



## Chemical weathering of monsoonal eastern China: implications from major elements of topsoil



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### ABSTRACT

Major element compositions of 36 bulk samples and 41 clay samples, which were obtained from 47 topsoils collected in monsoonal eastern China, were investigated with conventional wet chemistry and X-ray fluorescence (XRF) spectrometry, respectively. Based on major element analyses, the mobility of major elements and latitudinal distributions of  $\text{SiO}_2/\text{Al}_2\text{O}_3$  ratio, chemical index of alteration (CIA), chemical index of weathering (CIW) and weathering index of Parker (WIP) were analyzed. Meanwhile, the suitability of these chemical weathering indices to topsoils in monsoonal eastern China and its controls were discussed.

These investigations indicate that Na, K, Ca, Mg, and Si are relatively depleted, while Mn, P, Fe and Ti are relatively enriched in topsoils of the study area by comparison with their contents in the upper continental crust (UCC), and that alkali metal (Na, K) and alkaline earth metal (Ca, Mg) elements are generally easier to be depleted from their parent materials than other major elements during chemical weathering. The latitudinal distributions of CIA, CIW and WIP show that they are suitable to both bulk and clay samples, but  $\text{SiO}_2/\text{Al}_2\text{O}_3$  is only suitable to clay samples, not suitable in bulk ones. All these investigations indicate a significant dependence of grain-size in major element abundance and latitudinal distributions of  $\text{SiO}_2/\text{Al}_2\text{O}_3$ , CIA, CIW and WIP, but parent rock type has little effect on them, except its impact on the latitudinal distribution of WIP in clay samples. The significant grain-size dependence probably indicates the presence of unaltered minerals in bulk samples, thus we suggest that clay samples are more suitable to investigating chemical weathering of sediments on continents than bulk samples. The trivial effect of parent rock type probably indicates a relatively uniform chemical weathering on various parent rocks. Correlation analyses indicate that climate is the dominant control of chemical weathering of topsoils in the study area, and the significant latitude effect indicated by the spatial distributions of chemical weathering indices actually reflect the climate control on chemical weathering of topsoils.

Chemical weathering indices actually reflect the integrated weathering history in the study area. Besides the dominant control of climate, other factors like tectonics, parent rock, biology, landform and soil depth and age might also have some effect on the chemical weathering of topsoils in the study area, which needs further research.

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### 1. Introduction

Chemical weathering is an important surface process of the earth and related to many environmental changes caused by interactions and feedbacks among atmosphere, lithosphere, hydrosphere and biosphere. It has long been a research focus in geosciences because of its significance in the earth surface evolution (Nesbitt and Young,

1982; Allen et al., 2001; Chen et al., 2001; Minasny and McBratney, 2001; Dixon et al., 2009), global carbon cycle (Berner et al., 1983; Volk, 1987; Raymo et al., 1988; Brady, 1991; Berner, 1992, 1995; Amiotte Suchet and Probst, 1993; Kump et al., 2000), pedogenesis (Jackson and Sherman, 1953; Sverdrup and Warfvinge, 1988; Minasny and McBratney, 2001; Yoo et al., 2007), and civil engineering (Dearman et al., 1978; Steward and Cripps, 1983; Fookes et al., 1988; Lan et al., 2003).

Persistent investigations on the controls of chemical weathering have contributed a lot to our understanding on the links between chemical weathering and its controlling factors, such as tectonics (geological settings and topographical conditions) (Raymo et al.,

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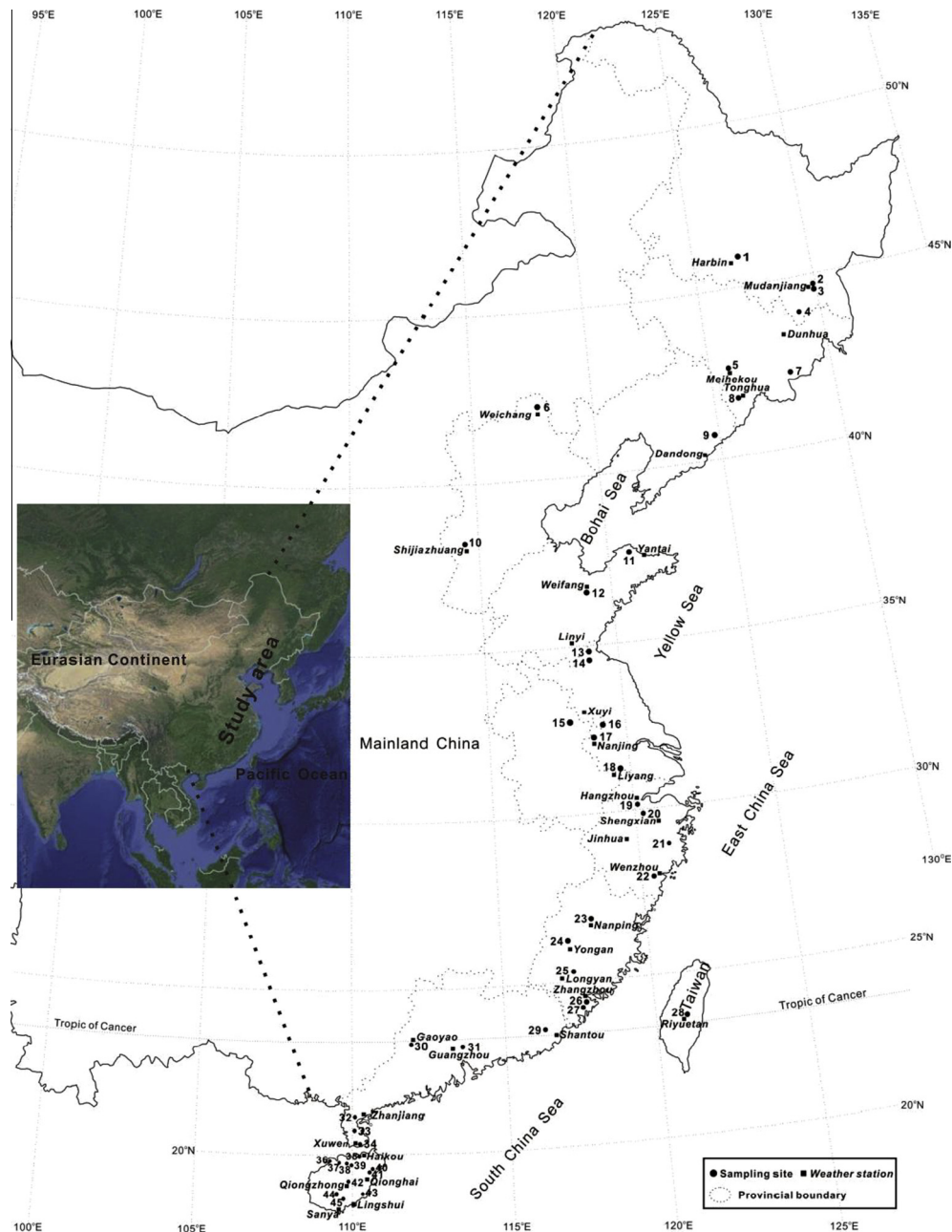
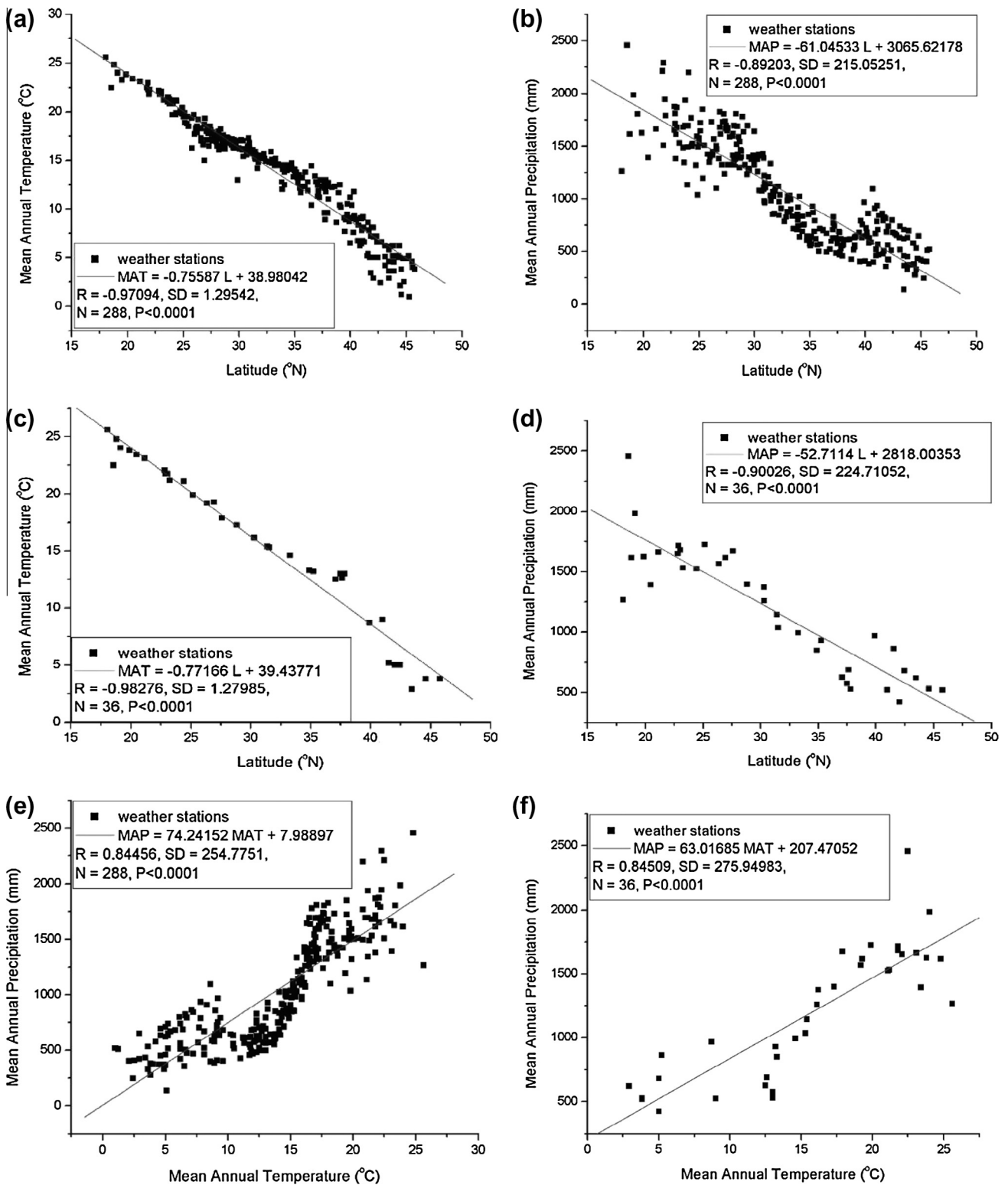


Fig. 1. A sketch map of the study area with sampling sites and weather stations.

1988; Drever and Zobrist, 1992; Raymo and Ruddiman, 1992; Berner and Berner, 1997; Riebe et al., 2001; Jacobson et al., 2003; West et al., 2005; Liu et al., 2007a,b; Moore et al., 2013); provenance (or lithology of source rock) (Sawyer, 1986; Nesbitt and Wilson, 1992; Le Pera et al., 2001; Dessert et al., 2003; Price and Velbel, 2003; Caspari et al., 2006); climate (temperature, precipitation and runoff) (Velbel, 1993; Brady and Carroll, 1994; White and Blum, 1995; Riebe et al., 2001, 2004; Yang et al., 2004; Deepthi and Balakrishnan, 2005; Singh et al., 2005; West et al., 2005; Liu et al., 2007a,b; Gislason et al., 2008; Gabet et al., 2010; Li and Yang, 2010); vegetation (Berner, 1992; Drever, 1994; Gislason et al., 1996); time (Grantham and Velbel, 1988; Taylor and Blum, 1995; Gislason et al., 1996; White and Brantley, 2003); and even human activities (Motuzova and Hong Van, 1999; Chetelat et al., 2008). Although much work has been done, controversies still remain on the controlling mechanisms of chemical weathering, especially

of silicate rocks (Stallard, 1995; Yang et al., 2004; West et al., 2005; Li and Yang, 2010; Willenbring and von Blanckenburg, 2010; Moore et al., 2013). Some conventional views about chemical weathering have even been challenged by new discoveries. For example, the famous "Uplift-Weathering Hypothesis" proposed a tectonic forcing of global cooling in Late Cenozoic, suggesting that the increases in chemical weathering driven by the uplift of the Tibet Plateau during that period may have resulted in the decreases of atmospheric  $\text{CO}_2$ , thus cooling the Late Cenozoic climate (Raymo and Ruddiman, 1992). A recent investigation questioned the tectonic forcing, however, with evidence on the long-term stability of global erosion and chemical weathering rates during the Late-Cenozoic cooling (Willenbring and von Blanckenburg, 2010). A most recent investigation also concluded that silicate weathering in uplifting mountain ranges does not control long-term climate change, based on Ca isotope analyses of silicate and carbonate



**Fig. 2.** Latitudinal distributions of MAT and MAP, and their correlations. Climatic data of (a), (b) and (d) are obtained from all the weather stations lower than 1000 m above seal level in the study area, and climatic data of (c), (d) and (e) were obtained from those closest to the sampling sites. Source: China Meteorological Data Sharing Service System (1961–1990).

weathering from rivers draining active mountain belts (Moore et al., 2013). Moreover, a recent investigation suggested that it is not the atmospheric CO<sub>2</sub> sink by silicate weathering but by carbonate weathering that is responsible for both long-term and short-term climate change (Liu et al., 2011). Another controversy dealt

with the relationship between physical denudation and chemical weathering. It is generally believed that chemical weathering is positively related to physical denudation (Gaillardet et al., 1999; Millot et al., 2002), but a recent case study doubted that by comparison of silicate weathering between the drainage basins of

**Table 1**  
Major element concentrations and LOI of bulk samples (in wt%).<sup>a</sup>

Bulk samples	SiO <sub>2</sub>	TiO <sub>2</sub>	Al <sub>2</sub> O <sub>3</sub>	Fe <sub>2</sub> O <sub>3</sub>	MnO	MgO	CaO	Na <sub>2</sub> O	K <sub>2</sub> O	P <sub>2</sub> O <sub>5</sub>	LOI	Total
HHBB <sub>B</sub>	61.60	0.77	12.89	3.74	0.05	1.15	1.42	2.07	2.63	0.09	13.18	99.59
HMHC <sub>B</sub>	54.26	1.48	15.24	7.28	0.07	2.09	1.45	2.36	1.61	0.11	13.26	99.21
HMWB <sub>B</sub>	50.31	1.24	13.88	6.25	0.14	2.93	3.22	1.73	2.31	0.19	17.31	99.51
HMJB <sub>B</sub>	48.03	1.04	14.02	6.22	0.10	1.92	3.36	1.39	1.21	0.40	22.05	99.74
JHFA <sub>B</sub>	62.24	1.14	13.33	6.08	0.09	2.05	1.42	1.90	2.11	0.17	9.19	99.72
HWYB <sub>B</sub>	62.13	0.65	12.47	3.68	0.05	1.64	2.02	1.72	3.00	0.09	11.96	99.41
HWYA <sub>B</sub>	75.45	0.18	12.20	1.22	0.01	0.74	0.10	2.85	5.16	0.01	1.71	99.63
JATD <sub>B</sub>	62.73	0.68	15.75	5.36	0.08	0.78	1.57	5.31	2.41	0.07	4.78	99.52
JTWA <sub>B</sub>	54.37	0.73	14.69	5.64	0.04	2.75	1.34	0.80	3.05	0.15	16.08	99.64
LKHA <sub>B</sub>	37.52	1.36	13.00	7.63	0.12	3.42	3.26	0.80	1.78	0.33	30.40	99.62
HSJA <sub>B</sub>	49.73	2.14	14.70	9.32	0.06	4.08	4.95	2.25	1.36	0.44	10.29	99.32
HSJC <sub>B</sub>	67.38	0.91	12.29	4.02	0.05	1.51	2.44	0.91	1.55	0.12	8.56	99.74
SPWA <sub>B</sub>	49.24	1.99	13.86	11.03	0.09	3.42	2.92	1.50	1.26	0.37	14.14	99.82
SLSA <sub>B</sub>	51.19	1.64	13.63	8.96	0.12	3.80	3.29	2.84	2.04	0.24	11.81	99.56
JDAAB <sub>B</sub>	51.72	1.79	14.80	11.52	0.09	2.15	2.10	1.33	1.25	0.37	12.62	99.74
ASNA <sub>B</sub>	45.99	1.94	15.58	12.69	0.13	6.55	4.98	0.87	0.43	0.68	9.53	99.37
JXYA <sub>B</sub>	47.88	2.36	13.24	11.88	0.24	2.46	2.57	1.00	1.22	0.27	16.11	99.23
JZWA <sub>B</sub>	59.30	2.09	14.09	8.72	0.13	1.71	0.07	0.53	1.37	0.07	11.48	99.56
ZHLB <sub>B</sub>	71.62	0.83	12.04	4.38	0.02	0.85	0.01	0.30	1.58	0.11	7.91	99.65
ZSPA <sub>B</sub>	34.95	3.90	14.05	21.00	0.25	6.53	3.74	0.38	0.42	0.85	13.25	99.32
ZLSA <sub>B</sub>	65.28	0.73	14.75	4.04	0.11	0.92	0.17	1.36	3.02	0.06	9.14	99.58
ZWWA <sub>B</sub>	55.68	1.23	18.36	5.87	0.01	1.19	0.01	0.25	1.91	0.12	14.98	99.61
FNSA <sub>B</sub>	72.93	0.76	10.98	2.51	0.01	1.01	0.21	0.19	1.24	0.06	9.87	99.77
FMSB <sub>B</sub>	34.96	3.01	20.27	18.08	0.11	1.49	0.12	0.19	0.46	0.77	20.18	99.64
FZC <sub>B</sub>	70.15	0.50	14.30	3.83	0.00	0.75	0.21	0.26	1.66	0.06	7.98	99.70
FLZA <sub>B</sub>	42.76	2.12	17.05	13.97	0.17	1.77	0.90	0.50	0.61	0.14	19.68	99.67
FLBB <sub>B</sub>	39.87	2.73	15.07	13.69	0.14	3.73	1.68	0.66	1.15	0.23	20.52	99.47
T <sub>B</sub>	78.66	0.71	8.37	3.86	0.05	0.47	0.16	0.36	1.51	0.07	5.46	99.68
GJHA <sub>B</sub>	60.19	0.69	17.44	5.17	0.03	1.20	0.12	0.51	3.04	0.08	11.21	99.68
GGZA <sub>B</sub>	70.29	0.39	15.08	1.88	0.01	1.20	0.12	0.16	0.71	0.08	9.61	99.53
GD <sub>B</sub>	56.50	2.06	14.69	12.73	0.14	1.11	0.17	0.32	0.28	0.13	11.33	99.46
HJJA <sub>B</sub>	50.73	2.30	18.85	13.61	0.04	1.18	0.12	0.13	0.21	0.10	12.26	99.53
HLGA <sub>B</sub>	34.38	4.49	17.58	21.58	0.30	1.90	0.83	0.68	0.62	0.25	17.21	99.82
HXYB <sub>B</sub>	52.51	2.52	13.88	11.39	0.24	2.51	1.87	0.84	0.79	0.28	12.94	99.77
HHLB <sub>B</sub>	68.06	1.45	9.15	7.74	0.13	1.95	0.45	0.38	1.11	0.17	9.03	99.62
HWN <sub>B</sub>	29.50	4.40	22.52	21.89	0.11	0.93	0.00	0.73	0.06	0.19	19.23	99.56

<sup>a</sup> LOI = loss on ignition, the subscript letter B in the table means bulk sample.

Changjiang and Huanghe, and suggested that strong physical denudation does not always result in intensive chemical weathering in drainage basins (Yang et al., 2004).

One of the major problems in chemical weathering investigation is the quantitative evaluation of chemical weathering intensity. To solve this problem, various solutions and chemical indices have been established and applied, most of which are based on major element analyses, such as SiO<sub>2</sub>/Al<sub>2</sub>O<sub>3</sub> ratio (Ruxton, 1968), WIP (weathering index of Parker) (Parker, 1970), CIA (chemical index of alteration) (Nesbitt and Young, 1982), CIW (chemical index of weathering) (Harnois, 1988), and ternary diagrams of chemical elements (Nesbitt and Young, 1989; Ohta and Arai, 2007). Good results have been obtained in their applications to the geochemistry of weathering profiles (Nesbitt and Young, 1989; Nesbitt and Wilson, 1992; Ng et al., 2001; Duzgoren-Aydin and Aydin, 2003; Price and Velbel, 2003; Deepthy and Balakrishnan, 2005; Singh et al., 2005; Caspari et al., 2006; Ohta and Arai, 2007; Shao et al., 2012) and river chemistry of dissolved loads (Liu et al., 2004, 2007a,b, 2009; Li and Yang, 2010; Shao et al., 2012). In fact, till present, most of our chemical weathering information is obtained from the geochemistry of weathering profiles and river chemistry of dissolved loads. Besides river sediments (Liu et al., 2004; Yang et al., 2004; Li and Yang, 2010; Shao et al., 2012) and weathering profiles of bedrock (Lan et al., 2003; Li et al., 2007a,b; Ma et al., 2007), eolian sediments have been another source of chemical weathering information in China (Gallet et al., 1998; Jun et al., 1998; Chen et al., 2001; Ding et al., 2001; Yang et al., 2006; Li et al., 2008; Hong et al., 2010; Xiong et al., 2010), particularly the loess–paleosol sequence from Chinese Loess

Plateau (Gallet et al., 1998; Chen et al., 1998; Chen et al., 2001; Yang et al., 2006). Unfortunately, the suitability of chemical weathering indices to topsoils developed on different rock types and spanning various climatic zones is rarely discussed.

Theoretically, the development of topsoils by in situ weathering of earthy and rocky materials on direct exposure to atmospheric agents in positive landforms without being altered by transportation, differentiation and deposition will probably result in a better preservation of chemical weathering information. Thus, here we present a chemical weathering investigation of topsoils from monsoonal eastern China with different chemical indices based on major elements analyses.

## 2. Study area, sampling and experiments

### 2.1. Study area

45 sampling sites, spanning 13 provinces along the east part of China (ca. 109°E–130°E, 18°N–46°N), were carefully selected to avoid contamination by alien sediments or human disturbance (Fig. 1 and supplementary Table 1). The study area is located on the eastern margin of the Eurasian continent, and western coast of the Pacific Ocean, topographically characterized by plains and hills lower than 1000 m above sea level (a.s.l.) on the average. The tectonic framework of the study area is primarily composed of the North China Craton, South China Block and Central China Orogenic belt, with very intense tectonic activities in Mesozoic, resulting in strong topographic contrasts and extensive crustal



**Table 2**  
Major element concentrations and LOI of clay samples (in wt%).<sup>a</sup>

Clay samples	Al <sub>2</sub> O <sub>3</sub>	CaO	Fe <sub>2</sub> O <sub>3</sub>	K <sub>2</sub> O	MgO	MnO	Na <sub>2</sub> O	P <sub>2</sub> O <sub>5</sub>	SiO <sub>2</sub>	TiO <sub>2</sub>	LOI	Total
HHBB <sub>C</sub>	18.02	1.15	7.94	2.78	2.29	0.17	0.59	0.26	45.96	0.91	20.27	100.33
HMHC <sub>C</sub>	16.55	2.44	11.92	1.62	2.58	0.18	0.37	0.27	43.17	1.24	19.82	100.18
HMWBC <sub>C</sub>	18.74	1.93	9.85	2.88	2.38	0.26	0.54	0.39	42.70	1.26	18.87	99.81
HMJB <sub>C</sub>	19.54	1.51	11.01	1.57	2.10	0.22	0.38	0.87	35.92	1.31	25.69	100.12
JHFA <sub>C</sub>	19.90	0.96	9.10	2.45	2.47	0.27	0.67	0.40	46.88	1.18	15.95	100.24
HWYB <sub>C</sub>	18.02	1.83	8.08	3.23	2.61	0.30	0.47	0.28	43.10	0.69	21.44	100.06
LKHA <sub>C</sub>	15.62	2.33	7.27	1.37	1.89	0.13	0.80	0.66	31.82	1.06	36.96	99.91
HSJC <sub>C</sub>	24.66	1.58	7.02	2.92	1.94	0.16	0.23	0.17	45.45	0.69	15.26	100.08
SPWA <sub>C</sub>	23.54	1.63	10.96	1.09	1.67	0.14	0.34	0.45	38.96	1.27	20.24	100.29
SLSA <sub>C</sub>	20.50	1.29	9.59	2.19	2.94	0.21	0.34	0.29	40.93	0.72	20.88	99.88
JDPA <sub>C</sub>	22.15	1.20	13.53	1.49	2.05	0.14	0.26	0.38	41.93	1.64	15.25	100.01
JDAAC <sub>C</sub>	22.86	1.36	13.03	1.29	2.29	0.28	0.17	0.83	40.58	1.48	16.09	100.27
ASNA <sub>C</sub>	23.84	2.42	9.47	1.17	1.99	0.21	0.37	1.16	41.85	1.26	16.73	100.46
JXYA <sub>C</sub>	17.44	1.45	13.86	1.18	2.21	0.13	0.45	0.30	41.71	1.85	19.41	99.99
JBTA <sub>C</sub>	19.11	1.28	14.24	0.78	3.57	0.33	0.36	0.31	39.30	0.76	20.22	100.27
JZWA <sub>C</sub>	24.96	0.43	13.26	1.89	1.70	0.11	0.14	0.11	44.31	1.15	11.72	99.77
ZHLB <sub>C</sub>	23.57	0.09	7.06	2.74	1.15	0.02	0.23	0.21	49.79	0.94	14.14	99.94
ZSPA <sub>C</sub>	23.81	1.06	13.26	0.44	1.19	0.24	0.17	0.55	40.63	2.73	16.18	100.25
ZLSA <sub>C</sub>	22.69	0.11	6.63	3.82	1.32	0.15	0.67	0.16	45.85	0.58	17.72	99.71
ZWWA <sub>C</sub>	28.12	0.04	8.18	1.70	0.74	0.02	0.13	0.18	42.49	0.54	17.48	99.63
FNSA <sub>C</sub>	24.51	0.04	4.00	2.49	0.99	0.03	0.00	0.07	53.33	0.78	13.42	99.65
FZC <sub>C</sub>	31.55	0.06	7.69	3.82	0.84	0.01	0.15	0.05	44.82	0.52	11.01	100.52
FLZA <sub>C</sub>	20.64	0.62	17.87	0.30	1.24	0.22	0.20	0.20	37.49	1.76	19.44	99.96
FLBB <sub>C</sub>	19.46	0.85	13.89	0.80	3.04	0.19	0.29	0.27	39.39	1.77	20.31	100.26
GJHA <sub>C</sub>	28.99	0.07	10.65	1.53	0.99	0.06	0.00	0.09	40.71	1.11	15.95	100.15
7-59 <sub>C</sub>	33.79	0.03	8.70	0.85	0.43	0.02	0.00	0.10	37.31	0.85	17.61	99.69
GGZA <sub>C</sub>	35.96	0.10	3.81	0.62	0.36	0.04	0.00	0.06	40.64	0.30	17.77	99.65
3-49 <sub>C</sub>	27.67	0.22	18.14	0.29	0.38	0.14	0.00	0.28	32.14	2.70	17.63	99.59
GD <sub>C</sub>	27.08	0.33	18.70	0.52	0.56	0.25	0.00	0.25	35.79	2.98	13.15	99.61
4-24 <sub>C</sub>	29.99	0.13	16.41	0.09	0.28	0.05	0.00	0.10	35.92	1.83	15.10	99.90
HHJA <sub>C</sub>	29.39	0.11	19.35	0.29	0.39	0.08	0.00	0.13	29.84	2.67	17.34	99.58
HLGA <sub>C</sub>	20.83	0.60	19.62	0.51	0.93	0.28	0.32	0.41	34.27	3.33	18.66	99.76
HHLA <sub>C</sub>	21.98	0.44	17.76	1.12	1.20	0.31	0.11	0.37	37.24	3.52	16.26	100.32
HXYB <sub>C</sub>	24.56	0.58	18.01	0.79	0.70	0.45	0.31	0.56	31.39	3.18	19.38	99.92
HHLB <sub>C</sub>	20.39	0.73	17.77	1.14	1.36	0.40	0.00	0.37	38.60	3.54	15.79	100.10
HWN <sub>C</sub>	28.62	0.08	19.34	0.08	0.31	0.10	0.00	0.24	30.74	2.30	17.88	99.69
HWPA <sub>C</sub>	28.02	0.05	18.11	0.05	0.24	0.11	0.00	0.24	32.49	1.09	19.54	99.94
HQWA <sub>C</sub>	32.43	0.03	5.99	1.93	0.52	0.02	0.00	0.06	40.68	0.36	17.61	99.63
HWA <sub>C</sub>	31.32	0.09	11.18	0.41	0.35	0.03	0.00	0.19	34.93	1.77	19.87	100.15
HWAA <sub>C</sub>	29.63	0.05	9.91	2.32	0.84	0.04	0.00	0.12	40.04	0.62	16.84	100.41
HBBA <sub>C</sub>	31.04	0.31	6.46	1.74	0.60	0.08	0.00	0.15	40.89	0.61	17.91	99.81

<sup>a</sup> LOI = loss on ignition, the subscript letter C in the table means clay sample.

movement in these regions (Zheng et al., 2013; Zhao and Zhai, 2013; Zhang and Zheng, 2013; Wang et al., 2013). Due to interactions between the largest continent and the largest ocean in the world, and seasonal variation of intertropical convergence zone (ITCZ), the study area is dominated by a monsoon climate of concurrent rainy and hot seasons, with a mean annual temperature (MAT) of 2.9–25.6 °C and a mean annual precipitation (MAP) of 400–2500 mm, both increasing southward and showing a latitude effect (supplementary Table 1 and Fig. 2).

## 2.2. Sampling

A total of 47 topsoil samples were collected from the top 2–4 cm of soils developed on various weathered silicate rocks (mostly basalt and granite) of the sampling sites along the east part of China between September and November in 2004 and 2005. The sampling intensity was one sample a site at 43 sites, and two samples a site at the rest two, but at different altitude and from different parent rock (supplementary Table 1). All the samples were collected in positive landforms, either on the top or gentle slope of a hill lower than 1000 m a.s.l., except HWY1 (1110 m) and HWY2 (1240 m), with identifiable parent rock, natural vegetation and far away from human activities. The geographic coordinate (longitude, latitude and altitude) of each sampling site was given by a handheld GPS device during sampling at the site. The climatic data (MAT and

MAP) of each site were obtained from the meteorological record of the nearest weather station during 1961–1990, which is available in China Meteorological Data Sharing Service System. Details of these samples, including parent rock, major element samples, longitude, latitude, altitude, MAT and MAP, are presented in supplementary Table 1.

## 2.3. Sample preparation and major element analyses

To investigate the major element geochemistry of all samples, 36 bulk samples were selected and 41 clay samples were separated from the 47 topsoils (supplementary Table 1, Tables 1 and 2) according to the following procedures.

First, plant residues, coarse sand and gravels were removed by sieving from the topsoil samples through a 200-mesh sieve before they were air-dried at room temperature. By this way, soil grains smaller than 75 μm were obtained. Then they were sub-sampled into two groups, one is for major element analysis, the other is for clay separation before chemical analysis. Clay grains smaller than 2 μm were separated at Guangdong Institute of Eco-environment and Soil Sciences with sedimentation method according to Stokes' Law. First, 30 g of dry bulk sample was weighed to make a 1:10 liquid suspension. Then the liquid suspension was put into an ultrasonic oscillation tank and disaggregated by stirring and ultrasonic oscillating at a power of 21.5 kHz and 300 mA for 30 min, and then

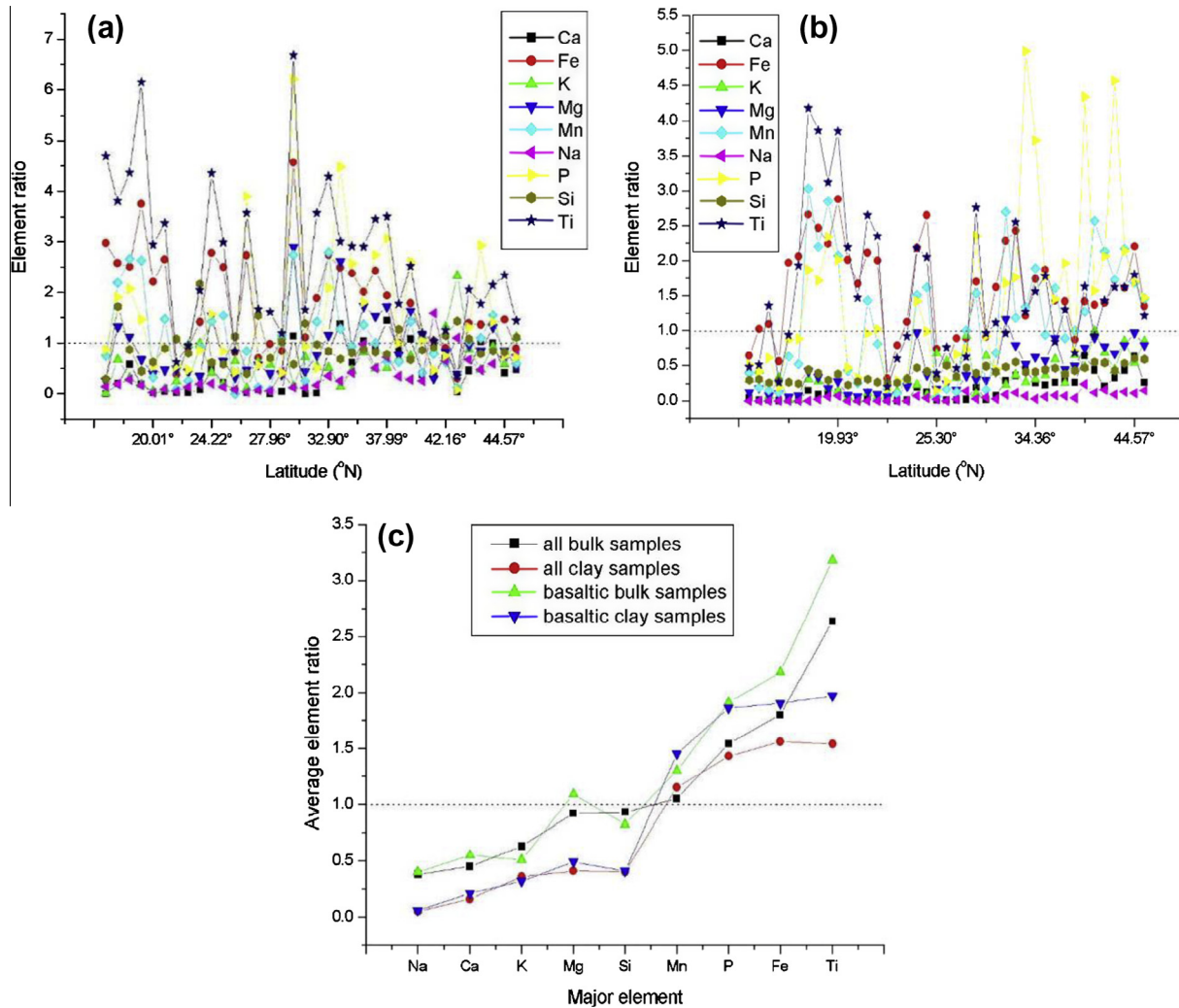


Fig. 3. Major element ratios of all bulk (a) and clay samples (b), and their averages (c) along the latitudinal gradient.

clays were separated by settling and centrifuging from the 1:10 liquid suspension. After the withdrawal time the liquid-suspension in the top 5 cm of the column was siphoned out, and air-dried into slices in an evaporating dish. Conventional chemical reagents, such as HCl, H<sub>2</sub>O<sub>2</sub> or NaO<sub>3</sub>P, were not used during clay separation in order to preserve terrigenous materials from dissolution of labile elements. A total of 36 bulk samples and 41 clay samples (in slices) were obtained from the 47 topsoil samples. These samples were first finely ground in an agate mortar. Then 2 g of powdered samples were precisely weighed with an electronic balance, and then treated with 20% acetic acid for over 12 h at room temperature to remove their carbonate fraction, followed by rinsing with distilled water for three times. After that, the carbonate-free samples were dried at 100 °C in an oven for 3 h. Finally, 0.7–1 g of these powder samples were precisely weighed and ignited in a clean ceramic crucible at 920 °C in a muffle furnace for at least 30 min to remove their organic and water components. By calculating the loss on ignition, the LOI values of these samples were obtained. The chemical compositions of bulk and clay samples were measured with different methods.

Chemical composition of bulk samples was determined by conventional wet chemistry at the Key Laboratory of Marginal Sea Geology in Guangzhou Institute of Geochemistry, Chinese Academy of Sciences. Concentrations of K<sub>2</sub>O, Na<sub>2</sub>O, CaO, MgO, MnO and P<sub>2</sub>O<sub>5</sub> in bulk samples were determined with an atomic absorption spectrophotometer of Shimadzu AA-6300C following the pro-

cedure of atomic absorption spectrometry (AAS) (Welz et al., 2008). The SiO<sub>2</sub> concentration of bulk samples was determined with wet chemical method. First, 0.25–0.5 g powder samples was precisely weighed and placed in a silver crucible. After alkali dissolution, water extraction and acid digestion, they were evaporated, treated by animal glue, and then the condensate was filtered and ignited. Finally, the precipitate of ignition was weighed to calculate the SiO<sub>2</sub> concentration. The Fe<sub>2</sub>O<sub>3</sub> content of bulk samples was measured with EDTA (ethylene diamine tetraacetic acid) volumetric method (Bennett and Reed, 1971). First, 50 ml of the filtrate was drawn after the separation of SiO<sub>2</sub>, and then it was neutralized with ammonia water till the appearance of yellow Fe precipitate. After that, the solution was diluted with distilled water to 100 ml, and then treated by 2–3 ml of 1 M HCL, and the Fe<sub>2</sub>O<sub>3</sub> content was estimated with titration method by titrating the indicator of salicylic acid into the standard EDTA solution. After the determination of Fe<sub>2</sub>O<sub>3</sub> content, excessive standard EDTA solution and buffered solution (pH = 4.5) were added to the filtrate, followed by boiling and cooling, then the Al<sub>2</sub>O<sub>3</sub> and TiO<sub>2</sub> concentrations were determined by titrating the indicators of nitroso R salt and standard CuSO<sub>4</sub> solution into the excessive EDTA solution. The Al<sub>2</sub>O<sub>3</sub> content was then calculated by a subtraction method in the Al<sub>2</sub>O<sub>3</sub> and TiO<sub>2</sub> contents.

Chemical composition of clay samples was determined by X-ray fluorescence (XRF) spectrometry with an XRF spectrometer of Rigaku 100e at the State Key Laboratory of Isotope Chronology and

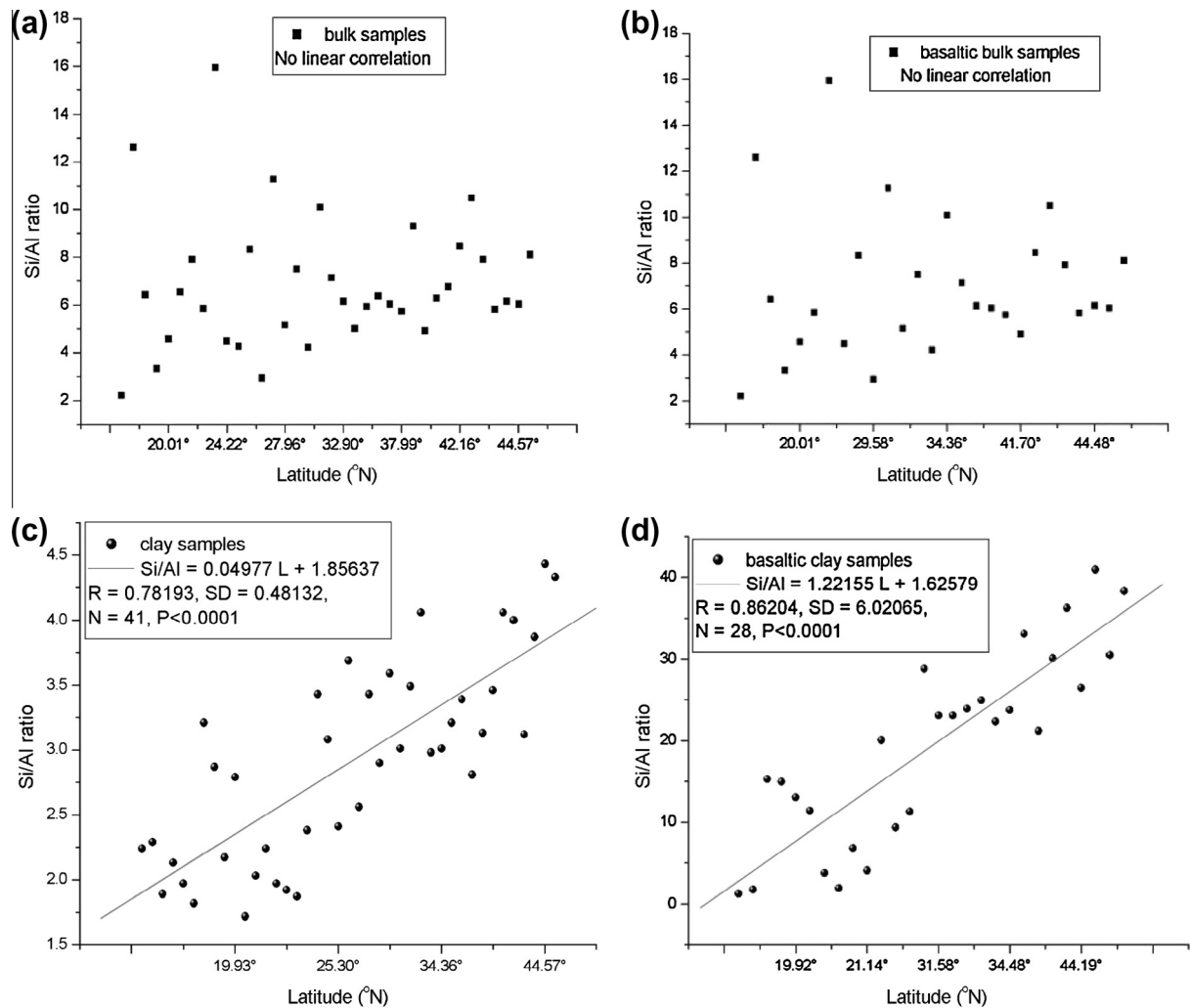


Fig. 4.  $\text{SiO}_2/\text{Al}_2\text{O}_3$  ratios of bulk (a), basaltic bulk (b), clay (c), and basaltic clay (d) samples along the latitudinal gradient.

Geochemistry in Guangzhou Institute of Geochemistry, Chinese Academy of Sciences. After the determination of LOI,  $0.6 \pm 0.02$  g of the carbonate-free samples and 8 times that amount of dry lithium tetraborate ( $\text{Li}_2\text{B}_4\text{O}_7$ ) were weighed accurately, and then mixed carefully in a plastic container. After that, they were put into a platinum crucible with two drops (about 0.1 g) of 2% LiBr and 1%  $\text{NH}_4\text{I}$  added as a co-solvent. The mixture was then fused in the platinum crucibles for 10 min with the fusion being carried out in an automatic fusion system. During the fusion, the crucible was regularly rotated and increasingly heated up to  $1160^\circ\text{C}$  to ensure complete mixing and melting. After cooling at room temperature, glass samples were placed in a sample container for major element analysis.

12 Chinese standard reference samples were used for quality control of the chemical measurements, including GSR-1, GSR-3, GSD-9, GSD-12, GSS-1, GSS-3, GSS-5, GSS-6, GSS-7, GBW07409, GBW07410 and GBW07411. The detection limit is  $\pm 0.01$  wt% and analytical uncertainties are  $\pm 2\%$  for all major oxides of bulk and clay samples. The analytical precision and accuracy of major element were monitored by stand reference materials, which are measured as “unknowns” with samples and sample duplicates. With few exceptions, there were no discrepancies between the analytical data obtained and the consensus data in the reference samples. The concentrations (weight %) of 10 elements traditionally listed as oxides in major element chemical analysis, Al, Ca, Fe, K, Mg, Mn, Na, P, Si and Ti have been determined in all samples,

and their results are presented sequentially from high latitudes to low latitudes in Tables 1 and 2, respectively.

### 3. Results and discussion

#### 3.1. Element mobility

Chemical weathering is a weathering process whereby rocks and minerals are transformed into new, fairly stable chemical combinations by such chemical reactions as hydrolysis, oxidation, ion exchange, and solution (Parker, 2003). During this process, mobile elements, such as K, Na, Ca, Mg and Si will be increasingly removed from parent rocks, while other elements, such as Al, Fe and Ti will be relatively enriched in the weathered materials (Ruxton, 1968; Parker, 1970; Nesbitt et al., 1980). To investigate the mobility of major elements for topsoils from eastern China, we employed the element ratio proposed by Singh et al. (2005) and defined as: element ratio ( $X$ ) =  $\frac{X/\text{Al}_2\text{O}_3(\text{sample})}{X/\text{Al}_2\text{O}_3(\text{UCC})}$ , where  $X$  is the analyzed element and UCC refers to the upper continent crust. The numerator refers to the content ratio of  $X$  oxide and  $\text{Al}_2\text{O}_3$  in the analyzed sample, while the denominator refers to their content ratio in UCC. The data source of UCC in our calculation was cited from Rudnick and Gao (2003). This element ratio refers to the relative enrichment or depletion of the analyzed element, i.e.  $>1$  indicates enrichment,  $<1$  indicates depletion and  $=1$  indicates no change in the

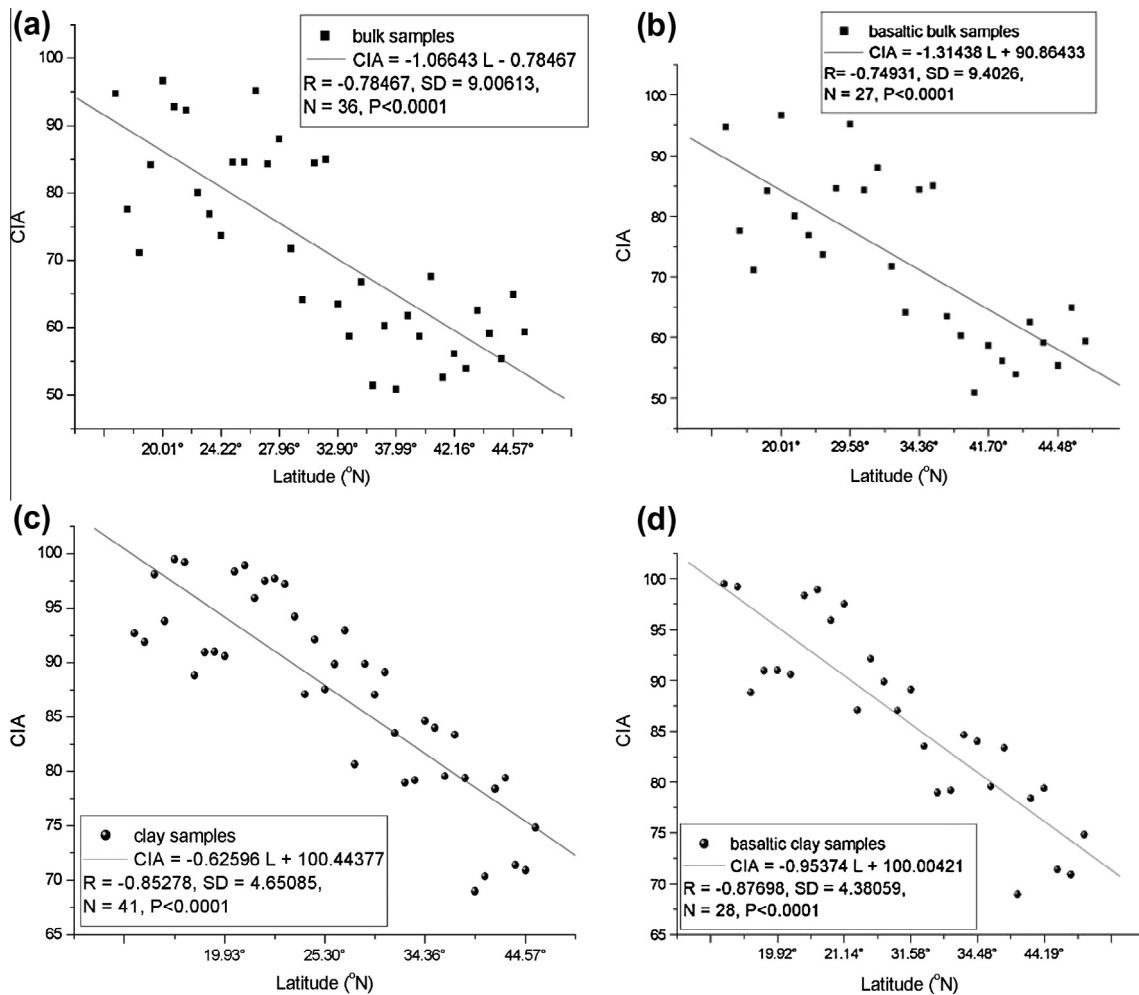


Fig. 5. CIA values of bulk (a), basaltic bulk (b), clay (c), and basaltic clay (d) samples along the latitudinal gradient.

relative abundance of element (Singh et al., 2005). Element ratios of bulk and clay samples were calculated as shown in supplementary Tables 2 and 3, respectively.

As shown in Fig. 3, the element ratios of bulk and clay samples display quite different variations along the latitudinal gradient, but both with relatively low element ratios of Na, Ca, K, Mg and Si, and relatively high contents of Mn, P, Fe and Ti. The average element ratios of Na, K, Ca, Mg, and Si in all samples are below 1, except the Mg content in basaltic bulk samples (1.09), indicating the relative depletion of these elements in topsoils by comparison with their corresponding contents in UCC (Rudnick and Gao, 2003). By contrast, the average element ratios of Mn, P, Fe and Ti in all samples are above 1, indicating the relative enrichment of these elements in topsoils by comparison with their corresponding contents in UCC (Rudnick and Gao, 2003). As shown in Fig. 3c, the average element ratios of bulk samples either developed on heterogeneous parent rocks or developed on homogeneous parent rock (basalt) display similar variations, and so do the average element ratios of clay samples, but the difference in the variations of element ratios between bulk and clay samples is a little bigger, generally with higher values of element ratios in bulk samples than in clay samples, except that of Mn, indicating that the element mobility of topsoils is less dependent on parent rock type than on grain-size. The intensity of relative depletion is:  $\text{Na} > \text{Ca} > \text{K} > \text{Mg} > \text{Si} > \text{Mn} > \text{P} > \text{Fe} > \text{Ti}$  for all bulk samples,  $\text{Na} > \text{Ca} > \text{K} > \text{Si} > \text{Mg} > \text{Mn} > \text{P} > \text{Ti} > \text{Fe}$  for all clay samples,  $\text{Na} > \text{K} > \text{Ca} > \text{Si} > \text{Mg} > \text{Mn} > \text{P} > \text{Fe} > \text{Ti}$  for basaltic bulk samples, and  $\text{Na} > \text{Ca} > \text{K} > \text{Si} > \text{Mg} > \text{Mn} > \text{P} > \text{Fe} > \text{Ti}$  for basaltic clay sam-

ples, indicating that alkali metal (Na, K) and alkaline earth metal (Ca, Mg) elements are generally easier to be depleted from their parent materials than other major elements during chemical weathering.

### 3.2. Chemical weathering intensity indicated by various chemical indices

Si/Al, WIP, CIA and CIW are the most widely used major element indices of chemical weathering, thus they are selected here to investigate the chemical weathering of topsoils in the study area. Weathering indices that include iron in their calculations were not included here because they do not distinguish between ferric and ferrous iron (Price and Velbel, 2003). The Si/Al ratio, WIP, CIA and CIW values of bulk and clay samples were calculated as shown in supplementary Tables 2 and 3, and plotted along the latitudinal gradient in Figs. 4–7, respectively.

The silica–alumina ratio was proposed by Ruxton (1968) to measure the intensity of desilicication and allitization, and it is defined as the molecular ratio of  $\text{SiO}_2/\text{Al}_2\text{O}_3$ , traditionally expressed as Si/Al. WIP is a weathering index of Parker (Parker, 1970) defined as  $\text{WIP} = 100 \times (2\text{Na}_2\text{O}/0.35 + \text{MgO}/0.9 + 2\text{K}_2\text{O}/0.25 + \text{CaO}/0.7)$ . It is an important weathering index based on the molecular proportions of alkali metal (Na, K) and alkaline earth metal (Ca, Mg) elements. Both Si/Al and WIP are negatively related to chemical weathering intensity, with Si/Al ratio and WIP value decreasing with the increase of chemical weathering intensity. CIA is a



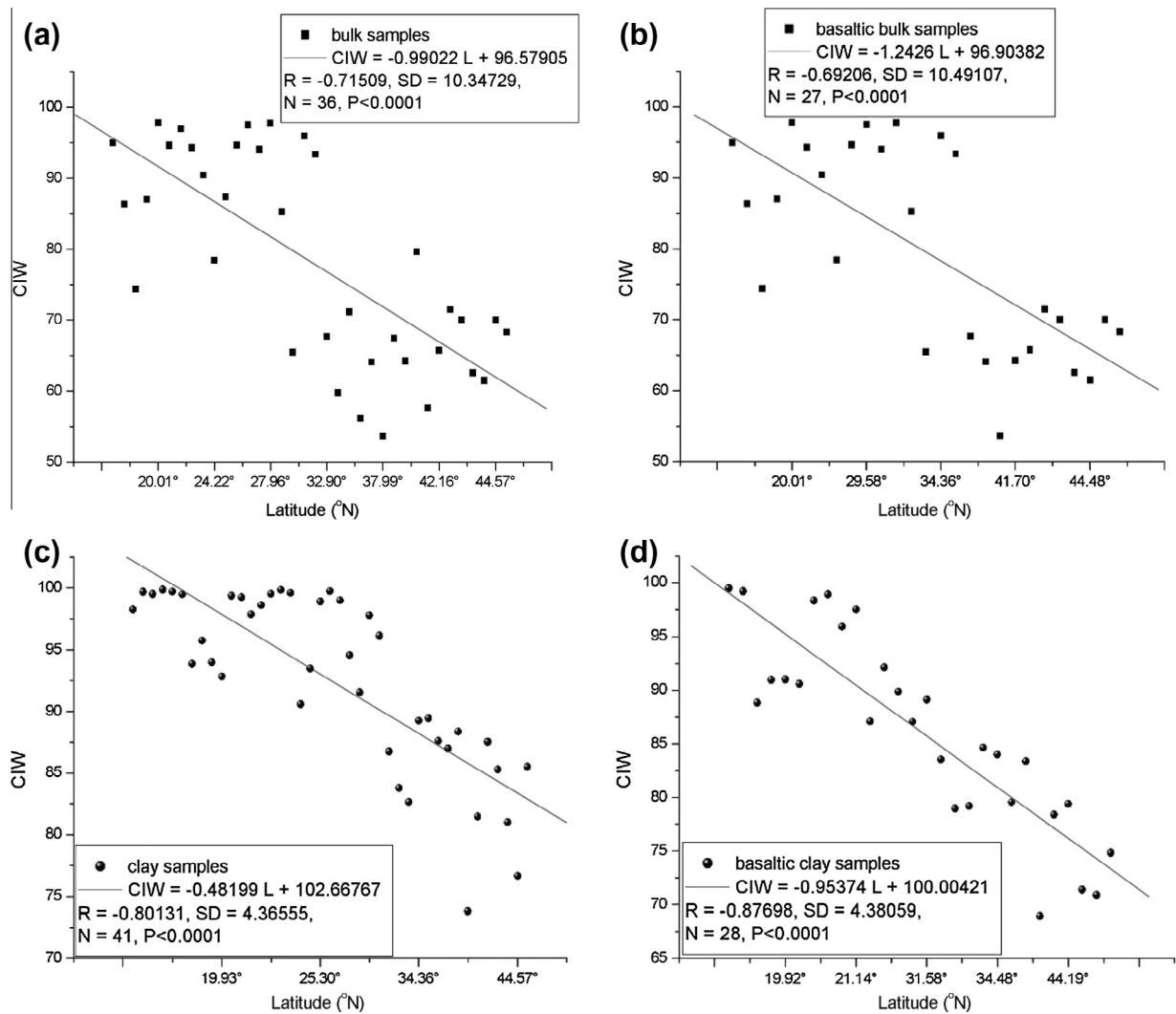


Fig. 6. CIW values of bulk (a), basaltic bulk (b), clay (c), and basaltic clay (d) samples along the latitudinal gradient.

chemical index of alteration proposed by Nesbitt et al (Nesbitt and Young, 1982), and defined as the molecular proportions of labile elements,  $CIA = 100 \times [Al_2O_3 / (Al_2O_3 + K_2O + Na_2O + CaO^*)]$ . CIW is a chemical index of weathering (Harnois, 1988), defined as the molecular proportions of Al, Na and Ca, i.e.  $CIW = [Al_2O_3 / (Al_2O_3 + CaO^* + Na_2O)] \times 100$ , without considering the proportion of K compared with CIA. Both CIA and CIW are positively related to chemical weathering intensity, with CIA and CIW values increasing with the increase of chemical weathering intensity. The  $CaO^*$  content of WIP, CIA and CIW is the amount of  $CaO$  incorporated in the silicate fraction of samples.

A chemical weathering investigation with these chemical indices applied to weathering profiles developed on heterogeneous felsic metamorphic parent rocks suggested that WIP was the most appropriate for application to weathering profiles on heterogeneous (and homogeneous) parent rock (Price and Velbel, 2003). A recent investigation has discussed the application of CIA to the river chemistry in China, and suggested that CIA does not reflect the instantaneous chemical weathering on continents, and that it is unrealistic to find a simple law of regulating chemical weathering in continents (Li and Yang, 2010). Another recent investigation on the river geochemistry of China reveals that integrated chemical weathering intensity in large latitudinal watersheds can be

quantitatively estimated using the proper geochemical proxies of river sediments with careful application (Shao et al., 2012). What about the applications of these chemical indices to chemical weathering of topsoils from monsoonal eastern China? For comparison, all the chemical indices were applied to four different types of samples, those developed on heterogeneous parent rocks, including 36 bulk samples and 41 clay samples, and those developed on homogeneous parent rock (basalt), including 27 basaltic bulk samples and 28 basaltic clay samples (supplementary Table 1, Tables 1 and 2).

### 3.2.1. $SiO_2/Al_2O_3$ ratio

As shown in supplementary Tables 2 and 3,  $SiO_2/Al_2O_3$  ratio is above 1 in all samples, and generally higher in bulk samples than in clay samples, with an average of 6.84 in all bulk samples, 6.96 in basaltic bulk samples, 2.90 in all clay samples, and 3.03 in basaltic clay samples, indicating that the topsoils of monsoonal eastern China are still undergoing processes of desilicification and allitization, and that parent rock diversity has little effect on the  $SiO_2/Al_2O_3$  ratios of both bulk and clay samples, but grain size has much effect on the  $SiO_2/Al_2O_3$  ratios of topsoil samples. As shown in Fig. 4, linear regression analysis shows no linear correlations between  $SiO_2/Al_2O_3$  ratios and their corresponding latitudes of all

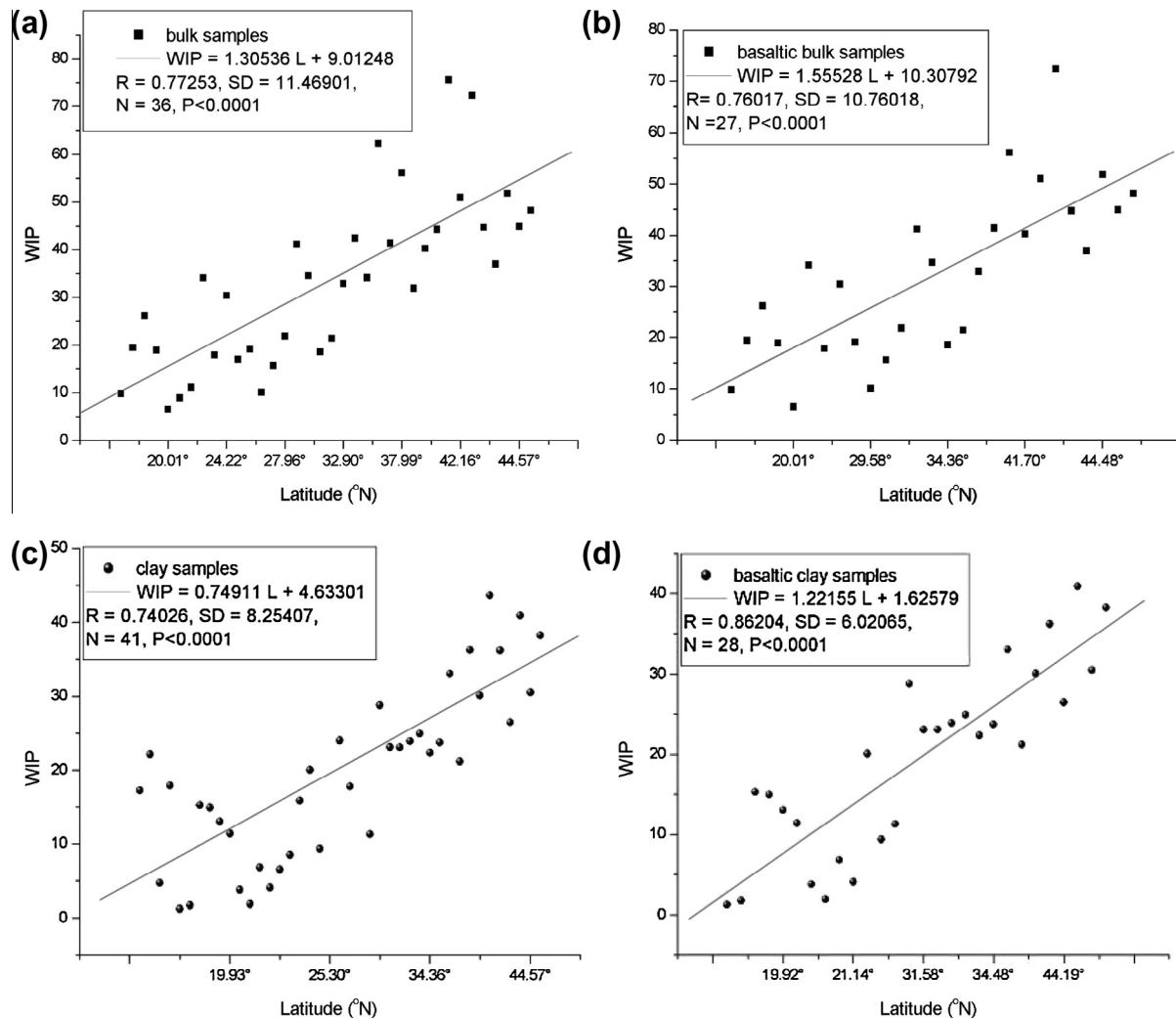


Fig. 7. WIP values of bulk (a), basaltic bulk (b), clay (c), and basaltic clay (d) samples along the latitudinal gradient.

bulk samples, no matter their parent rocks are heterogeneous or homogeneous, but a positive correlation is shown between  $\text{SiO}_2/\text{Al}_2\text{O}_3$  ratios and their corresponding latitudes of all clay samples, and the linear correlation is even better in clay samples developed on homogeneous parent rocks (basalt). All these results indicate that the latitudinal distribution of  $\text{SiO}_2/\text{Al}_2\text{O}_3$  ratio is more dependent on grain-size than on parent rock type, and that  $\text{SiO}_2/\text{Al}_2\text{O}_3$  ratio is not suitable to chemical weathering evaluation of bulk samples.

### 3.2.2. CIA

As shown in [supplementary Tables 2 and 3](#), CIA value is generally higher in clay samples than in bulk samples, with an average of 71.81 in all bulk samples, 72.46 in basaltic bulk samples, 87.30 in all clay samples, and 86.17 in basaltic clay samples, indicating the significant impact of grain-size on CIA values, and little impact of parent rock type on CIA values. [Fig. 5](#) shows the latitudinal distributions of CIA values for bulk and clay samples. A negative correlation is shown between the CIA values and their corresponding latitudes of both bulk and clay samples, and the linear correlation is even better in clay samples than in bulk samples, but parent rock diversity makes little difference in the linear correlations of them, indicating that CIA is suitable to both bulk and clay samples, and

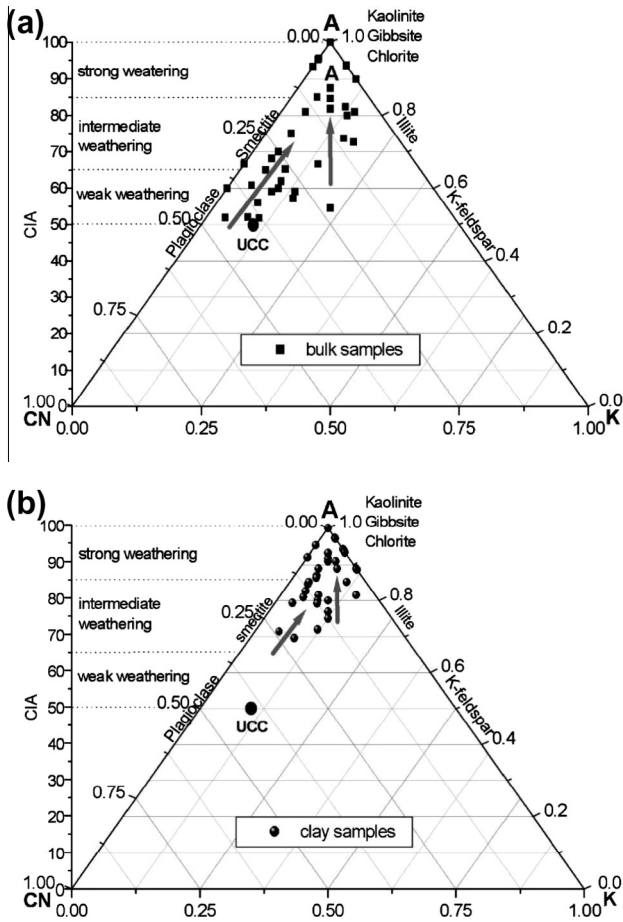
CIA values are more dependent on grain-size than on parent rock type.

### 3.2.3. CIW

As shown in [supplementary Tables 2 and 3](#), CIW values are generally higher in clay samples than in bulk samples, with an average of 78.26 in all bulk samples, 79.51 in basaltic bulk samples, 92.55 in all clay samples, and 90.58 in basaltic clay samples, indicating the significant impact of grain-size on CIW values, and little impact of parent rock type on CIW values. [Fig. 6](#) shows the correlation between CIW values and their corresponding latitudes of bulk and clay samples. Similar to the latitudinal distributions of CIA, the CIW values of both bulk and clay samples are negatively correlated with their latitudes, and the linear correlation is better in clay samples than in bulk samples, but parent rock diversity makes little difference in the latitudinal distributions of them, indicating that CIW is suitable to both bulk and clay samples, and CIW values are more dependent on grain-size than on parent rock type.

### 3.2.4. WIP

As shown in [supplementary Tables 2 and 3](#), WIP values are generally higher in bulk samples than in clay samples, with an average of 33.16 in all bulk samples, 32.08 in basaltic bulk samples, 20.36 in all clay samples, and 19.34 in basaltic clay samples, also



**Fig. 8.** The A–CN–K ternary diagrams of molecular proportions for bulk (a) and clay (b) samples of topsoils from eastern China. A =  $\text{Al}_2\text{O}_3$ ; C = CaO; N =  $\text{Na}_2\text{O}$  and K =  $\text{K}_2\text{O}$ . Also plotted is the UCC (Rudnick and Gao, 2003) as well as idealized mineral compositions (Nesbitt and Young, 1982, 1989). Arrows indicate predicted weathering trends exhibited by eastern China southward, which had experienced relatively weak to strong chemical weathering. The A–CN–K ternary diagrams probably indicate the presence of unaltered primary minerals in bulk samples.

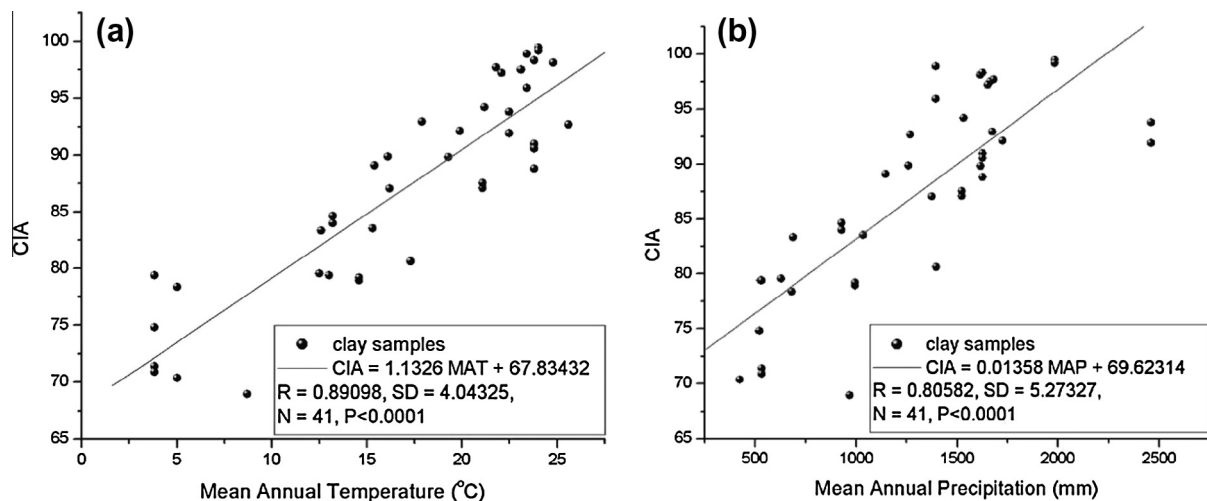
indicating the significant impact of grain-size on WIP values, and little impact of parent rock type on WIP values. Fig. 7 shows the latitudinal distributions of WIP values for bulk and clay samples.

WIP values are positively correlated with their latitudes for both bulk and clay samples, with little difference in the latitudinal distributions of WIP between all bulk samples and all clay samples, but the difference is a little bigger between clay samples developed on diverse rocks and those developed on the same rock type (basalt), indicating that WIP is also suitable to bulk and clay samples, and parent rock type plays an important role in the latitudinal distribution of WIP for clay samples.

As described above, grain-size has a significant impact on major element abundance and the latitudinal distributions of  $\text{SiO}_2/\text{Al}_2\text{O}_3$ , CIA, CIW and WIP. This is probably due to the presence of unaltered detrital minerals, such as quartz, plagioclase and K-feldspar in bulk samples, as indicated by the much higher  $\text{SiO}_2$  content in bulk samples (with an average of 55.0) than that in clay samples (with an average of 39.8) (Tables 1 and 2), and illustrated by the A–CN–K ternary diagrams (Fig. 8). Moreover, soil grain size itself implies the weathering intensity, i.e. the finer size meaning the stronger weathering, because weathering is one of the most important processes in soil formation. Compared with bulk samples, clay samples obtained from topsoils largely consist of secondary minerals which are produced by chemical weathering, particularly clay minerals, thus more directly and accurately reflect the chemical weathering of source sediments. In fact, the grain-size dependence of chemical weathering intensity has also been detected in Chinese loess-red clay deposit (Xiong et al., 2010). Consequently, we believe that clay samples are more suitable to investigating chemical weathering of sediments on continents than bulk samples. As for parent rock type, it has little impact on the latitudinal distributions of these indicators, except its impact on the latitudinal distribution of WIP in clay samples, probably indicating the relatively uniform chemical weathering on different rock types, and the difference in the latitudinal distributions of WIP between basaltic clay samples and clay samples of different source rocks may have resulted from the difference in enrichment of K, Na Ca and Mg between them.

### 3.3. Controls of chemical weathering

As indicated by the latitudinal distributions of  $\text{SiO}_2/\text{Al}_2\text{O}_3$ , CIA, CIW and WIP (Figs. 4–7), the chemical weathering intensity of topsoil samples increases with the decrease of latitude in monsoonal eastern China, displaying an obvious latitude effect. What is the control behind this phenomenon? To answer this question, we analyzed the latitudinal distributions of MAT and MAP in the study



**Fig. 9.** Correlation between CIA and MAT (a), and correlation between CIA and MAP (b), showing the climatic control on chemical weathering of topsoils in monsoonal eastern China.

area by correlating the MAT and MAP values of weathering stations located below 1000 m a.s.l. in the study area (ca. 109°E–130°E, 18°N–46°N) with their latitudes<sup>1</sup> (Fig. 2), then correlating the CIA values of all clay samples with their corresponding MAT and MAP along the latitudinal gradient (Fig. 9). The corresponding MAT and MAP for CIA values of all clay samples were estimated from the climatic record of the nearest weathering stations.

As shown in Figs. 2 and 9, the latitudinal distributions of MAT and MAP in the study area display a significant latitude effect like  $\text{SiO}_2/\text{Al}_2\text{O}_3$ , CIA, CIW and WIP of topsoils in the study area, particularly the latitudinal distributions of MAT, with MAT and MAP increasing southward along the latitudinal gradient in the study area, and the chemical weathering of topsoils gradually intensifying with the increase of MAT and MAP. Obviously, the latitudinal distributions of  $\text{SiO}_2/\text{Al}_2\text{O}_3$ , CIA, CIW and WIP of topsoils in monsoonal eastern China reflect those of MAT and MAP in the study area, indicating the dominant control of a monsoonal climate on the chemical weathering of topsoils in eastern China. This result is consistent with those obtained from river chemistry of China (Li and Yang, 2010; Shao et al., 2012). However we should keep in mind that the samples in our study were collected from different environments with various tectonics, parent rocks, climatic conditions, landforms, altitudes and vegetations in the study area. Soil formation is controlled by multiple factors like climate, biology, parent rock, landform, soil depth and age. In addition to the dominant control of climate, other factors might have some effect on the chemical weathering of topsoils in our study area, which needs further research. As pointed out by a recent investigation, chemical indices like CIA actually reflect the integrated weathering history in the study area (Li and Yang, 2010).

#### 4. Conclusions

Our chemical weathering investigations on topsoils from monsoonal eastern China based on major element analyses have obtained the following results.

1. Na, K, Ca, Mg, and Si are relatively depleted, while Mn, P, Fe and Ti are relatively enriched by comparison with the Al content. Alkali metal (Na, K) and alkaline earth metal (Ca, Mg) elements are generally easier to be depleted from their parent materials than other major elements during chemical weathering.
2. All the chemical weathering indices of  $\text{SiO}_2/\text{Al}_2\text{O}_3$ , CIA, CIW and WIP are suitable to bulk and clay samples, except the application of  $\text{SiO}_2/\text{Al}_2\text{O}_3$  in bulk samples. The  $\text{SiO}_2/\text{Al}_2\text{O}_3$  ratios of all samples indicate that the topsoils of the study area are still undergoing desilicication and alitization.
3. Grain-size has a significant impact on the mobility of major elements, and the latitudinal distributions of chemical weathering indices, but parent rock type has little effect on them, except its impact on the latitudinal distribution of WIP in clay samples. The significant grain-size dependence probably indicates the presence of unaltered minerals, such as quartz, plagioclase and K-feldspar in bulk samples, thus we suggest that clay samples are more suitable to investigating chemical weathering of sediments on continents than bulk samples. The trivial effect of parent rock type probably indicates a relatively uniform chemical weathering on various parent rocks. The difference in the latitudinal distributions of WIP between basaltic clay samples and clay samples of different source rocks may be attributed to the difference in enrichment of K, Na Ca and Mg between them.

4. Climate is the dominant control of chemical weathering of topsoils in monsoonal eastern China. In addition to the dominant control of climate, other factors like tectonics, parent rock, biology, landform and soil depth and age might have some effect on the chemical weathering of topsoils in our study area. Chemical indices like CIA actually reflect the integrated weathering history in the study area.

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#### Appendix A. Supplementary material

Supplementary data associated with this article can be found, in the online version, at <http://dx.doi.org/10.1016/j.jseaes.2013.12.004>.

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