



## Geochemical record of chemical weathering and monsoon climate change since the early Miocene in the South China Sea

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[1] The chemical index of alteration (CIA) and elemental ratios that are sensitive to chemical weathering, such as Ca/Ti, Na/Ti, Al/Ti, Al/Na, Al/K, and La/Sm, were analyzed for detrital sediments at Ocean Drilling Program Site 1148 from the northern South China Sea to reveal information of chemical weathering in the source regions during the early Miocene. High CIA values of  $\sim 80$ , coupled with high Al/Ti and Al/Na and low Na/Ti and Ca/Ti, are observed for the sediments at  $\sim 23$  Ma, indicating a high chemical weathering intensity in the north source region, i.e., south China. This was followed by gradual decreases in Al/Ti, Al/Na, La/Sm, and Al/K ratios, as well as the CIA values, and increases in Ca/Ti and Na/Ti ratios. These records together with other paleoclimate proxies, such as black carbon  $\delta^{13}\text{C}$  and benthic foraminifer  $\delta^{18}\text{O}$ , give reliable information on the climate changes in south China. Our results show that the climate in south China was warm and humid in the early Miocene ( $\sim 23$  Ma) according to the chemical weathering records. The humidity in south China decreased from the early Miocene to Present with several fluctuations centering at approximately 15.7 Ma, 8.4 Ma, and 2.5 Ma, coincident with the global cooling since the middle Miocene. These climate changes implied that the summer east Asian monsoon has dramatically affected south China in the early Miocene, whereas the influence of the summer monsoon on this region has decreased continuously since that time, probably because of the intensification of the winter monsoon. Such an evolution for the east Asian monsoon is different from that for the Indian monsoon.

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### 1. Introduction

[2] The global climate had changed significantly in the Cenozoic with the Earth's climate system continually drifting from extreme warmth with no ice cap at the poles to extreme cold with massive continental ice sheets and polar ice caps, which has been attributed to the gradual changes of some of the main boundary conditions on the Earth, including the geography and topography of continents [Zachos *et al.*, 2001]. The uplift of the Tibetan Plateau is believed to be an important driving force behind global cooling in the late Cenozoic [Raymo and Ruddiman, 1992] and the land-sea distribution resulted from plate motions in Eurasian may also have controlled the climate change during this period [Ramstein *et al.*, 1997]. In Southeast Asia and the southwest Pacific, a series of major tectonic events in the Cenozoic significantly changed the geography and topography [England and Houseman, 1986; Rowley, 1996], and had huge impact on the climate in this region [Hall, 2002]. For example, the Asian monsoon system is believed to have built up during the Cenozoic [An *et al.*, 2001; Kutzbach *et al.*, 1993; Ramstein *et al.*, 1997].

[3] Because monsoon controls the climate in the east and the south Asia, the commencement and the development of the monsoon climate is essential to understanding the climate change in these regions. Pedogenic carbonate  $\delta^{13}\text{C}$  records in Northern Pakistan suggest dramatic ecological change occurring at  $\sim 7.4$ – $7.1$  Ma, indicating the onset of the Indian monsoon [Quade *et al.*, 1989]. Chemical weathering in Himalayan region reconstructed in Bengal Fan also suggest an intensification of monsoon at  $\sim 7$  Ma [Derry and France-Lanord, 1996]. A number of studies on the Chinese Loess Plateau suggested that the onset of the east Asian monsoon occurred in the late Miocene (9–8 Ma), which is similar to the Indian monsoon [An *et al.*, 2001]. Recent results however, indicate that the winter monsoon has affected North China as early as 22 Ma [Guo *et al.*, 2002]. Moreover, the compiled palaeobotanical results in China indicate that the east Asian monsoon commenced in the latest Oligocene, much earlier than the late Miocene [Liu *et al.*, 1998; Sun and Wang, 2005]. All these facts suggest that the initiation and evolution of the east Asian monsoon is different from the Indian monsoon. However, this might be an artifact of the rarity of long and continuous profiles, because continuous records since the late Oligocene for the east Asian monsoon are scarce currently.

[4] In this respect, the South China Sea (SCS) is one of the best areas for this study. Having opened in about 30 Ma [Briais *et al.*, 1993; Clift and Lin, 2001], the SCS is now the largest marginal sea in the west Pacific. A continuous sediment core with age ranging from 32.8 Ma to the Present

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was drilled at ODP Site 1148 in the northern SCS [*Shipboard Scientific Party*, 2000]. This enables us to investigate the climate changes in this region during the last 30 Ma. The sediment core from ODP Site 1148 has been studied in terms of trace elements, isotopes and clay minerals [*Clift et al.*, 2002; *Jia et al.*, 2003; *Li et al.*, 2003]. The accumulation rates and clay mineral compositions of the sediment suggest that strong summer monsoon was likely influenced south China during the early and the early part of the middle Miocene [*Clift et al.*, 2002; *Clift*, 2006; *Clift and Sun*, 2006]. Also, black carbon  $\delta^{13}\text{C}$  records indicate that the summer monsoon has affected the terrestrial ecosystem of south China in the early Miocene [*Jia et al.*, 2003]. All these suggest that sediments at ODP Site 1148 are one of the best archives for studying the onset and the early evolution of the east Asian monsoon.

[5] Chemical weathering records in south China may provide a more definite answer for this question. The precipitation in south China is mainly supplied by summer monsoon [*Ding*, 1994], which controls the humidity in this region. As chemical weathering intensity is largely controlled by humidity [*Berner and Berner*, 1997], the variation of chemical weathering intensity in south China reflects the change of east Asian monsoon. Elemental and Nd isotopic results of the previous studies indicate that sediments deposited after  $\sim 23$  Ma at ODP Site 1148 were from south China [*Clift et al.*, 2002; *Li et al.*, 2003], and chemical composition of detrital components in marine sediments is sensitive to chemical weathering, and is helpful to trace climate changes in their source regions [*Tamburini et al.*, 2003; *Wehausen and Brumsack*, 2002; *Wei et al.*, 2003, 2004; *Zabel et al.*, 2001]. In this paper, we reassessed the major and trace element composition of the detrital components of sediments in the top 455 mcd (meters composite depth) at ODP Site 1148, focusing on the variations of selected element pairs that are sensitive to terrestrial chemical weathering. This may help to reconstruct the chemical weathering records in south China, and investigate the initiation and the early evolution of the east Asian monsoon.

## 2. Material, Methods, and the Data

[6] The sediment core recovered at ODP Site 1148 ( $18^{\circ}50.17'\text{N}$ ,  $116^{\circ}33.94'\text{E}$ , at a water depth of 3294 m) is 859.5 m long (Figure 1). Age model for these sediments were obtained from biostratigraphy of planktonic foraminifera and magnetostratigraphy [*Li et al.*, 2003], and revised by [*Li et al.*, 2005]. The upper sections, from 455 mcd to the top were continuously deposited since the early Miocene (from 23.6 Ma to the Present). They consist mainly of green-gray hemipelagic sediments. Biogenic carbonate accounts for 1% to 20% of the sediments from the top to about 190 mcd, and about 20% to 50% from 190 to 455 mcd. The biogenic carbonate contents decrease upward. Detailed description of these sediments is provided by [*Shipboard Scientific Party*, 2000].

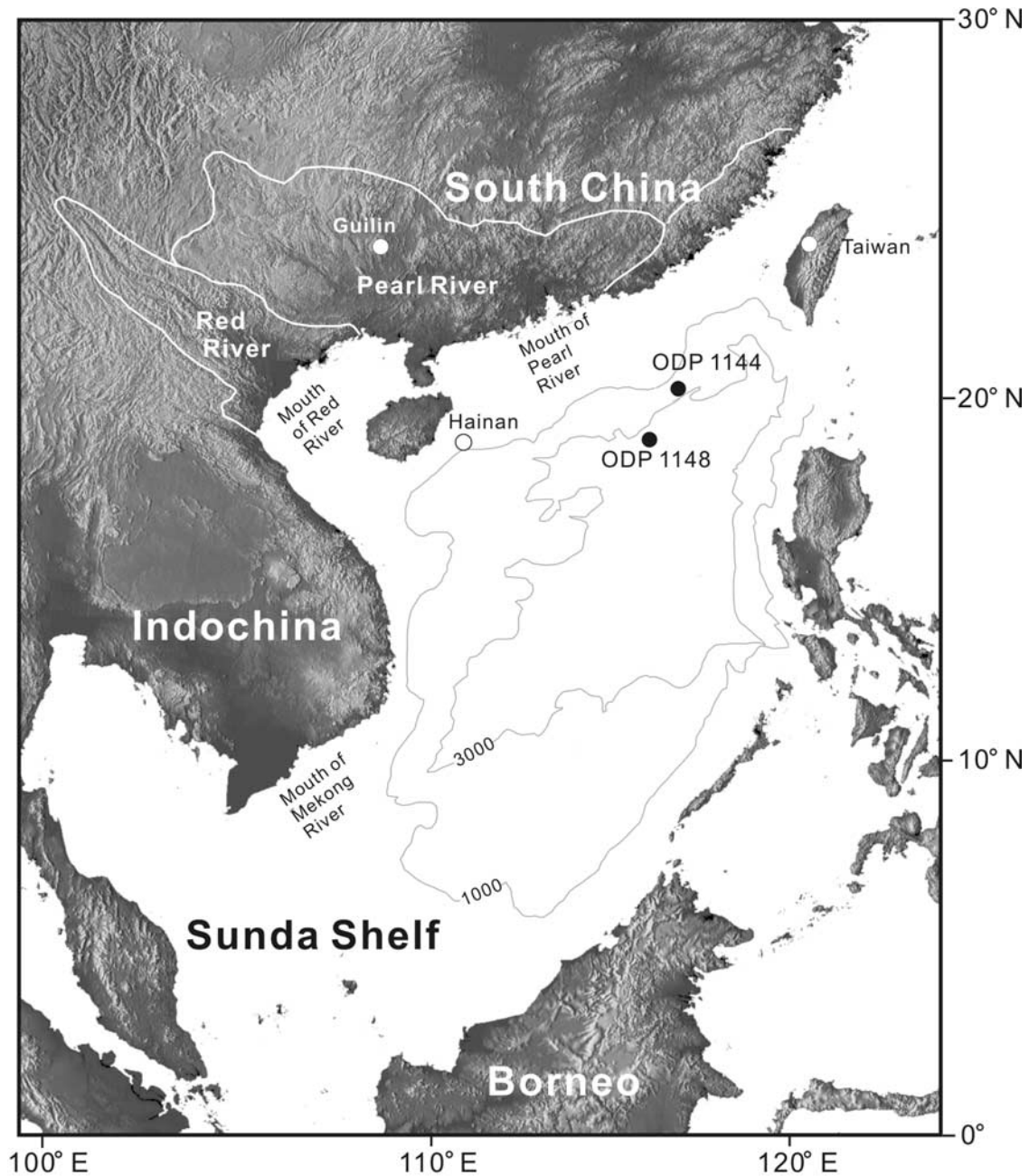
[7] The analyzed sediments were sampled in an interval of  $\sim 4$ – $5$  m, corresponding to an average time resolution of  $\sim 0.18$  m.y. The samples were first dried at  $50^{\circ}\text{C}$ , and then crashed into powders. This was followed by baking at

$670^{\circ}\text{C}$  for 30 min to destroy organic materials, and then reacting with 0.1 M HCl for 24 hours to remove carbonate and Fe-Mn oxides. Finally, the samples were rinsed with deionized water for 5 times and dried for elemental composition analysis. This chemical treatment can remove most of the authigenic materials in the sediments, such as carbonate, Fe-Mn oxides, organic materials and adhered materials except for biogenic opal [*Freydier et al.*, 2001]. However, the biogenic opal contents in the sediments above 455 mcd are very low [*Wang et al.*, 2001], and many of the elements, such as Al, K, Na, Ca, Mg, Ti and rare earth elements (REEs) are undetectable in biogenic opal. Thus the ratios between these elements are identical to the corresponding ratios of the detrital components [*Li et al.*, 2003]. Also, leaching by diluted HCl does not influence the chemical composition of detrital components in sediments from the SCS, which is in form of silicate. Similar treatment had been handled for the sediments at ODP Site 1144. Comparison between the chemical treated and bulk sediments indicates that potassium, one of the mobile elements during chemical weathering, was not leached from detrital components [*Wei et al.*, 2003], and most of the elements that are not concentrated in authigenic components show similar variation patterns both in treated and bulk sediments [*Wei et al.*, 2003, 2004]. Therefore this chemical treatment can effectively removed authigenic components, without changing the composition of the detrital component in the sediments.

[8] The sediments were digested by concentrated HF + HNO<sub>3</sub> mixture, and the major and trace elements were measured in the Key Laboratory of Isotope Geochronology and Geochemistry, Guangzhou Institute of Geochemistry, Chinese Academy of Sciences on an inductively coupled plasma atomic emission spectrometry (ICP-AES) and an inductively coupled plasma mass spectrometry (ICP-MS), respectively. Detailed analytical methods have been described by *Li et al.* [2002, 2003] and *Liu et al.* [1996]. Precision is generally 1–3% for major elements, and better than 3% for trace elements. The major and trace element data are given by *Li et al.* [2003] and ODP data archives, and the concentration of the elements discussed in this paper were presented in Table 1.

## 3. Chemical Weathering Records

[9] Chemical weathering can modify the major and trace element contents of rocks and sediments. The behaviors of elements are different during chemical weathering processes. Some elements, such as Na, Ca, and Sr, are very mobile and are easily removed from parent rocks and sediments, and thus are generally depleted in weathering products (solid residue) [*Nesbitt and Markovics*, 1997; *Nesbitt et al.*, 1980]. Other elements, such as K, Rb, Mg, Ba, are easily leached off primary minerals, but tend to be fixed by secondary clay minerals in weathering profiles [*Nesbitt and Markovics*, 1997; *Nesbitt et al.*, 1980], such that are usually enriched in the weathering products after moderate chemical weathering process [*Condie et al.*, 1995; *Nesbitt and Markovics*, 1997; *Nesbitt et al.*, 1980; *Peuraniemi and Pulkkinen*, 1993]. In contrast, elements, such as Al, Fe, Ti, Zr, Hf, are conservative during chemical weathering processes, and they tend to



**Figure 1.** Sketch bathymetric map of the South China Sea and surrounding regions showing the locations of ODP Site 1148 and ODP Site 1144. The topographic image shows the continents and islands. White lines range the modern drainage of the major rivers in South China that may transport sediments to the northern South China Sea. The mouths of the major rivers are also shown in the map. The white circles on land and the open circle on sea indicate the locations of the pollen records discussed in the text.

be enriched or kept constant in weathering products comparing with parent rocks and sediments [Condie *et al.*, 1995; Nesbitt and Markovics, 1997; Nesbitt *et al.*, 1980; Peuraniemi and Pulkkinen, 1993]. Importantly, the behaviors of the elements can change with the chemical weathering intensity. For example, K, Rb, Ti, Zr, REEs tend to be enriched in weathering products after moderate chemical weathering process, but become depleted after

extreme chemical weathering process [Condie *et al.*, 1995; Nesbitt, 1979].

[10] Weathering products compose the majority of the detrital components in sediments [Windom, 1976]. Thus the variations of element contents in weathering products can also be revealed in detrital sediments; that is, selected element contents and ratios in sediments can be used as tracers for the intensity of chemical weathering, and enable



**Table 1.** Concentrations of the Selected Elements in Detrital Sediments at ODP Site 1148<sup>a</sup>

Depth, mcd	Age, Ma	Major Elements, %						Trace Elements, $\mu\text{g/g}$				
		Al <sub>2</sub> O <sub>3</sub>	CaO	K <sub>2</sub> O	MgO	Na <sub>2</sub> O	TiO <sub>2</sub>	Sc	Zr	La	Sm	Th
49.11	0.87	16.60	0.38	3.33	2.12	1.33	0.89	17.01	150	34.14	4.64	12.02
55.46	0.93	15.27	0.46	3.47	1.48	1.55	0.92	14.34	159	25.95	3.65	9.99
60.21	0.98	16.09	0.46	3.17	1.70	1.44	0.98	16.60	151	33.50	4.67	12.41
61.41	0.99	16.20	0.46	3.64	1.84	1.55	0.95	15.73	164	28.66	3.91	10.56
65.91	1.03	16.51	0.21	3.72	1.68	0.92	0.90	15.81	160	27.37	3.31	10.06
68.68	1.06	16.35	0.56	3.30	2.15	1.53	0.89	16.45	180	30.16	4.09	9.11
70.18	1.08	16.23	0.60	3.30	2.15	1.52	0.92	16.86	153	32.32	4.60	12.11
75.83	1.13	16.92	0.36	3.63	1.84	1.39	0.94	15.89	163	28.70	3.70	10.75
79.98	1.17	16.02	0.53	3.39	1.54	1.58	0.99	15.62	157	34.66	4.92	13.18
85.18	1.25	16.49	0.45	3.62	1.86	1.48	0.92	15.45	167	27.99	3.83	9.71
89.48	1.31	17.18	0.48	3.59	1.74	1.57	1.06	16.51	164	37.74	5.21	14.45
95.43	1.39	16.56	0.42	3.57	1.98	1.43	0.95	15.30	166	33.26	4.47	11.30
100.03	1.45	16.83	0.51	3.40	1.86	1.45	0.95	16.54	165	35.96	4.98	12.55
105.93	1.53	16.10	0.33	3.47	1.48	1.39	1.00	14.92	168	20.53	3.25	4.19
110.48	1.61	17.00	0.43	3.09	2.13	1.30	0.90	18.74	172	28.63	3.79	9.96
116.19	1.79	16.26	0.46	3.52	1.81	1.43	1.01	15.38	161	34.16	4.49	11.38
121.41	1.97	16.97	0.54	3.25	1.92	1.36	0.98	17.25	151	30.42	3.91	11.07
126.31	2.13	16.22	0.44	3.42	1.75	1.47	0.97	15.90	154	20.26	3.36	5.76
126.86	2.14	16.33	0.53	3.27	1.67	1.40	1.01	15.70	151	31.54	4.03	12.05
131.36	2.29	17.17	0.41	3.23	2.02	1.31	0.99	17.12	173	30.08	3.98	9.57
136.31	2.45	16.86	0.31	3.41	1.97	1.32	0.96	16.10	153	31.48	4.01	9.07
141.31	2.52	16.76	0.35	3.01	1.85	1.21	0.95	16.04	153	29.19	3.87	7.99
146.86	2.59	15.88	0.45	3.34	1.69	1.36	0.98	15.20	164	14.42	1.86	5.03
151.03	2.70	17.23	0.44	3.25	2.13	1.37	1.04	17.90	156	31.98	4.05	10.86
156.03	2.84	16.86	0.43	3.09	1.97	1.31	1.08	17.65	146	33.99	4.35	11.81
160.53	3.19	16.22	0.44	3.00	1.91	1.38	1.10	17.48	152	31.81	4.24	11.49
165.63	3.58	15.75	0.41	2.85	1.86	1.34	1.07	17.24	152	27.70	3.74	9.50
170.13	4.03	15.53	0.40	2.59	1.97	1.38	1.00	17.69	149	27.48	3.82	9.51
175.23	4.58	14.63	0.54	2.80	1.62	1.36	1.15	16.19	158	29.78	4.06	11.00
179.73	5.04	14.97	0.36	3.09	1.71	1.27	1.08	15.85	158	30.67	4.18	11.54
184.83	5.17	15.56	0.38	3.14	1.68	1.38	1.19	16.69	162	32.08	4.41	11.34
189.33	5.33	16.37	0.42	3.76	1.52	1.54	1.30	16.83	193	29.28	3.94	10.58
194.53	5.64	15.92	0.37	3.35	1.80	1.33	1.04	16.75	158	29.56	4.08	11.48
199.03	6.02	15.16	0.34	3.23	1.78	1.20	1.00	15.84	154	26.30	3.36	8.32
204.23	6.33	14.69	0.34	2.99	1.65	1.16	1.04	15.45	155	28.69	3.81	11.65
208.73	6.61	16.61	0.38	3.84	2.12	1.37	1.06	16.16	167	30.48	4.02	11.78
213.83	6.91	15.68	0.31	3.62	1.92	1.22	0.97	14.81	169	31.99	3.94	11.21
218.33	7.18	16.74	0.22	3.52	2.00	1.01	1.03	16.61	168	32.69	3.96	11.10
223.53	7.41	16.23	0.21	3.38	1.84	1.07	1.08	16.14	175	33.12	4.09	10.95
228.03	7.60	16.30	0.29	3.49	1.85	1.09	1.06	16.36	177	32.05	3.88	11.81
233.13	7.81	15.17	0.23	3.40	1.60	1.08	1.04	15.20	165	27.46	3.37	11.15
237.63	7.99	15.37	0.22	3.30	1.74	1.07	1.04	15.53	172	23.28	3.08	7.58
242.73	8.20	15.72	0.21	3.19	1.89	0.91	0.96	15.91	155	24.67	3.05	7.91
247.23	8.35	16.15	0.21	3.20	1.88	0.91	0.96	16.10	158	28.59	3.46	12.03
252.33	8.52	16.07	0.16	3.89	1.89	0.91	0.93	15.70	152	29.42	3.50	11.86
256.83	9.42	15.99	0.22	3.36	1.79	1.02	0.95	15.64	157	25.72	3.20	8.33
261.93	9.64	16.21	0.21	3.33	1.80	0.99	0.94	15.86	159	27.97	3.44	12.89
266.43	9.68	16.49	0.23	3.30	1.95	1.00	0.86	15.22	166	27.85	3.43	10.99
271.53	10.04	15.57	0.22	3.22	1.68	1.02	0.94	15.08	163	28.15	3.45	12.43
276.03	10.42	16.23	0.21	3.23	1.76	0.96	0.92	15.83	163	27.39	3.31	11.59
281.13	10.80	16.32	0.22	3.27	1.67	0.98	0.94	15.86	159	27.19	3.37	12.84
285.63	11.61	16.44	0.22	3.24	1.70	0.97	0.90	15.72	152	28.00	3.36	11.43
290.73	12.05	15.46	0.22	3.18	1.58	0.98	0.92	14.91	157	28.88	3.47	11.82
295.23	12.40	16.29	0.21	3.47	1.71	1.04	0.93	15.57	156	29.83	3.52	13.23
300.33	12.94	15.60	0.21	3.15	1.60	0.98	0.96	14.79	153	22.88	2.76	7.88
304.83	13.32	15.71	0.21	3.27	1.70	0.95	0.91	14.92	174	28.25	3.43	11.61
310.03	13.68	15.41	0.20	3.32	1.64	0.92	0.91	14.93	152	22.71	2.73	8.01
314.53	14.07	15.73	0.17	3.21	1.80	0.84	0.86	14.90	151	26.74	3.15	9.66
319.63	14.52	16.63	0.15	3.47	1.85	0.87	0.91	15.49	146	23.45	2.74	7.21
324.13	14.92	16.59	0.14	3.24	1.82	0.82	0.86	15.21	152	27.24	3.24	11.67
329.33	15.38	17.03	0.12	3.08	1.87	0.70	0.82	15.22	131	25.59	3.15	9.26
333.83	15.73	15.26	0.20	2.72	1.63	0.79	0.80	13.80	140	22.94	2.84	9.91
338.93	16.08	15.50	0.21	2.60	1.72	0.84	0.82	14.16	145	21.79	2.66	8.10
343.43	16.38	16.10	0.15	2.80	1.83	0.78	0.74	14.74	122	22.35	2.75	8.11
348.53	16.73	15.84	0.17	2.84	1.86	0.74	0.76	14.29	135	24.41	2.95	8.48
353.03	17.04	15.24	0.22	2.70	1.81	0.86	0.77	13.85	151	22.83	2.72	8.44
358.03	17.38	15.68	0.20	2.74	1.77	0.85	0.74	14.24	142	22.67	2.79	8.97
362.53	17.68	15.48	0.19	3.12	1.92	0.96	0.84	14.19	152	25.33	3.02	9.80
367.63	18.03	15.78	0.19	3.20	1.81	0.94	0.79	14.17	146	25.84	3.01	9.88

Table 1. (continued)

Depth, mcd	Age, Ma	Major Elements, %						Trace Elements, $\mu\text{g/g}$				
		Al <sub>2</sub> O <sub>3</sub>	CaO	K <sub>2</sub> O	MgO	Na <sub>2</sub> O	TiO <sub>2</sub>	Sc	Zr	La	Sm	Th
372.13	18.33	15.72	0.14	3.06	1.97	0.86	0.81	14.00	154	26.25	3.02	9.39
377.47	18.57	15.37	0.17	3.14	1.84	1.01	0.84	13.39	158	26.20	3.06	10.79
380.47	18.71	15.86	0.19	3.14	1.71	0.97	0.85	13.91	162	26.94	3.29	10.68
387.17	19.02	16.29	0.19	3.01	1.69	1.01	0.84	13.97	170	25.17	2.98	9.03
390.17	19.16	15.50	0.13	2.77	2.05	0.90	0.73	13.10	155	24.69	2.94	11.91
396.87	19.57	16.16	0.15	2.90	1.97	1.07	0.79	13.47	145	24.55	2.91	9.54
399.87	19.76	15.33	0.27	2.62	1.62	1.00	0.72	12.06	147	24.29	2.86	10.31
406.47	20.18	15.55	0.18	2.68	1.93	1.03	0.73	12.87	144	23.13	2.70	8.02
409.47	20.37	16.88	0.29	2.80	1.90	1.03	0.75	13.40	142	23.06	2.66	7.38
416.17	20.79	15.87	0.22	2.65	1.84	0.93	0.71	13.14	135	22.44	2.60	7.12
419.17	20.98	15.83	0.21	2.71	1.77	0.92	0.71	13.28	146	24.24	2.74	8.02
425.87	21.40	15.75	0.18	2.74	1.65	0.78	0.72	12.38	131	25.21	2.98	7.78
428.87	21.59	16.30	0.22	2.75	1.64	0.86	0.71	12.98	129	23.14	2.71	8.01
438.47	22.20	16.73	0.07	2.59	1.70	0.81	0.75	13.12	123	21.92	2.66	7.76
445.17	22.62	16.43	0.02	2.58	1.79	0.72	0.74	12.75	129	25.36	3.00	7.42
448.17	22.81	15.92	0.01	2.55	1.80	0.80	0.74	12.97	136	24.39	2.91	7.72
454.77	23.24	12.10	0.00	2.12	1.51	0.60	0.59	7.20	127	23.94	2.79	7.08

<sup>a</sup>Data are from *Li et al.* [2003] and ODP data archive.

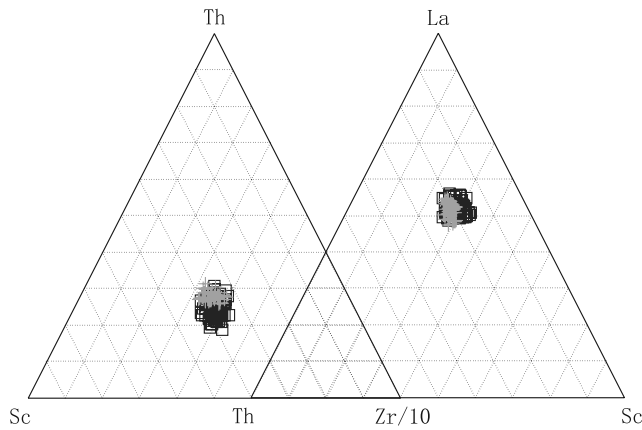
the reconstruction of climate and environment changes in source regions of the sediments [*Kronberg et al.*, 1986; *Nesbitt and Young*, 1982; *Zabel et al.*, 2001]. Preliminary studies indicate that element contents in detrital components of sediments from the SCS, in which terrestrial materials dominate, provide a good record of the chemical weathering intensity in the source region and exhibit obvious climate controlled cycles [*Wei et al.*, 2003, 2004].

[11] However, in addition to chemistry weathering in source regions, other factors may also influence the elemental ratios in detrital sediments, such as hydraulic sorting and changes of provenance [*Fralick and Kronberg*, 1997], as well as diagenesis after burial. Hydraulic sorting controls the grain sizes of the sediments and results in element discrimination [*Zhang et al.*, 2002], and diagenesis may change the distribution of some elements in sediments [*Froelich et al.*, 1979]. The elements we select here to reconstruct the chemical weathering records are Al, Ti, Na, Ca, K, Mg and REEs. The behaviors of these elements during chemical weathering have well been defined, and they are dominantly hosted in fine grain size fractions in sediment [*Zhang et al.*, 2002]. Considered that significant grain size change is not observed in the upper 455 mcd sediments at ODP Site 1148 [*Shipboard Scientific Party*, 2000], thus the ratios of these elements are not likely to be affected by hydraulic sorting. Furthermore, these elements are conservative during diagenesis [*Froelich et al.*, 1979], and their ratios seem not be affected by diagenesis.

[12] Provenance change of the sediments may be induced by following processes: change of source regions, consecutive weathering through heterogeneous source region and change of delivery patterns for sediments in marine basin. All these may change the elemental ratios in sediments and obscure the signals of chemical weathering. It is difficult to clarify the influence of each process in the South China Sea because the topography of the basin and surrounded regions has changed significantly since the early Miocene. Fortunately, some of the geochemical proxies, such as immobile trace elements and Nd isotopes can well track the provenance changes of sediments. As sediments from different

lithological units may have different values, change of provenance by any of the process mentioned above may result in change of the values of one or more of these geochemical proxies. Particularly, in the northern SCS, sediments are mainly supplied from south China, including Pearl River drainage and some coastal regions and islands, such as Taiwan and Hainan Islands [*Li et al.*, 2003]. We consider the general change of chemical weathering intensity in the whole south China in this study, thus using geochemical proxies can well monitor the sources out of this region, as well as significant change of lithological units for weathering within this region.

[13] Changes of the provenance for sediments deposited since ~23 Ma at ODP Site 1148 is very limited. Detailed Nd isotopic and geochemical studies have indicated that the major provenance change occurred at ~26 Ma (477 mcd) [*Clift et al.*, 2002; *Li et al.*, 2003]. Sediments below 477 mcd were mainly from the south provenance of Indochina-Sunda Shelf and Borneo, and sediments above 455 mcd (~23 Ma) were mainly from lands at the north margin of the SCS [*Li et al.*, 2003]. No significant provenance change is observed after 23 Ma according to the Nd isotopes [*Clift et al.*, 2002; *Li et al.*, 2003]. This is further supported by La-Th-Sc and Th-Sc-Zr/10 discrimination diagrams [*Bhatia and Crook*, 1986], which show that the ranges for the sediments deposited after 23 Ma (above 455 mcd) are concentrated, and overlap with the sediments from ODP Site 1144 (Figure 2), which is located very close to ODP Site 1148. The time span of the sediments gathered at ODP Site 1144 covers the last 1.1 Ma [*Shipboard Scientific Party*, 2000], and the their provenance has been well constrained to be from south China [*Shao et al.*, 2001]. Temporal change of provenance since 23 Ma can also be constrained by the variations of Th/Sc ratios and Nd isotopes (Figure 3). The Th/Sc ratios of these sediments change from 0.5 to 0.8, and from -13 to -11 for  $\epsilon\text{Nd}(0)$ , and they are all within the range of the north provenance [*Li et al.*, 2003], without any secular trend of increasing or decreasing. Therefore the selected element ratios are not resulted from changes in sediment provenance, but reveal change of chemical weathering intensity in their source regions.



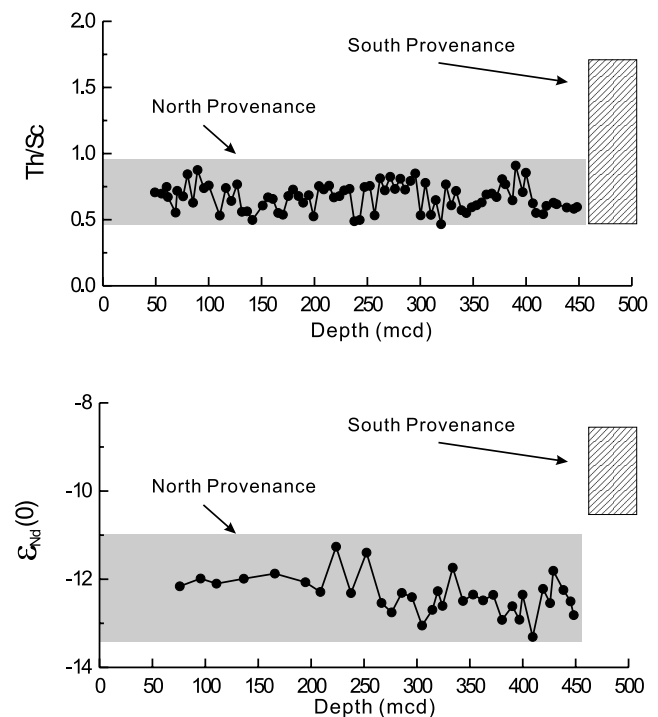
**Figure 2.** La-Th-Sc and Th-Sc-Zr/10 discrimination diagrams of the ODP Site 1148 sediments. The squares represent the sediments deposited after 23 Ma, and the crosses represent those from ODP Site 1144. These two groups overlap, indicating similar provenance.

[14] Here we focus on the chemical weathering information of the selected elemental ratios. The CIA defined as the percentage of  $\text{Al}_2\text{O}_3$  in  $[\text{Al}_2\text{O}_3 + \text{CaO} + \text{Na}_2\text{O} + \text{K}_2\text{O}]$  using molecular proportions [Nesbitt and Young, 1982] is generally used as a proxy for chemical weathering intensity. The Na/Ti and Ca/Ti ratios in the weathering products may decrease when chemical weathering is enhanced because Na and Ca are easily removed during chemical weathering [Nesbitt and Young, 1982]. In contrast, enhanced chemical weathering may result in higher Al/Ti and Al/Na ratios in sediments because Al tends to be enriched in weathering products [Nesbitt and Markovics, 1997]. Potassium tends to be enriched in weathering products after moderate chemical weathering but depleted after extreme chemical weathering [Condie et al., 1995; Nesbitt et al., 1980]. Therefore increasing Al/K ratios may indicate extensive strengthening in chemical weathering intensity. In addition to major elements, REEs are also discussed here because chemical weathering can alter the contents and the patterns of the REE in the weathering products [Condie et al., 1995; Nesbitt, 1979]. Because heavy REEs are depleted in the products after extreme weathering relative to light REEs [Nesbitt, 1979], the La/Sm ratios may trace whether extreme chemical weathering had occurred in the source regions. The variations of the Ca/Ti, Na/Ti, Al/Ti, Al/Na, Al/K, and La/Sm ratios, CIA values and mass accumulation rates (MAR) are shown in Figures 4a and 4b, respectively.

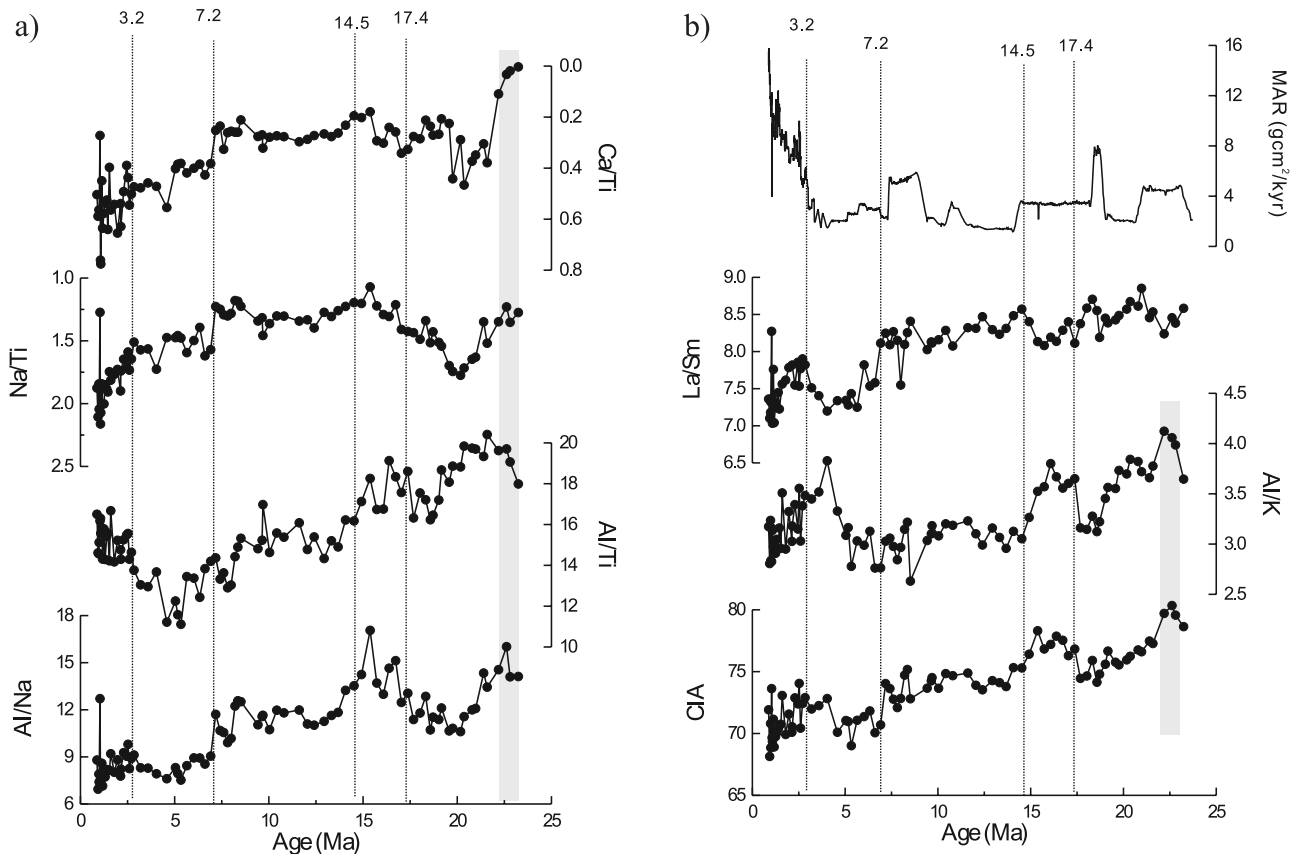
[15] Sediments at 455–445 mcd, with ages from 23.2 to 22.6 Ma, show very low Ca/Ti ratio, less than 0.05, and high CIA values, about 80. These values are similar to those in the sediments from the south provenance with tropical climate, where chemical weathering has been significantly enhanced since ~30 Ma [Li et al., 2003]. Meanwhile, the Na/Ti ratios of these sediments are low, whereas the Al/Na, Al/Ti, and Al/K ratios are high (Figures 4a and 4b). All these indicate that chemical weathering intensity in their source region, south China, was very high at ~23 Ma.

[16] From ~23 Ma to the present, a gradual decrease in chemical weathering intensity is clearly shown in Figures 4a and 4b. The CIA values decrease from about 80 at ~23 Ma to about 70 at 0.8 Ma. Both Ca/Ti and Na/Ti ratios show a continuous increasing trend, with Ca/Ti ratio rising from ~0.03 at ~23 Ma to ~0.8 at ~1 Ma, and from ~1.2 to ~2.2 from Na/Ti ratios. On the contrary, Al/Ti and Al/Na ratios decrease gradually (Figures 4a and 4b). Similar decreasing trend can also be seen in La/Sm and Al/K ratios. La/Sm ratio decreases from ~8.6 to ~7.0 and, Al/K ratio from ~4 to ~3 since ~23 Ma. Considering that chemical weathering intensity at ~23 Ma were high in south China, further enhancing of the chemical weathering intensity may result in increasing of La/Sm and Al/K ratios, because Sm and K are more easily be removed from weathering products during extensive weathering relative to La and Al [Nesbitt, 1979; Nesbitt and Markovics, 1997]. However, all these ratios show inverse trends, indicating that chemical weathering intensity in south China decreased continuously since the early Miocene.

[17] Several fluctuations of chemical weathering intensity can also be seen in the overall decreasing trend (Figures 4a and 4b). The major fluctuation occurred from ~17.4 Ma to 14.5 Ma with significantly increased Al/Na and CIA values, coupled with decreased Na/Ti ratios. The CIA value at 15.4 Ma was about 78, which is accompanied with maximum Al/Na and minimum Ca/Ti and Na/Ti values, revealing a second period of high chemical weathering



**Figure 3.** Temporal variations of (top) Th/Sc and (bottom)  $\epsilon_{\text{Nd}}(0)$  of the sediments at ODP Site 1148 since the early Miocene. The  $\epsilon_{\text{Nd}}(0)$  data are from Li et al. [2003]. Shaded areas represent the ranges for the south provenance and north provenance for the sediments at the South China Sea [Li et al., 2003].



**Figure 4.** Temporal variations of the selected element ratios, CIA values, and MAR: (a) Ca, Na, Al to Ti ratios and (b) La/Sm and Al/K ratios, MAR, and CIA values. Shaded bar and the vertical lines indicate the layers with significant variations of some of the element ratios. Numbers indicate the corresponding ages.

intensity in south China after 23 Ma. The chemical weathering intensity decreased rapidly after 15.4 Ma. The third high CIA value of about 75 occurred at 8.4 Ma. There was a rapid decrease of the chemical weathering intensity centering at  $\sim 7.2$  Ma with a significant jump of the Ca/Ti, Na/Ti, Al/Na and CIA values. Another fluctuation of the chemical weathering intensity occurred from 4.0 Ma to 2.3 Ma with a peak of Al/Ti and Al/K ratios at 4.0 Ma. Increase of La/Sm ratio was also shown during this period, which however occurred at  $\sim 3.2$  Ma. No fluctuation of Ca/Ti and Na/Ti ratios was shown during this period.

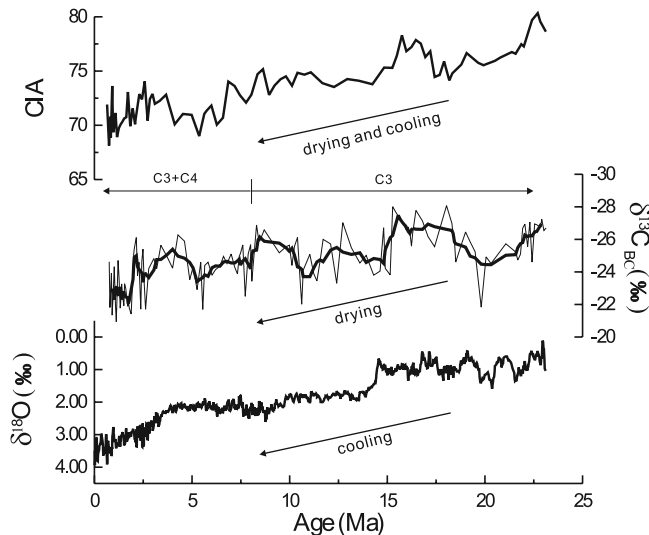
[18] MAR varied from 1.5 to  $8.0 \text{ gcm}^2/\text{kyr}$  during the period from  $\sim 23$  Ma to  $\sim 4$  Ma (455 mcd to 160 mcd) without any trend of increasing or decreasing (Figure 4b). As detrital component accounts for about 50 to 90 percent of these sediments, MAR variation are mainly resulted from changes of terrestrial input, which is related to physical erosion in source regions. However, MAR increased significantly after 3.2 Ma, from  $\sim 2 \text{ gcm}^2/\text{kyr}$  at 3.2 Ma to  $\sim 16 \text{ gcm}^2/\text{kyr}$  at 0.8 Ma (Figure 4b). The significant MAR increase corresponds to different variation patterns of the elemental ratios. Ca/Ti and Na/Ti ratios show increasing trend, accompanying with decreasing trend for Al/Na, Al/K, La/Sm ratios and CIA (Figures 4a and 4b). This seems to suggest a decrease of chemical weathering intensity. However, Al/Ti ratios show an

increasing trend (Figure 4a), implying enhanced chemical weathering intensity in south China since 3.2 Ma. Significant increase of MAR in sedimentary basins after 2–4 Ma are well recorded in global scale, and enhanced erosion induced by climate change is supposed to account for it [Zhang *et al.*, 2001]. Particularly, in the SCS, higher MAR is believed to correspond to stronger chemical weathering since the Miocene [Clift, 2006]. If this is true, the Al/Ti ratio provides the reliable information about chemical weathering in south China, while other elemental ratios were misread since 3.2 Ma. Such conflict between Al/Ti and other ratios may be attributed from significant increasing of MAR. Significant MAR increasing indicates enhanced physical erosion, which may transfer some detrital materials that has not been well weathered to the basin. As a result, the elemental ratios show contradict variation pattern. Despite of the high MAR period since 3.2 Ma, the elemental ratios show similar trends from  $\sim 23$  Ma to  $\sim 4$  Ma, indicating consecutive decreasing of chemical weathering intensity in south China during this period.

#### 4. Implications for Climate Changes in South China

[19] Climate and tectonics as well as other factors are important in controlling terrestrial weathering [Berner and





**Figure 5.** Secular variations of the CIA values, black carbon  $\delta^{13}\text{C}$ , and benthic foraminifer  $\delta^{18}\text{O}$  of the sediments at ODP Site 1148. The axis for  $\delta^{13}\text{C}$  and  $\delta^{18}\text{O}$  are in descending order. The black carbon  $\delta^{13}\text{C}$  ratios were from *Jia et al.* [2003]. The black carbon  $\delta^{13}\text{C}$  records during the last 0.5 Myr of *Jia et al.* [2003], which are from another core at ODP Site 1147, are not included in this diagram. The thin lines show the variations of the black carbon  $\delta^{13}\text{C}$ , and the bold lines are 5-point running averages. The time during which C4 plants flourished is also marked in Figure 5. The benthic foraminifer  $\delta^{18}\text{O}$  ratios refer to *Zhao et al.* [2001], and the lines for the  $\delta^{18}\text{O}$  ratios are 5-point running averages.

*Berner, 1997*]. Chemical weathering intensity is largely controlled by temperature and precipitation [*White and Blum, 1995*] [*Berner and Berner, 1997*]. High precipitation and warm temperatures can enhance the chemical weathering intensity, whereas either low temperature or precipitation can decrease the chemical weathering intensity [*White and Blum, 1995*].

[20] The south China, especially the drainage of the Pearl River and other rivers in the coast regions, as well as the offshore islands, such as Taiwan and Hainan, make up the source region for the sediments in the northern South China Sea [*Li et al., 2003*]. The high chemical weathering intensity at  $\sim 23$  Ma (Figure 4) indicates a warm and humid climate in south China in the early Miocene. This is supported by pollen records in this region. For example, several pollen records from middle and north Taiwan reveal abundant *Quercoidites*, common *Ulmaceae*, *Juglandaceae* and *Taxodiaceapollenites*, and scattered *Alnipollenites* in the early Miocene, suggesting a warm and damp climate [*Lei and Huang, 1997*]. Consistently, pollen records in Guilin, Guangxi Province [*Liu and Yang, 1997*], and in the east offshore of Hainan Island [*Li and Zhang, 1998*] all indicate a warm and humid climate in the early Miocene.

[21] The consecutive decreasing of chemical weathering intensity suggested by the elemental records indicates a trend of decreasing temperature or precipitation, or both, in

south China since the early Miocene. Decreasing temperature is in agreement with global climate cooling since the middle Miocene [*Zachos et al., 2001*], and this cooling trend is well exhibited in the benthic foraminifer  $\delta^{18}\text{O}$  ratios at ODP Site 1148 [*Zhao et al., 2001*]. Consistently, in south China, the pollen records all indicate such cooling trend [*Lei and Huang, 1997*; *Li and Zhang, 1998*; *Liu and Yang, 1997*]. Figure 5 shows the variations of the CIA values, black carbon  $\delta^{13}\text{C}$ , and benthic foraminifers  $\delta^{18}\text{O}$  at ODP Site 1148. The CIA decreasing agrees well with the cooling revealed by the variations of the benthic foraminifer  $\delta^{18}\text{O}$  ratios [*Zhao et al., 2001*] (Figure 5). Considering that decreasing temperature may result in decrease of chemical weathering intensity [*White and Blum, 1995*], the cooling from the middle Miocene may have contributed partly to the decrease of the chemical weathering intensity in south China.

[22] In addition to cooling, the humidity decline is also very important to the decreases of chemical weathering intensity. As revealed in Figure 5, the variation pattern of the CIA values is very similar to the black carbon  $\delta^{13}\text{C}$ , which shows an increasing trend from the early Miocene to Present. Also, the fluctuations of the CIA values observed for ca. 17–15 Ma, ca. 10–8 Ma, and 4–2 Ma, can be correlated to the low values of the black carbon  $\delta^{13}\text{C}$  ratios during these periods (Figure 5). These suggest that the changes of the chemical weathering intensity and the black carbon  $\delta^{13}\text{C}$  may have been driven by the same process. The change of the black carbon  $\delta^{13}\text{C}$  indicates secular adjustment of the ecosystem in south China in response to climate change, such as change of C3 plants and flourishing of C4 plants. The flourishing of C4 plants in the ecosystem in south China is believed to be in the late Miocene [*Jia et al., 2003*]. Thus from the early Miocene to the late Miocene, the gradual increase of the black carbon  $\delta^{13}\text{C}$  indicates secular change of the  $\delta^{13}\text{C}$  for C3 plants. Because C3 plants tend to increase their  $\delta^{13}\text{C}$  values in response to decreasing moisture [*Conte and Weber, 2002*], the increasing  $\delta^{13}\text{C}$  in the black carbon suggests decreasing humidity in south China from the early to late Miocene. After the late Miocene, the black carbon  $\delta^{13}\text{C}$  increase may indicate sustaining expansion of C4 plants in south China because C4 plants have much positive  $\delta^{13}\text{C}$  than C3 plants. The expansion of the C4 plants is closely related to  $\text{pCO}_2$  in atmosphere and climate [*Ehleringer et al., 1997*]. Beneath a threshold of atmospheric  $\text{pCO}_2$ , C4 expansion is largely controlled by moisture and temperature, with warm and arid climates favoring C4 over C3 plants [*Huang et al., 2001*]. Atmospheric  $\text{pCO}_2$  has been within the range that enables the expansion of C4 plants since the late Miocene [*Pagani et al., 1999*; *Pearson and Palmer, 2000*], whereas the temperature decreased [*Zachos et al., 2001*], which did not favor the expansion of C4 plants. Hence the C4 expansion [*Jia et al., 2003*] may have been induced by the climate factors other than temperature, i.e., decreasing humidity, which favors the expansion of C4 plants [*Huang et al., 2001*]. Therefore the increases of black carbon  $\delta^{13}\text{C}$ , together with decreases of chemical weathering intensity recorded by the elemental ratios indicate that humidity in



south China has decreased continuously since the early Miocene.

## 5. Implications for the Evolution of East Asian Monsoon

[23] Current precipitation in south China is controlled by east Asian monsoon, with moisture mainly transported from the nearby seas and oceans during summer [Ding, 1994]. Therefore the humidity change in south China is closely related to the evolution of the east Asian summer monsoon. Our elemental records, suggesting a warm and humid climate in south China in the early Miocene, support the idea that summer monsoon had significantly influenced the climate in south China as early as that time [Liu et al., 1998; Sun and Wang, 2005]. If, as we believe, humidity in the early Miocene was high, then the east Asian summer monsoon must have started before the early Miocene. The southeast summer monsoon, which is believed to have begun in the Oligocene [Liu et al., 1998; Sun and Wang, 2005], may have played a key role during this period. Consequently, the decreasing humidity indicates that the influence of summer monsoon on south China has been weakened consecutively since the early Miocene.

[24] Such evolution of the east Asian monsoon is different from that of the Indian monsoon system. Clay minerals and Sr isotopes in the sediments from Bengal Fan indicate that chemical weathering in Himalayan region was significantly enhanced in  $\sim 7$  Ma, whereas the fluctuation of weathering from  $\sim 20$  Ma to  $\sim 7$  Ma was relatively small [Derry and FranceLanord, 1996]. Some other geochemical records also suggest enhancement of chemical weathering in Himalayan region triggered by intensification of monsoon at  $\sim 8$  Ma [Filippelli, 1997]. This time seems concordant with the onset of the Indian monsoon [Quade et al., 1989]. However, the chemical weathering record in the sediments at ODP Site 1148, as well as other paleoclimate records in south China [Liu et al., 1998; Sun and Wang, 2005] indicate a early onset time, before the early Miocene, for the east Asian summer monsoon and consecutive decrease of the summer monsoon impact on south China since the early Miocene. Such difference suggests that the evolution of the east Asian monsoon prevailing in the east Asia and the Indian monsoon prevailing in the south Asia are different. Modern meteorological studies suggest that the Indian monsoon, which prevails in the region to the west of  $105^\circ\text{E}$ , and the east Asian monsoon, which prevails in the region to the east of  $105^\circ\text{E}$ , are different climate systems, and the interannual variation of the two systems are different because of different land-ocean configurations in these regions [Wang et al., 2003]. Considered that the evolutions of the land-ocean distribution in the east Asia and the south Asia are significantly different in Neogene [Hall, 2002], if the land-ocean distribution is essential for formation of monsoon system [Ramstein et al., 1997], the onset and the evolution of the two monsoon systems may not be necessarily the same. However, long and continuous paleoclimate records up to the early Miocene are scarce for both monsoon systems, such questions remain opening for further discussion.

[25] The mechanism for the consecutive decrease of the summer monsoon influence in south China as recorded by the chemical weathering variation is still not well known yet. Gradually intensifying of the east Asian winter monsoon may be one of the reasons, because winter monsoon is generally cold and dry, intensification of winter monsoon can result in less precipitation and low humidity in south China. In timescale of glacial/interglacial cycles, the variation of chemical weathering intensity in south China seem to be closely related to changes in winter monsoon [Wei et al., 2004]. The onset of the winter monsoon might be before 22 Ma according to the aridity records in North China [Guo et al., 2002]. However, the significant influence of the winter monsoon in south China seems late because the appearance of a large amount of black carbon occurred only after the late Miocene ( $\sim 7$  Ma) [Jia et al., 2003]. As variations of the east Asian winter monsoon from the early Miocene to present are not well known, more evidence is needed to determine whether the decreasing chemical weathering reflect gradual increases of the influence of the winter monsoon on south China.

## 6. Summary

[26] Element ratios that are sensitive to terrestrial chemical weathering, such as Ca/Ti, Na/Ti, Al/Ti, Al/Na, Al/K, La/Sm and CIA values of the detrital sediments at ODP Site 1148 were reassessed to reveal the chemical weathering intensity in the source regions since the early Miocene. This provides useful information about climate changes in the south China since that time.

[27] 1. The elemental ratios indicate a high chemical weathering intensity in south China in the early Miocene, indicating a warm and humid climate. The chemical weathering intensity in south China has decreased gradually since the early Miocene, indicating a decreasing of humidity accompanying with the global cooling since that time.

[28] 2. The changes of humidity implied from the elemental ratios indicate that the influence of summer monsoon on south China was significant in the early Miocene, supporting the hypothesis that the onset of the east Asian summer monsoon occurred before the early Miocene. The decreasing humidity indicates that the influence of summer monsoon on the climate in south China decrease gradually since the early Miocene. Such evolution for the east Asian monsoon is different from that of the Indian monsoon indicated by the chemical weathering records in sediments from the Indian Ocean.

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