



Geochemical characteristics of modern river sediments in Myanmar and Thailand: Implications for provenance and weathering



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ABSTRACT

The elemental composition of organic matter and the major and trace element compositions of stream sediments from Myanmar (Ayeyarwady and Sittaung rivers) and Thailand (Mekong and Chao Phraya rivers, and their tributaries) were determined to examine their distributions, provenance, and chemical weathering processes. Higher total organic carbon (TOC) and total nitrogen (TN) contents in the finer grained sediments indicate hydrodynamic energy may control their distributions. TOC/TN ratios indicate inputs of both aquatic macrophyte and higher vascular plant material to the river sediments. The major element abundances of the sediments are characterized by predominance of SiO₂ in coarser fractions and a marked negative correlation with Al₂O₃, representing primary grain size primarily control on SiO₂ content. Marked depletion of most labile elements (Na₂O, CaO, K₂O, Ba and Sr) relative to UCC (upper continental crust), indicate destruction of feldspar during chemical weathering in the source area or during transport. However, enrichment of some high field strength elements (Zr, Th, Ce and Y) relative to UCC and higher Zr/Sc ratios indicate moderate concentration of resistant heavy minerals in finer-grained samples. Discriminant diagrams and immobile trace element characteristics indicate that the Mekong, and Chao Phraya river sediments were largely derived from felsic sources with compositions close to typical rhyolite, dacite/granodiorite, UCC, I- and S-type granites. Relative enrichment of ferromagnesian elements (e.g. MgO, Cr, Ni) and high Cr/V and low Y/Ni ratios in Ayeyarwady and Sittaung sediments indicate the presence of a mafic or ultramafic component in their sources. The ICV (Index of Compositional Variability), CIA (Chemical Index of Alteration), PIA (Plagioclase Index of Alteration), α^{Al} , Rb/Sr and K₂O/Rb ratios indicate that the Ayeyarwady and Sittaung sediments record low to moderate degrees of chemical weathering in their source, compared to moderate to intense chemical weathering in the Mekong and Chao Phraya river basins. These results are compatible with existing major ion data for river waters collected at the same locations.

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1. Introduction

The Himalayan-Tibetan orogenic system has been developed by the collision of the Indian and Eurasian plates. This orogen has attracted the interest of geoscientists over the last few decades, owing to the large-scale landscape changes consequently produced in the climate patterns in Asian countries. The Himalayan-Tibetan orogenic belt forms the source of several large rivers in south

and southeast Asia, including the Ganges, Brahmaputra, Meghna, Ayeyarwady, Sittaung, Salween, Mekong, Chao Phraya, Red, and Pearl rivers (Fig. 1a). The Himalayan drainage complexes developed during rapid uplift and erosion of the Himalayan orogen, and transported melt waters and voluminous clastic detritus from the highlands through to the oceans (Milliman and Meade, 1983; Sarin et al., 1989; Gaillardet et al., 1999; Meybeck and Ragu, 2012; Bickle et al., 2015; Garzanti et al., 2016). These major rivers deliver approximately 10% of the total global riverine flux to the oceans (Chapman et al., 2015). The sum of global fluxes of riverine particulate organic carbon (POC) and dissolved organic carbon (DOC) together transported to the ocean is approximately 334.5–430 MtC yr⁻¹ (Schlünz and Schneider, 2000; Bird et al., 2008). Bird et al. (2008) also doc-

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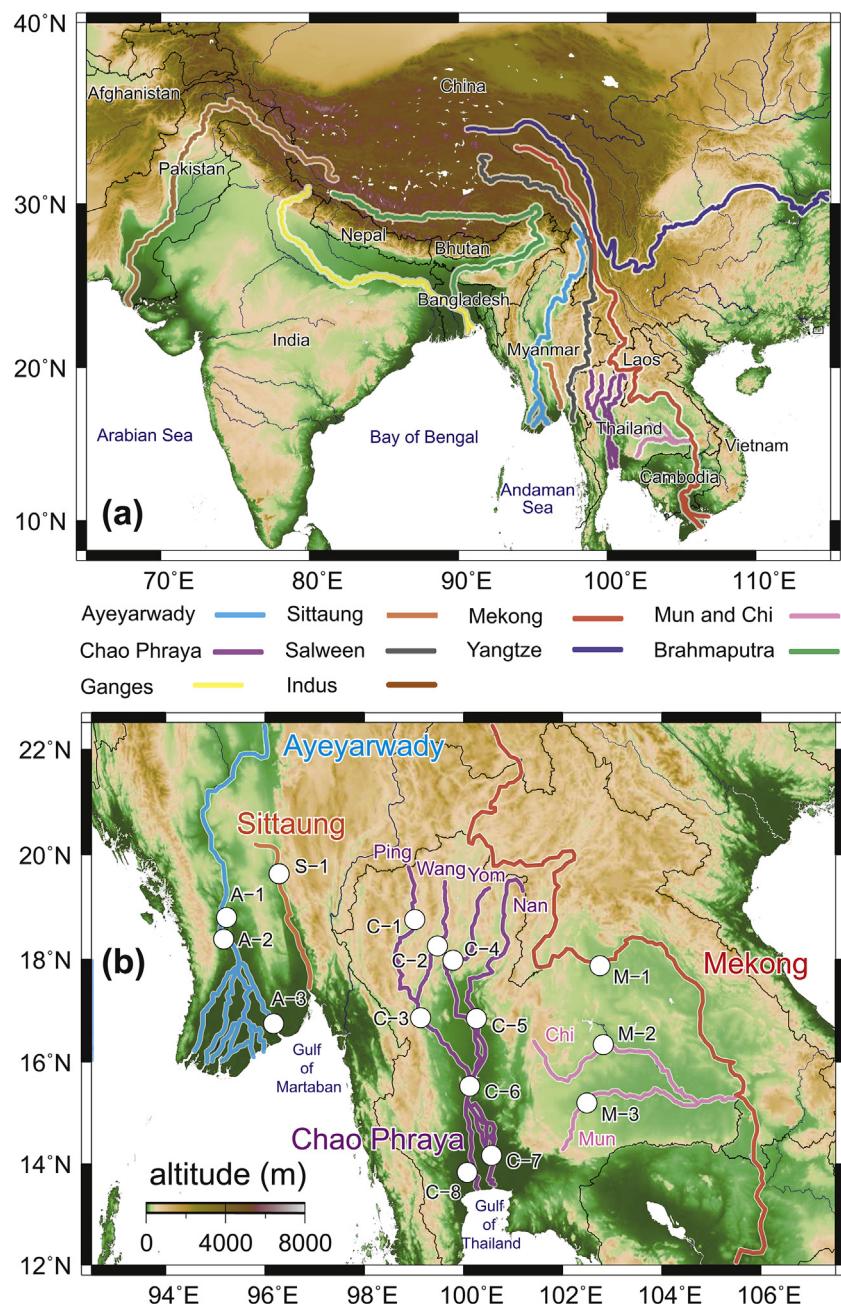


Fig. 1. (a) Locations of the major Himalayan rivers drainage basin (after Manaka et al., 2015), and (b) Location map of the studied rivers in Myanmar and Thailand, showing sampling sites (after Manaka et al., 2015).

umented that the Ayeyarwady River transports 2.2–4.3 MtC yr⁻¹ POC and ~0.9 MtC yr⁻¹ DOC, respectively to the Gulf of Martaban towards the Andaman Sea. In this study we have explored several major rivers (Ayeyarwady, Sittaung, Mekong, and Chao Phraya) and their main tributaries rising in the eastern Himalayan syntaxis and draining present-day Myanmar and Thailand (Fig. 1b). These rivers carry a large volume of chemical weathering flux and organic carbon flux to the Gulf of Martaban and the Gulf of Thailand, respectively. Moreover, the fluvial systems of these river basins represent varied climate conditions, elevations, landscape patterns, precipitation, and physical and chemical denudation, all of which are key controlling factors for input of terrigenous sediments and organic carbon to the oceanic environment.

The geochemical composition of clastic sediments depends on source rock type, relief, transportation, climate, weathering and

diagenesis, among other factors (Pettijohn et al., 1972; McLennan et al., 1993; Cox et al., 1995). The major and trace element compositions of detrital sediments are widely used to evaluate sediment type, maturity, chemical weathering intensity, sorting, recycling, and provenance (Nesbitt and Young, 1982, 1984; Taylor and McLennan, 1985; Wronkiewicz and Condé, 1987; Cutters et al., 1988; Herron, 1988; Roser and Korsch, 1988; Condé, 1993; McLennan et al., 1993; Cox et al., 1995; Fedo et al., 1995; Gaillardet et al., 1999; Selvaraj and Chen, 2006; Borges et al., 2008; Li and Yang, 2010; Lupker et al., 2012; Garzanti et al., 2013b, 2016; Awasthi et al., 2014; Garzanti and Resentini, 2016). Several authors have investigated riverine flux in the Ayeyarwady, Sittaung, Mekong, and Chao Phraya rivers from differing viewpoints (Gaillardet et al., 1999; Nozaki et al., 2000; Liu et al., 2007; Robinson et al., 2007, 2014; Bird et al., 2008; Borges et al., 2008; Furuichi et al., 2009; Gupta et al.,

2012; Licht et al., 2014; Chapman et al., 2015; Manaka et al., 2015; Garzanti et al., 2016). However, with the exceptions of Liu et al. (2007), Borges et al. (2008) and Garzanti et al. (2016), none of these studies utilize major and trace element compositions of the river sediments to examine sediment type, maturity, chemical weathering intensity and provenance. Liu et al. (2007) described only the chemical weathering of Mekong sediments, based on major element geochemistry, clay mineralogy, and Sr-Nd isotopes, whereas Borges et al. (2008) documented the petrographic and geochemical compositions of Mekong riverbed sediments draining the eastern Tibetan Plateau. Recently, studies of provenance and sediment fluxes in the Ayeyarwady River have been conducted by Garzanti et al. (2016). However, as yet no major and trace element geochemical studies have been made of Sittaung and Chao Phraya river sediments in Myanmar and Thailand. Distribution of total organic carbon (TOC), total nitrogen (TN) and ratios (TOC/TN) of the Ayeyarwady, Sittaung, Mekong, and Chao Phraya river systems are still limited (Bird et al., 2008; Ramaswamy et al., 2008). In this paper we use both major and trace element data for sediments from the Ayeyarwady and Sittaung rivers in Myanmar, and the Mekong and Chao Phraya and their main tributaries in Thailand to infer maturity, source area weathering, recycling and provenance in these river basins. A combination of major ions in these river waters and weathering proxies of the river sediments in the same region are presented to investigate ion exchange from parent-rock lithologies in the watersheds, which are a crucial aspect of global climate changes largely influenced by the Himalaya-Tibetan Plateau uplift. We also discuss TOC and TN concentrations, and TOC/TN ratios for the distribution and sources of organic matter in the sediments.

2. Geological setting

The overall geography, morphology, drainage areas, climate, water discharge, and suspended sediment loads for the major rivers studied in Myanmar and Thailand are summarized in Table 1.

2.1. The Ayeyarwady and Sittaung drainage basins

The Ayeyarwady River is the longest river in Myanmar (~2300 km long). It originates at the confluence of the Nmai and Mali rivers in Kachin State; these flow from the eastern syntaxis of the Himalayan-Tibetan Plateau system (Bird et al., 2008; Chapman et al., 2015). This river flows through Myitkyina in its upper reaches and Mandalay (central Myanmar) before meeting the Chindwin River, and finally discharges its water and sediment flux into the Gulf of Martaban (Fig. 1b). The headwaters of the Ayeyarwady River are still undergoing some of the highest rates of landscape adjustment in an area of active orogenesis (Robinson et al., 2007). The Nmai and Mali rivers rise in the Himalayan glaciers at about 28° N latitude, and flow over the Mogok Metamorphic Belt and Lohit Plutonic Complex, respectively. The Mogok Metamorphic Belt consists mainly of gneisses, granitoids, garnet and chlorite schists (Allen et al., 2008; Garzanti et al., 2016), whereas quartz-diorites, diorites, gabbro, gneiss and associated intrusive rocks are extensively exposed in the Lohit Plutonic Complex (Gururajan and Choudhuri, 2003; Garzanti et al., 2016). The Chindwin River rises in the Hukaung valley of Kachin State of Myanmar, and mainly drains through basement gneisses and arc-related volcanic rocks (Garzanti et al., 2016). The main rock types in the Ayeyarwady River catchment are Cretaceous to Middle Cenozoic flysch of the western Indo-Burman Ranges, Eocene to Miocene and Quaternary sediments of the Myanmar Central Basin, and Late Precambrian and Cretaceous to Eocene metamorphic, basic and ultrabasic rocks of the eastern syntaxis (Robinson et al., 2007; Bird et al., 2008; Awasthi et al., 2014; Chapman et al., 2015). The Ayeyarwady River

also drains Holocene lavas near Mandalay (Chapman et al., 2015). Climatic patterns in the Ayeyarwady River basin range from warm sub-tropical conditions in the upstream regions to humid tropical in the lower regions. These contrasts are largely influenced by South Asian monsoon circulation. Water discharge in the Ayeyarwady River is significantly greater (~89 km³/month) during the summer monsoon than in the winter monsoon (~12 km³/month) (Chapman et al., 2015).

The Sittaung River is located in south central Myanmar in Bago Division. The Sittaung originates from the edge of the Shan Plateau near Naypyitaw, and flows 420 km southward into the Gulf of Martaban. This river drains over slate and metamorphic belts of the Shan Plateau (Awasthi et al., 2014). The Sittaung River basin mainly has a tropical monsoon climate, and experiences heavy seasonal precipitation and high temperature during the summer monsoon period.

2.2. The Mekong and Chao Phraya drainage basins

The Mekong River is one of the longest rivers in southeast Asia (~4650 km long), rising in the Himalaya-Tibetan Plateau system and flowing southward through Myanmar, Thailand, Laos, Cambodia and Vietnam, before discharging into the South China Sea. The Mekong basin drainage area in Thailand (the Indochina terrain) is relatively flat land, but its upper reaches contain deep and narrow valleys (Liu et al., 2007; Manaka et al., 2015). The main rock types in the Mekong River and its tributary catchments (Mun and Chi rivers) are Paleozoic-Mesozoic sedimentary rocks (meta-sandstone, shale, slate, and phyllite), lesser intrusive igneous rocks and Precambrian metamorphic rocks in its upper and middle reaches, and alluvium with Triassic sedimentary rocks (typically sandstone and mudstone) and Neogene basalts in its lower reaches (Liu et al., 2007; Borges et al., 2008; Li et al., 2014). The Mekong River drainage basin is largely influenced by subtropical Asian monsoon climate, with precipitation ranging between ~1700 and 2000 mm during the summer months (Liu et al., 2007; Borges et al., 2008; Gupta et al., 2012). The headwaters of the Mekong River have humid and cold conditions during the summer monsoon, with average air temperature close to 1 °C (Borges et al., 2008), whereas the lower reaches have higher and mostly uniform temperature of 26–30 °C (Liu et al., 2007).

The Chao Phraya River originates at the confluence of the Ping, Wang, Yom and Nan rivers in Nakhon Sawan province, and runs southward over an alluvial plain to Bangkok, before finally feeding into the Gulf of Thailand (Fig. 1b). The Ping, Wang, Yom and Nan tributaries flow from the mountainous northwestern part of Thailand, and form part of the extensive Chao Phraya watershed, which has an area of approximately 1.1×10^5 km² (Meybeck and Ragu, 2012). The Chao Phraya basin is broadly known as the Shan-Thai terrain, and the geomorphologic features of this terrain are high mountains, a central plain, and an upland plateau. The Shan-Thai terrain consists of Proterozoic-Cretaceous sedimentary rocks on schist basement (Allen et al., 2008). The central plain is characterized by lowlands where the main stream of the Chao Phraya flows, and the region is largely influenced by wet monsoon climate. The catchment rock types include Paleozoic-Mesozoic metamorphic rocks and limestone (Manaka et al., 2015).

3. Materials and methods

Riverbank sediment samples ($n=15$) were collected from the Ayeyarwady and Sittaung rivers in Myanmar, and from the Mekong (Mun and Chi rivers) and Chao Phraya (Yom, Ping, Wang and Nan) river systems in Thailand between 12 and 21 July 2013 (Fig. 1b). Three samples (A-1 to A-3) were collected from the middle to lower

Table 1

Geographic, climatic, topographic, water discharge, and suspended sediment loads for the studied Myanmar and Thailand rivers (after Robinson et al., 2007; Ramaswamy et al., 2008; Li and Yang, 2010; Gupta et al., 2012; Meybeck and Ragu, 2012).

Rivers	Drainage basin area (10^3 km^2)	River length (km)	Source elevation (m)	Average air temperature ($^\circ\text{C}$)	Mean annual precipitation (mm/yr)	Water discharge (km^3/yr)	Sediment load (Mt/yr)
Ayeyarwady	414	2300	147	24	1970	486	260
Sittaung	48	420	115	27	2342	47	27–43
Mekong	795	4650	5224	21	1590	467	150
Chao Phraya	160	1200	25	29	1200	28	11

reaches of the Ayeyarwady River, and one sample (S-1) from the upper reaches of the Sittaung River. In the Mekong River watershed, one sample (M-1) was collected from middle part of the Mekong River main stream (~1500 km upstream from the river mouth), and two samples (M-2 and M-3) from the major tributaries Chi and Mun rivers, respectively (Fig. 1b). In the Chao Phraya watershed, five samples (C-1 to C-5) were collected from the upper reaches of major tributaries of the Chao Phraya River (Yom, Ping, Wang and Nan rivers), one sample (C-6) from the main stream in the middle part, and two samples (C-7 and C-8) from the lower main channel of the Chao Phraya. These large drainage basins are of geochemical interest because the influx of sediment and organic carbon yields from orogenic belts to the oceans is significant. These influxes are largely influenced by orogenic uplift and variable climatic conditions. We selected these sampling points to examine the variability of source materials input, weathering intensity, and organic carbon transport through large rivers.

Grain size analysis of the bulk river sediments was conducted at Kochi University, using a Mastersizer 2000 (Malvern) laser diffraction particle size analyzer. This instrument can simultaneously measure a range of grain size from 0.02 to 2000 μm . Organic matter was removed prior to particle size analysis using 30% hydrogen peroxide treatment, and clastic grains larger than 2 mm were removed by sieving. The samples were then treated with sodium hexametaphosphate solution to fully disperse the fine sediment fractions. Mean grain size (μm), sorting (σ_g), skewness (Sk_g) and Kurtosis (K_g) were estimated based on the geometric method of moments (Folk and Ward, 1957).

For the organic geochemical analysis, bulk samples were oven-dried at $<60^\circ\text{C}$ for more than two days, and then homogenized by grinding in an agate mortar and pestle to a fine powder. The TOC and TN contents were determined using a CHNS-O Elemental Analyzer (FISSION EA 1108) at Shimane University. Subsamples (ca. 10 mg) were acid-treated (1 M HCl) in Ag cups and dried to remove the carbonate fraction for the measurement of TOC content. The measured TOC and TN concentrations were calibrated against a BBOT standard [2,5-bis-(5-tert-butyl-benzoxazol-2-yl)-thiopene], and the analytical relative error (coefficient of variation) was $\pm 3\%$.

Prior to the inorganic geochemical analysis, powdered samples were dried at 110°C for more than 24 h. Loss on ignition (LOI) was measured by net weight loss after ignition in a muffle furnace at 950°C for more than 3 h. The concentrations of the major elements and 14 trace elements (Ba, Ce, Cr, Ga, Nb, Ni, Pb, Rb, Sc, Sr, Th, V, Y and Zr) were determined using a Rigaku RIX 2000 X-ray fluorescence spectrometer at Shimane University. Glass fusion beads were prepared with an alkali flux (80% lithium tetraborate ($\text{Li}_2\text{B}_4\text{O}_7$) and 20% lithium metaborate (LiBO_2)), in a sample to flux ratio of 1:2, following the methodology of Kimura and Yamada (1996). The accuracy of the major and trace element data were verified by analysis of ten Geological Survey of Japan (GSJ) standard rocks at run time.

To evaluate mineralogical maturity of sediments we use the Index of Compositional Variability, $ICV = (\text{Fe}_2\text{O}_3^* + \text{K}_2\text{O} + \text{Na}_2\text{O} + \text{CaO} + \text{MgO} + \text{TiO}_2)/\text{Al}_2\text{O}_3$ (Cox et al., 1995). However, weathering intensities can also be calculated using the Chemical Index of Alteration (CIA; Nesbitt and Young,

1982, 1984) and the Plagioclase Index of Alteration (PIA; Fedo et al., 1995). The degree of alteration of feldspar to clay minerals is calculated using molar proportions, from the formula $CIA = 100 \times [\text{Al}_2\text{O}_3]/(\text{Al}_2\text{O}_3 + \text{CaO}^* + \text{Na}_2\text{O} + \text{K}_2\text{O})]$, where CaO^* represents the Ca incorporated only in the silicate fraction. However, due to the lack of CO_2 data in our present study, correction has been made here only for Ca present in apatite (Fedo et al., 1995). PIA is also estimated using molecular proportions, using the formula $PIA = 100 \times [(\text{Al}_2\text{O}_3 - \text{K}_2\text{O})]/(\text{Al}_2\text{O}_3 + \text{CaO}^* + \text{Na}_2\text{O} - \text{K}_2\text{O})]$, where CaO^* is the amount of CaO contained in the silicate fraction only. Unweathered plagioclase feldspar has PIA values of 50, whereas aluminous clay minerals such as kaolinite, illite, and gibbsite have maximum PIA values of 100.

Weathering intensities can also be estimated for each mobile element during different weathering of silicates (Na, Ca, Sr, Mg, K, Ba, and Rb) by comparing its abundance to that of an immobile element (e.g. Al) with comparable magmatic compatibility in the analyzed sample and in average upper continental crust (Gaillardet et al., 1999; Garzanti et al., 2013b). The α^{Al} value for a given element E is calculated as $\alpha^{\text{Al}}\text{E} = [\text{Al}/\text{E}]_{\text{sample}}/[\text{Al}/\text{E}]_{\text{UCC}}$ (Garzanti et al., 2013b).

4. Results

TOC (wt.%), TN (wt.%), major and trace element (anhydrous normalized basis) concentrations of the Ayeyarwady, Sittaung, Mekong, Mun, Chi, and Chao Phraya river sediments are presented in Table 2. LOI values range between 0.41 and 8.23 wt.%. The major element data were normalized to 100% LOI-free for all geochemical plots and comparisons. The same normalization factors were also applied to the trace element data.

4.1. Variation in grain sizes

The mean grain sizes of the bulk sediment samples from the Myanmar and Thailand rivers are listed in Table 2. The samples range from well sorted to poorly sorted, and comprise clayey medium silts to coarse sand, with mean grain size varying from 62 to 196 μm in the Ayeyarwady, 57–192 μm in Mekong, and 10–47 μm in most Chao Phraya samples (Table 2), with differing volume percentages of sand, silt and clay. The single Sittaung River sediment has a mean grain size of 32 μm , while two samples from Chao Phraya tributaries (Ping, C-3; Wang, C-2) are coarser grained, with mean values of 634 and 283 μm , respectively.

4.2. TOC and TN contents

TOC and TN abundances in the river sediments range from 0.04 to 1.47 wt.% (average 0.62 wt.%) and 0.003–0.14 wt.% (average 0.06 wt.%), respectively (Table 2). TOC and TN contents are relatively high in the Chao Phraya River and its tributaries. Three (C-1, C-4, C-8) of the four Chao Phraya very coarse silt samples analyzed have high TOC (1.18–1.47 wt.%) and TN (0.12–0.14 wt.%) contents, whereas only one medium sand (C-2) has comparable contents (1.44 wt.% TOC, 0.14 wt.% N). Abundances in the Mekong

Table 2

Grain size parameters, total organic carbon (TOC, wt.%), total nitrogen (TN, wt.%), major element (wt.%) and trace element (ppm) compositions (anhydrous normalized) for the Myanmar and Thailand river sediments.

Locality	Ayeyarwady and Sittaung River System, Myanmar				Mekong River System, Thailand			Chao Phraya River System, Thailand							
	A-1	A-2	A-3	S-2	M-1	M-2 (Chi)	M-3 (Mun)	C-1 (Ping)	C-2 (Wang)	C-3 (Ping)	C-4 (Yom)	C-5 (Nan)	C-6	C-7	C-8
Latitude ($^{\circ}$ N)	18.82	18.82	21.13	18.93	17.89	16.36	15.22	18.78	18.27	16.88	18.00	16.87	15.55	14.19	13.95
Longitude ($^{\circ}$ E)	95.21	95.21	94.85	96.46	102.74	102.80	102.47	99.00	99.46	99.12	99.77	100.25	100.11	100.55	99.75
Grain size	VCSi	FS	FS	CSi	VCSi	VFS	FS	VCSi	MS	CS	VCSi	CSi	VCSi	MSi	VCSi
Mean grain size (μm)	62	196	129	32	57	78	192	35	283	634	39	27	47	10	39
Sorting (σ_g)	2.54	1.78	1.57	2.76	2.99	1.97	3.67	3.16	3.32	1.31	3.15	3.58	4.00	2.49	3.14
Skewness (Sk_g)	0.63	0.84	0.90	0.001	0.57	0.69	0.33	0.92	0.24	1.00	1.35	1.14	1.01	0.90	1.26
Kurtosis (K_g)	0.28	0.25	0.24	0.20	0.27	0.30	0.31	0.28	0.33	0.26	0.16	0.26	0.22	0.23	0.16
Clay (%)	10	2	2	20	12	8	11	18	9	0	27	26	23	38	24
Silt (%)	52	14	19	66	55	38	24	62	19	0	58	59	52	61	59
Sand (%)	39	84	79	14	33	54	65	20	72	100	14	15	25	1	17
TOC (wt.%)	0.46	0.84	0.19	0.68	0.21	0.11	0.19	1.47	1.44	0.19	1.18	0.04	0.52	0.29	1.44
TN (wt.%)	0.04	0.06	0.02	0.06	0.01	0.003	0.01	0.12	0.14	0.02	0.13	-	0.05	0.03	0.14
TOC/TN	12.48	15.15	12.46	11.56	22.41	38.97	16.24	12.76	10.15	12.03	9.09	-	10.37	9.62	10.58
TDS (mg/l) ^a	43.9	59.8	60.3	70.9	164.6	288.8	376.8	169.9	279.8	162.9	238.5	124.7	139.6	190.2	n.a.
<i>Major elements (wt.%)</i>															
SiO ₂	69.89	77.64	72.86	64.40	65.53	90.92	80.03	65.25	88.04	85.67	64.70	79.73	76.82	64.58	62.05
TiO ₂	0.68	0.66	0.72	0.76	0.86	0.41	0.49	0.92	0.23	0.07	0.95	0.65	0.50	0.98	0.72
Al ₂ O ₃	13.62	10.05	12.00	16.06	15.56	4.17	9.09	15.51	4.50	7.34	15.82	8.76	11.45	20.48	16.39
Fe ₂ O ₃ T	5.17	3.58	4.45	6.46	6.05	1.48	5.54	5.62	2.89	0.65	6.46	3.35	3.41	4.06	5.82
MnO	0.09	0.06	0.08	0.15	0.16	0.04	0.09	0.25	0.05	0.03	0.23	0.13	0.12	0.01	0.28
MgO	1.85	1.17	1.47	1.69	1.24	0.32	0.38	1.00	0.28	0.24	0.98	0.77	0.61	0.83	1.28
CaO	1.27	1.39	1.87	1.19	0.77	0.16	0.30	0.62	0.52	0.26	0.68	0.60	0.45	0.37	1.05
Na ₂ O	1.33	1.28	1.51	1.05	0.50	0.12	0.02	0.18	0.15	0.68	0.37	0.24	0.36	0.31	0.09
K ₂ O	1.85	1.64	2.11	2.02	2.12	0.24	0.28	2.70	0.78	3.31	2.08	0.87	2.14	1.78	3.06
P ₂ O ₅	0.09	0.06	0.09	0.12	0.14	0.03	0.03	0.17	0.04	0.04	0.15	0.25	0.09	0.03	0.18
LOI	3.81	1.65	2.07	6.02	6.90	1.49	3.78	7.58	1.67	0.41	7.08	3.92	3.94	6.85	8.23
Total	99.66	99.19	99.24	99.92	99.85	99.39	100.05	99.81	99.14	98.69	99.50	99.25	99.88	100.28	99.15
ICV	0.89	0.97	1.01	0.82	0.74	0.65	0.77	0.71	1.08	0.71	0.73	0.74	0.65	0.41	0.73
CIA	68.5	61.6	60.1	73.5	78.7	86.2	91.8	79.7	69.7	59.1	81.0	82.9	76.5	87.1	76.9
PIA	73.1	64.8	63.1	79.3	87.4	90.5	94.6	92.4	76.7	71.5	90.2	90.0	88.3	94.4	89.0
<i>Trace elements (ppm)</i>															
Ba	335	312	379	358	402	88	221	575	201	607	1904	185	437	229	535
Ce	62	86	107	85	78	68	81	112	56	24	62	72	66	54	109
Cr	248	400	317	205	73	25	51	88	51	1	72	75	46	87	66
Ga	15	11	13	17	17	4	10	20	4	7	18	10	12	24	22
Nb	12	12	14	14	15	9	11	24	7	5	18	13	13	20	26
Ni	122	60	63	113	34	4	20	49	5	1	36	23	21	29	37
Pb	23	17	25	25	30	9	20	38	18	32	44	16	27	12	61
Rb	92	73	105	116	114	30	45	182	58	173	123	60	139	146	247
Sc	10.8	8.7	7.9	14.9	12.5	2.1	5.3	8.8	2.2	0.5	11.4	6.0	4.4	12.7	9.7
Sr	157	159	189	138	97	26	30	52	33	78	113	53	69	66	43
Th	13	15	22	18	14	7	11	26	10	7	14	10	17	22	28
V	108	81	94	129	128	36	89	115	31	3	131	72	67	126	103
Y	25	21	26	30	33	16	24	40	18	10	33	22	24	22	38
Zr	239	423	528	238	262	452	327	334	269	44	246	400	227	171	235
Th/Sc	1.2	1.7	2.8	1.2	1.1	3.5	2.1	2.9	4.7	15.2	1.3	1.6	3.8	1.8	2.9
Zr/Sc	22.1	48.6	67.2	16.0	20.9	213.6	62.3	38.0	123.2	97.1	21.5	66.4	51.8	13.5	24.1
Cr/V	2.3	4.9	3.4	1.6	0.6	0.7	0.6	0.8	1.6	0.3	0.5	1.0	0.7	0.7	0.6
Y/Ni	0.2	0.3	0.4	0.3	1.0	4.4	1.2	0.8	3.9	10.3	0.9	0.9	1.1	0.7	1.0
Rb/Sr	0.59	0.46	0.55	0.84	1.17	1.16	1.51	3.51	1.76	2.21	1.09	1.14	2.03	2.20	5.76
K ₂ O/Rb	167	186	168	144	155	67	53	123	111	159	141	120	128	101	103

^aBelow detection limit; ^bn.a.not analyzed. LOI – loss on ignition. Grain size abbreviations: CS, coarse sand; MS, medium sand, FS, fine sand; VFS, very fine sand; VCSi, very coarse silt; CSi, coarse silt; MSi, medium silt.^a TDS = F⁻ + Cl⁻ + NO₃⁻ + SO₄²⁻ + Na⁺ + K⁺ + Mg²⁺ + Ca²⁺ + HCO₃⁻ (Manaka et al., 2015).

River and its Mun and Chi tributaries are lowest and the most uniform (TOC 0.11–0.21 wt.% and TN 0.003–0.01 wt.%; Table 2). The Ayeyarwady and Sittuang samples are mainly sandy, and have TOC and TN contents (0.19–0.84 wt.% and 0.02–0.06 wt.%, respectively; Table 2) intermediate between the Chao Phraya and Mekong sediments. TOC/TN ratios in the samples analyzed range between 9.09 and 38.97, with the three highest values in the Mekong River and its tributaries (>15). Despite the wide range in these ratios, TOC shows a marked linear trend and strong positive correlation with TN ($r=0.98$, Fig. 2) overall.

4.3. Major elements

The sandy Ayeyarwady sediments show a relatively narrow range in SiO_2 content (69.89–77.64 wt.%), with an average of 73.46 wt.%. The Mekong, Mun and Chi sediments have a wide range of SiO_2 (65.53–90.92 wt.%, average 78.83 wt.%), as does the Chao Phraya and its tributaries (Yom, Ping, Wang, and Nan), spanning a range from 62.05 to 88.04 wt.% (average 73.35 wt.%). The

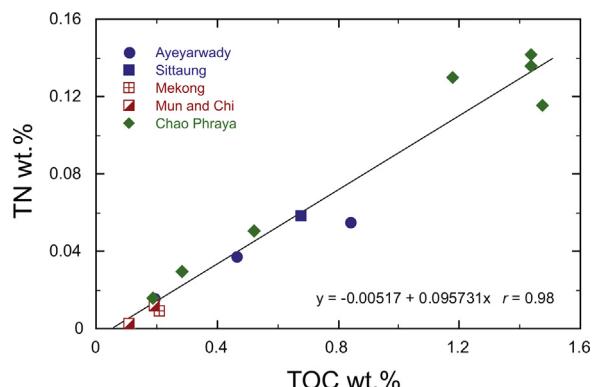


Fig. 2. TOC-TN variation diagram for the Myanmar and Thailand river sediments.

Ayeyarwady River sediments have relatively uniform Al_2O_3 contents (10.05–13.62 wt.%, average 11.89 wt.%), whereas the Mekong and Chao Phraya sediments show more variation, ranging from

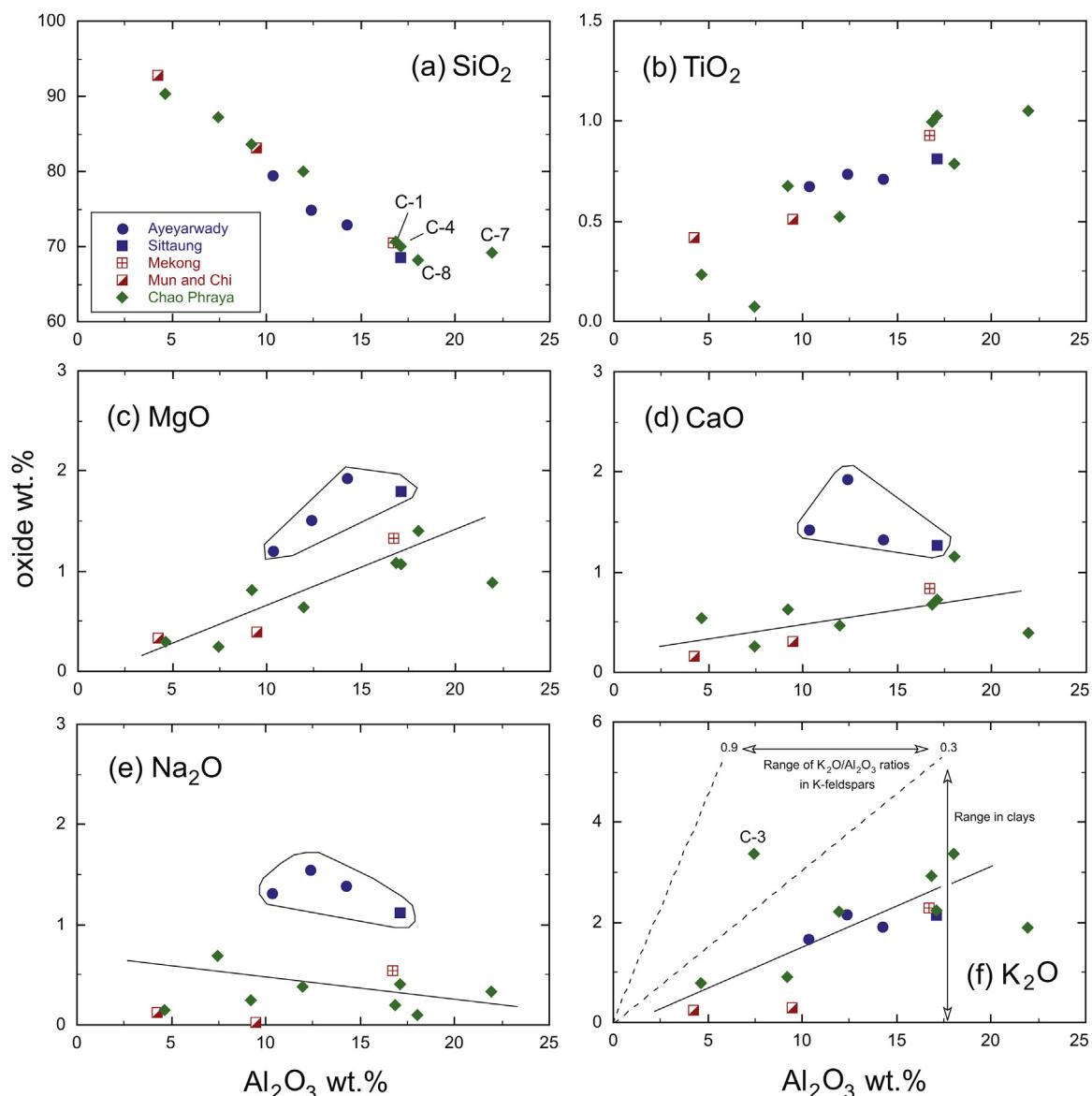


Fig. 3. Selected major element- Al_2O_3 variation diagrams for the Myanmar and Thailand river sediments. Ranges of $\text{K}_2\text{O}/\text{Al}_2\text{O}_3$ ratios in feldspars and clays from Cox et al. (1995). Solid lines are indicative detrital trends in the Thailand samples (fitted by eye). Fields on (c), (d) and (e) enclose the Myanmar data. Alpha-numeric labels are sample numbers (Table 2).

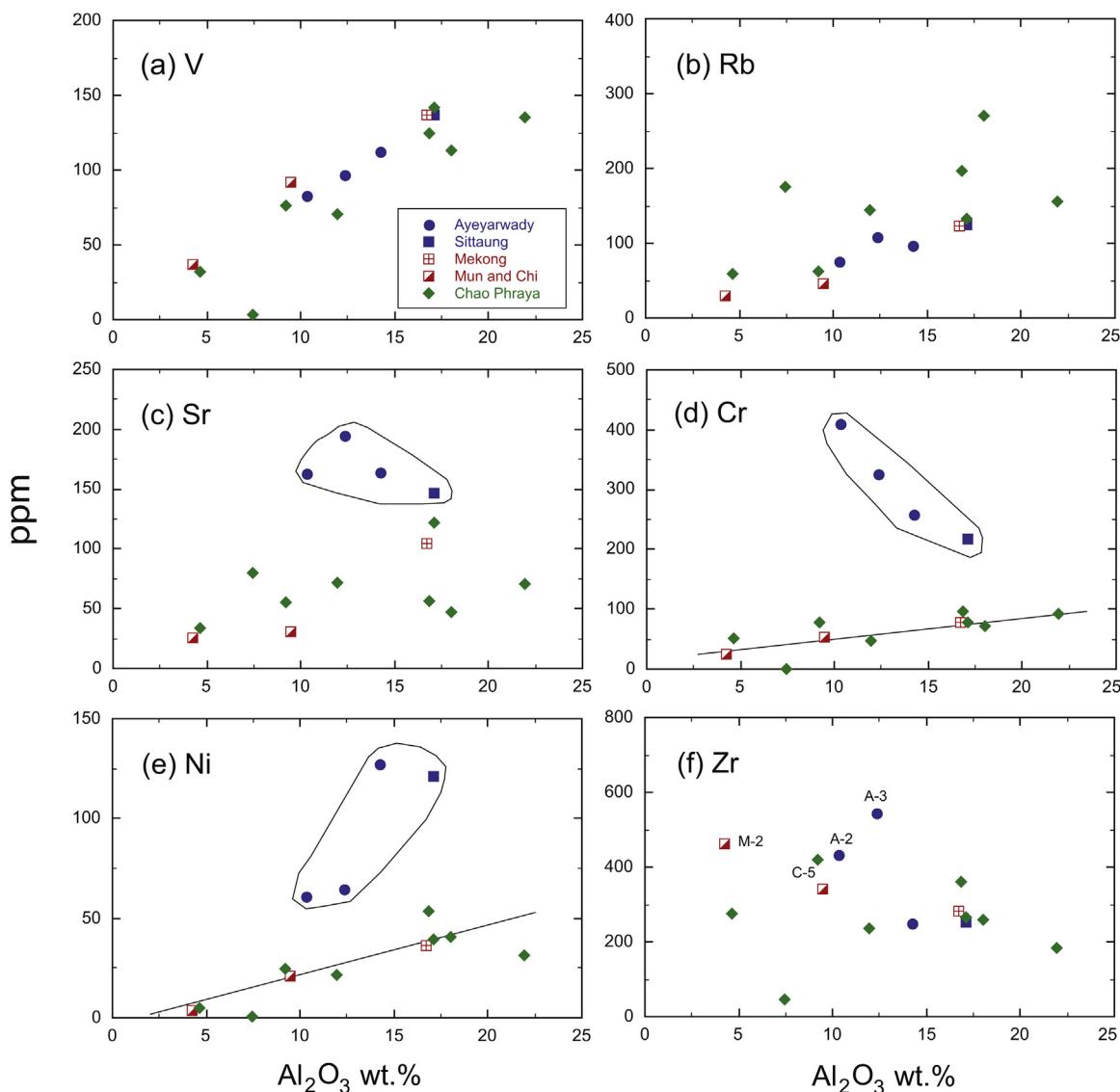


Fig. 4. Selected trace element- Al_2O_3 variation diagram for the studied rivers sediments in Myanmar and Thailand. Solid lines are indicative detrital trends in the Thailand samples (fitted by eye). Fields on (c), (d) and (e) enclose the Myanmar data. Alpha-numeric labels are sample numbers (Table 2).

4.17 to 15.56 wt.% (average 9.61 wt.%) and 4.50–20.48 wt.% (average 12.53 wt.%), respectively. The single Sittaung River sediment analyzed is a coarse silt, and has relatively low SiO_2 (64.40 wt.%) and high Al_2O_3 (16.06 wt.%) contents (Table 2). Four silt samples of different grades analyzed from Thailand (C-1, C-4, C-7 and C-8, Fig. 3a) have the highest anhydrous Al_2O_3 contents (>16.82 wt.%) and lowest SiO_2 (68.24–70.75 wt.%). As can be expected for the two most abundant oxides, SiO_2 is negatively correlated with Al_2O_3 , with a marked linear trend of decreasing SiO_2 with increasing Al_2O_3 (Fig. 3a).

Abundances of MgO , CaO , Na_2O and K_2O are highest in the Ayeyarwady and Sittaung rivers (>1 wt.%), and lower values are seen in the Mekong and Chao Phraya sediments (<1 wt.%). $\text{Fe}_2\text{O}_3\text{T}$ contents in the samples generally range from 1.48 to 6.46 wt.%, with the lowest concentration in Chao Phraya (Ping River) sample C-3 (0.65 wt.%). Four elements (TiO_2 (Fig. 3b); $\text{Fe}_2\text{O}_3\text{T}$, MnO , and P_2O_5) show broad positive correlation with Al_2O_3 , and little difference in trend between the sample suites.

In contrast to the above four elements, MgO forms a single broad trend in the Mekong and Chao Phraya suites, but all four Myanmar samples are displaced to higher MgO values (Fig. 3c).

A similar pattern is observed for CaO (Fig. 3d), with low values (all <1.16 wt.% CaO anhydrous) and a weak positive correlation with Al_2O_3 in the Mekong and Chao Phraya systems, and distinctly higher values (1.26–1.93 wt.% anhydrous) in the Myanmar samples. Abundances of Na_2O are also low in the Mekong and Chao Phraya suites (<0.69 wt.% anhydrous), and no correlation is evident with Al_2O_3 (Fig. 3e), but contents in the Myanmar samples are again higher (1.12–1.55 wt.%). Finally, K_2O shows a scattered positive correlation with Al_2O_3 , with most samples having $\text{K}_2\text{O}/\text{Al}_2\text{O}_3$ ratios of <0.3 (Fig. 3f), although one Chao Phraya River sample (C-3; coarse sand, Ping River) plots between $\text{K}_2\text{O}/\text{Al}_2\text{O}_3$ ratios of 0.3 and 0.9.

4.4. Trace elements

Trace element concentrations show wider variations. Abundances of Ba and Rb are relatively high in Chao Phraya sediments at 185–1904 ppm (average 584 ppm) and 58–247 ppm (141 ppm), respectively. Barium contents in the Ayeyarwady sediments are more uniform, ranging from 312 to 379 ppm (average 342 ppm). Strontium contents are uniformly higher (138–189 ppm, average 161 ppm) in the Ayeyarwady and Sittaung sediments than in

all other rivers (26–113 ppm, average 60 ppm). Average contents and ranges of the elements Ga (13 ppm; 4–24 ppm), Nb (14 ppm; 5–26 ppm), and Th (16 ppm; 7–28 ppm) are similar, and there is little difference in abundances between rivers. Cerium (average 75 ppm, 24–112 ppm), Pb (26 ppm, 9–61 ppm) and Y (26 ppm, 10–40 ppm) also show little systematic contrast between rivers. In contrast, the Ayeyarwady and Sittaung samples are characterized by considerably greater Cr (248–400 ppm, average 292 ppm) and Ni (60–122 ppm, average 89 ppm) contents than all other rivers, which together average 58 ppm Cr (1–88 ppm) and 24 ppm Ni (1–49 ppm). Vanadium is also a little more abundant in the four Ayeyarwady and Sittaung sediments (average 103 ppm; 81–129 ppm) than in the other rivers (average 82 ppm; 3–131 ppm), although overlap is considerable. The pattern for Zr is similar, averaging 357 ppm (range 238–528 ppm) in the four Ayeyarwady and Sittaung samples, compared to 270 ppm (44–452 ppm) in the remaining rivers. One Chao Phraya sediment (sample C-3, Ping River) has an unusually low content of 44 ppm (Table 2).

On trace element-Al₂O₃ variation diagrams (Fig. 4), most of the elements analyzed show positive correlations with Al₂O₃, but of varying strength. These can be divided into four groups. Elements in the first group (V (Fig. 4a), Ga, Nb and Y) are well correlated with Al₂O₃ in all suites. The second group of Rb (Fig. 4b), Ba, Pb, and Sr show moderate to weak correlations with Al₂O₃, with tendency for abundances to increase with increasing Al₂O₃ content. However, the correlation of Sr with Al₂O₃ is weak (Fig. 4c), with low (<~100 ppm) and relatively uniform values in the Mekong and Chao Phraya samples, and higher values in the four Myanmar (Ayeyarwady and Sittaung) sediments. The third group consists of the ferromagnesian elements Cr (Fig. 4d), Ni (Fig. 4e) and Sc. For these elements the Mekong and Chao Phraya samples fall on single linear trends. However, the Myanmar samples are strongly enriched (~2–4x) in Cr and Ni relative to these trends (Fig. 4d, e). Elements in the fourth group (Th, Ce and Zr) show considerable scatter and poor correlation with Al₂O₃. The highest abundances of Zr (Fig. 4f) occur in fine or very-fine grained sand samples (A-2, A-3, M-2; >400 ppm, Table 2). In contrast, very coarse silt samples in the Ayeyarwady and Mekong (A-1 and M-1, respectively) have relatively lower Zr concentrations (239 and 262 ppm, respectively).

4.5. Elemental ratios and indices

For comparative purposes, the average major and trace element abundances in each river were normalized against the composition of average upper continental crust (UCC), using the values compiled by Rudnick and Gao (2005). The UCC-normalized (UCC_N) plot (Fig. 5) shows that a few elements are enriched in the river sediments relative to UCC, but many are strongly depleted. Among the major oxides, SiO₂ and TiO₂ are generally enriched relative to UCC. In some averages (Ayeyarwady, Mun and Chi, Chao Phraya) Al₂O₃ is somewhat depleted. The most striking feature, however, is the marked depletion in the more mobile major elements Na₂O, K₂O, CaO and MgO (Fig. 5). Depletion of K₂O, CaO and Na₂O is especially marked in the Mun and Chi average (Fig. 5), with all three elements present at less than one-tenth of the UCC values. The mobile trace elements Sr and Ba are also significantly depleted relative to UCC. Two trace elements (Ni and Cr) often linked with ferromagnesian minerals are notably enriched in the Ayeyarwady and Sittaung (2–3 x UCC), compared to depletion in the other rivers.

The major element log ratios of SiO₂/Al₂O₃ and Na₂O/K₂O (Pettijohn et al., 1972), and log ratios of SiO₂/Al₂O₃ and Fe₂O₃/K₂O (Herron, 1988) are commonly used to classify terrigenous sediments. According to the geochemical classification scheme of Pettijohn et al. (1972), the Ayeyarwady and Sittaung sediments are characterized as litharenites, whereas the Mekong and Chao Phraya samples range from arkose through to sublitharenite. The

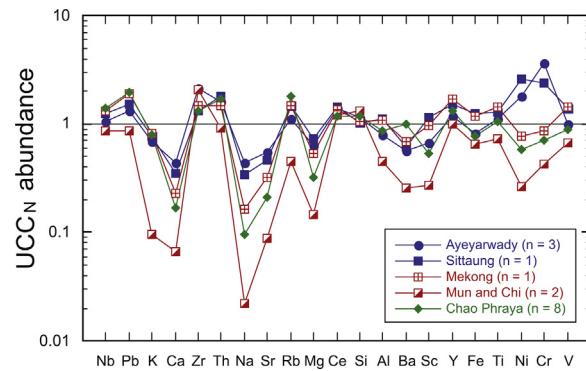


Fig. 5. Multi-element plot showing average river sediment compositions normalized against UCC (values of Rudnick and Gao, 2005). Elements are arranged from left to right in order of increasing normalized abundances in average Mesozoic-Cenozoic sandstone (Condie, 1993) relative to UCC, following the methodology of Dinelli et al. (1999). Major elements are normalized as oxides and trace elements as ppm.

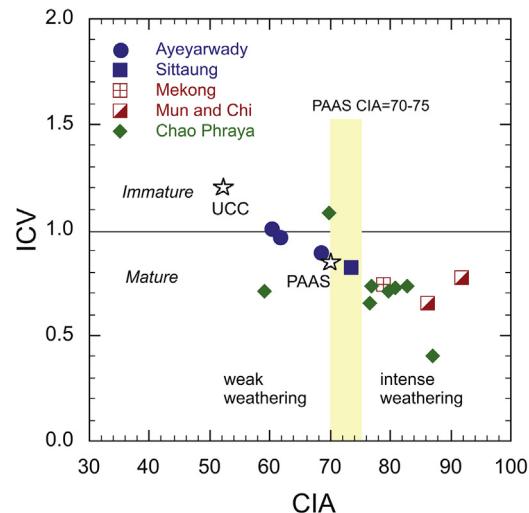


Fig. 6. ICV (Index of Compositional Variability) versus CIA (Chemical Index of Alteration) plot (after Long et al., 2012) for the Myanmar and Thailand river sediments. UCC (Upper Continental Crust) and PAAS (Post-Archean Australian Shale) values from Taylor and McLennan (1985).

Na₂O/K₂O ratios in Mekong and Chao Phraya sediments are distinctly lower than in the Ayeyarwady and Sittaung samples, due to their higher Al₂O₃ and K₂O and lower Na₂O contents (Table 2). On the SiO₂/Al₂O₃ and Fe₂O₃/K₂O classification scheme of Herron (1988), the finest-grained samples (clays) are classified as shales, whereas the three sandy Ayeyarwady samples (A1–A3) are classed as wackes and litharenite. The Mun and Chi samples are both classified as Fe-sands, and the Mekong very coarse silt (M-1) falls within the shale field. The single Chao Phraya coarse sand (C-3, Ping River) falls well within subarkose classification due to its very low Fe₂O₃/Al₂O₃ ratio of ~0.71, whereas all other samples have positive ratios (Table 2).

ICV values in the Ayeyarwady River system range from 0.89 to 1.01 (average 0.96), whereas the single samples from the Sittaung and Mekong rivers have ICV values of 0.82 and 0.74, respectively. Mun and Chi River ICV values range from 0.65 to 0.77 (average 0.71), and those in the Chao Phraya rivers from 0.41 to 1.08 (average 0.72). The Ayeyarwady River sediments plot between average Upper Continental Crust (UCC) and Post-Archean Australian Shale (PAAS) on an ICV-CIA plot (Fig. 6), whereas the Mekong, Mun and Chi, and most Chao Phraya samples fall around or beyond PAAS. Average ICV values in the Ayeyarwady samples are close to unity.

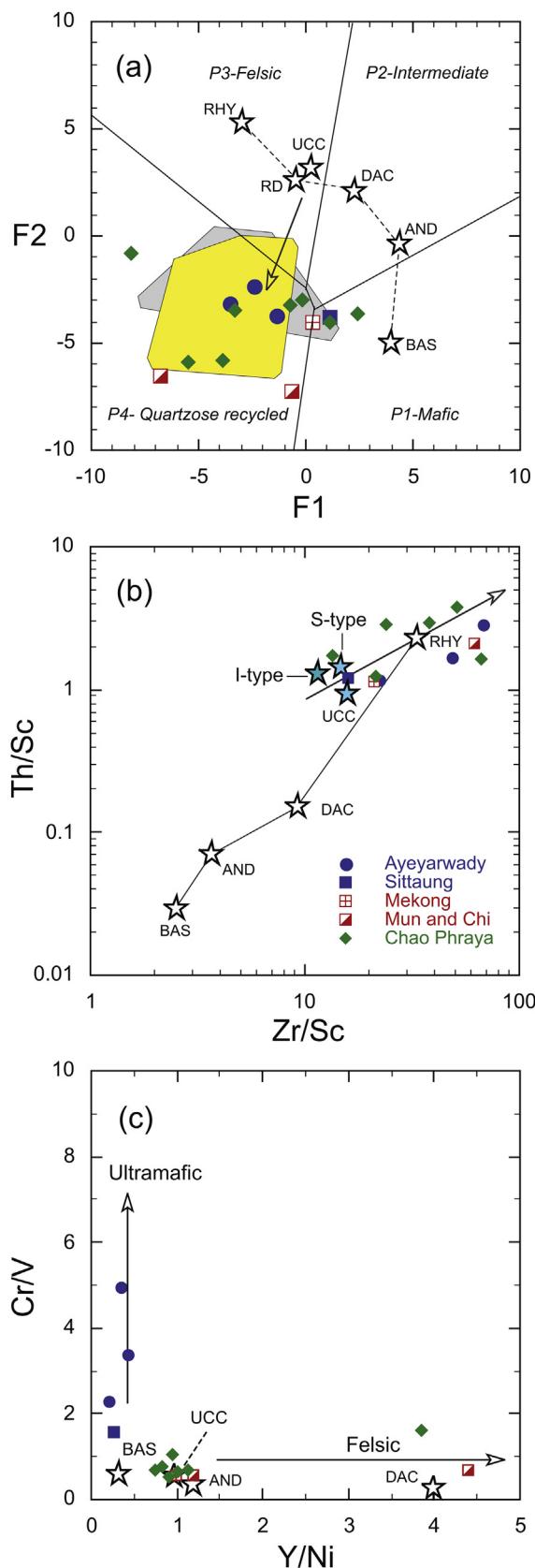


Fig. 7. (a) Major element provenance discriminant plot (Roser and Korsch, 1988); (b) Th/Sc-Zr/Sc plot (McLennan et al., 1993); (c) Cr/V-Y/Ni plot (modified from McLennan et al., 1993). Stars: UCC (average upper continental crust, Taylor and McLennan, 1985), BA, AN, DA, RD, RH, PHG/GR, FE – average basalt, andesite, dacite, rhyodacite, rhyolite, Phanerozoic granite and felsic volcanic (Taylor and McLennan, 1985; Condie, 1993), PCT- primary source composition trend, and average I-type and S-type granite averages from Whalen et al. (1987). Chao Phraya sample

4.6. Provenance indicators

The discriminant function diagram for sedimentary provenance studies proposed by Roser and Korsch (1988) distinguishes four provenance categories, namely mafic (P1), intermediate (P2), felsic (P3) and quartzose recycled (P4), based on the abundances of seven major elements. On this plot (Fig. 7a), the Ayeyarwady, Mun and Chi, and most of the Chao Phraya sediments fall within the quartzose recycled field (P4). The single Mekong sample analyzed falls on the P4-P1 boundary, whereas the Sittaung River and two Chao Phraya samples are displaced towards the P1 field (Fig. 7a).

$\text{Al}_2\text{O}_3/\text{TiO}_2$ ratios are also useful provenance indices (Hayashi et al., 1997). Ratios in the samples analyzed here range between 14 and 100, with high SiO_2 contents (~65–91 wt.%).

Abundances of relatively immobile trace elements such as Cr, Ni, V, Y, Th, Sc, Zr, and/or ratios formed from such elements (e.g. Th/Sc, Zr/Sc, Cr/V, Y/Ni) are commonly used to decipher mafic and felsic source rock provenance signatures in sediments and sedimentary rocks (Taylor and McLennan, 1985; Wronkiewicz and Condie, 1987; McLennan et al., 1993).

On a binary Th/Sc-Zr/Sc (McLennan et al., 1993) plot, all the analyzed samples fall near the primary compositional trend (PCT) close to the compositions of average rhyolite, UCC, and I- and S-type granites (Fig. 7b). Most of the Ayeyarwady, Mun and Chi, and Chao Phraya sediments fall close to the average rhyolite and granite compositions, but trend toward higher Th/Sc and Zr/Sc values. Abundances of Cr and Ni in the Ayeyarwady (248–400 ppm and 60–122 ppm, respectively) sediments are relatively high (Figs. 4 and 5), but Cr/Ni ratios fall between 2 and ~7. With one exception, the Cr/V ratios in the Mekong and Chao Phraya samples are <0.8, and Y/Ni ratios range up to 10.3 (Table 2). In contrast, Y/Ni ratios in the Ayeyarwady and Sittaung samples are uniformly low (0.2–0.4), whereas Cr/V ratios range from 1.6 to 4.9 (Table 2). The Myanmar samples thus form a distinctive vertical trend on the Cr/V-Y/Ni plot (Fig. 7c),

4.7. Weathering indices

CIA values for the Ayeyarwady and Sittaung sediments are relatively low, ranging from 60 to 69 (average 63) and 74, respectively (Table 2). In contrast, CIA values for the Mekong, Mun and Chi are 79, 92 and 86, respectively (Table 1). CIA values are also high in the sediments from the Chao Phraya and its tributaries, ranging between 59 and 87 (average 77). However, the lowest values (59.1, C-3; 69.7 C-2; Table 2) are recorded in the two sandy samples. Values in the clay-rich samples are more uniform (76.5–82.9) and greater than post-Archean shales.

On the ternary $\text{Al}_2\text{O}_3 - (\text{CaO}^* + \text{Na}_2\text{O}) - \text{K}_2\text{O}$ (A-CN-K) ternary diagram (Fig. 8a), the river sediments display a marked loss in K, Na, and Ca relative to UCC, and trend towards the A-apex. The Ayeyarwady, Sittaung and Mekong sediments follow the ideal weathering trend (IWT) from UCC, with lower K-corrected CIA in the Ayeyarwady (~68) than Sittaung (~74) and Mekong (~80) sediments (Fig. 8a). The Chao Phraya sediments also generally plot along the IWT, with the Mun and Chi falling near the A apex. On the (A-K)-C-N diagram (Fig. 8b), high PIA values (95) are seen in most of the Chao Phraya sediments, and Mun (95), Chi (91), and Mekong (87) samples, whereas relatively low PIA values occur in the Sittaung (79) and Ayeyarwady (<73) sediments. Most samples plot in the

C-2 ($\text{Zr/Sc} = 123.2$) and Mun River sample M-2 ($\text{Zr/Sc} = 213.6$) plot off-scale on Fig. 7b. Gray (Ayeyarwady) and yellow (Mekong) fields on (a): data from Garzanti et al. (2016) and Borges et al. (2008), respectively. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

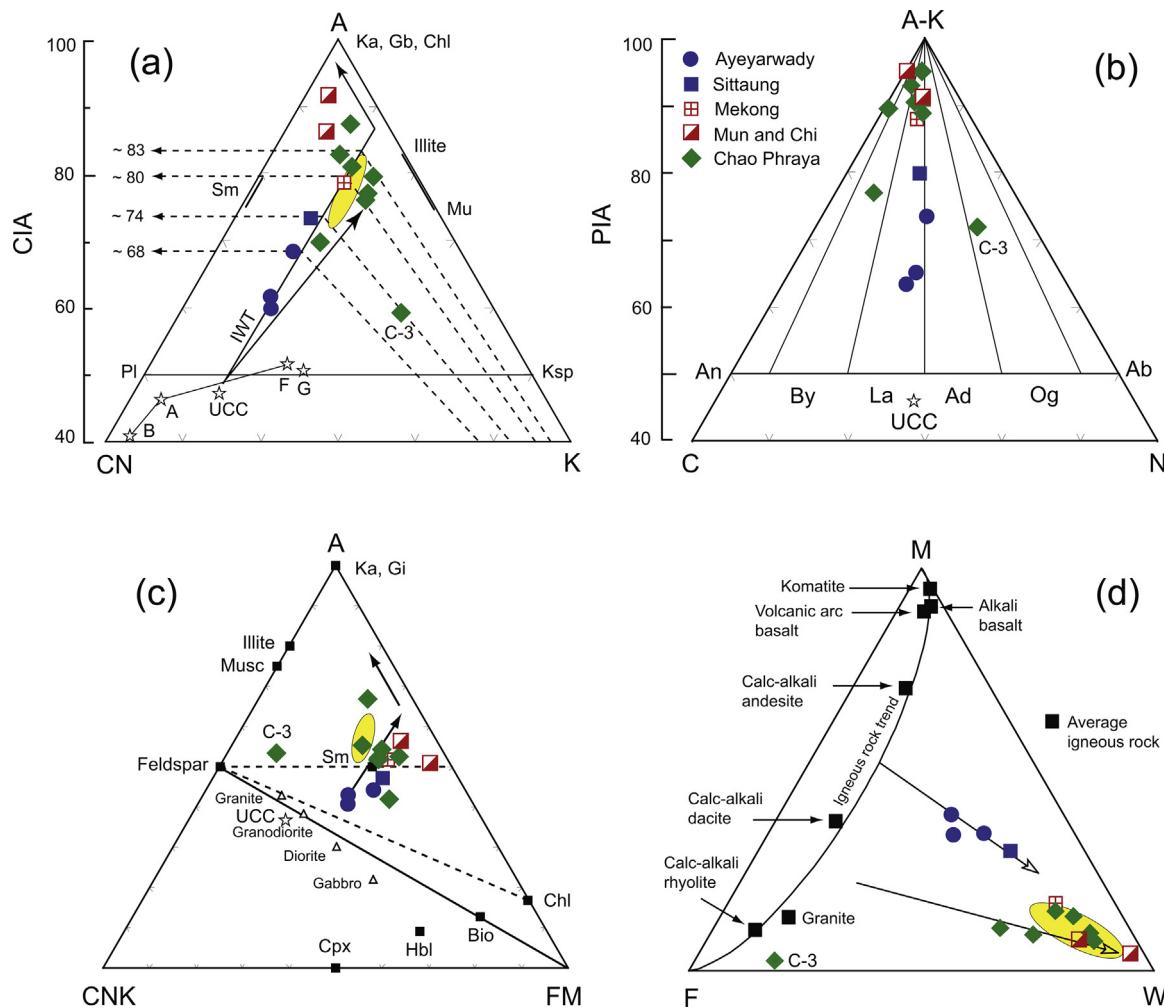


Fig. 8. (a) A-CN-K plot (Nesbitt and Young, 1984); (b) (A-K)-C-N plot (Fedo et al., 1997); (c) A-CN-K-FM plot (Nesbitt and Young, 1989); (d) M-F-W plot (Ohta and Arai, 2007) for river sediments in Myanmar and Thailand. Apices: A = Al_2O_3 , CN = $\text{CaO} + \text{Na}_2\text{O}$, A-K = $\text{Al}_2\text{O}_3 - \text{K}_2\text{O}$, C = CaO , N = Na_2O , CNK = $\text{CaO} + \text{Na}_2\text{O} + \text{K}_2\text{O}$, FM = $\text{Fe}_2\text{O}_3\text{T} + \text{MgO}$, M = Mafic, F = Felsic, W = Weathered material. Mineral abbreviations: Ka = kaolinite, Gb = gibbsite, Chl = chlorite, Mu = muscovite, Pl = plagioclase, Ksp = K-feldspar, An = anorthite, By = bytownite, La = labradorite, Ad = andesine, Og = oligoclase; Ab = albite, Bio = biotite, Hbl = hornblende, Cpx = clinopyroxene, Ga = gabbro, Di = diorite, Gd = granodiorite, B = basalt, A = andesite, F = felsic volcanic rock, G = granite (Condie, 1993); IWT = Ideal weathering trend; and UCC = upper continental crust composition (Taylor and McLennan, 1985). The solid arrowed line indicates the trend towards illite-muscovite. Yellow fields on (a), (c) and (d): field for Mekong River sediments (data from Liu et al., 2007). Rock averages on (d) as plotted by Ohta and Arai (2007). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

labradorite and bytownite fields, and trend towards the A-K apex. One Chao Phraya sample (C-3) falls in the oligoclase field.

Weathering intensity and approximate mineralogical composition in the river sediments can also be evaluated from the A-CNK-FM [$\text{Al}_2\text{O}_3 - (\text{CaO}^* + \text{Na}_2\text{O} + \text{K}_2\text{O}) - (\text{FeO}_t + \text{MgO})$] (Nesbitt and Young, 1989) ternary plot (Fig. 8c). The Ayeyarwady and Sittaung samples plot within the feldspar-smectite-chlorite triangle, near source compositions ranging between diorite to granite. The Mekong, Mun and Chi, and Chao Phraya data plot above the feldspar-smectite tie line and trend toward the A-apex. However, the Ayeyarwady and Sittaung samples plot below the feldspar-smectite line.

Ohta and Arai (2007) proposed a new system (MFW) to evaluate both source rock composition and the effects of weathering. This was based on the abundances of eight major elements (excluding MnO and P₂O₅), and multivariate statistical analyses and transformation to produce loadings for each element in individual samples. From these, values for three parameters are produced, where M = mafic, F = felsic, and W = weathering products. These are combined on a ternary diagram, where fresh source rocks plot in an arc between M and F, and weathered samples are displaced toward the

W apex (Fig. 8d). With the exception of the coarse sand from the Ping River (C-3), all Mekong and Chao Phraya samples plot near the W apex. The Thailand data overall also define a trend running away from igneous rock composition between dacite and granite.

5. Discussion

5.1. Distribution and source of organic matter

TOC refers to the amount of organic matter preserved within sediment which is likely to have originated from various sources, such as macrophytes, trees, algae/organism, and phytoplankton debris (Meyers, 1994; Goñi et al., 2013). Grain size is likely to influence the TOC contents of the sediments in this study, as fine-grained sediments generally have higher TOC contents than coarse-grained sediments (Keil et al., 1994). Clay and silt particles provide a large specific surface area and good binding sites for organic carbon preservation, and lower porosity for oxygen infiltration (Keil et al., 1994). As noted above (Section 4.2), TOC and TN contents in this study are greatest in the clays and silty clays, and lowest in the sandy samples, and a strong correlation exists between TOC and

TN (Fig. 2). These features suggest that abundances of TOC and TN in these river sediments are strongly influenced by hydrodynamic sorting (Carrie et al., 2009).

TOC/TN ratios are widely used to investigate the sources of organic matter in terrigenous sediments (Meyers, 1994; Hedges et al., 1997; Sampei and Matsumoto, 2001; Hossain et al., 2009). Low TOC/TN ratios (6–9) are mainly derived from planktonic/algae organisms, while values of 4–10 indicate nonvascular aquatic plant origin, and high ratios (>12) derivation from vascular plants (Meyers, 1994; Hedges et al., 1997; Sampei and Matsumoto, 2001). The high values and wide range in TOC/TN ratios (9.09–38.97; Table 2) observed in the samples analyzed here thus suggest influxes of both nonvascular and vascular plant-derived organic matter to the sediments. Furthermore, the marked linear trend and strong positive correlation between TOC and TN ($r=0.98$, Fig. 2) imply a comparable source, especially for the moderate to high TOC sediments, and probable hosting of the nitrogen in the organic matter.

5.2. Bulk geochemical compositions

Strong correlations on oxide-Al₂O₃ variation diagrams (Figs. 3 and 4) signify a relatively uniform source (Barovich and Foden, 2000), whereas moderate to poor correlations may suggest a mixed source (Ugidos et al., 1997). Hydrodynamic sorting during fluvial transport of sediments may affect mineralogical composition and thus chemical composition, as high density minerals are concentrated in coarser fractions, and low density minerals accumulate in finer fractions (McLennan et al., 1993; Garzanti et al., 2010; Lupker et al., 2012; Garzanti and Resentini, 2016). Settling velocity of different size grades can also influence elemental distributions in sedimentary environments, as elements such as Na, K, Rb, Ba, and Si are concentrated in tectosilicate-rich coarse to fine sand grades, whereas Y, Th, Ti, Zr, V, Nb, Cr, Fe, Ni, and Cu occur in heavy mineral-rich very-fine sands, except in very coarse silt grades, which tend to have relatively lower heavy mineral contents (Garzanti et al., 2010). The strong negative correlation seen between SiO₂ and Al₂O₃ (Section 4.3, Fig. 3a) suggests sorting effect and hydrodynamic separation of coarser quartz, feldspar, and lithic fragments from aluminous clays. Highest Al₂O₃ contents in the four Thailand silts (Section 4.3) and broad positive correlations between Al₂O₃ and TiO₂ (Fig. 3b), Fe₂O₃T, MnO, and P₂O₅ further suggest that abundances of these elements are allied with phyllosilicate fractions (e.g. illite, Dabard, 1990) and/or heavy minerals incorporated in the fine fractions. Three other elements (MgO, CaO and K₂O) show differing patterns, as noted in Section 4.3. The higher MgO and lower CaO contents of the Myanmar samples relative to the Mekong and Chao Phraya data suggest that some contrasts in source composition and source weathering exist between the suites. Based on the positive correlation between K₂O and Al₂O₃ and K₂O/Al₂O₃ ratios of <0.3 (Fig. 3f) in most samples indicates that K₂O resides mainly in aluminous clays (Cox et al., 1995). However, higher K₂O/Al₂O₃ in Chao Phraya River sample C-3 suggests that K-feldspar controls the K₂O content in that case (Cox et al., 1995).

As for TiO₂, Fe₂O₃T, MnO, and P₂O₅, positive correlations of V (Fig. 4a), Ga, Nb and Y with Al₂O₃ also suggest that abundances of these trace elements are primarily controlled by clay minerals, with the linear trend reflecting the effect of sediment sorting. The moderate to weak correlations of Rb (Fig. 4b), Ba, Pb, and Sr with Al₂O₃ indicate that their distribution is controlled by aluminosilicates, especially feldspars (Hossain et al., 2010). The weaker correlation of Sr with Al₂O₃ (Fig. 4c) is caused by low (<~100 ppm) contents in the Mekong and Chao Phraya samples and higher values in the Myanmar sediments, suggesting an input of fresh (or less weathered) plagioclase feldspar in the latter. This result is consis-

tent with the study of Garzanti et al. (2016), who suggested that sediments from the Ayeyarwady River headwaters are primarily of feldspatho-quartzose composition. Single linear trends observed for the ferromagnesian elements Cr (Fig. 4d), Ni (Fig. 4e) and Sc in the Mekong and Chao Phraya samples also indicate primary control by sorting. However, clear enrichment in Cr and Ni (~2–4x) relative to these trends (Fig. 4d, e) in the Myanmar samples, coupled with elevated MgO (Fig. 3c) abundances in those cases, suggest the presence of a mafic/ultramafic component in the Myanmar sediments. High concentrations of Cr and Ni in downstream sediments of the Ayeyarwady River reflect their source from a mafic and ultramafic suture-zone and volcanic-arc derived rocks (Garzanti et al., 2016). Lack of correlation of Th, Ce and Zr with Al₂O₃ suggests association with heavy minerals such as allanite, monazite and zircon. Allanite and to a lesser extent monazite and zircon are common heavy minerals in the lower Ayeyarwady River (Garzanti et al., 2016). The finest classes of sediments may be two or even three orders of magnitude richer in Zr content relative to the coarsest classes, and very-coarse silt fractions may be depleted in heavy minerals (Garzanti et al., 2010). Some such concentration of detrital zircon in specific size grades seems to have occurred in the samples of this study, as shown by the highest abundances of Zr (Fig. 4f) in fine or very-fine grained sand samples A-2, A-3, and M-2, and lower concentrations in very coarse silt grade samples A-1 (Ayeyarwady) and M-1 (Mekong).

On the UCC-normalized (UCC_N) plot enrichment in SiO₂ and TiO₂ relative to UCC (Fig. 5) indicates sorting effect during fluvial transport. Depletion of Al₂O₃ in the Ayeyarwady, Mun and Chi, and Chao Phraya averages is indicative of stronger quartz dilution in those cases. Strong depletion in mobile major elements (Na₂O, K₂O, CaO, MgO, Sr, and Ba; Fig. 5) suggests significant destruction of reactive feldspars and ferromagnesian minerals during chemical weathering in the sources of the sediments, although addition of recycled sedimentary clasts could also contribute to such depletion. Strongest depletion of K₂O, CaO and Na₂O in the Mun and Chi average (Fig. 5) implies more intense weathering in their catchments than in the other rivers. Among the trace elements, Zr, Th, Ce, and Y are moderately enriched relative to UCC, inferring their association with resistant heavy minerals (e.g. zircon, monazite, and apatite). Depletion of Ni and Cr in the Mekong, Mun-Chi and Chao Phraya averages relative to UCC contrasts with clear enrichment in the Ayeyarwady and Sittaung (2–3 x UCC). This depletion could be due to clay mineral control or a difference in provenance, as discussed further below.

5.3. Geochemical classification and maturity

As outlined above (Section 4.5) sediment classifications based on the major element log ratios SiO₂/Al₂O₃–Na₂O/K₂O (Pettijohn et al., 1972) and SiO₂/Al₂O₃–Fe₂O₃/K₂O (Herron, 1988) include litharenites (Ayeyarwady and Sittaung), and a range from arkose through to sublitharenite (Mekong and Chao Phraya). Overall, the classifications arising from these three major element ratios emphasize the relative textural and mineralogical immaturity of the sandier samples, with none being classified as quartz arenite under either scheme.

Major rock-forming minerals (feldspars, amphiboles and pyroxenes) have higher ICV values (>0.84) than clay minerals (<0.84, kaolinite, illite, and muscovite) (Cox et al., 1995). The high ICV values in the Ayeyarwady River and the single sample from the Wang River (a tributary in the Chao Phraya) reflect their sandy nature and textural immaturity. In contrast, lower ICV values indicate recycling and/or high contents of deeply weathered detritus (Cox et al., 1995; Gaillardet et al., 1999). The lower ICV values (<0.84) observed in the Sittaung, Mekong, Mun and Chi, and Chao Phraya rivers, coupled with higher CIA (Fig. 6) thus reflect a more mature character

and potentially more intensive source weathering, as discussed in Section 5.5.

5.4. Provenance

Major element abundances in sediments and sedimentary rocks are widely used to assess their provenance signatures, because they tend to at least partially reflect the average source composition (Taylor and McLennan, 1985; Roser and Korsch, 1988; Condie, 1993; Hayashi et al., 1997; Garzanti et al., 2009; Garzanti and Resentini, 2016). Plot position (Fig. 7a) of the Ayeyarwady, Mun and Chi, and most Chao Phraya sediments within the quartzose recycled field (P4) of Roser and Korsch (1988) is consistent with progressive increase in quartz and decrease in feldspar due to source weathering and/or sediment recycling. Enrichment of quartz and sedimentary lithics together with low feldspar in the downstream reaches of the Ayeyarwady River indicate that compositions were mostly controlled by fluvial transportation and/or addition of recycled sedimentary and metasedimentary detritus from the Indo-Burman Ranges (Garzanti et al., 2016). However, Allen et al. (2008) documented that high quartz and low feldspar and lithic fragments in the Ayeyarwady River sediments provide evidence of recycled orogenic provenance. Positioning of the Mekong sample on the P4-P1 boundary and displacement of the Sittaung and two Chao Phraya samples towards the P1 field (Fig. 7a) suggests that these sediments contain relatively high $\text{Fe}_2\text{O}_3\text{T}$ and TiO_2 concentrations. These features suggest that the analyzed samples were mostly derived from similar lithotypes, with strong influence from felsic sources, and minor inputs of intermediate/mafic source rocks, as also suggested by the UCC-normalized patterns (Fig. 5). Both $\text{Fe}_2\text{O}_3\text{T}$ and TiO_2 are common constituents of mafic rocks (Borges et al., 2008). Volcanic rocks are exposed on the Shan Plateau where the headwaters of the Sittaung River flows, and these likely contributed a significant mafic mineral fraction to the sediments.

Published major element datasets from the Ayeyarwady (Garzanti et al., 2016) and Mekong (Borges et al., 2008) rivers were also plotted on the F1-F2 diagram (Fig. 7a) for comparison. Both the Ayeyarwady (gray field) and Mekong (yellow field) data plot within the P4 field, and samples from the present study overlap them, further indicating a quartzose recycled provenance. Petrographic data for Mekong River sediments (mean $Q_{51}F_5R_{44}$) also indicates that they mainly originated from a quartzose recycled orogen provenance (Borges et al., 2008). This result also supports the study of Licht et al. (2014), who reported that high Zr/TiO_2 ratios in the Central Myanmar Basin demonstrated an increase in quartz, metamorphic and sedimentary rock fragments, due to advanced unroofing in the Indo-Burman Ranges, and/or by the progressive loss of mafic source materials. Sediments in modern rivers draining regions underlain by recycled rocks may have increased contents of highly stable minerals and consequently less stable minerals due to unroofing, abrasion, and weathering.

Information on mafic, intermediate, and felsic igneous source rock compositions are also provided by $\text{Al}_2\text{O}_3/\text{TiO}_2$ ratios, because these do not change significantly during chemical weathering, transportation, burial and the diagenesis of clastic sediments (Hayashi et al., 1997). The $\text{Al}_2\text{O}_3/\text{TiO}_2$ ratios for mafic, intermediate, and felsic igneous rocks range from 3 to 8 ($\text{SiO}_2 = 45\text{--}52\text{ wt.\%}$), 8–21 ($\text{SiO}_2 = 53\text{--}66\text{ wt.\%}$), and 21–70 ($\text{SiO}_2 = 66\text{--}76\text{ wt.\%}$), respectively (Hayashi et al., 1997). The high $\text{Al}_2\text{O}_3/\text{TiO}_2$ ratios observed in the samples analyzed here (14–100) coupled with significant SiO_2 contents (~65–91 wt.\%) thus indicate dominantly felsic sources.

$\text{Th}/\text{Sc-Zr}/\text{Sc}$ (McLennan et al., 1993) relationships also suggest primarily felsic sources, as all the analyzed sediments fall about the PCT, near the compositions of average rhyolite, UCC, and I- and S-type granites (Fig. 7b). Both I- and S-type granitic suites are present in the Central Myanmar Basin (Licht et al., 2014; Robinson et al.,

2014), and thus can contribute to the sediment flux in the Myanmar samples. The higher Th/Sc and Zr/Sc ratios in the Mun and Chi, and Chao Phraya sediments plotting at the upper end of the data distribution in Fig. 7b reflect zircon concentration during fluvial transport or inheritance from recycled sediment sources.

High contents of Cr (>150 ppm) and Ni (>100 ppm) coupled with low Cr/Ni ratios (between 1.3 and 1.5) in shale are consistent with ultramafic sources (Garver et al., 1996). Relatively high Cr and Ni contents in the Ayeyarwady (248–400 ppm and 60–122 ppm, respectively; Table 2; Figs. 4 and 5) and Cr/Ni ratios of 2 to ~7 suggest that these elements could be supplied from sources other than mafic/ultramafic rocks, as proposed by Garver et al. (1996). Mafic volcanic rocks and ophiolites are present in the Indo-Burman Ranges (Garzanti et al., 2013a, 2013b) where the Ayeyarwady River crosses the region, but contribution of these mafic components from those protoliths to the Ayeyarwady/Sittaung river sediment load were thought to be negligible (Garzanti et al., 2013a). The Ayeyarwady River sediments are characterized by feldspathic, lithic and quartzose detritus, with typically felsic volcanic/metavolcanic, chert, shale/slate to siltstone/metasiltstone rock fragments, minor serpentinite grains, micas, and heavy mineral suites typically rich in zircon, tourmaline, rutile, Cr-spinel, epidote and chloritoid (Garzanti et al., 2013a, 2016). However, the high Cr and Ni contents of the Ayeyarwady River sediments are probably controlled by detrital heavy minerals which include Cr-spinel, chromite, serpentinite and/or ultradense Fe-Ti-Cr oxides (Garzanti et al., 2009, 2013b). Some lower tributaries of the Ayeyarwady River join the Sittaung River. The moderately elevated Cr and Ni contents and UCC_N enrichments (Figs. 4 and 5) observed in the Sittaung sample (205 ppm and 114 ppm, respectively) may thus be a result of mixing between these river sediments, and/or influx of mafic components from nearby volcanic rock cover in the Shan Plateau which some headwaters of the Sittaung River cross. In contrast, the very low Cr and Ni abundances in the Mekong, Mun and Chi, and Chao Phraya sediments (<88 and <50 ppm, respectively; Figs. 4 and 5) are consistent with derivation primarily from felsic sources.

A clearer indication of the presence of an ophiolitic component in the Ayeyarwady and Sittaung sediments is given by Cr/V and Y/Ni ratios (Fig. 7c), which are sensitive to ophiolitic, mafic and felsic end member sources (Wronkiewicz and Condie, 1987; McLennan et al., 1993). Chromium is mainly hoisted in chromite (Hiscott, 1984), a mafic mineral typically present in ophiolitic sequences, and high Cr/V ratios indicate ultramafic source composition (Wronkiewicz and Condie, 1987; McLennan et al., 1993). Mafic to ultramafic sources tend to have high ferromagnesian mineral contents, resulting in high Cr/V ratios (>8) coupled with low Y/Ni (<0.5) ratios (McLennan et al., 1993). Cr/V ratios in Phanerozoic granite (Condie, 1993), UCC (Taylor and McLennan, 1985) and calcalkaline volcanic rock averages are typically <1, while Y/Ni ratios are variable, defining a flat trend towards higher Y/Ni (Fig. 7c). The horizontal trend of Y/Ni ratios at low Cr/V (<0.8) in the Mekong and Chao Phraya samples thus follows the calcalkaline rock averages (Fig. 7c), inferring a common felsic source. In contrast, the distinctive vertical trend in the Myanmar samples to high Cr/V (1.6–4.9) at uniformly low Y/Ni (Fig. 7c) signals the presence of a small but significant ultramafic component in their source that is lacking in the Mekong and Chao Phraya samples. This component is not clearly identified by the major element provenance discriminant (Fig. 7a) or Th/Sc-Zr/Sc systematics (Fig. 7b).

Contrasts in the abundances of several elements (Figs. 3 and 4), UCC_N abundances (Fig. 5) and provenance indicators (Fig. 7) described above thus reveal small differences in the provenance of the Ayeyarwady and Sittaung River sediments in Myanmar versus the dominantly felsic sources of the Mekong and Chao Phraya sediments in Thailand.

5.5. Source area weathering

The CIA (Nesbitt and Young, 1982, 1984) and PIA (Fedo et al., 1995) indices are widely used to assess source area weathering history and the source composition of sedimentary rocks. Weathering intensity and duration in the source area can be evaluated by examining the relationships among alkali and alkaline earth elements (Nesbitt and Young, 1982). Abundances of Ca, Na and K in residual soils and derived sediments decrease during progressive chemical weathering as the intensity or duration of weathering increases (Nesbitt and Young, 1982). These labile elements mainly reside in feldspars, which are easily altered into clay minerals. In general, fresh igneous rocks have CIA values near 50, typical shales range from 70 to 75, and residual clays (kaolinite, chlorite and gibbsite) have CIA values close to 100 (Nesbitt and Young, 1982, 1984).

As noted above (Section 4.7) CIA values for the Ayeyarwady (60–69) and Sittaung sediments (74) are low. Although the lower ratios in the Ayeyarwady samples are partly due to their sandy nature, these values indicate a low to moderate degree of chemical weathering in the Myanmar source area. Low silicate weathering in the Ayeyarwady River basin is also indicated by the study of total alkalinity budgets in that basin by Manaka et al. (2015). These authors estimated CO_2 consumption by silicate weathering flux in the Ayeyarwady River basin to be $63\text{--}145 \times 10^9 \text{ mol yr}^{-1}$, which was lower than carbonate weathering ($96\text{--}167 \times 10^9 \text{ mol yr}^{-1}$). Low silicate weathering in this river basin is attributed to lesser vegetation, soil cover and lower temperatures at its higher altitude where the river crossed silicate terrains (Meybeck, 1987). Higher CIA values for the Mekong, Mun and Chi (79, 92 and 86; Table 1), indicate moderately to intensely weathered source. The wide range (59–87; average 77) of CIA values in the Chao Phraya and its tributaries suggest variable degrees of chemical weathering in the source rocks, although the lowest values are recorded in the two sandy samples. In clay-rich samples, more uniform (76.5–82.9) CIA values greater than post-Archean shales are indicative of intense source weathering overall.

The CIA values are also presented graphically on the $\text{Al}_2\text{O}_3 - (\text{CaO}^* + \text{Na}_2\text{O}) - \text{K}_2\text{O}$ (A-CN-K) ternary diagram (Fig. 8a) to examine the weathering trends more precisely, and to evaluate bulk source composition (Nesbitt and Young, 1984; Fedo et al., 1995). Lower K-corrected CIA in the Ayeyarwady (~68) and Sittaung (~74) samples compared to the Mekong (~80) sediments (Fig. 8a) suggest low to moderate chemical weathering in the Ayeyarwady and Sittaung sources, and more intense chemical weathering in the latter. Moderate to intense chemical weathering and moderate hydrolysis in the Mekong River basin have also been reported by Liu et al. (2007), based on kaolinite/illite ratio, illite chemistry index, and CIA (72–83). The single sample from the Mekong River itself (M-1) falls within the field for Mekong sediments from Liu et al. (2007). Trend of the Chao Phraya sediments along the IWT, and position of the Mun and Chi samples near the A apex reflect more intense weathering in their source. However, small deviations from the IWT parallel to the A-CN edge may reflect mixing of varied source materials simultaneously affected by different degrees of weathering and/or the influence of partially modified Na, K or Ca in the silicate fraction (McLennan et al., 1993; Schoenborn and Fedo, 2011). Addition of recycled sedimentary clasts with high CIA may also contribute to scatter of individual samples, although the importance of that cannot be assessed here. The coarse sand sample C-3 (Ping River) is displaced well away from the IWT towards the K apex (Fig. 8a). This is probably due to case comparative enrichment in coarse detrital K-feldspar. The $\text{K}_2\text{O}/\text{Al}_2\text{O}_3$ ratio of this sample falls well within the range expected for K-feldspars (Fig. 3f), whereas the lower ratios in all other samples are typical of clays.

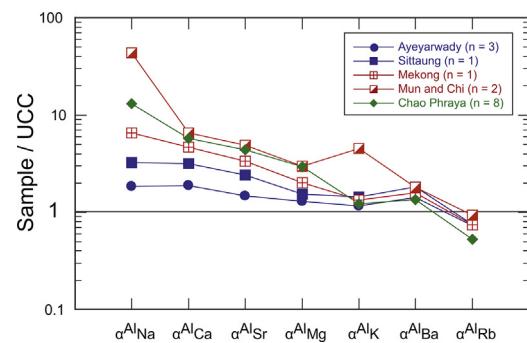


Fig. 9. Average α^{Al} values for alkali and alkaline earth metals relative to UCC (values of Rudnick and Gao, 2005) for each river sediments in Myanmar and Thailand (Garzanti et al., 2013b).

The degree of chemical weathering can also be evaluated using the PIA (Fedo et al., 1995). PIA measures the weathering of plagioclase after removal of the K_2O and Al_2O_3 incorporated in K-feldspar (Fedo et al., 1995, 1997; Schoenborn and Fedo, 2011). High PIA (95) values in most of the Chao Phraya and Mekong samples, contrasts with relatively low values occur in the Sittaung and Ayeyarwady sediments are compatible with the CIA ratios in these systems. The PIA ratios in the river sediments thus record moderate to high degrees of plagioclase weathering, with strong depletion in CaO and Na_2O . Labradorite or bytownite classification for most of the samples contrasts with oligoclase classification for Chao Phraya sample C-3, suggesting more albitic source in that case (Fedo et al., 1997).

Weathering paths on the A-CN-K-FM ternary plot (Fig. 8c) are less predictable than those in A-CN-K space (Nesbitt and Young, 1989). Concentration of the Ayeyarwady and Sittaung samples near source compositions ranging between diorite to granite and position of the Mekong, Mun and Chi, and Chao Phraya data above the feldspar-smectite tie line, with trend toward the A-apex, indicate a mostly felsic source composition for the river sediments. Spread toward aluminous clays is due to leaching of K_2O , Na_2O , and CaO within the weathering profiles. In contrast, only moderate chemical weathering is suggested for the Ayeyarwady and Sittaung samples plotting below the feldspar-smectite line. Mekong River sediment data from Liu et al. (2007) were also plotted on the A-CN-K-FM diagram (Fig. 8c). The field for these Mekong River sediments lies close to our data, inferring a similar weathering regime for the Mekong River basin and inputs of dominantly felsic detritus.

Weathering intensities of sediments can also be evaluated by depletion or enrichment of α^{Al} values for the alkali and alkaline earth metals relative to UCC (Borges et al., 2008; Garzanti et al., 2013b; Garzanti and Resentini, 2016). Mobile element concentrations in the sediments are compared to those of a non-mobile element (e.g. Al) and UCC. The chemical mobility indexes are denoted as α (Gaillardet et al., 1999), and α^{Al} values close to ~1 signify no chemical weathering, whereas values >1 are associated with depletion of elements relative to UCC (Gaillardet et al., 1999; Garzanti et al., 2013b). Average $\alpha^{\text{Al}}\text{E}$ values for the river sediments analyzed here show marked enrichment compared to UCC for the Chao Phraya and Mekong watershed areas, whereas values for the Ayeyarwady and Sittaung watersheds are close to UCC (Fig. 9), indicating variable degrees of weathering intensity. Average $\alpha^{\text{Al}}\text{Rb}$ (~1) values display are lower than UCC, and $\alpha^{\text{Al}}\text{K}$ (<2) and $\alpha^{\text{Al}}\text{Ba}$ (<2) values are almost identical for all rivers, except average Mun and Chi rivers sediments, which display higher values ($\alpha^{\text{Al}}\text{K}$, 3.16–5.85). The $\alpha^{\text{Al}}\text{E}$ values for the Ayeyarwady and Sittaung sediments follow the mobility sequence of $\alpha^{\text{Al}}\text{Na} \approx \alpha^{\text{Al}}\text{Ca} > \alpha^{\text{Al}}\text{Sr} > \alpha^{\text{Al}}\text{Ba} > \alpha^{\text{Al}}\text{Mg} \approx \alpha^{\text{Al}}\text{K} > \alpha^{\text{Al}}\text{Rb}$, whereas the Mekong, Chao Phraya and their tributaries follow sequence $\alpha^{\text{Al}}\text{Na} > \alpha^{\text{Al}}\text{Ca} > \alpha^{\text{Al}}\text{Sr} > \alpha^{\text{Al}}\text{Mg} > \alpha^{\text{Al}}\text{Ba} > \alpha^{\text{Al}}\text{K} > \alpha^{\text{Al}}\text{Rb}$. Val-

ues for highly mobile elements such as $\alpha^{\text{Al}}\text{Na}$ and $\alpha^{\text{Al}}\text{Ca}$ are high in the Mun and Chi samples (7.58–78.87 and 6.02–7.08, respectively), Chao Phraya (2.30–40.51 and 2.00–12.97, respectively) and in the Mekong (6.56 and 4.69, respectively) sediment, suggesting moderate to extremely weathered detritus. However, comparatively low $\alpha^{\text{Al}}\text{Na}$ and $\alpha^{\text{Al}}\text{Ca}$ values in the Sittaung (3.23 and 3.16, respectively) and Ayeyarwady samples (1.66–2.18 and 1.49–2.50, respectively) reflect low to moderate degree of chemical weathering. Similar low $\alpha^{\text{Al}}\text{Na}$ values are recorded in lower Ayeyarwady River sand ($\alpha^{\text{Al}}\text{Na}$, 1.6 ± 0.2) and silty levees ($\alpha^{\text{Al}}\text{Na}$, 1.8 ± 0.2), as reported by Garzanti et al. (2016). The α^{Al} values overall thus confirm the results from CIA and PIA described above, with lower weathering intensity in the Sittaung and Ayeyarwady watersheds compared to the Mekong and Chao Phraya systems.

The A-CN-K or (A-K)-C-N plots discussed above (Fig. 8a and b) are insensitive to the small differences in provenance noted between the Myanmar and Thailand suites. This is not the case with the MFW plot (Fig. 8d). With one exception, the Mekong and Chao Phraya samples plot near the W apex, reflecting their higher CIA and PIA indices, and define a trend away from felsic source composition (dacite to granite average). We also plotted published Mekong River sediment datasets (Liu et al., 2007) in order to assess the compatibility of the provenance and weathering signals within this Mekong River system. Our Mekong data matches the field for the data from Liu et al. (2007) in Fig. 8d, confirming a deeply weathered felsic source. The trend from the igneous rock average for the Ayeyarwady and Sittaung samples clearly differs from that of the Mekong and Chao Phraya data, originating from a position between andesite and dacite (Fig. 8d). This further confirms the presence of a small but significant mafic component in the Myanmar sediments. The Myanmar samples also plot closer to the igneous rock average curve, compatible with their lower CIA, PIA and α^{Al} values. The MFW plot thus highlights the more intense weathering in the Mekong and Chao Phraya source, and the provenance contrasts between the Myanmar and Thailand sediments.

The degree of chemical weathering in sediments can be also revealed by Rb/Sr and K₂O/Rb ratios (McLennan et al., 1993). During progressive chemical weathering, Rb/Sr ratios in soil profiles are strongly elevated (>0.5) while K₂O/Rb ratios (<250) decrease markedly relative to UCC (McLennan et al., 1993). Rb/Sr and K₂O/Rb ratios in UCC are 0.32 and 252, respectively (Taylor and McLennan, 1985). The Rb/Sr ratios in the analyzed river sediments are relatively high (0.46–5.76, average 1.73), and K₂O/Rb ratios are relatively low (53–186, average 128), reflecting moderate to intense chemical weathering, consistent with the CIA, PIA and ICV values. From the above observations, we conclude that silicate weathering is comparatively weak in the Ayeyarwady and Sittaung river basins, moderate to intense in the Mekong and Chao Phraya River basins, and most intense in the Mun and Chi river basins.

5.6. Relationship between sediment chemistry and water properties of the Myanmar and Thailand rivers

Variations of water discharge and sediment transfer into riverine regimes are sensitive to climate change. High rainfall and flooding during seasonal fluctuations also influence intense chemical weathering, soil/rock erosion and ion exchange in watersheds. The major and trace element geochemical data of the river sediments were compared with major ion data for river waters collected at the same locations in the Ayeyarwady, Sittaung, Mekong and Chao Phraya basins, to investigate controlling factors such as precipitation, rock weathering, and/or evaporation-crystallization contribution. The intensity of chemical weathering in the study area is also assessed based on solute concentrations in the river waters. Major ion compositions of the river waters were taken

from Manaka et al. (2015). Gibbs (1970) documented that low total dissolved solids (TDS) and high Na/(Na + Ca) ratios in river water are controlled by atmospheric precipitation, intermediate TDS and low Na/(Na + Ca) are controlled by rock weathering, and high TDS and high Na/(Na + Ca) ratios in river water are regulated by evaporation-crystallization processes. The CIA data (average CIA 66) show that the Ayeyarwady River watershed experienced a lower degree of chemical weathering, whereas the Mekong (average CIA 86) and Chao Phraya (average CIA 76) river sediments contain moderately to intensely weathered detritus. Average TDS values in the Ayeyarwady, Sittaung, Mekong and Chao Phraya rivers are 55 mg/l, 71 mg/l, 277 mg/l, and 187 mg/l, respectively, and average Na/(Na + Ca) ratios are 0.3, 0.4, 0.6, and 0.3, respectively (Manaka et al., 2015). The moderate TDS and Na/(Na + Ca) values in the Mekong and Chao Phraya rivers (Gibbs, 1970) indicate that silicate weathering was the potential source of major ions to the river waters, which is consistent with the moderate to high CIA and PIA values. Li et al. (2014) noted that low to moderate TDS in the lower Mekong River (53 mg/l to 198 mg/l) and low Na/(Na + Ca) ratios are consistent with significant rock chemical weathering in the watershed area. However, relatively low TDS and Na/(Na + Ca) values in the Ayeyarwady and Sittaung rivers water were probably controlled by low silicate weathering and/or precipitation. Ota et al. (2017) noted that low chemical weathering intensity (CWI, 57–66), low to moderate CIA (69–74), and low mode grain size values (5.7 μm –18.7 μm) in Andaman Sea core sediments suggest fluctuation of the Indian Summer Monsoon over the Ayeyarwady River basin was the controlling factor for changing the lower degree of chemical weathering, and that the climate was probably dry and cold to a little more warm and humid. Low chemical weathering could have influenced the lower degree of ion exchange from rock weathering to river solute in the Ayeyarwady watershed, owing to higher physical weathering than chemical weathering, hence decreasing CIA values. However, the relatively high TDS and CIA in the Mekong and Chao Phraya rivers imply warm and humid climatic conditions prevailed in their watersheds, thus releasing a significant portion of major ions from silicate weathering to the rivers.

6. Conclusions

The TOC, TN, major and trace element compositions of stream sediments from the Ayeyarwady and Sittaung rivers in Myanmar, and the Mekong and Chao Phraya rivers and their tributaries in Thailand were examined to identify their distributions, provenance, and chemical weathering processes. High TOC and TN concentrations in fine grained sediments of the rivers indicate hydrodynamic sorting may control these distributions. TOC/TN ratio suggests input of a mixture of aquatic macrophyte and higher vascular plant material to the sediments. The predominance of SiO₂ in coarser-grained samples and Al₂O₃ in finer-grained samples display obvious negative correlation, demonstrating primary grain size control on SiO₂ distributions due to sorting. The marked depletion of most labile elements such as Na₂O, CaO, K₂O, Ba and Sr relative to UCC suggest extensive destruction of feldspar during prolonged chemical weathering in the source areas or during river transport. Enrichment of several high field strength elements (Zr, Th, and Ce) indicate moderate concentration of resistant heavy minerals in all suites, and relative enrichment in ferromagnesian elements (especially Cr and Ni) in the Myanmar samples indicate the presence of a small mafic or ultramafic component in their source. Major element and immobile trace element characteristics suggest that the Mekong and Chao Phraya sediments were mostly derived from felsic sources compositions close to average rhyolite, dacite/granodiorite, UCC, I- and S-type granites. Higher

Zr/Sc and Th/Sc ratios in most of the silt or clay-rich river sediments reflect heavy mineral concentrations in the fine fractions, and high Cr/V and low Y/Ni ratios in the Ayeyarwady and Sittaung samples support the presence of a small ultramafic or mafic component in the Myanmar watersheds. ICV, CIA, PIA, α^{Al} , MFW, Rb/Sr and K₂O/Rb ratios and relationships in the river sediments indicate that the Ayeyarwady and Sittaung watersheds underwent low to moderate degrees of chemical weathering, compared to moderate to intense chemical weathering in the Mekong, Mun and Chi, and Chao Phraya basins. A-CNK–FM and MFW diagrams further indicate low to moderate chemical weathering and the mafic component in the Ayeyarwady and Sittaung source terrains, and moderate to intense weathering in the Mekong and Chao Phraya sediments sources, with major contribution from felsic source materials. The results from the weathering indices are compatible with published data for major ion data for river waters collected at the same locations.

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References

- Allen, R., Najman, Y., Carter, A., Barfod, D., Bickle, M.J., Chapman, H.J., Garzanti, E., Vezzoli, G., Andò, S., Parrish, R.R., 2008. *Provenance of the Tertiary sedimentary rocks of the Indo-Burman Ranges, Burma (Myanmar): Burman arc or Himalayan-derived?* *J. Geol. Soc. London* 165, 1045–1057.
- Awasthi, N., Ray, J.S., Singh, A.K., Band, S.T., Rai, V.K., 2014. Provenance of the Late Quaternary sediments in the Andaman Sea: implications for monsoon variability and ocean circulation. *Geochem. Geophys. Geosyst.* 15 (10), 3890–3906.
- Barovich, K.M., Foden, J., 2000. A Neoproterozoic flood basalt province in southern-central Australia: geochemical and Nd isotope evidence from basin fill. *Precambrian Res.* 100, 213–234.
- Bickle, M.J., Tipper, E., Galy, A., Chapman, H., Harris, N., 2015. On discrimination between carbonate and silicate inputs to Himalayan rivers. *Am. J. Sci.* 315, 120–166.
- Bird, M.I., Robinson, R.A.J., Oo, N.W., Aye, M.M., Lu, X.X., Higgitt, D.L., Swe, A., Tun, T., Win, S.L., Aye, K.S., Win, K.M.M., Hoey, T.B., 2008. A preliminary estimate of organic carbon transport by the Ayeyarwady (Irrawaddy) and Thanlwin (Salween) Rivers of Myanmar. *Quat. Int.* 186, 113–122.
- Borges, J.B., Huh, Y., Moon, S., Noh, H., 2008. Provenance and weathering control on river bed sediments of the eastern Tibetan Plateau and the Russian Far East. *Chem. Geol.* 254, 52–72.
- Carrie, J., Sanei, H., Goodarzi, F., Stern, G., Wang, F., 2009. Characterization of organic matter in surface sediments of the Mackenzie River Basin, Canada. *Int. J. Coal Geol.* 77, 416–423.
- Chapman, H., Bickle, M., Thaw, S.H., Thiam, H.N., 2015. Chemical fluxes from time series sampling of the Irrawaddy and Salween Rivers, Myanmar. *Chem. Geol.* 401, 15–27.
- Condie, K.C., 1993. Chemical composition and evolution of the upper continental crust: contrasting results from surface samples and shales. *Chem. Geol.* 104, 1–37.
- Cox, R., Lowe, D.R., Cullers, R.L., 1995. The influence of sediment recycling and basement composition on evolution of mudrock chemistry in the southwestern United States. *Geochim. Cosmochim. Acta* 59, 2919–2940.
- Cullers, R.L., Basu, A., Suttnar, L.J., 1988. Geochemical signature of provenance in sand size material in soils and stream sediments near the Tobacco Root batholith Montana, U.S.A. *Chem. Geol.* 70, 335–348.
- Dabard, M.P., 1990. Lower Bivorian formations (Upper Proterozoic) of the Armorican Massif (France): Geodynamic evolution of source areas revealed by sandstone petrography and geochemistry. *Sediment. Geol.* 69, 45–58.
- Dinelli, E., Lucchini, F., Mordini, A., Paganelli, L., 1999. Geochemistry of Oligocene-Miocene sandstones of the northern Apennines (Italy) and evolution of chemical features in relation to provenance changes. *Sediment. Geol.* 127, 193–207.
- Fedo, C.M., Nesbitt, H.W., Young, G.M., 1995. Unraveling the effects of potassium metasomatism in sedimentary rocks and paleosols, with implications for paleoweathering conditions and provenance. *Geology* 23, 921–924.
- Fedo, C.M., Young, G.M., Nesbitt, H.W., Hanchar, J.M., 1997. Potassic and sodic metasomatism in the Southern Province of the Canadian Shield: evidence from the Paleoproterozoic Serpent Formation Huronian Supergroup, Canada. *Precambrian Res.* 84, 17–36.
- Folk, R.L., Ward, W.C., 1957. Brazos River bar: a study in the significance of grain size parameters. *J. Sediment. Petrol.* 27, 3–26.
- Furuichi, T., Win, Z., Wasson, R.J., 2009. Discharge and suspended sediment transport in the Ayeyarwady River, Myanmar: centennial and decadal changes. *Hydro. Processes* 23, 1631–1641.
- Gaillardet, J., Dupré, B., Allègre, C.J., 1999. Geochemistry of large river suspended sediments: silicate weathering or recycling tracer? *Geochim. Cosmochim. Acta* 63, 4037–4051.
- Garver, J.I., Royce, P.R., Smick, T.A., 1996. Chromium and nickel in shale of the Taconic foreland: a case study for the provenance of fine-grained sediments with an ultramafic source. *J. Sediment. Res.* 66, 100–106.
- Garzanti, E., Resentini, A., 2016. Provenance control on chemical indices of weathering (Taiwan river sands). *Sediment. Geol.* 336, 81–95.
- Garzanti, E., Andò, S., Vezzoli, G., 2009. Grain-size dependence of sediment composition and environmental bias in provenance studies. *Earth Planet. Sci. Lett.* 277, 422–432.
- Garzanti, E., Andò, S., France-Lanord, C., Vezzoli, G., Censi, P., Galy, V., Najman, Y., 2010. Mineralogical and chemical variability of fluvial sediments: 1. Bedload sand (Ganga-Brahmaputra, Bangladesh). *Earth Planet. Sci. Lett.* 299, 368–381.
- Garzanti, E., Limonta, M., Resentini, A., Bandopadhyay, P.C., Najman, Y., Andò, S., Vezzoli, G., 2013a. Sediment recycling at convergent plate margins (Indo-Burman Ranges and Andaman–Nicobar Ridge). *Earth Sci. Rev.* 123, 113–132.
- Garzanti, E., Padoan, M., Andò, S., Resentini, A., Vezzoli, G., Lustrino, M., 2013b. Weathering and relative durability of detrital minerals in equatorial climate: sand petrology and geochemistry in the East African Rift. *J. Geol.* 121, 547–580.
- Garzanti, E., Wang, J.-G., Vezzoli, G., Limonta, M., 2016. Tracing provenance and sediment fluxes in the Irrawaddy River basin (Myanmar). *Chem. Geol.* 440, 73–90.
- Gibbs, R.J., 1970. Mechanism controlling world water chemistry. *Science* 170, 1088–1090.
- Goñi, M.A., O'Connor, A.E., Kuzyk, Z.Z., Yunker, M.B., Gobeil, C., Macdonald, R.W., 2013. Distribution and sources of organic matter in surface marine sediments across the North American Arctic margin. *J. Geophys. Res.: Oceans* 118, 4017–4035.
- Gupta, H., Kao, S.-J., Dai, M., 2012. The role of mega dams in reducing sediment fluxes: a case study of large Asian rivers. *J. Hydrol.* 464–465, 447–458.
- Gururajan, N.S., Choudhuri, B.K., 2003. Geology and tectonic history of the Lohit valley, eastern Arunachal Pradesh, India. *J. Asian Earth Sci.* 21, 731–741.
- Hayashi, K., Fujisawa, H., Holland, H.D., Ohmoto, H., 1997. Geochemistry of 1.9 Ga sedimentary rocks from northeastern Labrador, Canada. *Geochim. Cosmochim. Acta* 61, 4115–4137.
- Hedges, J.I., Keil, R.G., Benner, R., 1997. What happens to terrestrial organic matter in the ocean? *Org. Geochim.* 27, 195–212.
- Herron, M.M., 1988. Geochemical classification of terrigenous sands and shales from core or log data. *J. Sediment. Petrol.* 58, 820–829.
- Hiscott, R.N., 1984. Ophiolitic source rocks for Taconic-age flysch: trace element evidence. *Geol. Soc. Am. Bull.* 95, 1261–1267.
- Hossain, H.M.Z., Sampéi, Y., Roser, B.P., 2009. Characterization of organic matter and depositional environment of Tertiary mudstones from the Sylhet Basin, Bangladesh. *Org. Geochem.* 40, 743–754.
- Hossain, H.M.Z., Roser, B.P., Kimura, J.-I., 2010. Petrography and whole-rock geochemistry of the Tertiary Sylhet succession, northeastern Bengal Basin, Bangladesh: provenance and source area weathering. *Sediment. Geol.* 228, 171–183.
- Keil, R.G., Montluçon, D.B., Prahl, F.G., Hedges, J.I., 1994. Sorptive preservation of labile organic matter in marine sediments. *Nature* 370, 549–552.
- Kimura, J.-I., Yamada, Y., 1996. Evaluation of major and trace element XRF analyses using a flux to sample ratio of two to one glass beads. *J. Mineral. Petrol. Econ. Geol.* 91, 62–72.
- Li, C., Yang, S., 2010. Is chemical index of alteration (CIA) a reliable proxy for chemical weathering in global drainage basins? *Am. J. Sci.* 310, 111–127.
- Li, S., Lu, X.X., Bush, R.T., 2014. Chemical weathering and CO₂ consumption in the lower Mekong River. *Sci. Total Environ.* 472, 162–177.
- Licht, A., Reisberg, L., France-Lanord, C., Soe, A.N., Jaeger, J.-J., 2014. Cenozoic evolution of the central Myanmar drainage system: insights from sediment provenance in the Minbu Sub-Basin. *Basin Res.* 2014, 1–15.
- Liu, Z., Colin, C., Huang, W., Le, K.P., Tong, S., Chen, Z., Trentesaux, A., 2007. Climatic and tectonic controls on weathering in south China and Indochina Peninsula: clay mineralogical and geochemical investigations from the Pearl, Red, and Mekong drainage basins. *Geochim. Geophys. Geosyst.* 8, Q05005, <http://dx.doi.org/10.1029/2006GC001490>.
- Long, X., Yuan, C., Sun, M., Xiao, W., Wang, Y., Cai, K., Jiang, Y., 2012. Geochemistry and Nd isotopic composition of the Early Paleozoic flysch sequence in the Chinese Altai, Central Asia: evidence for a northward-derived mafic source and insight into Nd model ages in accretionary orogen. *Gondwana Res.* 22, 554–566.
- Lupker, M., France-Lanord, C., Galy, V., Lavé, J., Gaillardet, J., Gajurel, A.P., Guilmette, C., Rahman, M., Singh, S.K., Sinha, R., 2012. Predominant floodplain

- over mountain weathering of Himalayan sediments (Ganga basin). *Geochim. Cosmochim. Acta* 84, 410–432.
- Manaka, T., Otani, S., Inamura, A., Suzuki, A., Aung, T., Roachanakanan, R., Ishiwa, T., Kawahata, H., 2015. Chemical weathering and long-term CO₂ consumption in the Ayeyarwady and Mekong river basins in the Himalayas. *J. Geophys. Res.: Biogeosci.* 120, 1165–1175.
- McLennan, S.M., Hemming, S., McDowell, D.K., Hanson, G.N., 1993. Geochemical approaches to sedimentation, provenance, and tectonics. *Geol. Soc. Am. Spec. Paper* 284, 21–40.
- Meybeck, M., Ragu, A., 2012. GEMS-GLORI World River Discharge Database. Laboratoire de Géologie Appliquée, Université Pierre et Marie Curie, Paris France.
- Meybeck, M., 1987. Global chemical weathering of surficial rocks estimated from river dissolved loads. *Am. J. Sci.* 287, 401–428.
- Meyers, P.A., 1994. Preservation of elemental and isotopic source identification of sedimentary organic matter. *Chem. Geol.* 114, 289–302.
- Milliman, J.D., Meade, R.H., 1983. World-wide delivery of river sediment to the oceans. *J. Geol.* 91, 1–22.
- Nesbitt, H.W., Young, G.M., 1982. Early Proterozoic climates and plate motions inferred from major element chemistry of lutes. *Nature* 299, 715–717.
- Nesbitt, H.W., Young, G.M., 1984. Prediction of some weathering trends of plutonic and volcanic rocks based on thermodynamic and kinetic considerations. *Geochim. Cosmochim. Acta* 48, 1523–1534.
- Nesbitt, H.W., Young, G.M., 1989. Formation and diagenesis of weathering profiles. *J. Geol.* 97, 129–147.
- Nozaki, Y., Lerche, D., Alibo, D.S., Snidvongs, A., 2000. The estuarine geochemistry of rare earth elements and indium in the Chao Phraya River, Thailand. *Geochim. Cosmochim. Acta* 64, 3983–3994.
- Ohta, T., Arai, H., 2007. Statistical empirical index of chemical weathering in igneous rocks: a new tool for evaluating the degree of weathering. *Chem. Geol.* 240, 280–297.
- Ota, Y., Kawahata, H., Murayama, M., Inoue, M., Yokoyama, Y., Miyairi, Y., Aung, T., Hossain, H.M.Z., Suzuki, A., Kitamura, A., Moe, K.T., 2017. Effects of intensification of the Indian Summer Monsoon on northern Andaman Sea sediments during the past 700 years. *J. Quat. Sci.* 32, 528–539.
- Pettijohn, F.J., Potter, P.E., Siever, R., 1972. Sand and Sandstone. Springer-Verlag, New York (618 pp.).
- Ramaswamy, V., Gaye, B., Shirodkar, P.V., Rao, P.S., Chivas, A.R., Wheeler, D., Thwin, S., 2008. Distribution and sources of organic carbon, nitrogen and their isotopic signatures in sediments from the Ayeyarwady (Irrawaddy) continental shelf, northern Andaman Sea. *Mar. Chem.* 111, 137–150.
- Robinson, R.A.J., Bird, M.I., Oo, N.W., Hoey, T.B., Aye, M.M., Higgitt, D.L., Lu, X.X., Swe, A., Tun, T., Win, S.L., 2007. The Irrawaddy river sediment flux to the Indian ocean: the original nineteenth-century data revisited. *J. Geol.* 115, 629–640.
- Robinson, R.A.J., Brezina, C.A., Parrish, R.A., Horstwood, M.S.A., Oo, N.W., Bird, M.I., Thein, M., Walters, A.S.W., Oliver, G.J.H., Zaw, K., 2014. Large rivers and orogens: the evolution of the Yarlung Tsangpo-Irrawaddy system and the eastern Himalayan syntaxis. *Gondwana Res.* 26, 112–121.
- Roser, B.P., Korsch, R.J., 1988. Provenance signatures of sandstone–mudstone suites determined using discriminant function analysis of major-element data. *Chem. Geol.* 67, 119–139.
- Rudnick, R.L., Gao, S., 2005. Composition of the continental crust. pp. 1–64. In: *The Crust* (ed. R.L. Rudnick), Vol. 3, *Treatise on Geochemistry* (eds H.D. Holland and K.K. Turekian), Elsevier-Pergamon, Oxford.
- Sampei, Y., Matsumoto, E., 2001. C/N ratios in a sediment core from Nakaumi Lagoon, southwest Japan—usefulness as an organic source indicator. *Geochem. J.* 35, 189–205.
- Sarin, M.M., Krishnaswami, S., Dilli, K., Somayajulu, B.L.K., Moore, W.S., 1989. Major ion chemistry of the Ganga-Brahmaputra river system: weathering processes and fluxes to the Bay of Bengal. *Geochim. Cosmochim. Acta* 53, 997–1009.
- Schlünz, B., Schneider, R.R., 2000. Transport of terrestrial organic carbon to the oceans by rivers: re-estimating flux and burial rates. *Int. J. Earth Sci.* 88, 599–606.
- Schoenborn, W.A., Fedo, C.M., 2011. Provenance and paleoweathering reconstruction of the Neoproterozoic Johnnie Formation, southeastern California. *Chem. Geol.* 285, 231–255.
- Selvaraj, K., Chen, C.-T.A., 2006. Moderate chemical weathering of subtropical Taiwan: constraints from solid-phase geochemistry of sediments and sedimentary rocks. *J. Geol.* 114, 101–116.
- Taylor, S.R., McLennan, S.M., 1985. *The Continental Crust: Its Composition and Evolution*. Blackwell Scientific, Oxford (312 p.).
- Ugidos, J.M., Valladares, M.I., Recio, C., Rogers, G., Fallack, A.E., Stephens, W.E., 1997. Provenance of Upper Precambrian–Lower Cambrian shales in the Central Iberian Zone, Spain: evidence from a chemical and isotopic study. *Chem. Geol.* 136, 55–70.
- Whalen, J.B., Currie, K.L., Chappell, B.W., 1987. A-type granites: geochemical characteristics, discrimination and petrogenesis. *Contrib. Mineral. Petrol.* 95, 407–419.
- Wronkiewicz, D.J., Condle, K.C., 1987. Geochemistry of Archean shales from the Witwatersrand Supergroup, South Africa: source-area weathering and provenance. *Geochim. Cosmochim. Acta* 51, 2401–2416.