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ORIGINAL PAPER

Behavior of major and minor elements in a temperate river estuary to the coastal sea

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Abstract Water samples were collected for 23 different stations along a cross section profile of an estuary extending over to adjacent sea. The collected water samples were filtered and analyzed for major–minor ions and strontium isotope using the standard procedure to understand the geochemical behaviors of major and minor elements. The normalized values indicated that all riverine elements were entering to adjacent coastal sea with some significant variations at the estuary. The seawater dilution and regression lines explain about the overall patterns for seven elements. Removal processes were detected on calcium, magnesium, strontium and sulfate in the estuarine region. No significant mineral precipitation observed to release magnesium with respect to calcium. Minor variations of strontium and sulfate ions could be attributed to the presence of organic matter in the study area. Comparing seven elements with total suspended matters revealed that the total suspended matters played crucial role in either adsorption or absorption of all the elements in estuary before it reaches to coastal sea. Mixing patterns of strontium isotope showed minimal non-conservative with an evidence of active geochemical process in the estuary.

Keywords Adsorption and absorption · Changjiang river estuary · Major and minor elements · Strontium isotope · Total suspended matter

Introduction

Major and minor element concentrations are being used in the river and its drainage basin to understand the effect of weathering, erosion and anthropogenic process as a proxy because of high abundance in the earth crust. Rivers transport the major geochemical information from continents to ocean through estuary. Estuarine environments are an important part for the elemental cycles and also act as a marginal filter (Lisitsyn 1995), and regulate the amount of river-borne elements entering to the coastal environment and ultimately the deep ocean (Ramanathan et al. 1993). The estuaries dilute all types of dissolved and solid materials due to tidal influences over a period of residence time and generate green house gases through digenesis-autogenic process which affect the overall elemental compositions (BrichGF and Teutsch 1996). The estuarine turbidity zone (density driven circulation) interacts with these elements. Thus, the estuarine mixing and subsequent exchange with adjacent waters those are fundamental process governing biological, chemical and sediment interactions within coastal and estuarine system (Bell 1994).

If these ion constituents altered substantially in the estuarine region, it may not reflect the river elements transport flux precisely to the adjacent sea.

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This paper attempts to provide and quantify those altered processes made at Changjiang river estuary, the third largest river in the world (Wang et al. 2007). Numerous hydrology and biogeochemistry study had conducted and discussed (Hu et al. 1982; Zhang et al. 1990; Chen et al. 2002 Yang et al. 2002; Ding et al. 2004). In addition, turbidity distributions in relation with tidal fluctuations and export nature to coastal sea have been studied too (Zhou 1994; Zhang et al. 1995; Wang et al. 1995; Hu and Hu 1995; Shi and Ling 1999; Li et al. 2000a, b; Jiang and Yao 2002; Li and Zhang 2003). But the impact and influence of estuarine maximum turbidity to the behavior of major and minor elements have not been investigated. In this regard, an observation has been made during June 2005 to understand the geochemistry of major and minor elements, their variation and behavior during the transition from river to coastal sea.

Materials and methods

Study area

The Changjiang river originates in the Qinghai–Tibet Plateau and flows eastwards for 6,300 km and meets East China Sea, form a delta Chogming Island (Yi et al. 2003). The upper basin is characterized by complex rock compositions, including widely distributed Paleozoic carbonate rock in the south with an area of 400 × 10^3 km^2 (about 20 % of the total carbonate area in China); Jurassic red sandstone in the Sichuan Basin; and Mesozoic igneous rocks in the river source area. The middle–lower basin mostly consists of Paleozoic marine and quaternary fluviolacustrine sedimentary rocks, together with intermediate to felsic igneous rocks that are common but sporadic (Yang et al. 2004). The major part of the Changjiang basin is influenced by East Asian monsoon. Annual atmospheric precipitation ranges from less than 400 mm in the river source area to more than 2,000 mm in highlands of the middle–lower reaches, with an average of 1,100 mm (Yang et al. 2004).

The study area was selected at the lower reach of Changjiang river between Nanjing City and East China Sea (Fig. 1). The Huangpu River (station no. 7) passes through the Shanghai city and empties at the mouth. The Chongming Island divides the Changjiang river channel in the downstream area. The South branch downstream from the Liuhe is again divided in the North and South Channels by the Changxing and Hengsha Island, and finally the South Channel branches into the North and South passage downstream from Jiuduansha, thus the river has three points of bifurcation and four outlets before entering into the East China Sea (Wang and Liu 2003; Li et al. 2000a, b). The water mixes very forcefully in these long channels that lead to energetic resuspension of bottom sediments (Li et al. 2000a, b).

Sample collection and analytical procedure

A total of 34 water samples were collected engaging a mechanized boat (23 surfaces and 11 bottoms) during June 2005 along the Changjiang river. The surface water samples were collected using a clean plastic bucket and bottom water samples were collected using 5-L Niskin bottle. Parameters like pH, specific conductance (Ec), total dissolved solids (TDS) and alkalinity (HCO_3) were measured on board soon after every sampling. The pH was measured with a calibrated portable mobile pH-meter that had been calibrated (NBS 7 and NBS 10) at every 10 sample intervals. Specific conductance and TDS were measured using special HORIBA electrode. The HCO_3^- measurements were performed with a digital titrate on the 50 ml of the collected sample using 0.01 M HCl. Chlorinity (Cl ‰) was measured by argentometric titration method in the shore

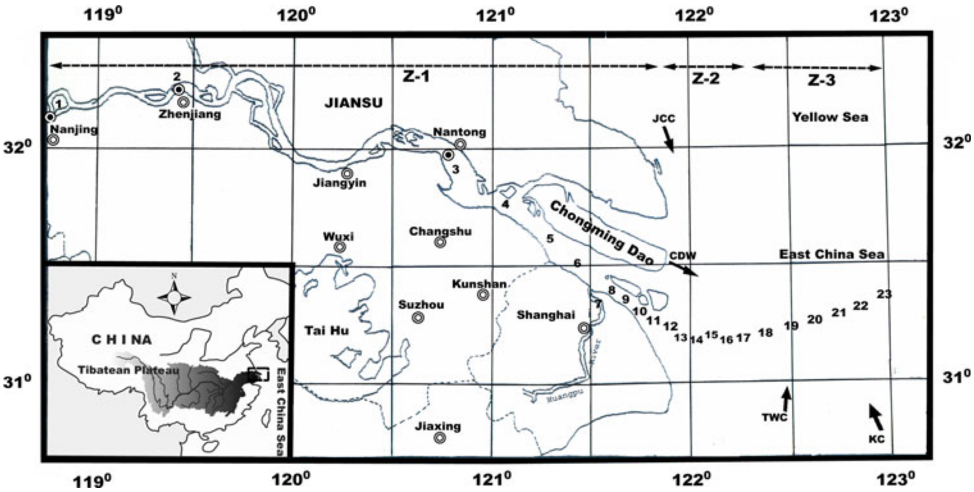

Fig. 1 Map of the sampling locations along the Changjiang river estuary from Nanjing City to East China Sea and the no. *7* represents the Huangpu River

laboratory, and the silver nitrate solution ($AgNO_3$) was standardized with standard seawater of chlorinity 19.374 ‰ obtained from Copenhagen, Denmark. The salinity calculations were made after correction from Knudsen's table (Knudsen 1902).

About 1 l of water samples filtered through pre-weighted acid-washed 0.45 µm cellulose acetate filter paper (Millipore). The filtered paper was warped with aluminum foil and kept in Petri-dish for total suspended matter (TSM) calculations. The filtered water sample was split into various analytical purposes, 50 ml of the filtered samples was stored separately in clean HDPE bottles for anion analysis, and another 50 ml of the samples was acidified with ultra-purified HNO_3 and stored in a clean HDPE bottle for cation measurements. Sufficient care was taken for the samples (i.e. collection, filtration and analysis) to avoid contamination and stored in a cool and dark place until analysis.

Cations were analyzed by inductively coupled plasma-optical emission spectrometry (ICP-OES) and the anions were analyzed by ion chromatography (ICS-90, DIONEX). To separate dissolved strontium from major element in the water for measuring Sr isotopic composition, the filtered water samples were directed to pass through an AG50 W-50 resin column through HCl medium. All strontium isotope ($^{87}Sr/^{86}Sr$) compositions were measured using thermal ionization mass spectrometry (TIMS: GV Instruments) at the State Key Laboratory, Institute of Geochemistry, CAS, Guiyang. The value $^{87}Sr/^{86}Sr$ for NSB987 strontium standard was 0.710236 ± 0.000012 ($n = 6$) during the period of sample measurement.

Results and discussion

General characteristics of the river

All stations chosen for sampling were divided into three Zones according to their salinity gradient. These are classified as Changjiang river downstream area (Stn. 1–11; Salinity <0.1 psu), Changjiang river estuary (Stn. 12–16; Salinity 0.1–14 psu) and Coastal sea (Stn. 17–23; Salinity 15–32.3 psu). The results of the chemical and isotopic compositions are summarized in Table 1. The pH varied from 7 to 8.7 with a nonsystematic variation from river to coastal sea. Mook and Koene (1975), Ramanathan et al. (1993) and Wang and Liu (2003) observed the unsystematic variations of pH in the river and coastal sea water mixing zone. The short-term variation of pH in the system was dominated by the carbonate equilibrium. As results of pH, the estuary did not show any consistent transition between riverine and seawater mixing. The Ec and TDS results showed sharp increase due to mixing of river water with seawater. The increasing trend of Ec and TDS was consistent with previous observation in Changjiang estuary by Wang and Liu (2003) and also by Ramanathan et al. (1993) in Cauvery estuary, India. The TSM varied from 0.5 to 518.0 (mg/L) in the entire study stations. The maximum concentration of TSM observed in estuary region (212 mg/L). However, the concentrations of TSM decreased as salinity increases.

Cation and anion

The bicarbonate concentration increased with increase in salinity, due to river dilution with seawater (Table 1). The fluctuation in the estuarine regions may be due to the consumption processes. Sulfate (SO_4) showed conservative mixing with seawater. The higher concentration of SO_4 in estuary and coastal sea could be due to the effect of remineralised organic matters (Schijf 2007) and also the consumption by local vegetation in the river area (Ramanathan et al. 1993). Whereas, the SO_4 removal process compensated in the estuary by the contributions of the organic matters (Ramanathan et al. 1993).

All the major cations represented wide variations with respect to increase in salinity. Such types of behavior of the major elements are very similar to the observation in the Indian Subcontinent rivers (Ray et al. 1984; Subramanian and Jha 1988; Ramanathan et al. 1993; Sharma et al. 2007). Sodium (Na) was lower in concentrations because of its preferential absorption and incorporation onto silicates (SiO_2). Calcium (Ca), magnesium (Mg) and Na showed their concentrations closer to seawater dilution line in river region, indicating the fluvial contributions. The dissolved strontium (Sr) followed a mixing line by two end-member concentration salinity plots. Few scatters and several points fall above the mixing line in the estuary (5-salinity), but the rest of the samples showed for conservative nature. In the plots of $^{87}Sr/^{86}Sr$ isotope versus inverse Sr concentrations, an acceptable linear correlation exhibited for river and seawater mixing. The river end member has high $^{87}Sr/^{86}Sr$ values and coastal sea has lower $^{87}Sr/^{86}Sr$ values ($r^2 = 0.87$). Though water samples have taken in the estuarine environment, it could be influenced by the diurnal mixing actions for short time period (Fig. 2).

Elemental concentrations normalizing to seawater

To avoid the dilution error and to visualize the data properly, all the cations (including Sr) were normalized to Na, and all anions were normalized to chlorine (Cl). Schijf and Byrne (2007) reported that about 2–3 % uncertainty in the Na and Cl concentrations was caused due to analytical precision and represented actual salinity fluctuations due to annual changes in coastal waters or a minor, but variable degree of mixing between the spring and sewage effluents

Table 1 Major and minor elements concentration (mg/l) and the $^{87}Sr/^{86}Sr$ ratios in the dissolved loads for water samples form Changjiang river estuary during 2005

Station	Salinity (psu)	Na (mg/l)	K (mg/l)	Ca (mg/l)	Mg (mg/l)	Sr (mg/l)	Cl (mg/l)	SO_4 (mg/l)	HCO_3 (mM)	pH	EC (ms/cm)	TSM (mg/l)	TDS (mg/l)	$^{87}Sr/^{86}Sr \pm 2$ s
Changjiang river (downstream area)														
1	0.00	6.35	2.06	26.2	6.14	0.15	4.42	13.2	1.51	7.5	0.22	59.6	107	0.710542 ± 0.000012
2	0.00	6.08	2.05	26.2	6.05	0.15	4.57	13.3	1.51	7.5	0.22	85.2	107	0.710577 ± 0.000012
3	0.00	6.91	2.07	26.3	6.20	0.15	5.43	13.6	1.51	7.4	0.22	57.4	109	0.710578 ± 0.000010
4	0.00	6.31	2.10	26.8	6.62	0.16	4.70	15.1	1.52	7	0.23	13.4	112	0.710495 ± 0.000013
5	0.00	6.88	2.31	26.7	6.61	0.16	4.77	15.3	1.58	7.5	0.23	17.6	112	0.710938 ± 0.000010
6	0.00	6.34	2.08	26.6	6.64	0.16	4.90	15.7	1.57	7.3	0.23	38.1	112	0.710599 ± 0.000014
7	0.00	63.6	11.4	33.1	12.4	0.23	76.8	76.3	2.00	7.2	0.77	120	376	Not analyzed
8	0.00	6.95	2.14	26.9	6.68	0.16	5.48	16.0	1.58	7.2	0.23	113	110	0.710506 ± 0.000015
9	0.00	28.9	2.07	26.2	9.29	0.17	40.7	20.3	1.56	7.5	0.23	49.6	111	0.710542 ± 0.000012
10-0	0.00	3.81	3.42	16.5	3.81	0.08	6.48	16.2	1.51	7.6	0.23	81.8	110	0.710545 ± 0.000012
10-7	0.00	7.27	1.37	26.4	7.26	0.16	5.54	16.2	1.52	7.7	0.23	284	111	0.710559 ± 0.000014
11	0.10	143	36.5	14.0	14.4	5.32	11.0	17.3	1.56	7.2	0.26	88.4	127	0.708655 ± 0.000016
Changjiang river estuary														
12	1.30	382	47.0	27.0	33.1	7.75	529	72.4	1.58	7.7	2.53	160	1,266	0.709661 ± 0.000013
13	4.40	1,446	87.9	67.1	173	1.43	2,149	242	1.63	8.2	8.00	173	4,260	0.709337 ± 0.000013
14	7.00	2,451	124	103	300	2.04	3,654	415	1.68	8.3	12.3	80.6	6,810	0.70927 ± 0.000013
15	9.60	3,149	161	136	410	2.58	5,013	573	1.66	8.2	16.3	211	9,230	0.709243 ± 0.000011
16-0	13.6	4,344	221	187	583	3.37	7,173	831	1.79	8.5	22.5	130	1,307	0.709178 ± 0.000011
16-7	5.95	4,220	221	188	582	3.39	7,273	851	1.79	8.3	22.8	518	1,324	0.709164 ± 0.000012
Coastal sea														
17	19.1	5,755	306	253	790	4.43	1,000	1,187	2.05	8.2	30.8	7.70	1,848	0.709186 ± 0.000013
18	17.1	5,223	278	239	728	4.17	9,564	1,133	2.07	8.3	27.8	12.4	1,646	0.709168 ± 0.000011
19-0	21.7	6,321	357	288	904	4.97	1,182	1,414	1.89	8.2	34.6	13.8	2,090	0.709164 ± 0.000013
19-6	25.2	6,974	410	363	1030	5.64	1,407	1,715	2.19	8.1	39.6	18.5	2,420	0.709121 ± 0.000012
19-16	29.5	7,964	483	424	1190	6.59	1,640	2,014	2.21	8.1	45.5	53.1	2,830	0.708619 ± 0.000084
20	19.3	5,792	311	273	810	4.69	1,094	1,305	2.07	8.2	31.1	7.40	1,859	0.709133 ± 0.000015
21-0	20.8	5,983	331	275	849	4.75	1,117	1,345	2.19	8.6	33.2	1.00	1,998	0.709158 ± 0.000014
21-10	27.8	7,584	447	392	1119	6.12	1,519	1,854	2.24	8.3	43.2	1.50	2,680	0.709141 ± 0.000014
21-20	31.8	8,323	512	440	1244	7.07	1,769	2,161	2.21	8.1	48.7	1.10	3,060	0.708849 ± 0.000080
21-30	31.9	8,458	527	455	1263	7.08	1,777	2,186	2.36	8	48.9	21.4	3,070	0.70914 ± 0.000011
22-0	21.9	6,337	366	309	922	5.12	1,264	1,526	2.14	8.6	34.8	0.30	2,110	0.709157 ± 0.000014
22-10	29.0	8,036	485	418	1181	6.75	1,682	2,629	2.24	8.3	44.8	0.70	2,790	0.709154 ± 0.000012
22-20	31.2	8,264	508	438	1223	6.81	1,788	2,198	2.29	8.1	47.9	1.00	3,000	0.709149 ± 0.000018
22-30	31.6	8,482	530	452	1,276	7.00	1,834	2,252	2.24	8.1	48.4	6.40	3,040	0.709152 ± 0.000013
22-50	32.3	8,439	511	439	1,244	7.05	1,834	2,246	2.17	8.2	49.4	15.9	3,100	0.709144 ± 0.000013
23	23.8	6,850	389	346	984	5.68	1,381	1,661	2.19	8.7	37.6	0.50	2,290	0.709177 ± 0.000012

Underlined values corresponds to Huangpu River

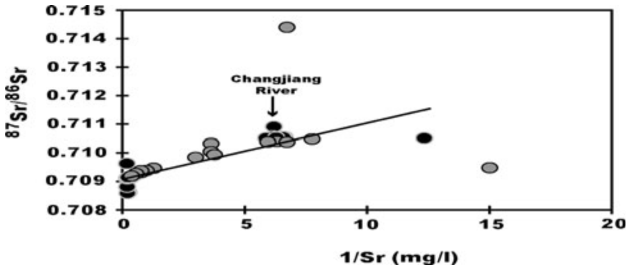

Fig. 2 $^{87}Sr/^{86}$ Sr versus 1/Sr (mg/l) isotopic mixing ratios diagram with sea water for the dissolved load in the Changjiang river estuary. The *gray solid circles* are the open data study samples adopted from Wang et al. (2001) and Wang and Liu (2003)

and ambient seawater, caused by tidal actions. Cation and anion are two separate groups, so errors arising from sample dilutions and instrument instability are always highly correlated within the each group.

Elemental behavior during mixing

The highly linear correlations of Na and Cl ions with density (Fig. 3f) indicated that Na and Cl behaved conservatively with respect to mixing with coastal sea water. It followed that a linear mixing plots of some other elements versus Na or Cl signify conservative behavior of that

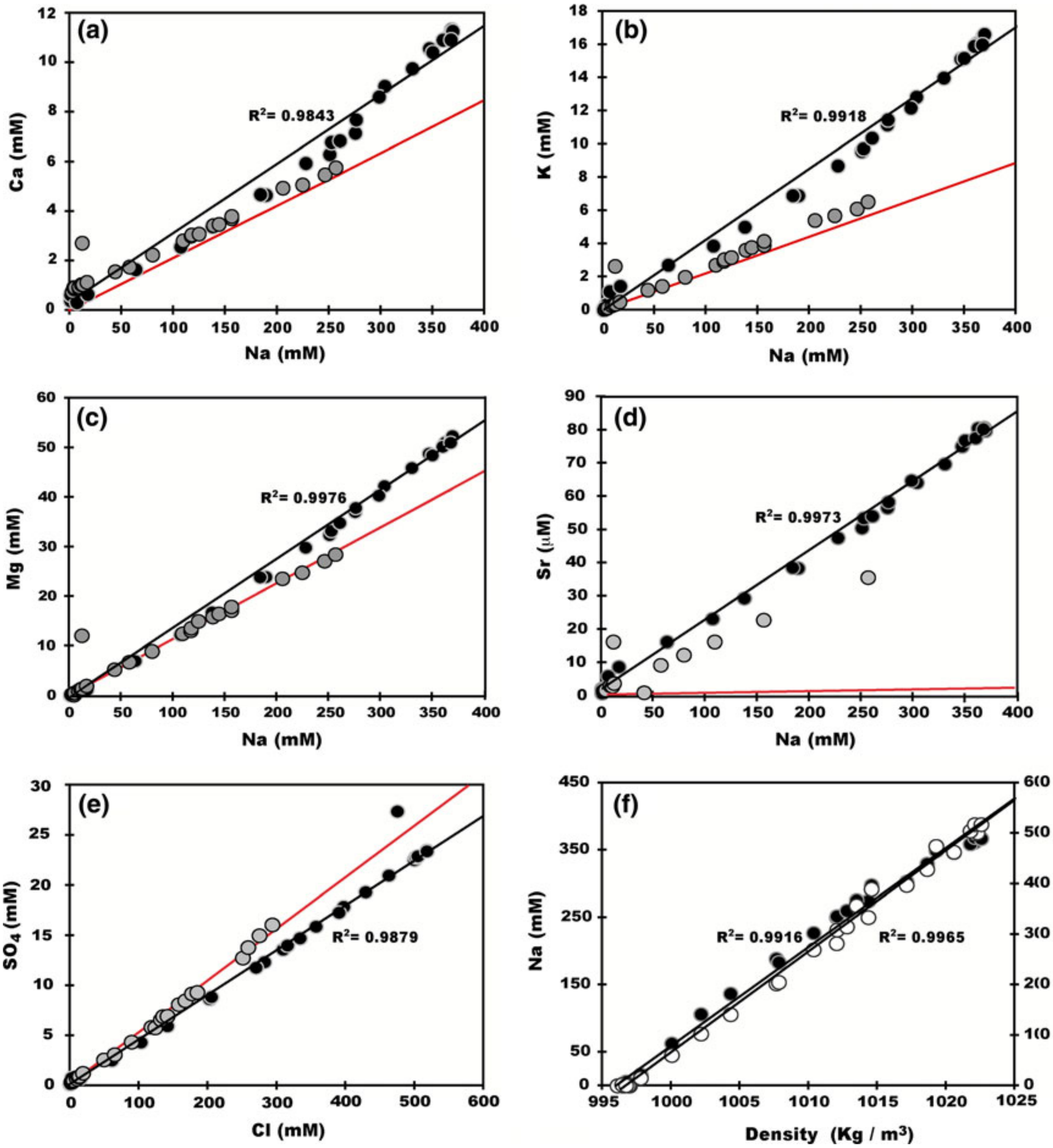

Fig. 3 Mixing plots **a** Na–Ca, **b** Na–K, **c** Na–Mg, **d** Na–Sr, **e** SO_4–Cl, **f** Density versus Na–Cl for the major and minor elements. The *solid straight* is the mixing line and the *red straight line* is the seawater dilution line. The *black solid circles* are our study samples and the *gray solid circles* are the open data study samples referred from Wang et al. (2001) and Wang and Liu (2003)

elements, whereas positive or negative deviations from linearity must be caused due to the enrichment or release at the corresponding salinity. The mixing plots of Na–Ca, Na–K, Na–Mg, Na–Sr and SO_4–Cl indicated that the major cations were linearly mixing only in higher salinity, but overall represented conservative behavior (Fig. 3a–e). It also observed that all major and minor elements deviated from the seawater water slope line in the intermediate salinity region. The present study observation was quite similar with the observation made by Wang and Liu (2003). However, shift could be attributed to pollution or Suess Effect and the systematic differences of elements occurred could be due to water-mass residence time before entering to coastal sea.

It is noteworthy that the substantial removal of Ca, Sr and K did not affect Mg concentrations in higher salinity, may likely to be removed of those elements due to distinct process effects. To understand the process, all the samples were normalized with standard seawater. Table 2 represents the average ionic ratio concentrations with respect to regions corresponding to standard seawater. These values were smaller or larger than one-indication depletion or enrichment of the numerator ion with respect to sea water, respectively. It showed that the river having higher values, 3–4 times more compared to estuary and coastal sea region. Comparing between estuary and coastal sea, it clearly prominent that coastal sea values were closer to standard seawater, but the values in river and estuary were higher than that of standard seawater. They showed linearity with other elements and also reported significant conservative behavior, whereas positive or negative deviations must be caused due to the enrichment or removal process at the corresponding salinity. Hunter (1983) proposed that, as the river water mix with seawater, the negative charge on suspended colloids is not reversed by adsorption of sea water cations, but remains uniformly negative throughout the entire salinity range, decreasing sharply in magnitude during the first few per mill salinity. This confirms that the adsorption process for positive ion particles on to suspended matters occurring continuously at lower salinity regions and the reaction may significantly pronounce in the estuary. And the suspended sediment concentration may reduce from estuary to sea through the principles of diffusive transport process. The mineral solubility had calculated for the three regions with respect to some common elements in the system namely, aragonite and calcite (with or without Mg). Saturation state of dolomite was also determined. The solubility product data have taken from the literatures. Nevertheless, both river and estuary have oversaturated, ascertain that removal processes are the major dominant process (Tables 3, 4).

Table 2 Major anion and cation ratios in Changjiang river, estuary and coastal seawaters with respect to ambient sea water (SW)

Zone	River	Estuary	Coastal sea
Without normalized elemental ratio compositions (mM)			
K/Na	0.189	0.0381	0.0343
Ca/Na	1.816	0.0278	0.0287
Mg/Na	0.748	0.1155	0.1377
Sr/Na	0.0057	0.00107	0.00021
SO_4/Cl	0.9059	0.0437	0.0455
Standard sea water normalized elemental ratio compositions			
K/Na (SW: 0. 0218)	8.69	1.75	1.57
Ca/Na (SW: 0. 0219)	82.96	1.27	1.31
Mg/Na (SW: 0. 1126)	6.65	1.026	1.23
Sr/Na (SW: 0. 00091)	31.0	6.00	1.10
SO_4/Cl (SW: 0. 0517)	17.52	0.84	0.881

Table 3 Correlation matrix of major and minor elements with total suspended matter (TSM)

	River	Estuary	Coastal sea
TSM versus Cl	0.09	0.49	0.14
TSM versus SO_4	0.17	0.50	0.07
TSM versus Na	0.06	0.43	0.16
TSM versus Mg	0.13	0.47	0.18
TSM versus Sr	0.04	0.47	0.15
TSM versus Ca	0.04	0.48	0.19
TSM versus K	0.04	0.48	0.17

Table 4 Saturation state of Changjiang river, estuary and coastal sea with respect to various form of calcium carbonate

Mineral	Ω		
	River	Estuary	Coastal sea
Calcite	0.34	0.52	1.32
Aragonite	0.16	0.28	1.12
Dolomite	−0.84	−0.85	−0.83

Saturation state Ω is the solubility product, where values larger and smaller than 1 represent super saturation and under saturation with respect to the corresponding mineral, respectively

Zone near or above saturation are underlined for emphasis

Sr isotopes along the salinity gradients

Past studies in estuary on Sr behavior revealed two component conservative mixing between sea and river water (Andersson et al. 1992; Ingram and Sloan 1992; Brunskill et al. 2003; Sharma et al. 2007). Recent data on groundwater mixing with seawater in a subterranean estuary indicated that Sr can behave non-conservatively under certain redox conditions (Charette and Sholkovitz 2006). That Sr isotope along salinity gradients was not affected by precipitation and desorption from estuarine sediments and thus behaved conservatively observed for the San Francisco Bay (Ingram and Sloan 1992; Ingram and DePaolo, 1993) and for the Baltic Sea (Andersson et al. 1992). On the contrary, in the present study, the Sr in Changjiang river with coastal sea is generally represented in a conservative manner (Fig. 3) and the minimal deviation is observed from the pure conservative line. These deviations may be accepted due to biological or microbial process in the estuary. Xu and Marcantonio (2007) reported that the Sr has three sources in the Mississippi River mixing zone. Apart from river and sea, the third source could be from (1) submarine ground water discharge, (2) radiogenic particulate Sr that is dissolved and released into the mixing zone and (3) may be the greater concentration of radiogenic colloidal organic and/or inorganic particles. The Sr suspended load for Changjiang river estuary has about 0.015-fold higher than the dissolved load in river zone reported by Wang et al. (2007). The major contributions of silicate and carbonate particles in suspended particulate matters are preferentially from dissolution of carbonate basin rock weathering. The estuary-mixing zone has the highest concentration of suspended particulate matter than river zone, which comes under (2) hypothesis reported by Xu and Marcantonio (2007). This signifies that the role of Sr in dissolved as well as in particulate forms could cause minor deviation from the conservative nature at the mixing zone by its adsorption–desorption mechanism.

Conclusion

Na, K, Ca, Mg, Sr, Cl, and SO_4 concentrations in the estuarine water obtained along salinity gradients in the Changjiang river display a general two component mixing between river and seawater. The minor deviation from theoretical mixing line especially in the estuary acted a minimal non-conservative behavior. Normalized values confirmed that river contributes higher amount of all ions to coastal sea, but it substantially reduced at the estuary. This behavior may arise by suspended particles interaction and active phase change in the quantity of both organic and inorganic components. Information on such process from the river region is highly essential to predict for other zone. The Sr isotope mixing patterns revealed for minimal of non-conservative and deviations of all the elements do not compromise the uses of Sr isotopic process.

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