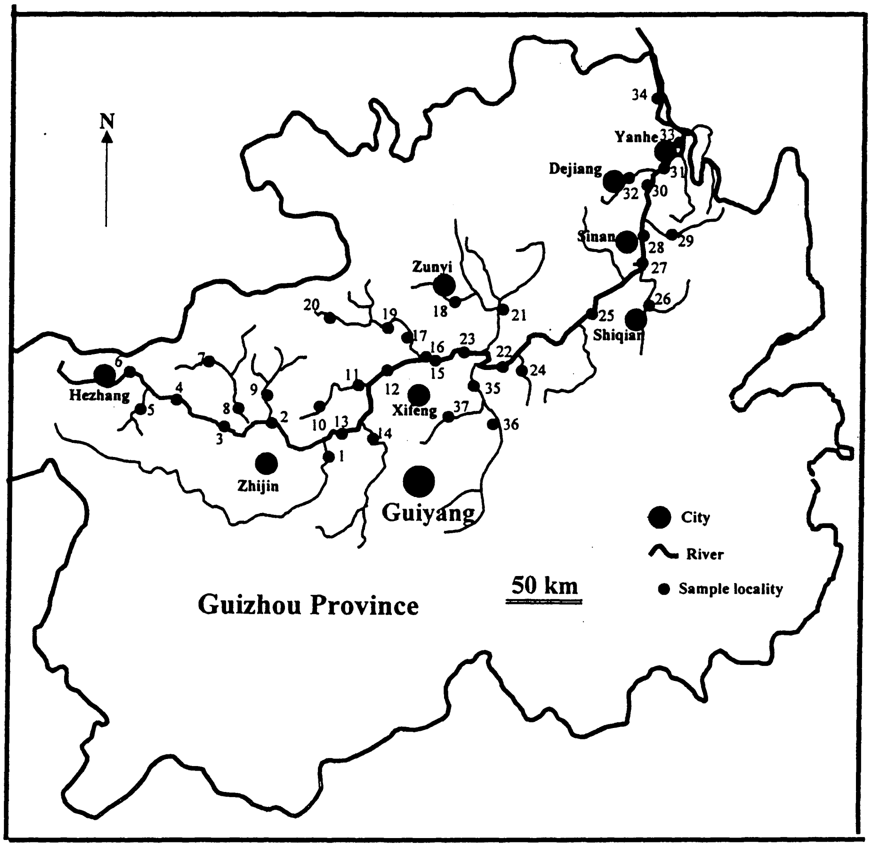

Fig. 1. The map showing the sample localities and the distribution of the Wujiang River System in Guizhou Province.

total cation charges ($TZ^+ = Na^+ + K^+ + 2Mg^{2+} + 2Ca^{2+}$) vary from 2.73 to 6.31 meq/L and the total anion charges ($TZ^- = Cl^- + 2SO_4^{2-} + HCO_3^- + NO_3^-$) vary from 2.98 to 6.31 meq/L. The total cation and anion charges are of balance within the limit of experimental errors. The reason why TZ^+ values are often higher than TZ^- values is the existence of organic anions in river water. TZ^+ values are close to those of the Changjiang River and Yellow River (Zhang, 1993, 1994; Huang, 1992), but are higher than those of world average river water ($TZ^+ = 0.725$ meq/L, Mebeck, 1981).

Variations in major ion composition

Anion and cation ternary diagrams provide a way to visualize the compositions and therefore the relative importance of different weathering regimes (Hu et al., 1982; Stallard and Edmond, 1983; Edmond et al., 1995, 1996). Fig.2 shows variations in major cation composition of Wujiang River water. HCO_3^- and SO_4^{2-} in the river water are the major anions and HCO_3^-/SO_4^{2-} ratio is greater than 6:4. Si accounts for less than 5% of the total ions and Cl^- less than 5% of the total anions. Fig. 2 also shows variations in chemical composition of water from global major rivers. The chemical composition of Wujiang River water is similar to that of water from the rivers flowing on the Siberian Craton Platform, but is different from that of the Amazon, Orinoco and Andean basin rivers. This shows that chemical weathering in carbonate areas controls the composition of river water. The compositional characters of cations in Wujiang River water are significantly different from those of other large rivers throughout the world. Ca + Mg accounts for more than 90% of the total cations in Wujiang River water (except sample No. 14), and Mg/Ca ratios range from 0.09 to 0.32 (mol∶mol), which is similar to what has been observed in the

Table 1. The major chemical components (in mmol/L otherwise noted) in the river water samples from Guizhou karst areas

Sample No.	SC(mS/cm)	DO(%)	pH	T(℃)	HCO_3^-	Cl^-	NO_3^-	SO_4^{2-}	Si	K^+	Na^+	Ca^{2+}	Mg^{2+}
990107-1	0.425	88.50	8.23	11.6	2.38	0.13	0.15	0.74	0.09	0.04	0.31	1.55	0.36
990107-2	0.285	90.30	8.34	11.0	2.45	0.03	0.05	0.23	0.13	0.01	0.05	1.00	0.33
990108-3	0.366	87.90	8.50	10.5	2.35	0.06	0.07	0.46	0.09	0.02	0.10	1.42	0.33
990108-4	0.396	94.40	8.59	9.5	2.60	0.06	0.07	0.45	0.10	0.02	0.10	1.46	0.34
990108-5	0.451	90.90	8.59	7.9	2.60	0.03	0.05	0.79	0.09	0.02	0.07	1.66	0.49
990108-6	0.389	87.80	8.35	9.1	2.70	0.09	0.08	0.46	0.10	0.05	0.10	1.42	0.34
990109-7	0.651	83.00	7.97	7.8	3.00	0.30	0.42	0.92	0.09	0.07	0.37	1.47	0.63
990109-8	0.603	83.10	7.98	8.2	2.10	0.26	0.48	0.93	0.09	0.05	0.30	1.62	0.59
990109-9	0.449	89.70	8.29	8.7	2.30	0.09	0.18	0.65	0.10	0.03	0.14	1.57	0.42
990109-10	0.439	81.00	8.22	9.8	2.60	0.05	0.07	0.68	0.09	0.03	0.12	1.81	0.33
990110-11	0.414	104.90	8.15	11.7	2.48	0.07	0.13	0.65	0.10	0.03	0.18	1.61	0.38
990110-12	0.420	96.10	8.14	12.1	2.38	0.08	0.13	0.70	0.09	0.03	0.17	1.63	0.40
990110-13	0.565	63.40	8.20	5.6	4.10	0.16	0.08	0.62	0.08	0.07	0.19	1.93	0.68
990110-14	1.269	55.70	8.09	5.4	3.38	1.35	0.08	3.01	0.06	0.50	4.87	2.35	0.64
990122-15	0.403	92.05	8.15	13.8	2.40	0.07	0.13	0.66	0.09	0.03	0.16	1.65	0.42
990122-16	0.413	82.16	8.22	12.9	2.60	0.08	0.12	0.70	0.06	0.04	0.17	1.65	0.43
990122-17	0.411	68.88	8.00	13.2	2.40	0.08	0.10	0.62	0.06	0.04	0.14	1.61	0.46
990122-18	0.565	79.10	7.98	10.1	4.05	0.15	0.14	0.70	0.15	0.05	0.13	1.51	0.72
990123-19	0.503	93.92	8.22	8.2	3.00	0.09	0.26	0.66	0.06	0.05	0.28	1.77	0.72
990123-20	0.519	98.49	8.27	10.0	3.80	0.06	0.07	0.53	0.21	0.04	0.06	1.25	1.00
990123-21	0.501	106.75	8.23	9.4	2.98	0.57	0.07	0.70	0.11	0.08	0.53	1.57	0.67
990123-22	0.437	110.15	8.28	12.6	2.95	0.13	0.10	0.64	0.09	0.04	0.21	1.70	0.53
990124-23	0.398	111.48	8.16	12.9	2.60	0.14	0.11	0.65	0.09	0.04	0.22	1.66	0.50
990124-24	0.346	101.99	8.17	10.7	3.28	0.06	0.04	0.14	0.04	0.03	0.10	1.02	0.65
990124-25	0.376	112.81	8.18	12.9	2.78	0.14	0.11	0.65	0.14	0.04	0.21	1.65	0.49
990124-26	0.33	118.31	8.37	11.0	3.08	0.07	0.02	0.14	0.16	0.02	0.09	1.05	0.63
990124-27	0.362	106.57	8.09	9.8	3.45	0.05	0.03	0.15	0.08	0.02	0.08	1.12	0.54
990125-28	0.436	98.70	8.16	12.6	2.30	0.19	0.11	0.63	0.12	0.04	0.28	1.65	0.48
990125-29	0.295	102.83	8.12	10.7	2.65	0.05	0.03	0.12	0.10	0.02	0.07	1.01	0.44
990125-30	0.404	134.82	8.73	13.3	2.65	0.12	0.11	0.68	0.06	0.04	0.20	1.66	0.48
990125-31	0.42	103.25	8.27	12.7	2.60	0.12	0.10	0.65	0.12	0.04	0.21	1.66	0.48
990125-32	0.32	106.62	8.38	11.9	2.73	0.04	0.03	0.14	0.09	0.02	0.07	1.21	0.42
990126-33	0.421	98.98	8.08	12.6	2.70	0.12	0.10	0.57	0.16	0.04	0.21	1.65	0.48
990126-34	0.419	103.99	8.07	12.6	2.70	0.13	0.10	0.58	0.06	0.04	0.21	1.62	0.48
990127-35	0.415	91.58	7.88	13.0	2.28	0.07	0.11	0.69	0.11	0.03	0.18	1.72	0.44
990127-36	0.549	107.05	8.34	12.0	3.60	0.34	0.20	0.81	0.17	0.08	0.44	2.11	0.79
990127-37	0.429	105.89	8.34	11.4	3.15	0.06	0.05	0.59	0.13	0.03	0.09	1.57	0.75

Changjiang River (0.25), Yokon (0.35) and carbonate karst streams in Germany (0.2 − 0.3), but different from what is reported from the Huanghe River (~1) (Kempe, 1982; Huh, 1998). Fig. 2 also indicates that river water from other major rivers throughout the world is rich in $Na^+ + K^+$ and Guayana Shield river water is particularly rich in $Na^+ + K^+$.

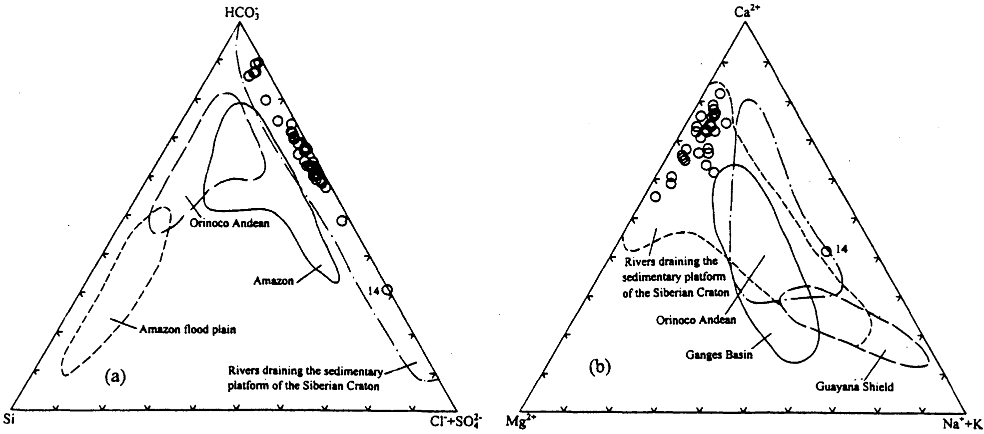

Fig. 2. Diagram showing variations in major anions (a) and cations (b) in Wujiang River water(μmeq/L). For the data of world rivers, see Gibbs, 1972; Stallard, 1983; Sarin, 1989; Huh, 1998.

Chemical Weathering in Carbonate Areas and Chemistry of River Water

Weathering characteristics of carbonate rocks

Weathering of carbonate minerals is ubiquitous in karst areas. The main weathering procedure is described as follow:

$$Ca_xMg_{1-x}CO_3 + H_2CO_3 = xCa^{2+} + (1-x)Mg^{2+} + 2HCO_3^- \tag{1}$$

$$Ca_xMg_{1-x}CO_3 + H_2SO_4 = xCa^{2+} + (1-x)Mg^{2+} + SO_4^{2-} + H_2CO_3 \tag{2}$$

On the anion diagram, pure carbonate weathering yields only HCO_3^- with no Si, so the data points fall at the apex of HCO_3^-. The products of evaporite weathering would fall at the apex of $(Cl^- + SO_4^{2-})$. Silicate weathering results in HCO_3^- and Si, so the data points fall in the central part of the ternary diagram. On the cation diagram, the products of evaporite mineral weathering fall at the $(Na^+ + K^+)$ apex and those of limestone weathering fall at the Ca-Mg joint, the exact location varying with the Mg enrichment of the limestones with dolomites in the middle (Ca:Mg = 1:1). Silicate weathering shows a trend from the Ca-Mg joint toward the $Na^+ + K^+$ apex. Evaporite weathering shows a trendency toward the $(Na^+ + K^+)$ apex. From Fig.2, we can see that the composition of major elements in the Wujiang River system by enrichments in $Mg^{2+} + Ca^{2+}$ with almost no $Na^+ + K^+$ and in $HCO_3^- + Cl^- + SO_4^{2-}$ with almost no Si. HCO_3^- and Ca^{2+} are the dominant ions. The composition of major elements is different from that of Guayana Shield and Amazon flood plain river water, but similar to that of water from rivers on the Siberian craton sedimentary platform. This shows the chemical composition of river water is largely controlled by carbonate weathering.

In the diagram showing variations in $Mg^{2+} + Ca^{2+}$ relative to HCO_3^- (Fig.3a), a number of data points fall off the 1:1 equivalent line, indicating pure carbonate weathering cannot explain

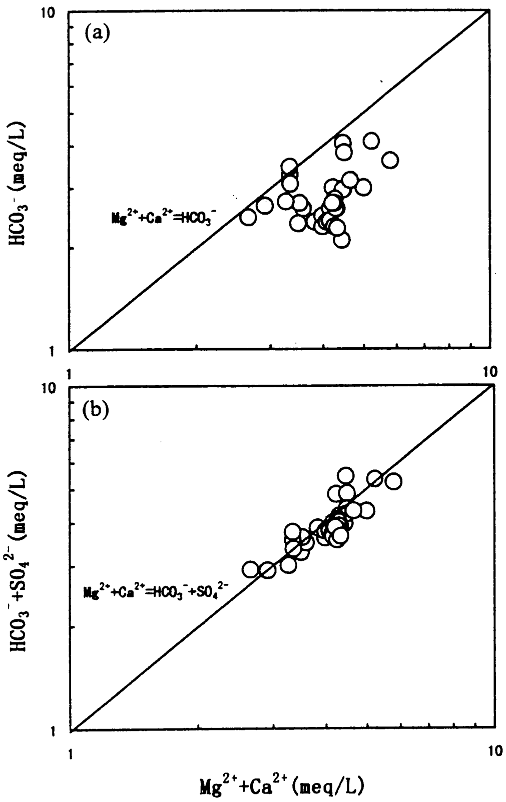

Fig. 3. Diagram showing variations in $Mg^{2+} + Ca^{2+}$ relative to HCO_3^- (a) and $HCO_3^- + SO_4^{2-}$ (b) in Wujiang River water (except sample No. 14).

completely the relevant chemical composition of river water. Calcium and magnesium derived from sulfate and evaporite weathering are the major components of river water. Although sodium and potassium derived from silicate mineral and evaporite weathering are the minor components of river water, a number of data points still fall off the 1:1 equivalent line in Fig. 3b. From the fact, we think some $Na^+ + K^+$ come from silicate mineral and evaporite weathering. In this work the saturation indices for calcite, aragon spar and dolomite have not been calculated. But according to the calculations of Huh et al. (1998) under similar temperature and similar concentrations of ions dissolved in river water, we can say with certainty that Wujiang River water is oversaturated relative to these carbonate minerals. The existence of CO_2 from soils in underground water and the lack of nuclear crystal for calcite precipitation may be the main factor leading to such over-saturation.

The contribution of evaporite and silicate mineral weathering to water chemistry

Cl^-/Na^+ would be 1:1 when evaporite is dominant in river water. The concentrations of Na^+ and Cl^- in Wujiang River water are lower than those of Huanghe River water and higher than those of river water from the Siberian Craton sedimentary platform. A number of samples fall above the 1:1 equivalent line in Fig. 4. This indicates Na^+ comes from other sources, for example Na-feldspar and Na-clay mineral weathering. The data points of the samples with high Na^+ concentrations are close to the 1:1 equivalent line. This indicates the importance of evaporite weathering.

No correlation is found between SO_4^{2-} and Cl^- (Fig. 5) in Wujiang River water. SO_4^{2-} doesn't vary with Cl^- in the case of $Cl^- > 0.05$ meq/L. SO_4^{2-} and Cl^- are often associated with each other in evaporite, but there doesn't exist a proper correlation. Moreover, it is very important to know the source of SO_4^{2-} because the concentrations of Cl^- are lower than those of SO_4^{2-}. If Ca and Mg from sulfate weathering are the main sources of SO_4^{2-} in river water, $M_{(Ca+Mg)}$ [$M_{(Ca+Mg)} = Ca^{2+} + Mg^{2+} - HCO_3^-$] would be directly proportional to SO_4^{2-} and be equivalent after deducting the Ca and Mg from carbonate weathering. Fig. 5 shows this correlation doesn't exist. So we think Ca^{2+} and Mg^{2+} from sulfate weathering are not the decisive factors controlling $M_{(Ca+Mg)}$ and SO_4^{2-} in river water.

In all river samples, Na^+ and K^+ from silicate weathering account for 10% of the total cations, and K^+ is less than 2% of the total cations. It is difficult to quantitatively calculate the concentrations of Ca derived from silicate weathering because of the influence of carbonate. Si

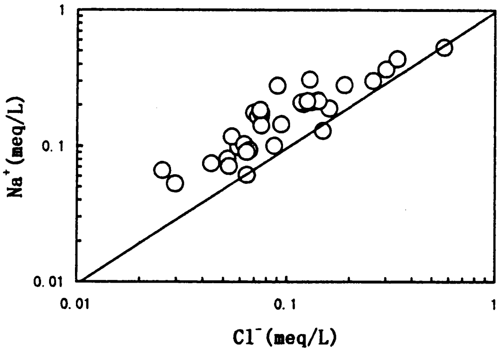

Fig. 4. Diagram showing variations in Na^+ relative to Cl^- in Wujiang River water (except for sample No. 14).

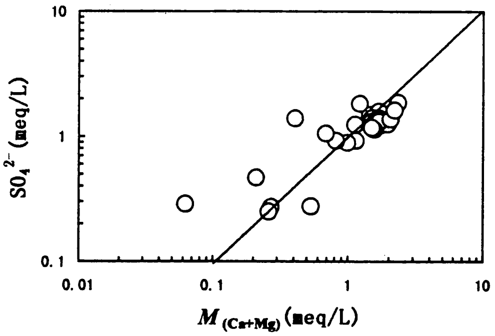

Fig. 5. Diagram showing variations in $M_{(Ca+Mg)}$ relative to SO_4^{2-} in Wujiang River water. $M_{(Ca+Mg)} = Ca^{2+} + Mg^{2+} - HCO_3^-$ (except for sample No. 14)

contents vary from 0.02 - 0.2 (meq/L) in Wujiang River water with a mean value of 0.1 meq/L. Such low Si contents of river water in the world catchment are also reported. The reason is that there is no soil cover or no soluble Si in carbonates. This explanation may be suitabe to the low Si contents of karst river water because silicate weathering would contribute little to dissolved matter in river water.

The impact of human activities on water chemistry

The products of human activities may be directly input into river water as waste or by way of atmosphere. Besides anthropogenic discharge, the atmosphere input also contains marine and terrestrial components (biological emissions, products from vegetation combustion and soil dust) (Stallard and Edmond, 1981). Studies (Stallard and Edmond, 1981; Negrel et al., 1993) have shown that the marine input may not significantly influence the chemistry of river water if the rivers are far apart from the ocean. Moreover, the Wujiang River System was characterized by minimum discharge and minimum rainfall in winter during the sampling period. The river is supplied by underground water and this part of underground water is greatly influenced by rocks and soils at the catchment. So we don't think the atmosphere inputs would effect the solutes in river waters. The anthropogenic

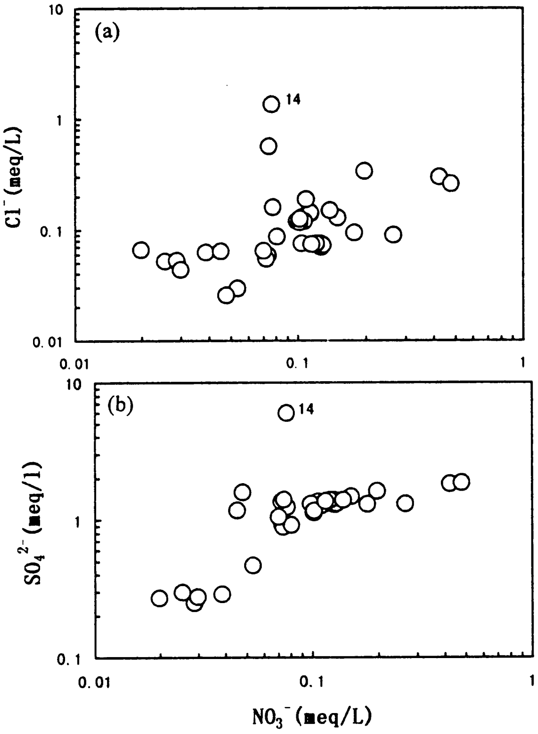

Fig. 6. Diagram showing variations in NO_3^- relative to Cl^- (a) and SO_4^{2-} (b) in Wujiang River water.

source is characterized by the dominance of K, Ca, S, Cl and N (Stallard and Edmond, 1981; Sarin et al., 1989), but K, Ca, S, Cl are derived from rock/soil weathering. So variations in NO_3^- can indicate the impact of human activities on river water chemistry.

NO_3^- in river water comes from nitrogenous fertilizers applied in agricultural activities and industrial pollution because it is advantageous to produce nitrogen in the condition of artificial pollution and lack of oxygen. In all river water samples, NO_3^- and Cl^- are linearly correlated with SO_4^{2-} and not with HCO_3^-. This indicates NO_3^-, Cl^- and SO_4^{2-} come from the same source (Chen Jingsheng, 1998). The total hardness and total N concentrations of Wujiang River water had increased three times in the ten years from 1980 to 1990. Their studies further indicated that the total N is linearly correlated with the amount of nitrogenous fertilizer applied. SO_4^{2-} in river water is correlated with the amount of coal combustion.

Conclusions

The hydrogeochemical characteristics of Wujiang River water are different from those of global major rivers: the river and its tributaries have high total dissolved solid concentrations, with Ca^{2+} and HCO_3^- being dominant, Mg^{2+} and SO_4^{2-} coming next. Both $Na^+ + K^+$ and $Cl^- +$ Si account for 5% − 10% of the total cations and anions, respectively. These common features show the chemical composition of river water is largely controlled by carbonate weathering, with the impact of silicate and evaporite weathering being of less importance.

The concentrations of NO_3^- in the river water indicated the impact of human activities on the chemical composition of river water. Variations in SO_4^{2-} and Cl^- contents are positively correlated with NO_3^-, so the river water polluted by industrial waste discharge has higher SO_4^{2-}, Cl^- and Na^+ concentrations. So it is considered that SO_4^{2-}, Cl^- and NO_3^- seem to come from human activities such as agricultural production, mining practice and industrial pollution.

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