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Chemical weathering under mid- to cool temperate and monsoon-controlled climate: A study on water geochemistry of the Songhuajiang River system, northeast China

Baojian Liu [a,b,c], Cong-Qiang Liu [b,*], Gan Zhang [a], Zhi-Qi Zhao [b], Si-Liang Li [b], Jian Hu [b], Hu Ding [b], Yun-Chao Lang [b], Xiao-Dong Li [b]

[a] State Key Laboratory of Organic Geochemistry, Guangzhou Institute of Geochemistry, Chinese Academy of Sciences, Guangzhou 510640, China
[b] State Key Laboratory of Environmental Geochemistry, Institute of Geochemistry, Chinese Academy of Sciences, Guiyang 550002, China
[c] Graduate University of Chinese Academy of Sciences, Beijing 100049, China

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ABSTRACT

For the first time, the river water geochemistry of the Songhuajiang Basin, the third largest river system in China, has been studied in order to understand chemical weathering and associated CO_2 consumption rate in a mid- to cool temperate and monsoon-controlled climatic zone. The major ion compositions of the river waters are characterized by the dominance of Ca^{2+} and HCO_3^-, accounting for 46% and 74% of major cations and anions, respectively. The average total dissolved solids (TDS, 116 mg L^{-1}) and total cation concentration (TZ^+, 1388 μEq L^{-1}) of the river waters are similar to those of global major rivers. The chemical weathering rates of carbonate, silicate and evaporites in the whole Songhuajiang Basin are estimated to be approximately 5.15, 2.23 and 0.40 t km^{-2} a^{-1}, respectively. The total rock weathering rate for the whole Songhuajiang Basin is approximately 7.78 t km^{-2} a^{-1}, which is at the lower end of the spectrum for global major rivers, and is comparable with that of the Amur and the Congo-Zaire River. The estimated CO_2 consumption rates for the whole Songhuajiang Basin are 53.4×10^3 mol km^{-2} a^{-1} and 66.6×10^3 mol km^{-2} a^{-1} by carbonate and silicate weathering, respectively.

As a sub-basin, the 2nd Songhuajiang has the highest (18.9 t km^{-2} a^{-1}) while the Nenjiang River Basin has the lowest total rock weathering rate (5.03 t km^{-2} a^{-1}), which indicates important controls of regional climate and lithology. Compared to the large rivers of China, total rock weathering rates increase from north to south, supporting the idea that the climate acts as major control on global chemical weathering. The CO_2 consumption rate by silicate weathering within the whole Songhuajiang River Basin, though under mid- to cool temperate climate, is in the same order of magnitude as that of Huanghe (82.4×10^3 - mol km^{-2} a^{-1}) and is not much lower than that of Changjiang (112×10^3 mol km^{-2} a^{-1}), which suggests that the role of Songhuajiang River weathering in long-term climate change cannot be neglected compared to those of the large rivers of China.

1. Introduction

The chemical composition of river water mainly depends on sources of solutes and chemical equilibrium in the water. So, geochemical research on river water will provide important information on chemical weathering rates of the different rocks, and CO_2 consumption fluxes of a river basin. Also, the river water chemistry reveals the effects of atmospheric precipitation and human activities on the ecological environment in a basin (Gibbs, 1970, 1972; Hu et al., 1982; Stallard and Edmond, 1983; Goldstein and Jacobsen, 1987; Meybeck, 1987, 2003; Elderfield et al., 1990; Gaillardet et al., 1997, 1999b; Roy et al., 1999; Dupré et al., 2003; Millot et al., 2003; Das et al., 2005; Qin et al., 2006; Liu, 2007). Carbonate dissolution has an effect on the CO_2 balance of the atmosphere only on a timescale similar to or shorter than the residence time of HCO_3^- in the oceans (100 ka), while silicate weathering results in a net consumption of CO_2 (Viers et al., 2004). Chemical weathering of silicates, therefore, is known as the dominant long-term sink for atmospheric CO_2 and thus the dominant regulator of the green-house effect over geological time-scales (Dessert et al., 2003; Godderis et al., 2003). Accordingly, estimation of the present-day rates of silicate weathering and characterization of its controlling factors are crucial in interpreting and predicting past and future changes in global climate (Chetelat et al., 2008; Moon et al., 2009).

* Corresponding author at: State Key Laboratory of Environmental Geochemistry, Institute of Geochemistry, Chinese Academy of Sciences, Guiyang 550002, China. Tel.: +86 851 5891164; fax: +86 851 5891609.
E-mail address: liucongqiang@vip.skleg.cn (C.-Q. Liu).

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Chemical weathering is controlled by multiple factors such as climate (temperature, runoff) (White and Blum, 1995; Liu et al., 2012), lithology, tectonics (Goudie and Viles, 2012), vegetation and human activities (Roy et al., 1999; Li et al., 2009; Xu et al., 2011), etc. However, the controlling mechanisms that these factors have on chemical weathering are still uncertain. The dominant factor differs under different climates. For example, weathering may respond rapidly to high-frequency climatic fluctuations under a tropical climate (Dosseto et al., 2006). Liu et al. (2012) showed that temperature and precipitation were the principal factors while tectonics was only a subordinate factor controlling the chemical weathering in tropical SE Asia. However, strong tectonic and weak climatic control could be observed under temperate climate (Riebe et al., 2001) and temperature may have a negative action on silicate weathering for river basins in a cold climate (Millot et al., 2003). In China, chemical weathering has been studied for river basins under subtropical climate, warm temperate climate and also a plateau climate (Han and Liu, 2004; Wu et al., 2005; Xu and Liu, 2007, 2010; Chetelat et al., 2008), but information on chemical weathering under mid- or cool temperate climate is still insufficient. So, more work should be carried out to understand the controlling mechanisms under different climatic regimes. The Songhuajiang River, located in the mid- to cool temperate zone, is the biggest tributary of the Amur River. It is also the third largest river in China in terms of either its annual discharge (73.3×10^9 m^3 a^{-1}) or drainage area (55.7×10^4 km^2). The basin is characterized by great climatic and ecological diversities. Wetland, primeval forests, and typical black soil are widely distributed in this area. Therefore, it offers the possibility of investigating chemical weathering rates under a mid- to cool temperate climate, and exploring the different controlling factors on chemical weathering.

In this study, a systematic investigation has been carried out on the hydrogeochemistry of the whole Songhuajiang River system, in particular, to characterize aqueous geochemistry and its controlling factors. The purposes of this study are to decipher the different sources of solutes in the Songhuajiang River system, to quantify rock weathering and associated CO_2 consumption rates, and finally to explore the mechanisms controlling chemical weathering under different climates and geologic conditions.

2. Natural setting of the Songhuajiang River Basin

2.1. Geography and geology

The Songhuajiang River Basin, located in NE China, lying between 41°42′–51°38′N latitude and between 119°52′–132°31′E longitude, covers a total area of 55.72×10^4 km^2, which is about 30% of the Amur River's total drainage area (1.86×10^6 km^2) and about 6% of the land area of China. The basin can be divided into four structural areas: The Changbai Mountain (with an elevation of 200–2700 m) on its eastern part; the Daxinganling Mountains (with an elevation of 700–1700 m) on its western part and the Xiaoxinganling Mountains (with an elevation of 600–1000 m) on the northern part; the center and southern part of the basin is occupied by the Songnen plain (with an elevation of 50–200 m). The Songhuajiang River has two headstreams, the Nenjiang River (1370 km long) originating from the Daxinganling Mountains and the 2nd Songhuajiang River (958 km long) from Tianchi in the Changbai Mountain. After their confluence, the river is called the Songhuajiang River (main stream, 939 km long) and joins the Amur River at Tongjiang city (Figs. 1 and 2).

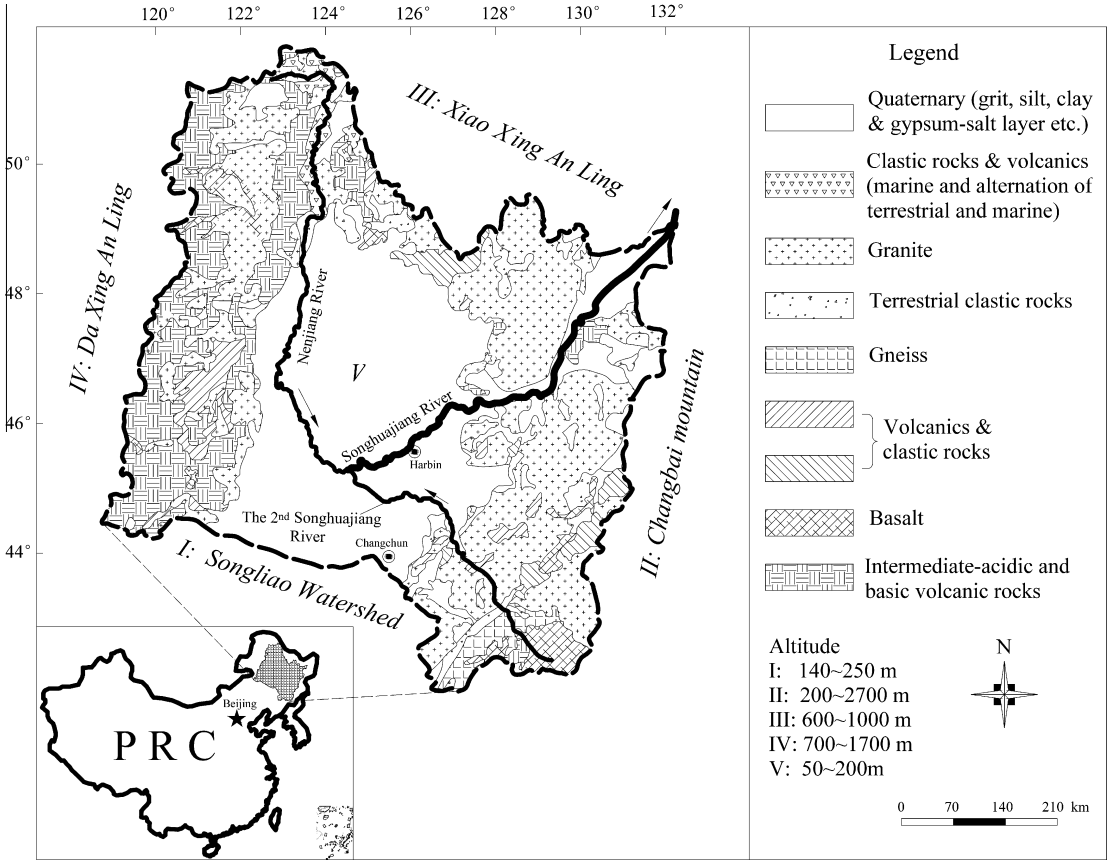

Fig. 1. Geological map of the Songhuajiang River Basin.

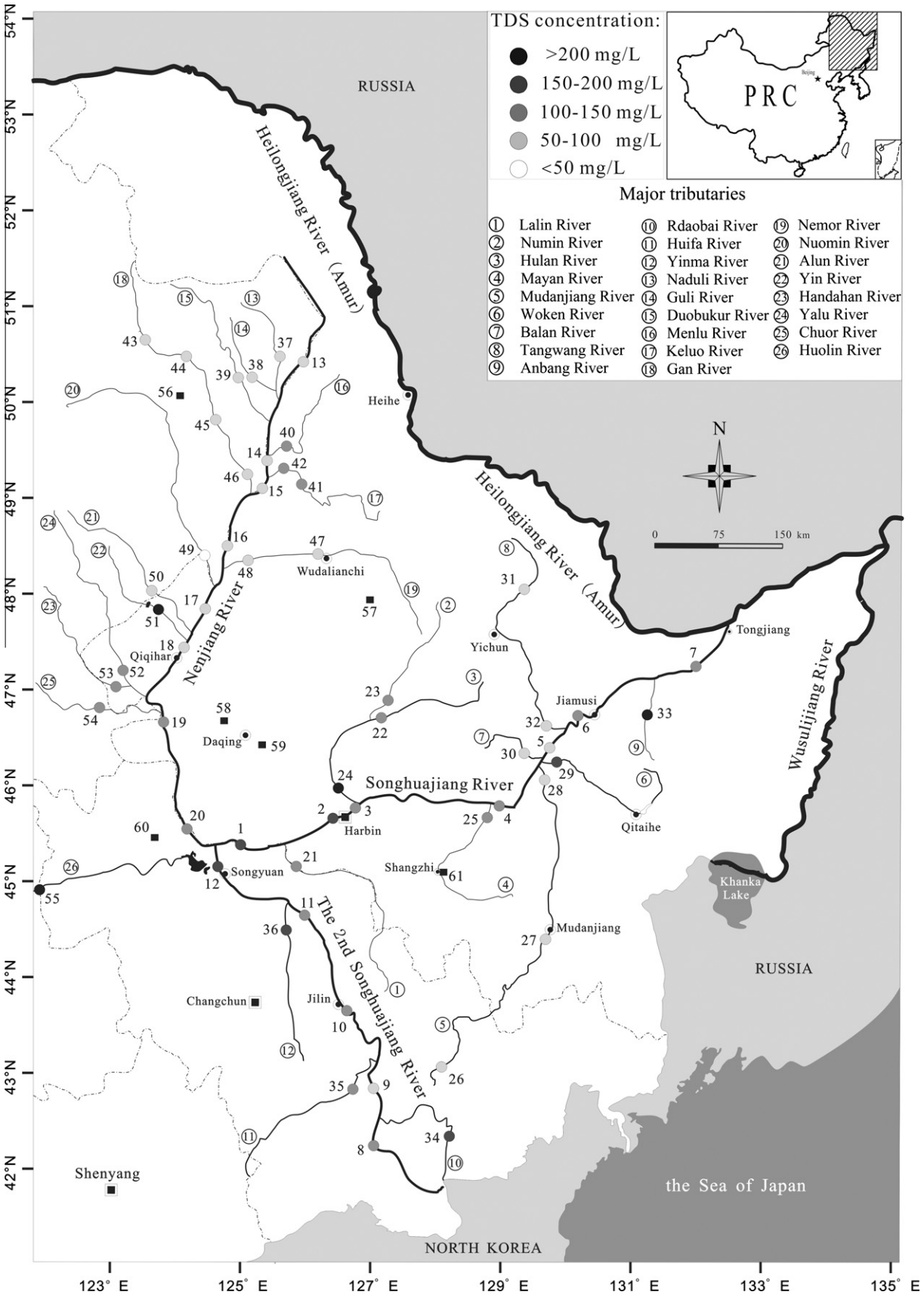

Fig. 2. Sampling locations in the Songhuajiang River Basin and spatial distribution of the TDS concentrations.

Geologically, Quaternary sediments (grit, silt, clay and gypsum-salt layer, etc.) are widely distributed in the middle of the basin, covering about 36% of the total basin area. The upper reaches of the Nenjiang River are covered by intermediate-acid/basic volcanic rocks and granites. The 2nd Songhuajiang Basin is covered mainly by basalt and gneiss in its upper reaches and by granites in its middle reaches. The Songhuajiang main stream flows on granites in its lower reaches (Fig. 1).

2.2. Climate, land cover and human activities

The Songhuajiang Basin climate is controlled by monsoon climate of medium latitudes. The annual air temperature is about 4–5 °C (average for years 1956–2000) and the mean precipitation received by the basin is about 400–700 mm a^{-1}. A gradient of these climatic parameters is observed from the southeastern part to the northwestern part with annual temperature from 5 to 2 °C and annual precipitation from 700 to 400 mm a^{-1} (Qian, 2007). From November to the following March, most of the rivers are frozen and the water discharge is very low during that time, only a few hundredths of the annual flow. Heavy forests are mainly distributed in the east, west and north parts of the basin. The Songnen Plain is an important agricultural region. Within the plain, the confluence area of Nenjiang and the 2nd Songhuajiang is a corn producing region. Crops in the upper reaches of Nenjiang and the lower reaches of the Songhuajiang main stream are dominated by wheat. Rice is mainly cultivated in the upper reaches of the 2nd Songhuajiang. One of the three black soil terrains in the world is located here, distributed mainly in the middle reaches of the Nenjiang River, the lower reaches of the 2nd Songhuajiang and around Harbin City. Average population density in the whole river basin is about 97 indiv km^{-2}, mainly concentrated on a line between Harbin and Changchun city.

3. Sampling and analytical methods

Main streams of Songhuajiang, the 2nd Songhuajiang, Nenjiang and their major tributaries were sampled during July 2010. The sampling locations are shown in Fig. 2. Samples were collected from the river bank at depths ranging from 50 cm to 100 cm or from the middle of the river when a boat was available. In total, 54 river water samples (20 from the main streams, 34 from the tributaries) were collected together with 13 samples from other types of water (rainwater, groundwater, sewage, stream and lake water).

For each sample, 10–20 L of water were collected using high density polyethylene (HDPE) containers previously acid-washed and cleaned with ultrapure water. Water samples were filtered a few hours after sample collection through pre-washed 0.22 μm Millipore membrane filters. The first liter of filtrate was discarded to clean the filter, and then a small portion was stored for measuring anions. Another portion was acidified with ultra-pure 6 M HCl to pH < 2 and stored in HDPE bottles (pre-washed with double-distilled ultra-purified HCl and rinsed with Milli-Q 18.2 MΩ water) for measuring cations. All of the bottles were sealed and kept in a dark place.

Temperature, pH and electric conductivity (EC) were measured on site using a portable EC/pH meter (WTW, pH 3210/Cond 3210 Germany) and the alkalinity was determined by titration with HCl before filtration. The accuracies were ±0.01 for the determination of pH, ±0.1 (°C) for temperature, ±0.5% for EC and ±0.01 (mL) for the amount of HCl consumed in the titration of alkalinity. Major cations (Ca^{2+}, Mg^{2+}, Na^{+}, K^{+}) and silicic acid concentrations were measured by Inductively Coupled Plasma Optical Emission Spectrometry (ICP-OES, Vista MPX, USA) with a precision of better than 5%. Anion (Cl^{-}, NO_3^{-} and SO_4^{2-}) concentrations were determined by ionchromatography (Dionex, ICS-90, USA) with a precision of 5%. Regent and procedural blanks were determined in parallel to the sample treatment, and the national standard reference materials of China were used in the determination of cations and anions. The national standard reference materials are: GBW(E)080118 for Ca^{2+}, GBW(E)080126 for Mg^{2+}, GBW(E)080127 for Na^{+}, GBW(E) 080125 for K^{+}, GBW(E)080272 for SiO_2, GBW(E)082048 for Cl^{-}, GBW(E)082050 for SO_4^{2-} and GBW(E)082049 for NO_3^{-}.

4. Results

Solution parameters (pH, temperature and EC), major cation and anion concentrations of the samples are presented in Table 1. The river water samples are ca. neutral with pH values ranging from 6.66 to 8.87. Total dissolved cations ($TZ^{+} = K^{+} + Na^{+} + Ca^{2+} + Mg^{2+}$, in μEq) range from 481 to 3636 μEq L^{-1}, with an average of 1388 μEq L^{-1}. The total dissolved solid (TDS, mg L^{-1}), expressed here as the sum of major inorganic species concentration ($Na^{+} + K^{+} + Ca^{2+} + Mg^{2+} + HCO_3^{-} + Cl^{-} + SO_4^{2-} + NO_3^{-} + SiO_2$), is also given in Table 1. For all the river water samples, TDS values range from 48.1 to 306 mg L^{-1}, with an average of 116 mg L^{-1}, which is comparable with the global average of 100 mg L^{-1} (Gaillardet et al., 1999b). The Nenjiang River has the lowest average TDS value (98.1 mg L^{-1}), followed by the Songhuajiang River (128 mg L^{-1}), and the 2nd Songhuajiang has the highest TDS values (average 142 mg L^{-1}). There is a significant correlation between TDS and TZ^{+} ($R^2 = 0.97$). EC varies from 32.5 to 418 μs cm^{-1} with an average of 133 μs cm^{-1} for all the river water samples. The Songhuajiang main stream has the highest EC value (mean 176 μs cm^{-1}) and Nenjiang has the lowest one (mean 94 μs cm^{-1}). The extent of inorganic charge imbalance, characterized by the normalized inorganic charge balance ($NICB=(TZ^{+} - TZ^{-})/TZ^{+}$, where $TZ^{-} = HCO_3^{-} + Cl^{-} + SO_4^{2-}$ in μEq), was within ±10% for most of the samples. From NIBC it can be estimated that the contribution of organic ligands is not significant to the charge balance.

Major ion compositions are shown in the anion and cation ternary diagrams (Fig. 3). As shown in the figure, Ca^{2+} is the dominant cation, accounting for about 46% (in μmol L^{-1}) of the total cation composition in most of the river water samples (Fig. 3a). The concentration of Ca^{2+} ranges from 127 to 826 μmol L^{-1} in the rivers and has average values of 417, 437 and 317 μmol L^{-1} for Songhuajiang main stream, the 2nd Songhuajiang, and Nenjiang, respectively. The next highest cation is Na^{+}, which accounts for more than 30% (in μmol L^{-1}) of the total cation concentrations in river waters. The K^{+} concentration is relatively low in the whole basin and is estimated to be approximately 5% (in μmol L^{-1}) of the total cation composition. However, in some tributaries of Nenjiang river system like the Nemor River (sample 47 and 48) and the Keluo River (sample 41 and 42), K^{+} concentration is relatively high, probably because these rivers flow through the Wudalianchi volcanic area, where the volcanics are rich in K. For instance, the K^{+} concentrations are 533 μmol L^{-1}, 388 μmol L^{-1} and 255 μmol L^{-1} for the Wudalianchi samples 1, 2 and 3, respectively. Concentrations of Mg^{2+} and K^{+} in the river water samples from Daxinganling and Xiaoxinganling Mountains are very low: Mg^{2+} concentrations are 8.64 and 1.12 μmol L^{-1}; K^{+} concentrations are 16.1 and 8.79 μmol L^{-1}, for samples 31 and 43, respectively.

On the anion ternary diagrams, most of samples cluster toward the HCO_3^{-} apex (Fig. 3b) and HCO_3^{-} accounts for approximately 74% (in μmol L^{-1}) of the total anion concentrations, ranging from 273 to 3544 μmol L^{-1} with an average of 927 μmol L^{-1} for all the river water samples. The second dominant anions are SO_4^{2-} and Cl^{-}, each accounting for about 10% (in μmol L^{-1}) of the total anion composition. The highest HCO_3^{-} concentration was observed in sample 51 from the Yin River (tributary number 22, Fig. 2) and most was

Table 1
The chemical compositions of major ions in the river waters from the Songhuajiang River system, NE China (units: μmol L^{-1}).

Rivers	Date-sample number	pH	EC(μs cm^{-1})	Ca^{2+}	Mg^{2+}	Na^{+}	K^{+}	HCO_3^-	SO_4^{2-}	Cl^-	NO_3^-	SiO_2	TDS(mg L^{-1})	TZ^+ (μEq L^{-1})	TZ^- (μEq L^{-1})	NICB (%)
Mainstreams																
Songhuajiang River	14–1	7.81	174	494	193	581	57.6	1355	211	315	103	116	168	2011	2092	−4.0
	20–2	7.61	222	525	209	593	67.5	1088	228	327	128	115	157	2128	1872	12.0
	20–3	7.68	215	515	197	562	63.1	1074	224	329	118	114	154	2051	1850	9.8
	19–4	8.05	162	386	119	398	47.6	737	185	233	24.0	71.9	106	1456	1341	7.9
	19–5	7.41	124	303	59.1	249	36.2	667	140	170	0.91	91.6	86.4	1010	1116	−10.5
	19–6	7.20	145	350	108	371	43.7	807	168	214	57.1	78.4	108	1331	1356	−1.9
	18–7	7.48	191	428	161	464	58.6	983	187	253	99.7	100	133	1701	1609	5.4
The 2nd Songhuajiang River	24–8	7.54	138	426	168	224	42.6	913	198	71.1	84.3	199	122	1455	1379	5.3
	24–9	7.30	89.4	301	116	204	46.8	667	147	74.4	84.8	196	95.9	1085	1036	4.6
	24–10	7.19	112	361	134	247	56.4	702	179	174	125	172	110	1293	1233	4.6
	20–11	7.66	201	484	189	533	82.9	793	304	326	213	172	152	1962	1727	12.0
	14–12	7.70	208	573	220	676	88.7	1194	315	456	210	133	188	2351	2279	3.1
Nenjiang River	25–13	6.93	54.3	215	37.8	83.1	15.1	534	17.4	6.61	2.56	137	54.9	605	575	4.9
	17–14	7.62	81.2	314	89.3	172	19.8	953	55.5	13.9	0.76	144	92.1	998	1078	−8.1
	18–15	7.62	79.6	306	83.8	162	23.7	858	55.6	15.3	1.26	138	85.5	965	984	−2.0
	18–16	7.46	92.4	329	91.2	169	28.6	949	66.7	38.7	2.26	111	92.9	1037	1121	−8.1
	21–17	7.49	85.3	300	89.1	185	31.0	830	69.3	37.0	2.01	108	84.9	994	1006	−1.1
	21–18	7.39	107	365	150	240	33.9	870	87.0	71.3	20.3	171	101	1304	1115	14.5
	20–19	7.80	112	401	136	247	29.9	1260	81.2	72.6	13.0	118	121	1349	1495	−10.8
	14–20	7.81	138	459	167	321	30.7	1402	117	91.0	20.5	117	139	1604	1727	−7.7
Tributaries of Songhuajiang River																
Lalin River	20–21	7.08	224	498	177	614	89.1	913	284	427	77.1	93.4	150	2054	1906	7.2
Hulan River	14–22	7.23	176	436	128	359	47.7	737	226	224	3.96	168	116	1534	1413	7.9
Numin River	14–23	7.07	174	415	131	351	41.9	737	221	229	0.09	147	113	1483	1408	5.1
Hulan River	14–24	7.37	336	653	297	1023	88.5	1925	315	402	186	134	242	3011	2957	1.8
Mayan River	19–25	7.94	169	387	107	393	57.6	632	219	304	2.50	138	108	1438	1374	4.5
Mudanjiang River	22–26	7.29	92.6	329	77.3	137	26.9	702	115	59.8	29.2	209	89.6	976	992	−1.7
	13–27	7.24	133	321	141	232	54.2	702	140	176	59.9	160	100	1211	1158	4.4
	19–28	7.45	107	282	76.2	196	41.6	583	145	159	80.2	149	88.3	954	1032	−8.2
Woken River	19–29	7.41	261	512	294	761	47.7	1545	202	332	12.8	155	183	2421	2280	5.8
Balan River	19–30	—	—	216	60.8	98.0	24.1	421	98.2	44.4	71.4	165	64.4	676	662	2.0
Tangwang River	17–31	6.88	55.1	232	8.64	105	16.1	421	60.3	38.2	13.8	191	57.7	603	580	3.8
	19–32	7.17	86.7	269	31.3	157	26.5	513	116	70.1	34.0	185	74.3	785	814	−3.7
Anbang River	18–33	8.87	418	789	520	845	173	1755	332	797	337	90.5	264	3636	3217	11.5
Tributaries of the 2nd Songhuajiang River																
Rdaobai River	23–34	7.77	127	197	98.7	967	79.4	1159	77.9	193	12.9	520	153	1638	1508	7.9
Huifa River	24–35	6.96	171	413	132	256	80.0	632	195	253	284	117	120	1426	1276	10.5
Yinma River	20–36	7.54	290	737	260	586	103	1313	337	538	77.7	48.9	193	2680	2526	5.8
Tributaries of Nenjiang River																
Naduli River	25–37	6.86	50.1	159	87.5	94.8	12.0	490	23.2	11.3	1.58	172	54.1	599	548	8.5
Guli River	25–38	7.33	32.5	251	43.9	118	8.83	590	42.4	3.52	0.27	180	65.2	716	678	5.3
Duobukur River	25–39	7.40	50.8	164	62.9	93.1	7.63	420	57.6	14.6	0.04	172	52.5	555	550	0.9
Menlu River	17–40	7.68	130	414	224	400	29.9	1531	126	24.7	9.37	173	150	1707	1807	−5.9
Keluo River	17–41	7.65	108	326	146	304	75.5	1146	84.1	82.6	26.0	135	117	1324	1396	−5.5
	17–42	7.87	130	409	190	356	76.2	1436	103	87.7	2.31	125	140	1630	1730	−6.2
Gan River	24–43	7.46	47.8	191	1.12	105	8.79	406	56.0	16.8	0.19	164	51.0	497	535	−7.6
	24–44	7.66	51.7	217	10.8	111	9.06	436	74.5	17.6	5.45	155	55.9	576	602	−4.6
	23–45	7.18	60.6	250	7.15	127	13.3	530	66.3	29.7	0.84	172	63.8	656	692	−5.6
	18–46	7.60	80.7	282	96.5	172	21.2	800	80.8	64.7	15.4	143	86.8	950	1027	−8.1
Nemor River	17–47	7.86	79.2	260	47.9	154	54.6	701	72.5	64.5	16.2	79.3	75.1	825	911	−10.4
	18–48	7.17	90.1	289	81.7	255	56.9	815	84.9	92.6	7.72	93.6	88.9	1053	1078	−2.3
Nuomin River	18–49	6.66	56.4	127	25.6	159	17.2	273	87.0	32.5	27.9	169	48.1	481	480	0.3
Alun River	26–50	7.57	102	353	89.8	222	17.2	792	133	70.8	59.5	219	103	1125	1130	−0.4
Yin River	26–51	7.79	151	826	586	561	41.9	3544	76.1	164	0.06	241	306	3428	3860	−12.6
Yalu River	21–52	7.58	102	353	102	273	21.3	810	129	107	17.0	194	102	1204	1175	2.3
Handahan River	21–53	7.45	84.5	280	94.0	317	23.4	906	136	69.4	114	116	107	1089	1247	−14.6
Chuor River	21–54	8.00	109	408	113	240	21.7	1129	136	46.5	1.38	83.7	114	1305	1447	−10.9
Other types of water sample																
Huolin River (continental river)	08–55	8.17	411	1125	544	1208	31.9	4284	355	239	1.76	191	403	—	—	—
Streamlet from Daxinganling	23–56	7.10	53.2	188	34.8	122	3.68	404	78.3	7.47	0.68	201	55.9	—	—	—
Streamlet from the black soil	17–57	7.83	504	930	730	1345	98.5	4423	26.0	705	0.56	235	401	—	—	—
Talahong Nur	20–58	9.18	347	288	783	7815	147	8564	245	1073	0.14	44.8	803	—	—	—
Wanghua Nur	19–59	9.05	877	281	829	10655	60.6	9339	620	1913	0.17	50.2	979	—	—	—
Groundwater	14–60	7.95	220	666	261	763	20.3	2679	84.6	26.0	0.76	396	248	—	—	—
Sewage (Shangzhi City)	27–61	7.50	645	893	266	1725	391	2387	393	1374	6.46	224	343	—	—	—
Wudalianchi 1 (Huoshaoshan)	15–	6.23	1107	5259	645	778	533	11164	503	293	20.0	771	1052	—	—	—
Wudalianchi 2 (Jingpo)	16–	7.28	211	337	198	728	388	1790	69.2	203	8.42	91.1	179	—	—	—
Wudalianchi 3 (The 3rd pool)	16–	7.31	173	358	212	647	255	1832	81.5	167	2.40	36.6	172	—	—	—
Rainwater (Jiamusi City)	18–	5.87	27.3	26.8	3.95	6.94	2.18	158	27.7	14.0	47.3	2.92	17.3	—	—	—
Rainwater (Shangzhi City)	26–	—	—	8.71	3.82	15.7	12.0	52.7	11.9	29.9	15.5	2.49	7.80	—	—	—
Rainwater (Wudalianchi City)	16–	7.29	24.4	8.92	4.08	22.1	2.06	74.1	66.3	4.70	63.9	17.8	17.1	—	—	—
Rainwater (Daqing City)	18–	7.07	26.5	25.5	4.21	7.81	3.73	—	58.9	6.60	77.0	1.64	—	—	—	—

TDS: total dissolved solid; NICB = 100%×($TZ^+ - TZ^-$)/TZ^+.

balanced by $Ca^{2+} + Mg^{2+}$ [(Ca + Mg)/HCO_3 = 0.4]. Perhaps it is affected by the Yinhe reservoir located a few kilometers upstream. Samples 33 and 36 have the highest Cl^- and SO_4^{2-} concentrations. Both of these sample sites are surrounded by cropland and are probably polluted by agricultural activities.

Fig. 4 shows the equivalent charge balance of $Ca^{2+} + Mg^{2+}$ versus HCO_3^- and versus $HCO_3^- + SO_4^{2-}$. For the Nenjiang River samples, HCO_3^- is well balanced by $Ca^{2+} + Mg^{2+}$, and SO_4^{2-} is not significant in the chemical equilibrium. But for the 2nd Songhuajiang and Songhuajiang River samples, the sum of $Ca^{2+} + Mg^{2+}$ is in excess relative to HCO_3^- but smaller than $HCO_3^- + SO_4^{2-}$. It can also be seen from Fig. 3b that SO_4^{2-} plays a relatively more important role in the Songhuajiang and the 2nd Songhuajiang than in the Nenjiang. Probably because the rivers of the former two basins drain a larger

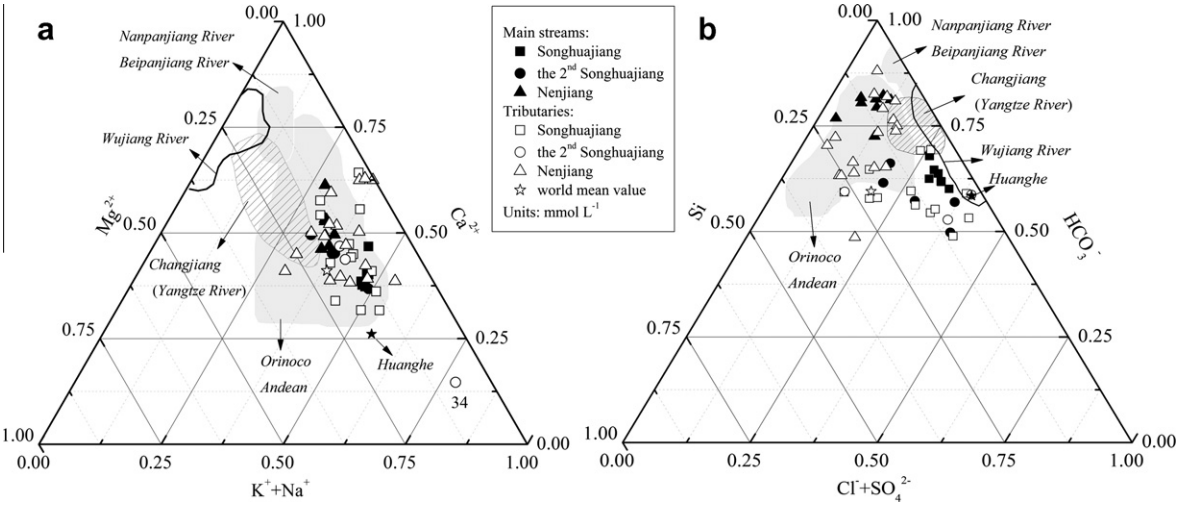

Fig. 3. Ternary diagrams showing cation (diagram a) and anion-Si compositions (diagram b). Also shown are chemical compositions of major ions of the large rivers of the world for a comparison. Data source for the Orinoco is Edmond and Palmer (1996), for Wujiang, Han and Liu (2004), for Xijiang, Xu and Liu (2007), for Changjiang, Chetelat et al. (2008), and for Huanghe, Gaillardet et al. (1999b).

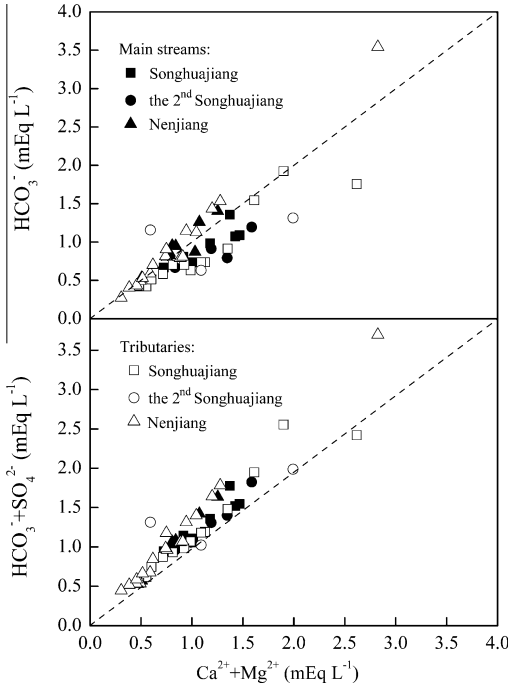

Fig. 4. Equivalent charge balance of $Ca^{2+} + Mg^{2+}$ versus $HCO_3^- + SO_4^{2-}$ (lower diagram) and versus HCO_3^- (upper diagram).

area of Quaternary sediments in which the "gypsum-salt layer" could yield SO_4^{2-}, in addition the population density is greater than that of the Nenjiang Basin.

5. Discussion

5.1. Source of dissolved load

Dissolved species in the river water are mainly derived from dry and wet atmospheric deposition, weathering of various rocks/minerals and anthropogenic inputs. So, in order to derive chemical weathering rates and associated CO_2 consumption occurring within the basin, it is important to constrain the contributions from these sources to the dissolved load in the river waters.

5.1.1. Atmospheric inputs

Chloride, as shown in many studies, is the most useful reference to evaluate atmospheric inputs to rivers, because it is conservative and is not involved in biogeochemical cycling, except for the case in small basins where biota play a dominant role (Gaillardet et al., 1997; Viers et al., 2001). Two complementary methods can be used for the correction of atmospheric inputs. When there are no salt-bearing rocks, no hydrothermal inputs in the river basin and anthropogenic inputs can be neglected, the river water sample with the lowest Cl^- concentration is assumed to have obtained its dissolved [Cl^-] exclusively from the atmosphere. This approach for estimating the atmospheric contribution has the advantage that it does not require corrections for evapo-transpiration if the samples are collected at the same time as its effect is already factored in by determining the Cl^- abundance of rivers (Rai et al., 2010), while dry atmospheric inputs are also accounted for. The other method consists of dividing the mean Cl^- concentration in rainwater by the evapo-transpiration factor (f_{et}), which is calculated as the ratio of the annual discharge over the annual precipitation received by the basin (Gaillardet et al., 1999b; Moquet et al., 2011).

In the Nenjiang River Basin, the lowest [Cl^-] concentration was found in sample 38. According to the geologic map (Fig. 1), there are no salt-bearing rocks and no hydrothermal inputs in this tributary basin. In addition, population density is less than 10 indiv km^{-2} there, much lower than the average population density of China (approximately 135 indiv km^{-2}) and NE China (average 82 indiv km^{-2}). For reference, the Amur Basin has a population density of <7 indiv km^{-2}, and pristine rivers such as the Amazon, Mackenzie and Siberian basins have population density <5 indiv km^{-2} (Moon et al., 2009). So, anthropogenic inputs of [Cl^-] to sample 38 can be neglected. Therefore, atmospheric inputs in the Nenjiang River Basin were calculated assuming that the lowest [Cl^-] concentration found among the samples (3.52 μmol L^{-1}, sample 38) is entirely from rain. This value is comparable with the one Moon et al. (2009) assigned for the Amur (about 4 μmol L^{-1}).

Meanwhile, in the Songhuajiang main stream and the 2nd Songhuajiang River systems, the lowest [Cl^-] concentrations for river water were found to be 38.2 (sample 31) and 71.1 μmol L^{-1} (sample 8), respectively. Considering relatively greater human activities in this area, using these values as exclusively rain-derived Cl^- in the correction of atmospheric inputs would overestimate the effect of precipitation (especially for the 2nd Songhuajiang River). So, atmospheric contributions of [Cl^-] to

these two basins were calculated using the second method. Mean [Cl^-] concentration of precipitation in July from the year 2003 to 2009 is used in the calculation (11.2 μmol L^{-1}, data obtained from EANET: Acid Deposition Monitoring Network in East Asia, http://www.eanet.cc, Primorskaya Russia), and evapo-transpiration factors were 0.33 and 0.30 for the 2nd Songhuajiang and the Songhuajiang main stream, respectively [(calculated from Qian, 2007]. The atmospheric contribution of [Cl^-] to river water was calculated to be 34.0 μmol L^{-1} for the 2nd Songhuajiang and 37.1 μmol L^{-1} for the Songhuajiang main stream, respectively. Although atmospheric [Cl^-] concentrations in large rivers do not exceed 30 μmol L^{-1} as described by Gaillardet et al. (1999b), there are other studies that determined the atmospheric [Cl^-] concentrations to be >this value, such as Shin et al. (2011) for the six major rivers in South Korea (49 μmol L^{-1}) and Li et al. (2009) for the upper Han River (34.7 μmol L^{-1}). Consequently, the values of atmospheric [Cl^-] concentrations calculated with the evapo-transpiration factors were used in the correction of atmospheric inputs for the 2nd Songhuajiang and the Songhuajiang main stream.

The atmospheric contribution of element X ($X = Ca^{2+}$, Mg^{2+}, Na^+, K^+ and SO_4^{2-}) to river water can be derived from the following equations:

For the Nenjiang River system:

$$X_{rr} = (X/Cl)_{rain} \times [Cl^-]_{atmospheric};$$

For rivers from the Songhuajiang main stream and the 2nd Songhuajiang:

$$X_{rr} = (X/Cl)_{rain} \times [Cl^-]_{rain}/f_{et}.$$

where X_{rr} is the contribution of element X from rain (in μmol L^{-1}) to rivers; f_{et} is the evapo-transpiration factor; $[Cl^-]_{atmospheric}$ is the atmospheric contribution of [Cl^-] to river water; $(X/Cl)_{rain}$ is the molar ratio of element X over Cl in rainwater. The X/Cl ratios of volume-weighted mean concentrations of precipitation in July from the year 2003 to 2009 were used as $(X/Cl)_{rain}$ in the calculation (data obtained from EANET, Primorskaya Russia), which are Mg/Cl = 0.31, Ca/Cl = 0.60, K/Cl = 0.49, Na/Cl = 0.99 and SO_4/Cl = 2.45. They are also in the ranges of the values calculated from the few rainwater samples analyzed in this study.

5.1.2. Anthropogenic inputs

Human activities generate both diffuse contamination (e.g., atmospheric pollutants and fertilizer application) and local contamination including sewage and industrial wastewaters. It is well known that PO_4^{3-}, NO_3^- and Cl^- are mostly derived from the agricultural fertilizers, animal waste and sewage (Grosbois et al., 2000). Calcium, Mg and HCO_3^- are conventionally considered to be insensitive to human pollution (Roy et al., 1999). Moreover, the content of TDS reflects the different lithologies in the basin and can be used as an index of land use and the effects of human activities on the water quality (Gaillardet et al., 1999a,b; Han and Liu, 2004; Chetelat et al., 2008). Variation of the NO_3^- concentration in the three main streams is shown in Fig. 5. In the main stream of the Nenjiang River, NO_3^- concentration is relatively low because the basin is sparsely populated and the agricultural activity is less intense in the western part than in the eastern part of the river basin. In contrast, the NO_3^- concentration in the 2nd Songhuajiang River is significantly high, with an average of 137 μmol L^{-1} which is about 10 times that in the Nenjiang River system (14.1 μmol L^{-1}). The NO_3^- concentrations in both the Nenjiang and the 2nd Songhuajiang rise when they flow into the Songnen Plain which is the main agricultural region. After the confluence of Nenjiang and the 2nd Songhuajiang, NO_3^- concentration in the river water is steadily high in the agricultural region and shows a sharp decrease after the confluence with the Mudanjiang River (annual discharge, 89.9×10^8 m^3 a^{-1}) due to the dilution effect. Then when the river flows into the Sanjiang plain which is an agricultural region for wheat cultivation, NO_3^- concentration increases again.

As discussed in Section 2.2, most of the population lives between Harbin and Changchun city. This area generates many domestic, agricultural and industrial wastewaters, about half of which are discharged into the rivers nearby: the total wastewater input to the rivers has been estimated to be about 15.8×10^8 t a^{-1} for the whole Songhuajiang River Basin (Water resources bulletin of the SongLiao basin 2005, 2006, 2007 and 2008). Fig. 6 shows the relationships between the molar ratios NO_3/Na, SO_4/Na, and Cl/Na for the river waters. The relatively low NO_3^-/Na^+ and Cl^-/Na^+ ratios of Nenjiang waters indicate less anthropogenic inputs into the river compared to those of the Songhuajiang main stream and the 2nd Songhuajiang River. Estimated anthropogenic contributions of [Cl^-] ranges from 1.08 to 760 μmol L^{-1} for the rivers of the Songhuajiang main stream and 37.1 to 504 μmol L^{-1} for the 2nd Songhuajiang, respectively (see Section 5.2.1). The lowest values were found for sample 31 (1.08 μmol L^{-1}) which is in the forests of Xiaoxinganling Mountains which have less human activity. Accordingly, the SO_4^{2-} concentration of sample 31 (60.3 μmol L^{-1}) is also the lowest one for samples from this region, and the concentration of NO_3^- (13.8 μmol L^{-1}) is relatively low. The highest values were found in samples 21, 33, 12 and 36, and they also have high values of NO_3^- and SO_4^{2-}. Most of the samples with high anthropogenic [Cl^-] concentrations are in the densely populated area and/or the agricultural zone with black soil (Fig. 5).

5.1.3. Rock weathering inputs

Sodium normalized molar values of each element (Ca/Na, K/Na, Mg/Na, Cl/Na, SO_4/Na, HCO_3/Na) were used instead of absolute concentrations in the determination of rock weathering. In all river water samples, the averages of these molar ratios are 1.40, 0.15, 0.42, 0.41, 0.48 and 3.40, respectively. The relationship between Ca/Na and HCO_3/Na ($r = 0.813$, $n = 54$, $p < 0.01$) is shown in a log–log space in Fig. 7a. All the samples show a cluster within a mixing area mainly between silicate and carbonate weathering.

For the rivers draining carbonate areas, like Wujiang, a karst-dominated terrain, chemical composition shows at least three end-members which are limestone, dolomite and silicate source as shown in Fig. 7b (Han and Liu, 2004). For the river water samples studied here, obvious silicate-dominant signatures could be recognized (Fig. 7a and b). The average values of the world's large rivers are also shown for comparison in the Fig. 7b. The distribution of the

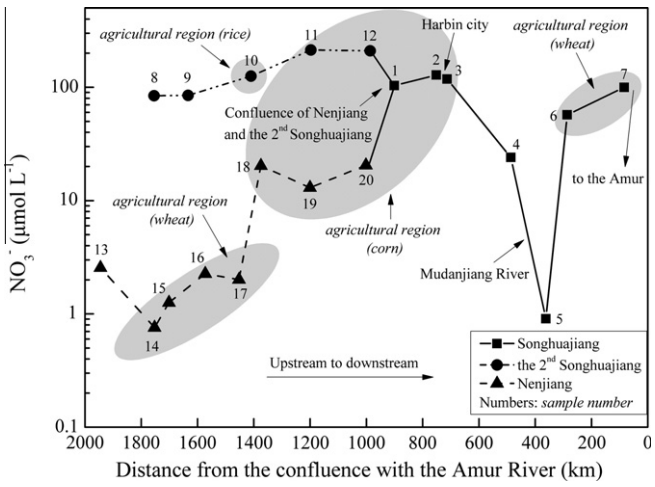

Fig. 5. Variation of NO_3^- concentration from upper to lower reaches in the mainstream of Nenjiang, the 2nd Songhuajiang, and main channel of Songhuajiang.

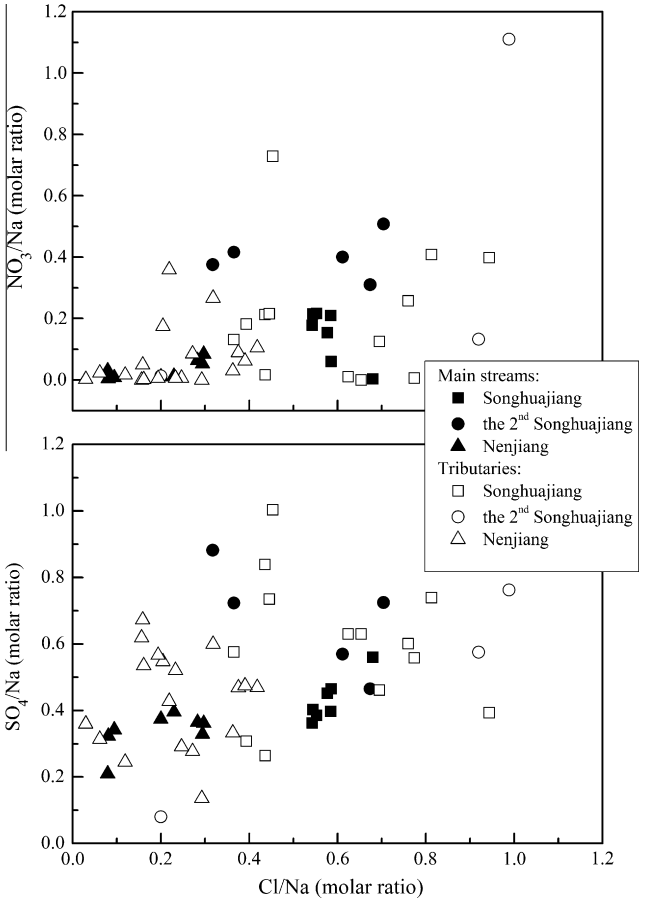

Fig. 6. Plots showing variations of SO_4/Na and NO_3/Na with Cl/Na.

present samples on the figure is in accordance with that of the world's large rivers, forming a major mixing trend of carbonate and silicate end-members, with some of the samples from the Songhuajiang main stream and the 2nd Songhuajiang River system affected by evaporites.

5.1.4. Hydrothermal sources

In the study area, primary volcanic activities could be observed at two sites: the Wudalianchi area and the Changbaishan Tianchi region (Wang et al., 1999). Most springs of the Wudalianchi area are cold ones (mostly under 15 °C). The sample Wudalianchi1 (Huoshaoshan) was collected at a relative hot spring in the Wudalianchi volcanic area and it has significantly high concentrations of some ions such as HCO_3^-, SO_4^{2-}, Cl^- and cations. The greatest effect should be observed between samples 47 and 48. However, there is no significant variation of ionic compositions between these two samples and those of the Nenjiang main stream before and after the confluence of water from the Wudalianchi volcanic area (sample 16–17). In addition, the capacity of hot springs of the Wudalianchi area is small and they flow into the lakes prior to the river. So, the effect of hydrothermal sources in the Wudalianchi volcanic area on the dissolved loads of the river is not significant. However, the springs from the Changbaishan Tianchi are hot (up to 83 °C) and and these spring waters have abundant Cl^-, HCO_3^-, and Si but low SO_4^{2-} concentrations (Gao, 2004). Sample 34 is obviously affected by hot spring water. Using the spring flow, their contribution to the total dissolved loads of the 2nd Songhuajiang at the river mouth (sample 12) was estimated It was found that solutes from the hot spring consitute <1% of the total dissolved loads, and are even less at the river mouth of Songhuajiang main stream (sample 7). Therefore, the contribution

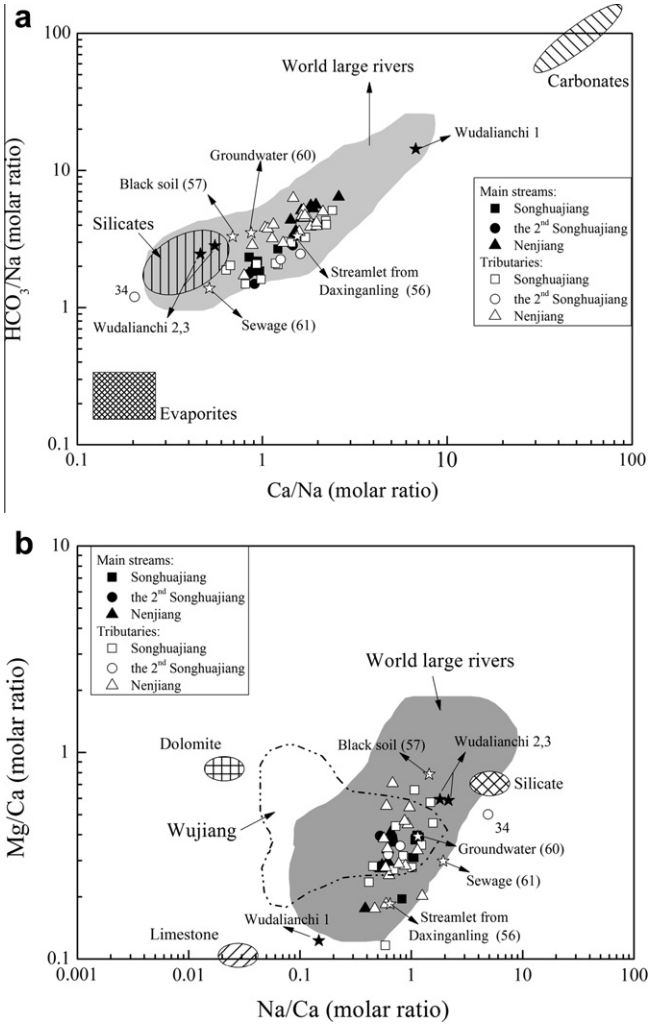

Fig. 7. Plots of Ca/Na versus HCO_3/Na (a) and Na/Ca versus Mg/Ca (b) ratios. Data for Wujiang and world large rivers are from (Han and Liu, 2004) and (Gaillardet et al., 1999b), respectively. End-members in Fig. 7a are from Gaillardet et al. (1999b), and those of Fig. 7b are from Han and Liu (2004). Sample numbers are shown by the figure.

of hydrothermal sources to the solutes of Songhuajiang River is negligable.

5.2. Chemical budget and chemical weathering rate estimation

5.2.1. Chemical budget

The silicate-derived fraction of solutes in river water is an important parameter for the calculation of long term CO_2 uptake rate by chemical weathering (Galy and France-Lanord, 1999; Bickle et al., 2005). In this study, a forward modeling approach was used to estimate the relative contribution of the different sources to the river (Galy and France-Lanord, 1999; Krishnaswami et al., 1999; Mortatti and Probst, 2003; Han and Liu, 2004; Moon et al., 2007; Xu and Liu, 2007).

The solutes in rivers are a mixture of atmospheric inputs, anthropogenic inputs and weathering of silicate, carbonate and evaporites, as well as sulfide minerals. So, for any element X in the river water the budget equation can be written as follows:

$$[X]_{river} = [X]_{atmospheric} + [X]_{anthropogenic} + [X]_{carbonate} + [X]_{silicate} + [X]_{evaporite} + [X]_{sulfide} \quad (1)$$

The subscripts stand for the different sources of element X.

The calculations are based on some straightforward simplifications of the budget equation. First, it is assumed that Cl^- in the river water is only derived from atmospheric and anthropogenic inputs. As described in Section 5.1.1, $[Cl]_{atmospheric}$ were determined to be 34.0 μmol L^{-1} for the 2nd Songhuajiang, 37.1 μmol L^{-1} for the Songhuajiang main stream, and 3.52 μmol L^{-1} for Nenjiang, respectively. So, the excess of $[Cl^-]$ over $[Cl]_{atmospheric}$ is assumed to be anthropogenic and balanced by Na (Li et al., 2009); Second, according to the geological map, all SO_4^{2-} after rain correction is from evaporite (gypsum) dissolution and sulfide oxidation (Fig. 1); Third, the dissolution of carbonate does not contribute to the dissolved Na and K and evaporite dissolution yields only $Ca^{2+} + SO_4^{2-}$; Fourth, the anthropogenic contribution to the cations such as Ca^{2+}, Mg^{2+} and K^+ is neglected; Moreover, all Na after the atmospheric and anthropogenic corrections and all K after the rain correction were assumed to be from silicate weathering.

Based on these assumptions, for element *X*, Eq. (1) can be simplified as follows:

$$[Cl]_{atmospheric\text{-}Nenjiang} = 3.52\ \mu mol\ L^{-1} \quad (2.1)$$

$$[Cl]_{atmospheric\text{-}the\ 2nd\ Songhuajiang} = 34.0\ \mu mol\ L^{-1} \quad (2.2)$$

$$[Cl]_{atmospheric\text{-}Songhuajiang\ main\ stream} = 37.1\ \mu mol\ L^{-1} \quad (2.3)$$

$$[Cl]_{river} = [Cl]_{atmospheric} + [Cl]_{anthropogenic} \quad (3)$$

$$[SO_4]_{river} = [SO_4]_{atmospheric} + [SO_4]_{evaporite} + [SO_4]_{sulfide} \quad (4)$$

$$[Na]_{river} = [Na]_{atmospheric} + [Na]_{silicate} + [Na]_{anthropogenic} \quad (5)$$

$$[K]_{river} = [K]_{silicate} + [K]_{atmospheric} \quad (6)$$

$$[Ca]_{river} = [Ca]_{carbonate} + [Ca]_{silicate} + [Ca]_{evaporite} + [Ca]_{atmospheric} \quad (7)$$

$$[Mg]_{river} = [Mg]_{carbonate} + [Mg]_{silicate} + [Mg]_{atmospheric} \quad (8)$$

As discussed by many authors (Galy and France-Lanord, 1999; Han and Liu, 2004; Xu and Liu, 2007), Eqs. (7) and (8) are difficult to solve because of the uncertainty about silicate versus carbonate contributions. In most river systems, Sr isotopic composition is an additional useful constraint, but unfortunately Sr isotope data are not available at the moment. So, the median value of $(Ca/Na)_{sil} = 0.44 \pm 0.23$, $(Mg/Na)_{sil} = 0.16$ and $(Ca/Mg)_{carb} = 2.5$ (calculated from the medians of local carbonate rocks, $Ca_{carb}/Na_{carb} = 50 \pm 20$, $Ma_{carb}/Na_{carb} = 20 \pm 8$) according to (Moon et al., 2009) for the Amur was used, since Songhuajiang is a tributary of the Amur and the geologic environment of the two basins is similar. The ratio of $(Ca + Mg)_{sil}/Na_{sil}$ was assigned to be 0.6, which is similar to the global ratio and within the range of the local rock composition (Frost et al., 1998) and also within the range described by Gaillardet et al. (1999b) for world rivers (0.59 ± 0.17). Then Eqs. (7) and (8) can be further simplified as follows:

$$[Ca]_{river} = 2.5 \times [Mg]_{carbonate} + 0.44 \times [Na]_{silicate} + [Ca]_{evaporite} + [Ca]_{atmospheric} \quad (9)$$

$$[Mg]_{river} = [Mg]_{carbonate} + 0.16 \times [Na]_{silicate} + [Mg]_{atmospheric} \quad (10)$$

The silicate to carbonate budget can be expressed by the ratio of dissolved cations from silicates over the sum of dissolved cations from silicates plus carbonates. This can be expressed as the ratio of equivalent cationic charge by combining Eqs.)((5)–(10):

$$X_{sil} = \frac{[Na]_{sil} + [K]_{sil} + 2 \times [Ca]_{sil} + 2 \times [Mg]_{sil}}{[Na]_{sil} + [K]_{sil} + 2 \times [Ca]_{sil} + 2 \times [Mg]_{sil} + 2 \times [Ca]_{carb} + 2 \times [Mg]_{carb}} \quad (11)$$

where the subscripts sil and carb stand for silicate and carbonate, respectively.

Results of the relative contributions from different sources to the cationic TDS (mg L^{-1}) for the Nenjiang River and both Songhuajiang and the 2nd Songhuajiang River system are illustrated in Figs. 8 and 9. Generally, the dissolved cation load of the rivers in the study area is dominated by both carbonate and silicate weathering, which together account for about 70% for the whole basin.

It is notable that the contribution of carbonate weathering to the dissolved cation load averages 39% even though no distinct distribution of carbonates could be found in the basin. Blum et al. (1998) found that carbonates provide more than 90% of the weathering-yield Ca even though carbonates represent only about 1% of fresh glacial till. It was estimated that this result was valid for a period of 55 ka, and then the mechanical exposure of the rock and the flux derived from carbonate will decrease while the flux derived from silicate weathering will increase (Viers et al., 2004). In the Nenjiang River Basin, X_{sil} ranged from 0.20 to 1.00, averaging 0.51; the 2nd Songhuajiang Basin X_{sil} ranged from 0.08 to 1.00, averaging 0.33, and for the basin of the Songhuajiang main stream X_{sil} ranged from 0.07 to 1.00, averaging 0.39. This corresponds with the distribution of silicate rocks. The Nenjiang River Basin is mainly covered by granites and volcanic rocks and the value of X_{sil} is relatively high. As for the other two basins, they have more tributaries draining Quaternary sediment areas in which secondary carbonate may exist. High X_{sil} values are observed in samples 43, 44 and 45 from the Gan River, sample 31 from the Tangwang River and sample 34 from the Rdaobai River. These rivers flow through areas covered by granites or volcanic rocks, no carbonates or Quaternary sediments could be found in their basins. In addition, diagenetic carbonate minerals or secondary carbonates may exist in the widely distributed Quaternary sediments which are more susceptible to weathering compared to granites and volcanic rocks.

5.2.2. Chemical weathering and CO_2 consumption rate

Together with surface area and hydrological data available for each basin, chemical weathering rates of silicates and carbonates in the basins were estimated from the budget of silicate/carbonate weathering. The chemical weathering rate of silicates (TDS_{sil}) is calculated using the Na^+, K^+, Ca^{2+} and Mg^{2+} concentrations from silicate weathering and assuming that all dissolved SiO_2 is derived from silicate weathering.

$$TDS_{sil} = [Na]_{sil} + [K]_{sil} + [Ca]_{sil} + [Mg]_{sil} + [SiO_2]_{river} \quad (12)$$

The dissolution of carbonate minerals in a catchment may take place due not only to the attack by H_2CO_3 derived from dissolution of CO_2 in the water, but also to attack by H_2SO_4 produced by the input of atmospheric SO_2 or the oxidation of sulfides (Han and Liu, 2004). Eqs. (13) and (14) describe the oxidation of sulfide and dissolution of carbonate minerals attacked by H_2SO_4, respectively. Together with Eq. (4) HCO_3^- yield by the dissolution of carbonate minerals attacked by H_2SO_4 could be estimated. However, the carbonate-yield TDS (TDS_{carb}) are equal whether the process involves only H_2CO_3 or both H_2CO_3 and H_2SO_4. Likewise, silicate minerals could also be attacked by H_2SO_4 and silicate weathering might be coupled with physical erosion by the generation of sulfuric acid (Calmels et al., 2007). However, the effect of SO_4^{2-} in the study area is not significant and the reaction between sulfuric acid and silicate minerals is less probable due to the lowest reactivity of silicates compared to carbonates (Beaulieu et al., 2011), so its effect on the dissolution of silicate minerals was neglected.

$$FeS_2 + 7/2O_2 + H_2O \rightarrow Fe^{2+} + 2SO_4^{2-} + 2H^+ \quad (13)$$

$$2Ca_xMg_{1-x}CO_3 + H_2SO_4 = 2xCa^{2+} + 2(1-x)Mg^2 + +2HCO_3^- + SO_4^{2-} \quad (14)$$

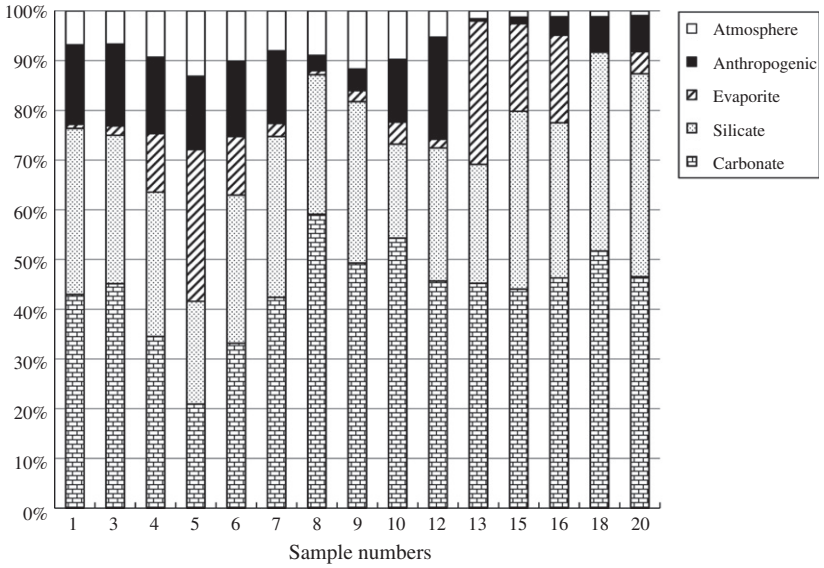

Fig. 8. Calculated contributions (in%) of the different reservoirs to the cationic TDS (mg L^{-1}) for the main streams of the rivers. Cationic TDS is equal to the sum of K^+, Na^+, Ca^{2+} and Mg^{2+} from the different reservoirs. Samples 1–7 are from the Songhuajiang main stream; samples 8–12 from the 2nd Songhuajiang; samples 13–20 from Nenjiang.

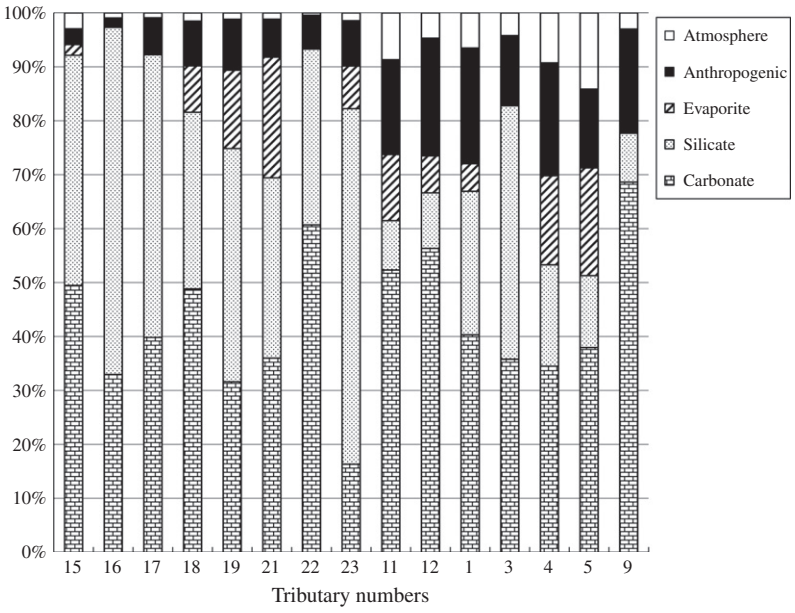

Fig. 9. Calculated contributions (in%) of the different reservoirs to the cationic TDS (mg L^{-1}) for the tributaries. Cationic TDS is equal to the sum of K^+, Na^+ Ca^{2+} and Mg^{2+} from the different reservoirs. Tributaries 15–23 are from the Nenjiang River system; tributaries 11 and 12 from the 2nd Songhuajiang River system; tributaries 1–9 from the Songhuajiang main stream.

The rate of carbonate weathering (TDS_{carb}) is calculated using the dissolved Ca and Mg from carbonate weathering and HCO_3^-.

$$TDS_{carb} = [Ca]_{carb} + [Mg]_{carb} + 1/2[HCO_3]_{carb} \quad (15)$$

During the dissolution of silicates, all yield [HCO_3^-] is derived from the atmosphere, so CO_2 consumption from silicate weathering ($CO_{2\text{-sil}}$) is calculated according to the equivalent charge balance between silicate-derived alkalinity and silicate-derived cations, and is independent of aqueous silica concentration.

$$CO_{2-sil} = [HCO_3]_{sil} = [Na]_{sil} + [K]_{sil} + 2 \times [Ca]_{sil} + 2 \times [Mg]_{sil} \quad (16)$$

While the amount of atmospheric CO_2 consumed by carbonate weathering ($CO_{2\text{-carb}}$) attacked by only H_2CO_3 equals one half of the carbonate-yield [HCO_3^-], which can be calculated as follows:

$$\begin{aligned} CO_{2\text{-carb}} &= 1/2 \times [HCO_3]_{carb} \\ &= 1/2 \times (2 \times [Ca]_{carb} + 2 \times [Mg]_{carb}) \\ &= [Ca]_{carb} + [Mg]_{carb} \end{aligned} \quad (17)$$

The last sample of each basin was used to calculate the rock weathering rates (sample 20 for the Nenjiang River, sample 12 for the 2nd Songhuajiang and sample 7 for the whole Songhuajiang River Basin, respectively). The results of the calculation for each basin are listed in Table 2. As shown in the table, silicate weathering rate is lower than carbonate weathering rate both in the main streams and most of the tributaries. Mudanjiang River, the biggest tributary of the Songhuajiang main stream, has a higher total rock weathering rate (10.6 t km^{-2} a^{-1}) than that of the Nenjiang River (5.03 t km^{-2} a^{-1}) probably due to the higher temperature and

Table 2
Chemical weathering and CO_2 consumption rates for the Songhuajiang River Basin, NE China.

Rivers	No. of sample used	Discharge (10^8 m^3 a^{-1})	Drainage area (10^3 km^2)	Silicates				Carbonates			Evaporites	Total rock weathering TDS (t km^{-2} a^{-1})
				$Cation_{sil}$[a] (t km^{-2} a^{-1})	TDS_{sil}[b] (t km^{-2} a^{-1})	$(Ca + Mg)_{sil}$[c] (10^9 mol a^{-1})	CO_2 cons.[d] (10^3 mol km^{-2} a^{-1})	$Cation_{carb}$[e] (t km^{-2} a^{-1})	TDS_{carb}[b] (t km^{-2} a^{-1})	CO_2 cons.[d] (10^3 mol km^{-2} a^{-1})[g]	TDS_{eva}[f] (t km^{-2} a^{-1})	
Tributaries of the Nenjiang River												
(15) Duobukur River	39	12.3	5.76	0.80	3.01	0.06	38.3	1.31	3.56	36.9/32.1	0.13	6.69
(18) Gan River	46	40.7	19.6	1.15	2.94	0.26	53.4	2.02	5.49	57.0/53.3	1.03	9.46
(20) Nuomin River	49	46.4	25.5	1.13	2.98	0.35	53.5	0.10	0.26	2.71/0.89	1.45	4.69
(21) Alun River	50	9.02	6.30	1.05	2.93	0.08	50.0	1.14	3.11	32.3/32.2	2.40	8.44
(25) Chuor River	54	23.7	17.4	1.28	1.96	0.28	60.7	1.37	3.73	38.7/38.0	2.19	7.88
(16) Menlu River	40	8.89	5.46	2.89	4.59	0.20	139	3.29	8.97	93.0/83.5	0.00	13.6
(17) Keluo River	42	10.7	8.40	1.88	2.83	0.17	84.2	2.30	6.27	65.0/59.0	0.00	9.10
(19) Nemor River	48	17.0	13.9	1.15	1.84	0.17	50.8	0.83	2.26	23.5/23.5	1.32	5.42
Tributaries of the 2nd Songhuajiang River												
(11) Huifa River	35	35.0	15.1	0.61	2.23	0.01	16.4	3.47	9.45	98.0/95.2	2.77	14.4
(12) Yinma River	36	11.4	7.52	0.83	1.27	0.03	28.8	4.53	12.32	128/115.4	1.87	15.5
Tributaries of the Songhuajiang River Main stream												
(3) Hulan River	24	47.9	32.1	4.54	5.74	1.79	215	3.45	9.40	97.5/80.8	0.00	15.1
(8) Tangwang River	32	58.2	20.5	1.20	4.36	0.31	57.1	0.21	0.56	5.84/5.8	7.47	12.4
(5) Mudanjiang River	28	89.9	37.7	0.61	2.75	0.20	25.1	1.74	4.75	49.2/49.2	3.12	10.6
Mainstreams												
Nenjiang	20	225	297	0.86	1.39	3.11	40.6	1.22	3.31	34.3/31.4	0.32	5.03
The 2nd Songhuajiang	12	169	73.4	2.91	4.74	2.24	128	4.96	13.5	140/116	0.65	18.9
The whole Songhuajiang River Basin	7	733	557	1.44	2.23	9.30	66.6	1.89	5.15	53.4/48.6	0.40	7.78
Other rivers of China												
Huanghe (Yellow River)[h]	—	410	752	2.13	3.46	19.0	82.4	—	4.65	39.9/—	6.65	14.8
Changjiang (Yangtze River)[i]	CJ-36	8650	1,705	2.40	2.35	43.0	112	14.0	36.4	379/—	4.11	42.8
Xijiang (from Pearl River)[j]	49	2300	352	3.26	7.45	13.9	154	30.0	78.5	807/—	0.20	86.1

[a] $Cation_{sil}$: the sum of cation (Na + K + Ca + Mg) concentrations derived from silicate weathering.
[b] See text for TDS_{sil} and TDS_{carb}.
[c] $(Ca + Mg)_{sil}$: the sum of Ca + Mg derived from silicate weathering.
[d] CO_2 cons.: the CO_2 consumption rates based on the cation contribution.
[e] $Cation_{Carb}$: the sum of cation (Ca + Mg) concentrations derived from carbonate weathering.
[f] TDS_{eva} values are calculated based on Na and Ca + Mg estimated from the mass budget and their stoichiometric equivalent of Cl and SO_4, respectively.
[g] The numbers (a/b), a stands for CO_2 consumed by the weathering of carbonate attacked by only H_2CO_3; b stands for CO_2 consumed by the weathering of carbonate attacked by both H_2CO_3 and H_2SO_4.
[h] Gaillardet et al. (1999b).
[i] Chetelat et al. (2008).
[j] Xu and Liu (2010).

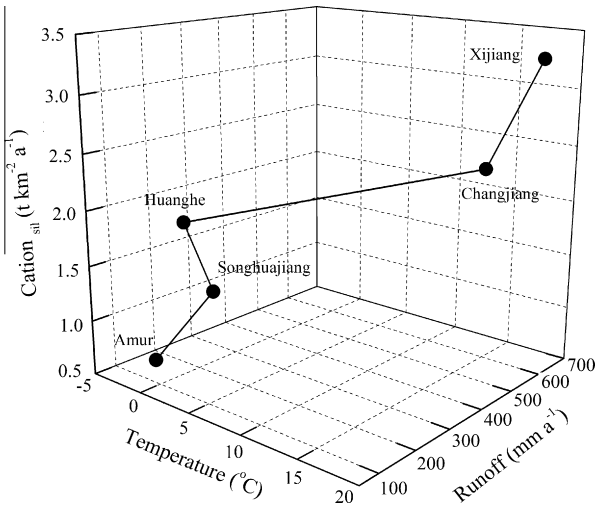

Fig. 10. Three-dimensional representation of the Cationsil flux variations along with annual temperature and runoff in the large river basins of China. Temperature and runoff are the mean values in each basin. Data sources are Moon et al. (2009) for the Amur, Gaillardet et al. (1999b) for Huanghe, Chetelat et al. (2008) for Changjiang, and Xu and Liu (2010) for Xijiang.

precipitation within its basin. For the two headstreams of the Songhuajiang River, both silicate and carbonate rock weathering rates are higher in the 2nd Songhuajiang River Basin. This can be attributed primarily, to the lithology which is the most important parameter controlling chemical weathering rates since the upper reaches of the 2nd Songhuajiang are mainly covered by basalt which is more easily weathered than acidic rocks. In addition, the 2nd Songhuajiang drops 1556 m along its 958 km long channel while Nenjiang drops 442 m from its source along a length of 1370 km, and high relief may enhance mechanical erosion by increasing the surface contact between solution and solid (Viers et al., 2004). In addition, the 2nd Songhuajiang Basin is located in the SE part of the study area, annual precipitation and temperature is higher in this basin than those in the Nenjiang River Basin. In addition, runoff in the 2nd Songhuajiang Basin (230 mm a^{-1}) is much higher than that in the Nenjiang Basin (76 mm a^{-1}). So chemical weathering in the 2nd Songhuajiang Basin may also be enhanced by climatic parameters like temperature, precipitation and runoff (Oliva et al., 2003). Finally, organic matter (vegetation) is probably another parameter since the upper and middle reaches of the 2nd Songhuajiang are mainly covered by forests and plants which may enhance the chemical weathering process in a steep topography, even the effect of vegetation on the chemical weathering rates is complex and difficult to investigate (Berner, 1992; Drever, 1994; Lucas, 2001).

The CO_2 consumption rates of silicate and carbonate weathering (only H_2CO_3) in the Nenjiang Basin are 40.6×10^3 and 34.3×10^3 mol km^{-2} a^{-1}, respectively. In the 2nd Songhuajiang River Basin, the rates are 128×10^3 and 140×10^3 mol km^{-2} a^{-1} for silicate and carbonate weathering, respectively. Within the whole Songhuajiang Basin, CO_2 consumption fluxes from both carbonate and silicate weathering are comparable, being 29.8×10^9 mol a^{-1} and 37.1×10^9 mol a^{-1} and account for ~0.24% and ~0.43% of the annual global CO_2 consumption by carbonate and silicate rocks, respectively [the annual global CO_2 consumption has been estimated to be 12300×10^9 mol a^{-1} from carbonate weathering and 8700×10^9 mol a^{-1} by continental silicate rock weathering, Gaillardet et al., 1999b]. With the addition of H_2SO_4 in the weathering of carbonate rocks, CO_2 consumption reduced to 48.6×10^3 mol km^{-2} a^{-1}. Compared with the rivers originating in the Qinghai-Tibet Plateau, CO_2 consumption flux by the weathering of silicate rocks within the 2nd Songhuajiang Basin (128×10^3 mol km^{-2} a^{-1}) is comparable with that of the Huanghe (120×10^3 mol km^{-2} a^{-1}, Wu et al., 2005), Brahmaputra (150×10^3 mol km^{-2} a^{-1}, Gaillardet et al., 1999b) and Dadu He (180×10^3 mol km^{-2} a^{-1}, Wu et al., 2008). The CO_2 consumption from the weathering of silicate rocks within the whole Songhuajiang Basin (66.6×10^3 mol km^{-2} a^{-1}) is on the same order of magnitude with that of the MeKong (70×10^3 mol km^{-2} a^{-1}, Wu et al., 2008). The value for Nenjiang Basin (40.6×10^3 mol km^{-2} a^{-1}) is relatively lower, but is comparable with that of the Indus (60×10^3 mol km^{-2} a^{-1}, Gaillardet et al., 1999b). When compared with other mountain/orogenic area rivers, CO_2 consumed by the weathering of silicate rocks within the whole Songhuajiang Basin (66.6×10^3 mol km^{-2} a^{-1}) is much lower than that of the Amazon rivers draining Andes and sub-Andes areas (300×10^3 mol km^{-2} a^{-1}, Moquet et al., 2011), but is comparable with that of the Amazon Basin (52.4×10^3 mol km^{-2} a^{-1}) and is higher than that of the Yukon (34.2×10^3 mol km^{-2} a^{-1}) (Gaillardet et al., 1999b).

5.3. Climate (temperature and runoff) as a main control on chemical weathering

It has been agreed that many factors, such as climate (temperature, runoff), lithology, tectonics, vegetation and human activities, control chemical weathering of river basins. However, the dominant factors have different controlling mechanisms under different climates, as reported by previous authors (Riebe et al., 2001; Dosseto et al., 2006; Liu et al., 2012). Within the study area, chemical weathering rates of silicate and carbonate in the 2nd Songhuajiang River Basin are much higher than that of the Nenjiang River Basin. This is probably because of the different climatic factors of these two basins as discussed in Section 5.2.2.

When compared with the nearby rivers, the flux of Cation$_{sil}$ in the Songhuajiang Basin (1.44 t km^{-2} a^{-1}) is higher than those of the Amur River [0.67 t km^{-2} a^{-1}, Moon et al., 2009] and of Siberian rivers draining platform regions [0.39 t km^{-2} a^{-1}, Huh et al., 1998]. Cation$_{sil}$ values for various rivers vary in the order Songhuajiang (1.44 t km^{-2} a^{-1}, T = 4–5 °C) < Huanghe [2.13 t km^{-2} a^{-1}, T = −4 to 14 °C, Gaillardet et al., 1999b] < Changjiang [2.40 t km^{-2} a^{-1}, T = 16–18 °C, Chetelat et al., 2008] < Xijiang [3.26 t km^{-2} a^{-1}, T = 14–22 °C, Xu and Liu, 2010] < Amazon rivers draining orogenic regions [5.34 t km^{-2} a^{-1}, T = 25–27 °C, Edmond and Palmer, 1996; Moon et al., 2009]. The silicate weathering rate (TDS$_{sil}$ = 2.23 t km^{-2} a^{-1}) of Songhuajiang Basin is far lower than that of the Nethravati River [42 t km^{-2} a^{-1}, Gurumurthy et al., 2012], which is also a monsoon-controlled river basin draining granite areas but with a much higher mean temperature (around 30 °C). Fig. 10 shows the variation of Cation$_{sil}$, runoff and temperature in the large river basins of China. The relationship illustrates that the fluxes of Cation$_{sil}$ for these basins increase linearly with annual temperature (for all the river basins) and runoff (with the exception of Huanghe). This is in agreement with Meybeck (1994) and White and Blum (1995). Compared with Songhuajiang, Huanghe has a lower runoff (54 mm a^{-1}) but higher values of Cation$_{sil}$ (2.13 t km^{-2} a^{-1}), suggesting that only temperature and runoff cannot fully explain the weathering mechanisms.

Of all the factors controlling chemical weathering rates, lithology is the most important one, since different kinds of rocks have different properties and different weathering susceptibility, e.g. Viers et al. (2004). The lithology of the Songhuajiang Basin is dominated by silicate rocks, and hence the contribution from silicate weathering to the dissolved cations is relatively high (about 30%, Figs. 8 and 9) compared with the world's large rivers. However, the calculated silicate weathering rate (TDS$_{sil}$, 2.23 t km^{-2} a^{-1}) is much lower even when compared with rivers draining carbonate dominated basins, such as Wujiang [6.0 t km^{-2} a^{-1}, (Han and Liu, 2004)] and Xijiang

[7.45 t km^{-2} a^{-1}, (Xu and Liu, 2010)]. This may be attributed to the importance of annual temperature and runoff, though silicate weathering rates are not governed by any single parameter (White and Blum, 1995; White and Brantley, 1995; West et al., 2005). As for the large rivers in China, Huanghe has the largest evaporite yield TDS (6.65 t km^{-2} a^{-1}) and Xijiang has the largest CO_2 consumption rates both from silicate weathering (154×10^3 mol km^{-2} a^{-1}) and carbonate weathering (807×10^3 mol km^{-2} a^{-1}). Carbonate-yield TDS in the Xijiang and Changjiang basins (78.5 and 36.4 t km^{-2} a^{-1}, respectively) are much higher than those in the Songhuajiang and Huanghe basins (5.15 and 4.65 t km^{-2} a^{-1}, respectively).

6. Conclusions

The chemical compositions of major ions in the Songhuajiang River water show that weathering of silicate and carbonate provides the majority of solutes in the river water. Calcium is the dominant cation, which accounts for approximately 46% of the total cation composition, followed by Na^+, accounting for more than 30% of the total cations. Bicarbonate, followed by $SO_4^{2-} + Cl^-$, are the dominant anions, accounting for approximately 74% of the total anion composition.

The chemical budget and weathering rates, as calculated according to a simplified forward model, show that the dissolved cation loads in the Songhuajiang River originated mostly from carbonate (about 40%) and silicate (about 30%) weathering. Cations in Songhuajiang main stream are most affected by human activities (average 17%) and those of Nenjiang are least affected (average 4%). The chemical weathering rates are estimated to be approximately 5.15 and 2.23 t km^{-2} a^{-1} for carbonate and silicate rocks, respectively. Total rock weathering rate (7.78 t km^{-2} a^{-1}) is at the lower end of the spectrum for world rivers. The rates of CO_2 consumption by carbonate and silicate weathering, respectively, are estimated to be 53.4×10^3 and 66.6×10^3 mol km^{-2} a^{-1} within the whole Songhuajiang River Basin. Within the whole Songhuajiang Basin or Songnen Plain, the 2nd Songhuajiang River had the highest while the Nenjiang River Basin had the lowest weathering rate, which indicates an important role for regional climate and lithology variation among the major controls on chemical weathering.

For the large rivers of China, the total rock weathering rate increases from north to south, which supports the idea that the climate acts as a major control on global chemical weathering. For the whole of the Songhuajiang River Basin, CO_2 consumption by silicate weathering is 66.6×10^3 mol km^{-2} a^{-1}, and is comparable with that of the Huanghe (82.4×10^3 mol km^{-2} a^{-1}). In this aspect, the Songhuajiang River Basin plays an important role in long-term climate change and cannot be neglected.

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