

Geochemical characterization of clay minerals in surface sediments of three major rivers along the east coast of Peninsular Malaysia

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Abstract: Clay mineral assemblages and major-element geochemistry of surface sediments of the tropical river-estuary system of Kelantan, Terengganu and Pahang Rivers were investigated. Clay minerals in these three major rivers mainly consist of kaolinite (72%-75%), illite (13%-20%), chlorite (7%-10%) and minor smectite (<1%). A change in clay mineral contents (chlorite+illite) from upstream to the downstream (kaolinite) pointed to the alteration sequence of more stable mineral under the hot and humid conditions. The geochemical study indicated that the concentrations of major elements decrease from the middle course to estuary. Elemental ratios suggest that CaO and Na₂O are the most chemically mobile elements while Fe₂O₃ is the least for the three investigated rivers. Formation of clay minerals (Al-rich) occur with enrichment of quartz (Si-rich) and feldspar (Na-rich). The illite chemistry index in these river basins averages 0.49 and is supported by the high chemical index of alteration (CIA) (>80) which shows that intensive chemical weathering had occurred in the Kelantan River, Terengganu River and Pahang River basins. The CIA values increase both north and southward directions from the Terengganu River pointing to the localized variations. The CIA values in clay fraction (<2 μ m) generally increased from the middle to the lower watershed.

Keywords: clay minerals, geochemistry, chemical weathering, tropical rivers, Peninsular Malaysia

INTRODUCTION

Geochemistry and mineralogy research on clay minerals are useful for evaluating the continental weathering process and mechanism through their geochemical and mineralogy compositions (Singh *et al.*, 2005; Liu *et al.*, 2007a, 2009). Distinct clay mineral composition can also be used as a tool to identify terrigenous contributions to estuarine and coastal deposits (Singh *et al.*, 2005).

The east coast of Peninsular Malaysia has three main rivers that flow into the South China Sea, i.e. Kelantan, Terengganu and Pahang Rivers. The Kelantan River basin is located at the north-eastern part of Peninsular Malaysia. The river is about 248 km long and drains an area of 13,100 km² (Ibbitt *et al.*, 2002). The Terengganu River basin covers approximately 5,000 km² of the State of Terengganu (Sultan & Shazili, 2010). The Pahang River is the largest river basin in Peninsular Malaysia. Its length is approximately 440 km and its basin area is about 25,600 km² (Tachikawa *et al.*, 2004). Among these rivers, the Pahang River basin produces the highest suspended solid load in Peninsular Malaysia as a result of active chemical weathering and erosion processes induced by high rainfalls and temperature and also its basin size (Sathiamurthy, 2008). This study compared the surface sediments clay mineralogy and major-element geochemistry between the middle course and estuary of the Kelantan, Terengganu and Pahang Rivers. Earlier papers were limited to estuary data only (Wang *et al.*, 2011; Liu *et al.*, 2012) and explained regional distribution of clay mineral assemblages around the South China Sea.

Geologically, Peninsular Malaysia is divided into three belts; Western, Central, and Eastern Malaya, each of which is

distinct in geology and tectonic history (Hutchinson, 1989). The study area is located in the central and eastern belts, an amalgamation of continental terranes, and is underlain with clastics, carbonates, granitoid bodies and volcanics ranging in age from Carboniferous to Quaternary. The Main Range Granite acts a natural divider of upper catchments of these rivers. The upper catchments of Kelantan and Pahang Rivers are dominated by interbedded sandstone, siltstone, shale and volcanics. Phyllite, slate and shale are the common lithologies of the Terengganu River catchment area. Granites are abundant forming elongated north-south trending bodies. The river drainage is characterized by a dense network of streams, a manifestation of tropical (humid and hot) conditions and a variability in lithology. Seasonal flooding in the East Coast rivers remove and transport sediments to the South China Sea, especially during the Northeast Monsoons.

METHODOLOGY

The Ponar grab sampler was used for the collection of surface sediments (penetration depth of 10 to 15 cm) from a boat in the middle and also near to the bank of a river. Sediment samples were collected at three major river sites in August 2009 and sampling locations are shown in Figure 1. The sediment samples were wet sieved (Hathway, 1955) into two sizes; <63 μ m (bulk-fraction) and <2 μ m (clay-fraction). Bulk-fraction sediments were de-carbonated using 1% HCl and washed repeatedly to neutral pH. Clay-fraction sediments were obtained from deflocculated suspensions, according to Stoke's Law and concentrated using the centrifugation technique. The resultant pastes were mounted onto glass

slides and dried. Their clay minerals composition was identified using X-ray diffraction (XRD) technique. XRD analyses were performed on each mount after different pre-treatments; air-drying, 24-hour ethylene-glycol salvation and 2-hour heating at 460°C. Identification of clay minerals on each mount was done by referring to the position of (001) series of basal reflections on XRD diagrams obtained under different treatments. Semi-quantitative computations of smectite (including mixed layers) (15-17 Å), illite (10 Å) and kaolinite/chlorite (7 Å) peak areas were done based on glycolated curve using the MacDiff software. Relative percentages of kaolinite and chlorite were distinguished according to the ratio from 3.57/3.54 Å peak areas. The illite

chemistry index which referred to the ratios of 0.5 and 1 nm peak areas, was also determined from the glycolated curve.

Major elements were measured in clay (<2 µm) and bulk-fraction (<63 µm) of sediments using the inductively coupled plasma-optical emission spectrometer (ICP-OES). The de-carbonated and separated clay (<2 µm) and bulk-fraction (<63 µm) of the sediments were oven dried at 60°C. Dried samples were further heated at 600°C for obtaining the loss on ignition (LOI) values. The same samples were digested with HNO₃ and HF (USEPA method 3052, 1996). Percentages of SiO₂ were determined by subtracting concentrations of other major elements and loss on ignition (LOI) values. Elemental ratio and chemical index of alteration (CIA) were determined from major element analysis.

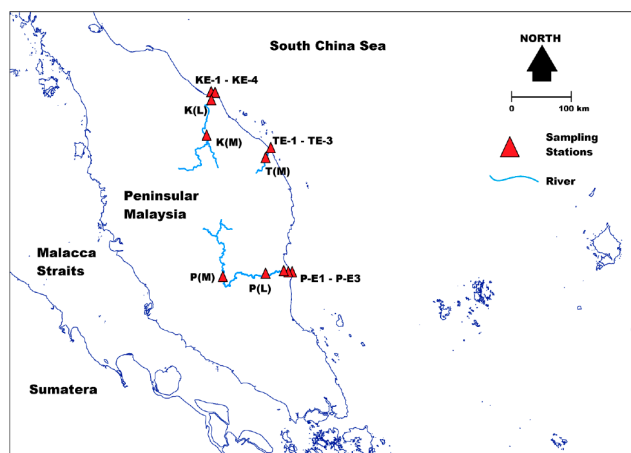


Figure1: Sampling area of the Kelantan River, Terengganu River and Pahang River along the East Coast Peninsula Malaysia. Note: K, T and P – Kelantan, Terengganu and Pahang River; M, L and E – middle course, lower course and estuary.

Table1: Clay mineral distribution and illite chemistry index of sediments of Kelantan, Terengganu and Pahang Rivers. Distance (km) is from estuary to the sampling location.

	Location	Distance (km)	Smectite	Illite	Chlorite	Kaolinite	Illite chemistry index
Pahang River	P (M)	174	0.31	21.42	26.57	51.70	0.42
	P (L)	43	0.64	20.18	16.05	63.13	0.36
	P-E1	0	1.00	16.00	2.00	81.00	0.49
	P-E2	0	1.00	16.00	2.00	81.00	0.44
	P-E3	0	1.00	18.00	0.00	81.00	0.39
	Average	-	0.79	18.32	9.32	71.57	0.42
	STDEV	-	0.31	2.45	11.59	13.54	0.05
Kelantan river	K (M)	97	0.05	20.74	21.30	57.91	0.41
	K (L)	8	0.02	24.75	19.30	55.93	0.39
	K-E1	0	0.00	17.00	1.00	82.00	0.41
	K-E2	0	1.00	18.00	0.00	81.00	0.45
	K-E3	0	0.00	13.00	1.00	85.00	0.49
	K-E4	0	0.00	22.00	1.00	77.00	0.38
	Average	-	0.18	19.25	7.27	73.14	0.42
STDEV	-	0.40	4.14	10.12	12.84	0.04	
Terengganu River	T (M)	25	0.26	12.20	30.57	56.97	0.50
	T-E1	0	0.00	11.00	8.00	81.00	0.55
	T-E2	0	0.00	17.00	1.00	81.00	0.52
	T-E3	0	0.00	9.00	1.00	80.00	0.48
	Average	-	0.07	12.30	10.14	74.74	0.51
	STDEV	-	0.13	3.40	14.01	11.86	0.03

RESULT AND DISCUSSION

Clay minerals

Table 1 shows the percentages of clay minerals in the Kelantan River, Terengganu River and Pahang River. The settling of kaolinite and smectite is associated with their particle size (Whitehouse *et al.*, 1960; Edzwald & O'Melia, 1974; Patchineelam & Neto, 2007). Kaolinite starts to flocculate with the increase of salinity at the lower course of a river. Flocculated kaolinite particles would be deposited within river system because of its larger particle size. In comparison, smectite has higher stability (lower tendency to flocculate) than kaolinite. Hence, it remains suspended in the estuary. Smectite is flocculated when salinity reaches marine values (Hover *et al.*, 1999) (around 35 ppt) and

gets deposited on the sea bed resulting in a smectite rich composition. With reference to Table 1, >70% kaolinite were deposited at the lower course to estuary section.

Tectonics, landform, climatic conditions and lithology of parent rocks in drainage basins play a significant role in the weathering processes (Wang *et al.*, 2011). Table 2 shows a significant difference in clay minerals composition between the Malay Peninsula and Borneo. Although both regions are experiencing the East Asian Monsoon, the weathering products are significantly different due to the variation in lithologies and different weathering mechanism (Liu *et al.*, 2012). The studied river basins (Rajang, Baram & Trusan) of Borneo are dominated by Cenozoic siliciclastic sediments (sandstone and shale; Tate, 2002). The dominant bedrock types underlying the catchments of investigated rivers are granite and granodiorite for Terengganu and Pahang Rivers and some additional sedimentary rocks such as shale, sandstone and limestone (Tate *et al.*, 2008) for Kelantan River. The bedrocks of these three rivers on the Malay Peninsula experience a generally stable tectonic setting since the Mesozoic and have low relief landscape. However, Borneo has been tectonically active since the Mesozoic and dominated with a more rugged terrain (Commission for the Geological Map of the World, 1975). Hence, the rocks of Borneo undergo more physical weathering whereas in the Malay Peninsula, chemical weathering predominates by generating high contents of SiO_2 , which is the main component for kaolinite formation. Relatively higher physical weathering in Borneo is possibly also due to the nature of bedrocks that are predominantly sedimentary rocks (Tate, 2002) and are mechanically weaker than the granites and metasediments (slate, phyllite and schist) in Peninsular Malaysia. High kaolinite is also due to the contribution from granites, in particular feldspars. The plagioclase and K-feldspar content of granites in Peninsular Malaysia average about 50% to 60% (Cobbings *et al.*, 1992), and they weather to form kaolinite in tropical climate and the residual minerals are mainly quartz. This is supported in research (Sultan and Shazili, 2010) that showed sediments from the Terengganu River basin are sandy loam to sand in texture (72.11%) and consisted mostly quartz.

The illite chemistry index value of <0.40 generally represents Fe-Mg-rich illite which is characteristic of

Table 2: Average clay mineral composition in surface sediments of the study area and other rivers in Malaysia. *Estuarine samples only.

Location	Clay minerals (%)				Source
	Kaolinite	Illite	Chlorite	Smectite	
Kelantan River	73.1	19.3	7.3	0	This study
Terengganu River	74.7	12.3	10.1	0	
Pahang River	71.3	18.3	9.3	0	
Kelantan River	81.25	17.5	0.75	0.25	*Wang <i>et al.</i> , 2011
Terengganu River	80.67	15.67	3.33	0	
Pahang River	81	16.67	1	0.75	
Rajang River	16	50.4	32.8	0.6	
Trusan River	13	78	9	0	
Baram River	11.67	77.33	11	0	

physical weathering while >0.40 indicates Al-rich illite released from strong hydrolysis (Wang *et al.*, 2011). The illite chemistry index of the samples from the three rivers are 0.42-0.51 (Table 1). However, values of the index decrease in the samples from Northwest and Northeast Borneo, i.e. 0.39 and 0.22, indicating decreased chemical weathering and increased physical weathering (Wang *et al.*, 2011). The results show that the Malay Peninsula experienced intensive chemical weathering.

Major elements

Major element oxide contents are given in Table 3. Clay-fraction of sediments generally contained higher concentrations of Al_2O_3 , Fe_2O_3 and P_2O_5 but lower SiO_2 and Na_2O than the corresponding bulk sediments (Figure 2). Correlations between Al_2O_3 (%) (X-axis) and plotted elements (Y-axis) were determined. CaO , Fe_2O_3 , MnO and P_2O_5 in the clay fraction, and Na_2O and SiO_2 in the bulk fraction showed negative correlations. This suggests mineralogical control on CaO and Fe_2O_3 concentrations and the leaching of mobile P_2O_5 and Na_2O elements in weathering processes. On the contrary, K_2O , TiO_2 and MgO in the clay and bulk fraction and Na_2O and SiO_2 in the clay fraction of sediments showed positive correlations (Figure 2). These patterns represent the enrichment of elements from middle course to estuary. Enrichment of Al_2O_3 by fixation to the secondary minerals implies clay minerals formation from upstream to the river mouth.

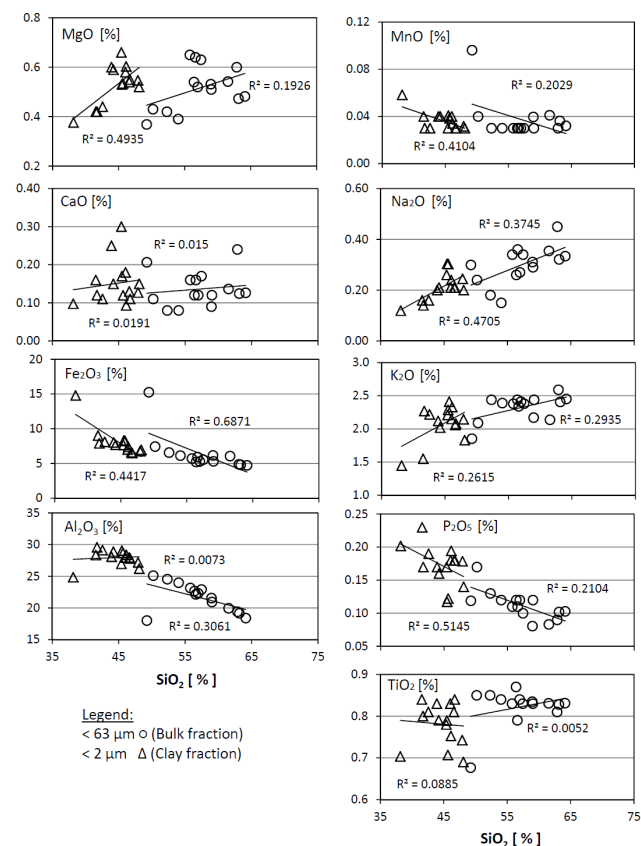


Figure 2: Plots of various element oxides against SiO_2 (%) concentrations in sediments of the clay and bulk size fractions.

Elemental ratio

The mobility of geochemical elements can be known by calculating the ratio of particular elements with reference to the least mobile element such as Al. The ratio of element X and Al_2O_3 in this study divided by the ratio of the same element contained in the Upper Continental Crust (UCC) gives the following equation (Singh *et al.*, 2005):

$$\text{Element ratio (X)} = [X/Al_2O_3 \text{ (this study)}] / [X/Al_2O_3 \text{ (UCC)}]$$

Ratio of >1 indicates enrichment; <1 indicates depletion; $=1$ indicates no changes in the relative abundance of element. Chemical mobility of major elements in the clay and bulk fractions of sediments in this study showed insignificant difference, except for TiO_2 .

The major elements were depleted in the trend of $CaO > Na_2O > MgO > MnO > K_2O > SiO_2 > P_2O_5 > TiO_2 > Fe_2O_3$ in clay-fraction as well as in bulk fraction. As implied,

Fe_2O_3 and SiO_2 were depleted while other elements were enriched from the middle course to estuary in the Kelantan River and the Pahang River. In contrast, Fe_2O_3 was enriched in the Terengganu River for the clay fraction while in bulk fraction, Fe_2O_3 in the Terengganu River showed a drastic decrease from the middle course to the estuary. TiO_2 was clearly enriched ($X=1.16$) through the weathering process for this river.

As a whole, CaO and Na_2O were the most chemically mobile major element oxides in the clay and bulk fractions of sediments whereas Fe_2O_3 was the least mobile. The chemical mobility of Ti, Al, and Fe are mostly stable. This mobility pattern indicates the resistance of each element against chemical weathering intensity in the formation of clay minerals.

Table 3: Element oxides (wt. %), LOI (%) and Chemical index of alteration (CIA) values in sediments of Kelantan, Terengganu and Pahang Rivers. Note: K, T and P – Kelantan, Terengganu and Pahang Rivers; M, L and E – middle course, lower course and estuary.

Particle size $< 2\mu m$ (%)												
Location	Al_2O_3	CaO	Fe_2O_3	K_2O	MgO	MnO	Na_2O	P_2O_5	TiO_2	SiO_2	LOI	CIA
K (M)	28.68	0.12	8.24	2.41	0.53	0.04	0.30	0.12	0.71	45.65	0.13	92.07
K (L)	29.04	0.17	8.32	2.29	0.53	0.04	0.30	0.12	0.79	45.48	0.13	92.41
K-E1	26.21	0.15	6.96	1.83	0.52	0.03	0.20	0.14	0.69	48.09	15.18	93.27
K-E2	28.09	0.25	8.02	2.12	0.60	0.04	0.20	0.17	0.83	43.91	15.78	92.70
K-E3	28.90	0.15	7.69	2.02	0.59	0.04	0.21	0.16	0.79	44.20	15.25	93.31
K-E4	26.98	0.30	7.65	2.21	0.66	0.03	0.26	0.17	0.78	45.40	15.57	91.96
P (M)	28.40	0.09	7.40	2.33	0.60	0.03	0.24	0.19	0.75	46.13	0.14	92.38
P (L)	27.19	0.13	6.76	2.14	0.55	0.03	0.24	0.18	0.74	47.91	0.14	92.53
P-E1	27.81	0.11	6.49	2.08	0.55	0.03	0.21	0.18	0.84	46.72	14.98	92.96
P-E2	27.97	0.13	6.61	2.06	0.54	0.03	0.21	0.18	0.81	46.60	14.86	93.02
P-E3	27.98	0.18	7.01	2.15	0.58	0.04	0.21	0.18	0.83	46.03	48.81	92.69
T (M)	24.86	0.10	14.79	1.45	0.38	0.06	0.12	0.20	0.70	38.16	0.19	94.46
T-E1	28.35	0.16	9.01	1.55	0.42	0.04	0.16	0.23	0.84	41.55	17.69	94.62
T-E2	29.58	0.12	7.90	2.27	0.42	0.03	0.14	0.17	0.80	41.73	16.83	92.95
T-E3	29.14	0.11	8.12	2.22	0.44	0.03	0.16	0.19	0.81	42.57	16.21	92.97
Particle size $< 63\mu m$ (%)												
K (M)	19.97	0.14	6.06	2.14	0.54	0.04	0.35	0.08	0.83	61.54	13.20	89.83
K (L)	21.57	0.09	6.16	2.17	0.53	0.04	0.31	0.08	0.83	58.95	12.90	90.60
K-E1	22.93	0.17	5.55	2.38	0.63	0.03	0.34	0.10	0.83	57.46	9.58	90.19
K-E2	19.42	0.24	4.88	2.59	0.60	0.03	0.45	0.09	0.81	62.87	8.02	87.43
K-E3	23.16	0.16	5.72	2.38	0.65	0.03	0.34	0.11	0.83	55.77	10.86	90.30
K-E4	22.15	0.16	5.91	2.34	0.64	0.03	0.36	0.11	0.79	56.61	10.89	89.99
P (M)	18.42	0.13	4.73	2.45	0.48	0.03	0.33	0.10	0.83	64.12	13.80	87.92
P (L)	19.17	0.12	4.81	2.41	0.47	0.04	0.32	0.10	0.83	63.15	14.10	88.54
P-E1	22.67	0.12	5.21	2.44	0.54	0.03	0.26	0.12	0.87	56.40	11.33	90.17
P-E2	22.32	0.12	5.29	2.41	0.52	0.03	0.27	0.12	0.84	56.96	11.12	90.11
P-E3	20.95	0.12	5.31	2.44	0.51	0.03	0.29	0.12	0.83	59.03	10.39	89.37
T (M)	18.03	0.21	15.26	1.85	0.37	0.10	0.30	0.12	0.68	49.23	19.20	90.00
T-E1	24.00	0.08	6.13	2.39	0.39	0.03	0.15	0.12	0.84	54.02	11.86	91.12
T-E2	25.11	0.11	7.44	2.09	0.43	0.04	0.24	0.17	0.85	50.19	13.33	92.17
T-E3	24.55	0.08	6.57	2.44	0.42	0.03	0.18	0.13	0.85	52.31	12.44	91.09

Chemical index of alteration (CIA)

Chemical weathering intensity was estimated using chemical index of alteration (CIA) stated in the following equation (Nesbitt & Young, 1982):

$$\text{CIA} = [\text{Al}_2\text{O}_3 / (\text{Al}_2\text{O}_3 + \text{CaO}^* + \text{Na}_2\text{O} + \text{K}_2\text{O})] \times 100$$

All the element oxides used for CIA computation are in molecular proportions and CaO^* represents the amount of CaO incorporated in the silicate fraction of sediment. The CIA quantifies the state of chemical weathering of rocks by referring to the loss of alkali and alkaline earth elements such as Na, K and Ca. In general, CIA of 45-55 indicates no weathering (Table 4); less than 60 indicates low chemical weathering; 60-80 indicates moderate chemical weathering; more than 80 indicates strong chemical weathering (Singh *et al.*, 2005). The CIA of the clay and bulk fractions of sediments in this study are shown in Table 3. The results showed CIA of more than 80 for all the clay and bulk fraction of sediments, suggesting strong chemical weathering in the study area. From the middle course to the estuary, the values of CIA were increasing which suggest the chemical weathering intensity was increasing along the rivers. The CIA values for the Pahang River surface sediment is between 87 to 93; the Kelantan River (89-93) and the Terengganu River (90-94). Overall, the chemical weathering degrees are gradually strengthened from Northwest Borneo to Malay Peninsula (Wang *et al.*, 2011) and could be stronger than south China and Indochina Peninsula. The chemical index of alteration increased both north and southward directions from the Terengganu River which registered the highest CIA values. This is mostly likely due to the localized variations in the monsoonal rain intensity and frequency along the east coast of Peninsular Malaysia. The geometric characteristics and the peculiar shape of Peninsular Malaysia with narrowing in the north and south along with other factors (e.g. wind direction, local relief) might be the additional factors contribution to the higher CIA values. The CIA values generally increased from the middle of watershed to the estuarine settings for all the three rivers. (Figure 3). A value of more than 90 for all the clay fractions ($< 2\mu\text{m}$) of sediments was recorded.

A simplified mineral alteration pattern observed in the tropical setting of east coast Peninsular Malaysia is given below (Figure 4). The gradual decrease of illite and chlorite from the upstream watershed to the downstream (Table 1) along the flow path is compensated by increase in kaolinite mineral contents. This apparent systematic change in clay mineral contents indicate alteration with kaolinite as stable product.

However, the most stable end products under the tropical environment are the metal oxides of Al, Fe and Mn (e.g. hematite Fe_2O_3 , gibbsite $\text{Al}(\text{OH})_3$, birnessite MnO_2) which is the result of Si leaching (Sposito, 2008) in kaolinite (and other silicate minerals). While parent rock composition is of importance in the type of clay minerals formed, the climate influence (intense monsoonal rainfall in the east coast Peninsular Malaysia) seems to be decisive factor in controlling the mineral alteration sequence. Among the three

major controls (lithological, tectonic and climatic) upon the clay mineral formations, the tectonic mechanism is the least.

CONCLUSION

The main components of clay mineral assemblages from middle course to estuary of the Kelantan River, the Terengganu River and the Pahang River were dominantly kaolinite minerals followed by illite, chlorite and smectite (scarce). This composition implied active chemical weathering in east coast of the Malay Peninsula. Intensive chemical weathering of parent rocks under the tropical climate resulted in a high abundance of kaolinite. The systematic increase in kaolinite and decrease in chlorite+illite mineral contents along the flow path indicated an alteration to more stable end products towards the South China Sea. The intensity of weathering increased from the upper watershed towards downstream as indicated by increasing Chemical index of alteration (CIA) values. The CIA values increase both north and southward direction from the Terengganu River indicating localized variations. Major element oxides of riverbed sediment were characterized by high contents of SiO_2 , Al_2O_3 , and Fe_2O_3 , accounting for $\geq 80\%$ of the total composition. Elemental ratios suggested that CaO and Na_2O were the most chemically mobile elements whereas Fe_2O_3 was the least mobile element along the rivers. The illite chemistry index and CIA indicated the increase of

Table 4: Chemical index of alteration (CIA).

CIA Value	Weathering intensity
<45	None to very low
45-55	Low
55-60	Moderate
60-80	Moderate to High
>80	Strong

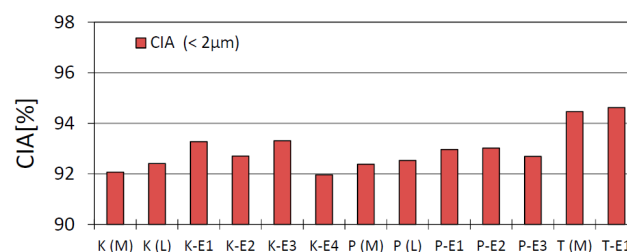


Figure 3: Chemical index of alteration (CIA) of the clay fraction ($< 2\mu\text{m}$) of sediments from the middle (M), lower (L), and estuary (E) locations of the rivers. Note: K, T and P – Kelantan, Terengganu and Pahang Rivers.

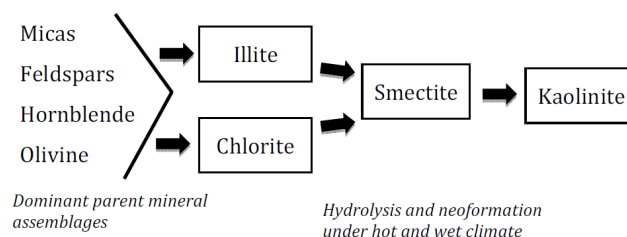


Figure 4: Simplified mineral alteration pattern (Barnhisel & Bertsch, 1989).

chemical weathering intensity from the middle to the lower course of Kelantan, Terengganu and Pahang river basins.

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