Chemical weathering in central Vietnam from clay mineralogy and major-element geochemistry of sedimentary rocks and river sediments

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Abstract

Clay mineralogy and major-element geochemistry of Miocene sedimentary rock and modern river sediment samples collected from the Ba River basin in central Vietnam are used to evaluate the chemical weathering processes during the Miocene and the present time. The results show that Miocene andesitic sedimentary rocks consist of high smectite (average 72%) with moderate kaolinite (24%), while Miocene felsic sedimentary rocks display abundant kaolinite (65%) with moderate smectite (25%). In comparison, modern river sediments are characterized by moderate smectite (43%) and kaolinite (37%). The typical distribution of clay minerals in the Ba River basin can be resulted from abundant occurrence of felsic intrusive rocks and volcanic rocks along with weak tectonic uplift and the tropical East Asian monsoon climate during the Miocene and in the present time. Despite their different clay mineralogical compositions, major elements of these Miocene sedimentary rocks and modern river sediments display stronger depletion of Ca, Na and Mg than of K and Si during the
chemical weathering. The chemical index of alteration (CIA) combined with kaolinite/illite ratio demonstrated moderate chemical weathering during the Miocene and in the present time in central Vietnam, demonstrating similar tectonic activity and climatic conditions occurred during these two periods.

Keywords: Geology, Geochemistry, Geography

1. Introduction

Weathering products of aluminosilicate rocks are widely utilized for assessing continental chemical weathering, because their mineral assemblage and geochemical composition can provide useful information on the mechanisms of chemical weathering (Galy and France-Lanord, 1999; Singh et al., 2005; Colin et al., 2006; Liu et al., 2007). The South China Sea is the largest marginal sea in the western Pacific, and it is supplied with more than $700 \times 10^6$ t/yr suspended sediments mainly by adjacent rivers (Liu and Stattegger, 2014; Liu et al., 2016, Fig. 1). These weathering products formed in different lithological and tectonic settings as well as under different regional climatic conditions. The study of these river sediments may contribute in understanding the alteration of parent rocks and land-sea interactions. The mechanism of weathering processes in adjacent river drainage basins relates strongly to the variation of terrigenous sediments in the South China Sea (Boulay et al., 2003; Liu et al., 2004; Colin et al., 2010; Clift et al., 2014). As a result, many studies have been carried out on river sediments surrounding this area to evaluate weathering processes. These studies include the Pearl, Red, Mekong, and Gianh rivers in South China and Indochina Peninsula (Liu et al., 2007; Jonell et al., 2016), major rivers in Hainan Island (Hu et al., 2014), mountainous rivers in Taiwan (Selvaraj and Chen, 2006; Liu et al., 2008), major rivers in Luzon, Philippines (Liu et al., 2009), Malay Peninsula, Borneo and Sumatra (Wang et al., 2011; Liu et al., 2012). Nevertheless, few research has been conducted on weathering products in central Vietnam, which is characterized by a complex lithological setting, including abundant intrusive and volcanic rocks, along with the tropical East Asian monsoon climate with humid climate and warm temperature. Only small mountainous rivers develop in central Vietnam but they play a significant role in contributing terrigenous sediments to the central Vietnam shelf (Schimanski and Stattegger, 2005). Consequently, central Vietnam is a fascinating area to evaluate the mechanism of chemical weathering on igneous rocks.

The Ba River is the largest river in central Vietnam with $14 \times 10^3$ km² drainage area and 390 km length (Figs. 1 and 2). The Ba River supplies $1 \times 10^6$ t/yr suspended sediments to the South China Sea (Milliman and Farnsworth, 2011). The highland part of the Ba River drainage basin lies mainly on the central highlands of Vietnam, which includes series of contiguous plateaus with an average elevation of about
800 m. In this area, granitization processes took principally place during the Paleo-proterozoic and Mesoproterozoic (Lan et al., 2003). The lithology of the Ba River drainage basin consists mainly of intrusive and extrusive igneous rocks and minor sedimentary rocks (Fig. 2A). Precambrian metamorphic rocks contain various granulitic rocks, and they are characterized by variably deformed or undeformed Paleo-Mesozoic granitic intrusions, which are mainly granite, granodiorite, and diorite (Luong and Bao, 1988; Nam, 1998; Osanai et al., 2001; Lan et al., 2003; Nakano et al., 2007; Lepvrier et al., 2008). Mesozoic extrusive rocks cover widely this research area during Late Triassic-Early Jurassic and Cretaceous, including mostly rhyolite, dacite, andesite, and felsite (Nam et al., 2001). Sedimentary rocks appear

Fig. 1. Fluvial drainage systems and their annual suspended sediment discharge to the South China Sea (modified from Liu et al., 2016), showing the location of the Ba River basin (squared) in central Vietnam. Arrows with numbers indicate observed fluvial sediment discharge (Mt/yr).
restrictively along the Ba River during the Middle-Late Miocene, and they contain conglomerate, grit-stone, sandstone, siltstone, clay-stone and lignite (Nielsen et al., 2007, Fig. 2A). Miocene sedimentary rocks are characterized by three facies associations: (1) fluvial channel and terminal lobe association, (2) mouth bar association, and (3) lake-mire association. Basaltic rocks largely cover this area during the Neogene-Quaternary (Hoang and Flower, 1998; Carter et al., 2000; Hoang et al., 2013). Holocene sediments have distributed mainly along rivers and coastline. The Ba River drainage basin is characterized by the tropical East Asian monsoon climate, with alternative wet and dry seasons. Temperature and rainfall variations in Nha Trang (Fig. 1) are available from the world climate data (http://www.worldclimate.com) with an average annual rainfall of 1300 mm (Fig. 3). The rainy season lasts from September to December with average monthly rainfall of 249 mm (nearly 75%), and temperature of 26.0 °C. The dry season is represented by low rainfall (monthly average ~41 mm, nearly 25%) and similar temperature (monthly average 26.9 °C) between January and August.

In this study, Miocene sedimentary rocks and modern river sediments were collected from the Ba River basin in central Vietnam to investigate clay mineralogy and major-element geochemistry. Clay-mineralogical proxies (clay mineral contents

Fig. 2. (A) Geological map of the Ba River drainage basin in central Vietnam, modified from the 1:500,000 Geological Map of Vietnam (Luong and Bao, 1988). 1. Holocene deposits; 2. Neogene-Quaternary basalts; 3. Miocene sedimentary rocks; 4. Mesozoic extrusive rocks; 5. Paleo-Mesozoic felsic intrusive rocks; 6. Precambrian metamorphic rocks; 7. rivers. (B) Locations of samples used in this study and their average clay mineral assemblages; shaded area shows the Ba River drainage basin. VS. Miocene andesitic sedimentary rock samples; FS. Miocene felsic sedimentary rock samples; RS. modern river sediment samples.

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and kaolinite/illicite ratio) and major-element results were used to assess chemical weathering processes during the Miocene and the present time in central Vietnam.

2. Materials & methods

Twenty-seven sedimentary rock samples and seven modern river sediment samples were collected from the Ba River basin in central Vietnam in November 2015 (Table 1; Fig. 2B). The sedimentary rock samples were collected from Miocene sedimentary rocks (Song Ba formation), and they were separated into two types: Miocene andesitic sedimentary rock samples (clastic grains mainly derived from andesitic-rhyolitic volcanic rocks-VS), and Miocene felsic sedimentary rock samples (clastic grains mainly derived from felsic intrusive igneous rocks-FS). The modern river sediment samples (muddy channel deposits-RS) were collected at different locations along the Ba River in order to represent typical sub-environments (Fig. 2B). Among them, two samples (MN13 and MN14) from the lower reach of main channel represent average compositions of the Ba River sediments; while three samples (MN04, MN06, and MN15) from the main channel and other two samples (MN16 and MN29) from the branch of the middle reach represent the highland environment of the Ba River basin. Contamination from riverbank sediments and potential man-made pollution were avoided when collecting these river sediment samples. Clay mineralogy and major-element geochemistry were analyzed for all sediment samples at the State Key Laboratory of Marine Geology, Tongji University.
Table 1. Geographic locations and clay mineral assemblages of Miocene sedimentary rock and modern river sediment samples in the Ba River basin.

| No. | Sample | Location                     | Latitude (N) | Longitude (E) | Smectite (%) | Illite (%) | Chlorite (%) | Kaolinite (%) | Smectite crystallinity (°2θ) |
|-----|--------|------------------------------|--------------|---------------|--------------|------------|--------------|---------------|-----------------------------|
| 1   | MN01   | Phu My                       | 13°21.354'   | 109°13.117'   | 82           | 0          | 0            | 17            | 1.01                        |
| 2   | MN02   | Kien Thiet                   | 13°05.971'   | 108°52.828'   | 97           | 1          | 0            | 1             | 0.88                        |
| 3   | MN03-2 | Chu Se mountain pass         | 13°36.531'   | 108°14.896'   | 61           | 1          | 0            | 37            | 0.72                        |
| 4   | MN05   | Cay Sung bride               | 13°20.866'   | 108°28.784'   | 84           | 1          | 0            | 15            | 0.75                        |
| 5   | MN17   | Phu Can bridge               | 13°11.085'   | 108°39.297'   | 72           | 5          | 0            | 23            | 0.89                        |
| 6   | MN18   | Phu Can bridge               | 13°11.085'   | 108°39.297'   | 64           | 6          | 0            | 31            | 0.90                        |
| 7   | MN19   | Phu Can bridge               | 13°11.085'   | 108°39.297'   | 63           | 5          | 0            | 31            | 1.08                        |
| 8   | MN20   | Phu Can bridge               | 13°11.085'   | 108°39.297'   | 71           | 4          | 0            | 25            | 0.88                        |
| 9   | MN21   | Phu Can bridge               | 13°11.085'   | 108°39.297'   | 79           | 4          | 0            | 17            | 0.94                        |
| 10  | MN22   | Phu Can bridge               | 13°11.085'   | 108°39.297'   | 59           | 9          | 0            | 32            | 0.99                        |
| 11  | MN23   | Phu Can bridge               | 13°11.085'   | 108°39.297'   | 68           | 5          | 0            | 28            | 0.96                        |
| 12  | MN24   | Phu Can bridge               | 13°11.085'   | 108°39.297'   | 75           | 4          | 0            | 21            | 0.97                        |
| 13  | MN25   | Phu Can bridge               | 13°11.085'   | 108°39.297'   | 61           | 3          | 0            | 36            | 1.21                        |
| 14  | MN26   | Phu Can bridge               | 13°11.085'   | 108°39.297'   | 62           | 4          | 0            | 34            | 1.07                        |
| 15  | MN27   | Phu Can bridge               | 13°11.085'   | 108°39.297'   | 65           | 3          | 0            | 32            | 1.11                        |
| 16  | MN28   | Phu Can bridge               | 13°11.085'   | 108°39.297'   | 66           | 3          | 0            | 31            | 1.17                        |

(continued on next page)
| No. | Sample | Location             | Latitude (N) | Longitude (E) | Smectite (%) | Illite (%) | Chlorite (%) | Kaolinite (%) | Smeectite crystallinity (°Δ2θ) |
|-----|--------|----------------------|--------------|---------------|--------------|------------|--------------|--------------|-------------------------------|
| 16  | MN02   | Phu Tuc              | 13° 10.925'  | 108° 42.000'  | 17           | 7          | 0            | 76           | 1.70                          |
| 17  | MN03-1 | Chu Se mountain pass | 13° 36.531'  | 108° 14.896'  | 48           | 2          | 0            | 50           | 0.85                          |
| 18  | MN07   | Phu Tuc              | 13° 10.925'  | 108° 42.000'  | 2            | 9          | 16           | 75           | 0.61                          |
| 19  | MN08   | Phu Tuc              | 13° 10.925'  | 108° 42.000'  | 17           | 7          | 0            | 76           | 1.70                          |
| 20  | MN09   | Phu Tuc              | 13° 10.925'  | 108° 42.000'  | 19           | 5          | 0            | 77           | 1.46                          |
| 21  | MN10   | Phu Tuc              | 13° 10.925'  | 108° 42.000'  | 33           | 7          | 0            | 60           | 0.96                          |
| 22  | MN11   | Phu Tuc              | 13° 10.925'  | 108° 42.000'  | 34           | 4          | 0            | 62           | 1.30                          |
| 23  | MN12   | Phu Tuc              | 13° 10.925'  | 108° 42.000'  | 46           | 4          | 0            | 50           | 0.97                          |
| 24  | MN30   | Cay Sung bride       | 13° 20.884'  | 108° 27.644'  | 4            | 18         | 0            | 78           | 0.31                          |
| 25  | MN31   | Quynh Phu            | 13° 17.965'  | 108° 37.111'  | 8            | 6          | 13           | 74           | 0.67                          |
| 26  | MN32   | Quynh Phu            | 13° 17.965'  | 108° 37.111'  | 46           | 4          | 0            | 49           | 1.80                          |
| 27  | MN33   | Quynh Phu            | 13° 17.965'  | 108° 37.111'  | 24           | 10         | 0            | 66           | 0.94                          |
|     |        |                      |              |               |              |            |              |              |                               |
|     |        | Modern river sediments (RS) |           |               |              |            |              |              |                               |
| 28  | MN04   | Buon Hiao            | 13° 22.266'  | 108° 27.907'  | 46           | 8          | 6            | 41           | 1.16                          |
| 29  | MN06   | Buon Toat            | 13° 18.332'  | 108° 35.922'  | 47           | 10         | 9            | 35           | 1.01                          |
| 30  | MN13   | Dinh Tho             | 13° 01.431'  | 109° 13.761'  | 35           | 12         | 12           | 41           | 1.14                          |
| 31  | MN14   | Hoa Hoi              | 13° 00.308'  | 109° 07.036'  | 32           | 14         | 12           | 43           | 1.01                          |
| 32  | MN15   | Phu Can              | 13° 11.139'  | 108° 39.378'  | 50           | 9          | 0            | 41           | 1.11                          |
| 33  | MN16   | Phu Tuc              | 13° 20.056'  | 108° 37.782'  | 56           | 11         | 10           | 23           | 1.07                          |
| 34  | MN29   | Kim Tan              | 13° 30.711'  | 108° 28.140'  | 39           | 13         | 11           | 37           | 1.03                          |
Clay minerals were identified using the PANalytical X’Pert PRO X-ray Diffractometer on oriented mounts of non-calcareous clay-sized (<2 µm) particles. Bulk sediment samples were treated with 0.2 N HCl and 10% H2O2 to remove carbonate and organic matter, respectively. This study used the method described in Liu et al. (2004) to make the oriented mounts. Three XRD runs were carried out following air drying, ethylene-glycol solvation for 24 hours, and heating at 490 °C for 2 hours. The position of the (001) series of basal reflections on the three XRD diagrams were used to identify clay minerals. Base on the glycolated curve, the MacDiff software (Petschick, 2000) was used to perform semi-quantitative estimates of peak areas of the basal reflections for the main clay mineral groups of smectite (including mixed layers) (15−17 Å), illite (10 Å), and kaolinite/chlorite (7 Å) (Holtzapfel, 1985). The ratio of the 3.57/3.54 Å peak areas was used to determine relative proportion of kaolinite and chlorite. Base on XRD diagrams, the smectite crystallinity was obtained from half height width of the 17 Å peak on the glycolated curve. The smectite crystallinity values were classified into the categories well crystalline (<1.5°Δ20), moderately crystalline (1.5−2.0°Δ20), and poorly crystalline (>2.0°Δ20) (Ehrmann et al., 2005).

Major elements were measured by using a PANalytical AxiosmAX wavelength dispersive X-ray Fluorescence (XRF) spectrometer. Samples were dried and crushed to fine powder by using an agate mortar. Each sample was then coned to ensure that the powdered sample was well mixed, and subsequently compressed with phosphoric acid powder (H3PO4) into a disc for analysis. The phosphoric acid is used as a cover on the outside of the sediment sample. Chinese rock and sediment standards GSR06 and GSD15 were used to control the analytical precision and accuracy. Major element oxides (i.e., SiO2, Al2O3, Fe2O3, MgO, CaO, K2O, Na2O, P2O5, TiO2, and MnO) were obtained. Besides, CaO associated with the silicate fraction was obtained through correcting bulk CaO using P2O5 (McLennan, 1993).

3. Results

Miocene andesitic sedimentary rock samples (VS) consist of high smectite (59−97%, average 70%), moderate kaolinite (1−37%, average 26%), and low illite (0−9%, average 4%) (Table 1; Fig. 2B). Miocene felsic sedimentary rock samples (FS) are represented by abundant kaolinite (49−78%, average 65%) and moderate smectite (2−48%, average 25%), with low illite (2−18%, average 7%) and scarce chlorite (0−16%, average 3%) (Table 1; Fig. 2B). Modern river sediment samples (RS) are characterized by moderate smectite (32−56%, average 43%) and kaolinite (23−43%, average 37%), with low illite (8−14%, average 11%) and chlorite (0−12%, average 9%) (Table 1; Fig. 2B). The smectite crystallinity shows low values in Miocene andesitic sedimentary rock samples (0.72−1.21°Δ20), Miocene felsic sedimentary rock samples (0.31−1.80°Δ20), and modern river sediment samples (1.01−1.16°Δ20) (Table 1).
Major-element compositions of Miocene andesitic sedimentary rock samples (VS), Miocene felsic sedimentary rock samples (FS), and modern river sediment samples (RS) are characterized by dominant SiO$_2$ and Al$_2$O$_3$ (their total average content is 88%), and by low K$_2$O, Fe$_2$O$_3$, Na$_2$O, MgO, CaO, P$_2$O$_5$, TiO$_2$, and MnO (their total average content is 8.5%) (Table 2). The major-element results indicate that Miocene sedimentary rocks usually contain higher SiO$_2$ and K$_2$O, and lower Al$_2$O$_3$, Fe$_2$O$_3$, TiO$_2$, MgO, Na$_2$O, MnO, CaO, and P$_2$O$_5$ than modern river sediments. The diagrams of Al$_2$O$_3$ versus SiO$_2$ and K$_2$O display relatively moderate to strong negative correlations in most of the samples (Fig. 4), suggesting an increase of Al$_2$O$_3$, but decreases of SiO$_2$ and K$_2$O during chemical weathering. The weathering products tend to gradually increase clay minerals (Al-rich mostly) and decrease quartz (Si-rich mostly) and K-feldspar (K-rich mostly) particles. Furthermore, moderate to strong positive correlations in most of the samples are found in diagrams of Al$_2$O$_3$ versus Fe$_2$O$_3$, MgO and TiO$_2$, implying enrichment of immobile elements Fe, Ti and Mg during chemical weathering. The diagrams of Al$_2$O$_3$ versus MnO and CaO are moderate positive correlations in modern river sediment samples, while no correlations in both Miocene andesitic and felsic sedimentary rock samples.

4. Discussion

4.1. Formation of clay minerals

Clay mineral assemblage in a river basin is generally related to the weathering process, which is principally influenced by climatic condition (rainfall and temperature), rock composition, and tectonic activity (Chamley, 1989; Liu et al., 2007). Clay minerals are weathering products of parent rocks on the Earth’s surface. The chemical weathering changes the composition of parent rocks by hydrolyzing the minerals. Smectite and kaolinite are typical in weathering products of this process. Smectite is usually related to chemical weathering of volcanic rocks. Volcanic rocks are weathered faster than most other rocks under warm and humid climate conditions (Bluth and Kump, 1994; Dessert et al., 2001). Volcanic rocks usually lead to high smectite that forms easily on basic materials as Fe-Mg species, and on rhyolitic materials rather as Al species (Chamley, 1989). Like the upper part of middle reach and the lower reach of the Mekong River drainage basin and Luzon in tropical climates (Liu et al., 2004, 2009), where volcanic rocks are dominant, smectite is yielded abundantly (Fig. 5). Kaolinite formation can be referred to as monosialitization of parent rocks, representing the intensive hydrolysis under warm and humid climate (Chamley, 1989). Kaolinite can be easily produced by parent rocks, which contain rich alkali and alkaline elements (e.g., granite, granodiorite and rhyolite) (Chamley, 1989). The Pearl River drainage basin in subtropical climate, Malay Peninsula in tropical climate, and Hainan in tropical climate contain abundant felsic intrusive rocks and/or felsic extrusive rocks, and kaolinite is formed prevaiingly
Table 2. Major element composition (%) and CIA of Miocene sedimentary rock and modern river sediment samples in the Ba River basin.

| No. | Sample | Al₂O₃ | CaO  | Fe₂O₃ | K₂O  | MgO  | MnO  | Na₂O  | P₂O₅ | TiO₂ | SiO₂ | CIA |
|-----|--------|-------|------|-------|------|------|------|-------|------|------|------|-----|
|     |        |       |      |       |      |      |      |       |      |      |      |     |
| 1   | MN01   | 14.60 | 0.145| 2.54  | 3.29 | 0.64 | 0.01 | 0.40  | 0.03 | 0.56 | 74.43| 77  |
| 2   | MN02   | 17.14 | 0.606| 4.52  | 2.54 | 1.35 | 0.08 | 1.17  | 0.02 | 0.50 | 67.19| 75  |
| 3   | MN03-2 | 29.81 | 0.326| 3.18  | 1.86 | 0.59 | 0.01 | 0.12  | 0.02 | 0.50 | 61.97| 93  |
| 4   | MN05   | 18.64 | 1.561| 4.13  | 1.94 | 0.59 | 0.01 | 0.60  | 0.02 | 0.76 | 70.48| 82  |
| 5   | MN17   | 11.92 | 0.278| 1.06  | 3.76 | 0.35 | 0.01 | 0.96  | 0.02 | 0.27 | 79.12| 66  |
| 6   | MN18   | 16.20 | 0.417| 1.87  | 3.51 | 0.50 | 0.02 | 0.92  | 0.02 | 0.60 | 74.14| 73  |
| 7   | MN19   | 12.38 | 0.314| 1.25  | 4.36 | 0.34 | 0.02 | 1.49  | 0.02 | 0.26 | 75.54| 62  |
| 8   | MN20   | 16.80 | 0.46 | 1.87  | 3.43 | 0.52 | 0.01 | 0.95  | 0.02 | 0.63 | 71.49| 73  |
| 9   | MN21   | 14.04 | 0.336| 1.51  | 4.03 | 0.44 | 0.02 | 1.22  | 0.01 | 0.38 | 74.82| 67  |
| 10  | MN22   | 10.23 | 0.274| 1.58  | 3.84 | 0.27 | 0.02 | 1.38  | 0.04 | 0.20 | 77.29| 60  |
| 11  | MN23   | 13.79 | 0.32 | 1.28  | 3.85 | 0.39 | 0.02 | 1.58  | 0.02 | 0.35 | 74.36| 65  |
| 12  | MN24   | 15.98 | 0.373| 2.00  | 3.66 | 0.60 | 0.02 | 1.27  | 0.03 | 0.60 | 70.99| 71  |
| 13  | MN25   | 14.48 | 0.246| 1.13  | 4.22 | 0.28 | 0.01 | 1.16  | 0.01 | 0.26 | 74.31| 68  |
| 14  | MN26   | 13.16 | 0.197| 0.87  | 3.81 | 0.24 | 0.01 | 1.01  | 0.01 | 0.28 | 77.12| 68  |
| 15  | MN27   | 16.13 | 0.333| 1.38  | 3.91 | 0.35 | 0.02 | 0.92  | 0.02 | 0.53 | 71.75| 72  |
| 16  | MN28   | 15.73 | 0.27 | 1.56  | 4.19 | 0.29 | 0.01 | 0.98  | 0.02 | 0.28 | 72.86| 70  |

Miocene felsic sedimentary rocks (FS)

| 17  | MN03-1 | 30.09 | 0.439| 2.22  | 2.14 | 0.41 | 0.01 | 0.09  | 0.03 | 0.61 | 62.71| 92  |
| 18  | MN07   | 22.54 | 0.173| 4.39  | 2.77 | 0.34 | 0.01 | 0.27  | 0.03 | 0.79 | 66.21| 86  |
| 19  | MN08   | 12.04 | 0.219| 1.43  | 3.82 | 0.20 | 0.01 | 0.15  | 0.02 | 0.75 | 80.16| 72  |
| 20  | MN9    | 17.96 | 0.227| 3.82  | 3.09 | 0.31 | 0.01 | 0.32  | 0.03 | 0.68 | 71.71| 81  |
| 21  | MN10   | 16.22 | 0.187| 0.82  | 3.97 | 0.29 | 0.01 | 0.20  | 0.02 | 0.57 | 76.68| 77  |
| 22  | MN11   | 13.57 | 0.161| 0.69  | 4.04 | 0.25 | 0.01 | 0.19  | 0.01 | 0.34 | 79.33| 73  |
| 23  | MN12   | 19.73 | 0.268| 1.88  | 3.49 | 0.38 | 0.01 | 0.26  | 0.02 | 0.61 | 72.17| 81  |
| 24  | MN30   | 8.54  | 0.077| 1.06  | 3.08 | 0.15 | 0.01 | 0.09  | 0.00 | 0.28 | 85.24| 70  |
| 25  | MN31   | 11.35 | 0.099| 0.56  | 2.92 | 0.19 | 0.01 | 0.13  | 0.01 | 0.70 | 82.43| 76  |
| 26  | MN32   | 13.21 | 0.203| 1.22  | 4.23 | 0.26 | 0.01 | 0.21  | 0.01 | 0.31 | 78.80| 71  |
| 27  | MN33   | 15.07 | 0.188| 0.75  | 5.17 | 0.23 | 0.01 | 0.20  | 0.02 | 0.46 | 76.42| 71  |

Modern river sediments (RS)

| 28  | MN04   | 18.45 | 1.299| 7.98  | 2.59 | 1.20 | 0.18 | 0.99  | 0.24 | 1.51 | 56.72| 75  |
| 29  | MN06   | 14.85 | 0.89 | 3.94  | 3.19 | 0.67 | 0.12 | 0.96  | 0.13 | 0.80 | 67.40| 70  |
| 30  | MN13   | 16.50 | 1.088| 4.33  | 3.04 | 0.75 | 0.15 | 1.32  | 0.19 | 0.82 | 61.53| 70  |
| 31  | MN14   | 18.78 | 1.00 | 6.94  | 2.55 | 0.97 | 0.24 | 0.83  | 0.21 | 1.08 | 60.32| 78  |
| 32  | MN15   | 16.41 | 0.868| 4.87  | 3.38 | 0.73 | 0.15 | 1.03  | 0.14 | 0.99 | 66.73| 71  |
| 33  | MN16   | 18.44 | 1.128| 5.83  | 3.33 | 0.88 | 0.29 | 1.73  | 0.13 | 0.87 | 62.90| 69  |
| 34  | MN29   | 17.31 | 0.877| 4.90  | 3.84 | 0.73 | 0.10 | 1.29  | 0.15 | 1.01 | 63.60| 70  |
Fig. 4. Variation diagrams of major elements of Miocene andesitic sedimentary rocks (VS), Miocene felsic sedimentary rocks (FS), and modern river sediments (RS) in the Ba River basin. Dashed lines indicate correlation with a coefficient $R^2_{VS}$ for VS, $R^2_{FS}$ for FS, and $R^2_{RS}$ for RS. Data of upper continental crust (UCC) from Taylor and McLennan (1985) were plotted as a reference.

(Liu et al., 2007, 2012, 2016, Fig. 5). Nevertheless, physical weathering leads to rock fragmentation without much change in the chemical components, and this can form illite and chlorite (Chamley, 1989). Illite and chlorite are considered primary minerals, and they are related strongly to the weak hydrolysis and/or strong physical erosion of bedrocks under relatively cold and dry climatic conditions. Similar situation occurs in highland part of the Mekong and Red River drainage basins in subtropical climate, Taiwan in tropical climate, and North Borneo in tropical climate (Liu et al., 2004, 2007, 2008, 2012), where the bedrocks yield abundant illite and chlorite under strong physical erosion.
The high-land part of the Ba River drainage basin is situated on the series of contiguous plateaus, where soil has developed strongly. Furthermore, this area characterized by relatively weak tectonic activity since the Miocene (Lan et al., 2003; Lepvrier et al., 2008). These indicate that the parent rocks in the Ba River drainage basin have not been eroded significantly since the Miocene. Besides, this area presents the tropical East Asian monsoon climate with heavy rainfall during September-December and warm temperatures throughout the year (Fig. 3). The East Asian monsoon climate during the Miocene was warmer in temperature than in the present time (Wei et al., 2006; Wan et al., 2007). The morphology, tectonic activity, and climate conditions during the Miocene and in the present time allow to develop mainly chemical weathering in the Ba River drainage basin, leading to the increase of secondary clay minerals (e.g., smectite and kaolinite). The smectite content is high in Miocene andesitic sedimentary rock samples (70%), moderate in modern river sediment samples (43%), and low in Miocene felsic sedimentary rock samples (25%) (Figs. 2B and 5). The abundant occurrence of Mesozoic extrusive rocks and Neogene-Quaternary basaltic rocks in the Ba River drainage basin could be weathered to form mainly smectite for Miocene sedimentary rocks and modern river sediments.

Fig. 5. Ternary diagram of clay mineral assemblages of Miocene andesitic sedimentary rocks (VS), Miocene felsic sedimentary rocks (FS), and modern river sediments (RS) in the Ba River basin. Average data of Pearl, Red and Mekong rivers (Liu et al., 2007), Taiwan rivers (Liu et al., 2008), Luzon rivers (Liu et al., 2009), North Boneo and Malay Peninsula rivers (Liu et al., 2012), and Hainan rivers (Liu et al., 2016) are plotted for comparison.
sediments. Volcanic rocks can generate abundantly fairly well to very well crystallized smectite (Chamley, 1989). Our samples display low values of smectite crystallinities (0.31–1.80°Δ20, average 1.03°Δ20) (Table 1), indicating the fairly-well to very-well crystallized smectite (Ehrmann et al., 2005). This suggests that smectite in the Ba River drainage basin was formed from volcanic rocks under temperate-warm climate. Therefore, the widespread distribution of volcanic rocks along with weak tectonic activity and warm climate conditions since the Miocene could result in abundant smectite content in the Ba River basin sediments. Besides, the kaolinite content is abundant in Miocene felsic sedimentary rock samples (65%), moderate in modern river sediment samples (37%), and low in Miocene andesitic sedimentary rock samples (26%) (Figs. 2B and 5). The Ba River drainage basin contains prevailing felsic intrusive rocks (e.g., granodiorite and granite) and felsic extrusive rocks (e.g., rhyolite, dacite, and felsite) (Fig. 2A), which can be potential parent rocks for producing high kaolinite in the weathering products. The combination of climatic condition and geological setting encourages significant chemical weathering processes of monosialitization and alitization. Therefore, the study area contains adequate climate conditions (warm and humid climate), weak tectonic activity and parent rocks enriched alkali and alkaline elements for resulting abundant kaolinite in the Ba River basin sediments. Nevertheless, all Miocene sedimentary rock samples and modern river sediment samples are represented by low illite and chlorite contents (Figs. 2B and 5). The weak tectonic activity and warm climate conditions since the Miocene have strongly decreased the physical erosion, preventing the formation of high illite and chlorite contents in the Ba River basin sediments. Consequently, the low contributions of illite and chlorite in all samples are likely related to the weak physical erosion in the Ba River drainage basin during the Miocene and the present time.

Indochina Peninsula supplies a large number of fluvial sediment to the central Vietnam shelf and the western South China Sea by the world’s largest rivers (e.g., Mekong and Red rivers) and central Vietnam mountainous small rivers (Liu et al., 2007, 2016; Milliman and Farnsworth, 2011). The distribution of clay minerals in the Ba River shows different compositions from those of the Mekong and Red rivers in the Indochina Peninsula (Fig. 5). The Ba River displays moderate smectite (43%) and kaolinite (37%), with low illite (11%) and chlorite (9%). In the Mekong and Red rivers, the clay mineral assemblages contain mainly illite (35–43%), with similar moderate kaolinite and chlorite (24–28%) and low smectite (6–11%) (Liu et al., 2007). The Vietnamese shelf and the western South China Sea contain large amounts of terrigenous sediments from the Indochina Peninsula (Liu et al., 2013; Schroeder et al., 2015; Liu et al., 2016). As a result, this studied area can be an end-member for clay mineral source analysis in the central Vietnam shelf as well as in the western South China Sea.
4.2. Chemical weathering trend

During chemical weathering, mobile elements (e.g., K, Na, and Ca) are gradually depleted from parent rocks, but other elements (e.g., Al, Fe, and Ti) are relatively enriched in the weathering products (Nesbitt et al., 1980). Consequently, the major element composition of weathering products of silicate rocks can be utilized to identify the altered pathway and to evaluate chemical weathering in a river basin (Nesbitt et al., 1980; Vital and Stattegger, 2000; Dessert et al., 2001; Singh et al., 2005). To identify and evaluate the major element mobility, we use elemental ratios calculated with respect to the least mobile element Al. The ratios of the content of element X and Al₂O₃ in studied samples divided by the ratio of the same element content in upper continental crust (UCC) can be obtained by the following elemental ratio (Singh et al., 2005):

\[
\text{Elemental ratio (X)} = \frac{X/\text{Al}_2\text{O}_3 (\text{sample})}{X/\text{Al}_2\text{O}_3 (\text{UCC})}
\]

The ratio refers to the relative enrichment or depletion of an element, i.e., >1 indicates enrichment, <1 indicates depletion, and =1 indicates no change in the relative abundance of the element (Singh et al., 2005).

The average elemental ratios of Miocene andesitic sedimentary rock samples and Miocene felsic sedimentary rock samples show similar chemical mobility (Fig. 6). However, major elements from two kinds of Miocene sedimentary rock samples present different chemical mobility in modern river sediment samples. Miocene sedimentary rock samples are more strongly depleted in all major elements than

![Fig. 6. Elemental ratios of Miocene andesitic sedimentary rock samples (VS), Miocene felsic sedimentary rock samples (FS), and modern river sediments (RS) in the Ba River basin, calculated from average major element concentrations normalized to UCC (Taylor and McLennan, 1985) with respect to Al₂O₃. The error bar for each element refers to the standard deviation of all samples.](https://doi.org/10.1016/j.heliyon.2018.e00710)
modern river sediment samples, except Si and K, implying that Miocene sedimentary rocks may contain minerals with high Si and K contents (e.g., quartz, micaeous, and K-feldspar). This conclusion is consistent with the low illite in all samples because of K usually is rich in illite. Additionally, Miocene sedimentary rock samples show lower elemental ratios of Ca, Mg, and Na than modern river sediment samples, suggesting that Miocene sedimentary rocks were derived more materials from felsic rocks (Paleo-Mesozoic felsic intrusive igneous rocks and Mesozoic extrusive rocks) than modern river sediments. The Ti and Mn are enriched, the Fe is stable, and P is slightly depleted in modern river sediment samples, while all of these elements are leached in Miocene sedimentary rock samples, implying that modern river sediments contain ferromagnesian minerals. These conclusions fit very well with the occurrence of parent rocks in the Ba River drainage basin. Miocene sedimentary rocks could be only generated from materials of Precambrian metamorphic rocks, Paleo-Mesozoic felsic intrusive igneous rocks, and Mesozoic extrusive rocks, while modern river sediments can be derived from materials not only from these rocks but also from Miocene sedimentary rocks and Neogene-Quaternary basaltic rocks (Fig. 2A). In this study area, the intensity of relative depletion is: Ca > P > Mg > Na > Mn > Fe > Ti > K > Si for Miocene sedimentary rock samples, and Ca > Na > Mg > P > K > Si > Fe > Ti > Mn for modern river sediment samples. These observations imply that alkali metal (Na) and alkaline earth metal (Ca, Mg) elements are more easily depleted from parent materials during chemical weathering than other major elements. According to similar strong depletions of Na, Ca, and Mg in all samples along with little leaching of Si and K in modern river sediment samples, and the stable Si and K contents in Miocene sedimentary rock samples suggest that the difference of chemical weathering states during the Miocene and the present time is not significant in the Ba River drainage basin.

The chemical index of alteration (CIA) is used to evaluate the degree of chemical weathering (Nesbitt and Young, 1982). The CIA has been utilized by many previous studies (McLennan, 1993; Galy and France-Lanord, 1999; Singh et al., 2005; Selvaraj and Chen, 2006; Liu et al., 2007). The CIA values are calculated as the formula (Nesbitt and Young, 1982):

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

where CaO* is the amount of CaO incorporated in the silicate fraction of parent rocks (Nesbitt and Young, 1989; Nesbitt et al., 1996). Based on the loss of mobile elements such as Na, Ca, and K against immobile Al, the CIA values are calculated to assess chemical weathering states for the rocks (Nesbitt and Young, 1982). The CIA values between of 45–55 infer no chemical weathering, values less than 60 mean low chemical weathering, values between 60-80 imply moderate chemical weathering, and values more than 80 indicate intensive chemical weathering.

Results from this study show that CIA values are 60–93 (average 71) for Miocene andesitic sedimentary rock samples, 71–92 (average 77) for Miocene felsic
sedimentary rock samples, and 69–78 (average 72) for modern river sediment samples (Table 2). Two kinds of Miocene sedimentary rock samples display wider variations in CIA values than modern river sediments. Generally, volcanic rocks are weathered more easily than intrusive rocks, implying higher CIA values in volcanic weathering products. However, Miocene andesitic sedimentary rock samples show an average 6 lower CIA values than Miocene felsic sedimentary rock samples. This situation can be caused by different ages of their source rocks because weathering products of older rocks may be weathered longer. Nevertheless, the averages of CIA values are not significantly different between Miocene andesitic sedimentary rock samples (average 71) and Miocene felsic sedimentary rock samples (average 77), suggesting that the degree of chemical weathering in this area was not strongly influenced by source rocks. Besides, moderate to strong linear correlations of CIA with kaolinite/illite and Al/Rb occur for all Miocene sedimentary rock samples and modern river sediment samples (Fig. 7). A higher CIA value corresponds to higher kaolinite/illite and Al/Rb, implying stronger chemical weathering. The results indicate moderate chemical weathering during the Miocene as well as in the present time. During the Miocene, the degree of chemical weathering is higher than in the present time. This may be correspondent to the warmer climate conditions during the Miocene than in the present time (Wei et al., 2006; Wan et al., 2007). To get a good general observation on chemical weathering surrounding the South China Sea, we compared our data with those from adjacent areas in the region. In the Ba River basin, the chemical weathering intensity displays a lower degree than in the Pearl River (Liu et al., 2007) and Sumatra rivers (Liu et al., 2012), where the chemical weathering is intensive. On the other hand, the Ba River basin shows quite similar chemical weathering states as in the Mekong and Red rivers (Liu et al., 2007), where the chemical weathering is moderate. Nevertheless, our study area has much stronger chemical weathering than those in Taiwan (Selvaraj and Chen, 2006; Liu et al., 2008) and Luzon (Liu et al., 2009), where the major elemental and clay mineralogical data present low to moderate chemical weathering.

Weathering trends in both Miocene sedimentary rocks and modern river sediments in the Ba River basin can be clearly investigated on Al₂O₃-(CaO* + Na₂O)-K₂O (A-CN-K) and Al₂O₃-(CaO* + Na₂O + K₂O)-(Fe₂O₃# + MgO) (A-CNK-FM) ternary diagrams (CaO* is the amount of CaO incorporated in the silicate fraction; Fe₂O₃ presents total Fe in the sample) (Nesbitt and Young, 1989; Nesbitt and Young, 1984, Fig. 8). Most of our samples plot less close to the Al₂O₃ apex than reference data of the Pearl and Mekong (Liu et al., 2007) and Sumatra rivers (Liu et al., 2012), while closer to the Al₂O₃ apex than reference data of Taiwan (Liu et al., 2008) and Luzon rivers (Liu et al., 2009), but similar to reference data of the Red River (Liu et al., 2007) (Fig. 8A). These observations indicate that the weathering trend in the Ba River basin is different from most of adjacent river basins surrounding the South China Sea, excluding the Red River basin. All Miocene felsic sedimentary
rock samples (FS) are plotted linearly parallel to the A-K or A-CNK line, and much closer to the A-K or A-CNK line and Al$_2$O$_3$ apex, implying enrichment of Al$_2$O$_3$ and abundant K$_2$O, while CaO, Na$_2$O, MgO and Fe$_2$O$_3$ contents (less abundant) are constant (Fig. 8). The distribution of Miocene andesitic sedimentary rock samples (VS) is linearly parallel to the A-CN or A-CNK line, closer to the Al$_2$O$_3$ apex and the A-K or A-CNK line, indicating the leaching of CaO and Na$_2$O, enrichment of Al$_2$O$_3$ and dominant K$_2$O, but MgO and Fe$_2$O$_3$ (less abundant) are stable (Fig. 8). These features indicate that Miocene sedimentary rocks could be created principally by material of felsic igneous rocks (Paleo-Mesozoic felsic intrusive igneous rocks and

![Graph](https://example.com/graph.png)

**Fig. 7.** Correlations of chemical index of alteration (CIA) with (A) Al/Rb molar ratio and (B) kaolinite/illite of Miocene andesitic sedimentary rocks (VS), Miocene felsic sedimentary rocks (FS), and modern river sediments (RS) in the Ba River basin. The graphs show linear correlations of mineralogical and element geochemical proxies of chemical weathering.
Mesozoic extrusive rocks). However, all modern river sediment samples are linearly parallel to the A-CN or A-CNK line and closer to FM apex, showing the leaching of CaO and Na₂O, less abundant K₂O, while MgO and Fe₂O₃ are constant (Fig. 8). These observations indicate that modern river sediments can include weathering products of mafic igneous rocks (Neogene-Quaternary basaltic rocks) and felsic igneous rocks (Paleo-Mesozoic felsic intrusive igneous rocks and Mesozoic extrusive rocks).

Fig. 8. (A) CIA, A-CN-K, and (B) A-CNK-FM ternary diagrams of Miocene andesitic sedimentary rock samples (VS), Miocene felsic sedimentary rock samples (FS), and modern river sediments (RS) in the Ba River basin are plotted for comparison. A = Al₂O₃; C = CaO*; N = Na₂O; K = K₂O; F = total Fe; M = MgO. Average data for Pearl, Red and Mekong rivers (Liu et al., 2007), Taiwan rivers (Liu et al., 2008), Luzon rivers (Liu et al., 2009), and Sumatra rivers (Liu et al., 2012) are plotted for comparison; UCC (Taylor and McLennan, 1985) was also plotted as a reference. Arrows indicate weathering trends exhibited by these sediments.
extrusive rocks). Besides, Miocene sedimentary rock samples and modern river sediment samples show much different weathering trends, implying that modern river sediments may not impact strongly by sedimentary contribution of Miocene sedimentary rocks. The conclusions on source rocks of Miocene sedimentary rocks and modern river sediments are also confirmed by comparison with weathering trend of UCC, the Pearl, Red and Mekong rivers, and Taiwan and Luzon rivers (Liu et al., 2007, 2008, 2009, Fig. 8). The weathering trends of Miocene sedimentary rocks are much close to the A-K and A-CNK line, while much different the Pearl, Red, and Mekong rivers, and Taiwan and Luzon rivers, suggesting abundant weathering products of felsic igneous rocks. However, the weathering trends of the river sediments are much far from the A-K and A-CNK line, but close to the Pearl, Red, and Mekong rivers, and Taiwan and Luzon rivers, coinciding with appearance of weathering products from both mafic and felsic igneous rocks in river sediments.

5. Conclusions

Clay mineralogy and major-element geochemistry have been studied on Miocene sedimentary rocks and modern river sediments in the Ba River basin to estimate the chemical weathering processes in central Vietnam.

1. The clay mineral assemblages of Miocene andesitic sedimentary rocks display abundant smectite (average 70%), moderate kaolinite (average 26%), and low illite (average 4%). Miocene felsic sedimentary rocks characterize dominant kaolinite (average 65%), moderate smectite (average 25%), with low illite (average 7%) and chlorite (average 3%). Moderate smectite (average 43%) and kaolinite (average 37%) with low illite (average 11%) and chlorite (average 9%) occur in modern river sediments.

2. The abundant Paleo-Mesozoic felsic intrusive rocks, Mesozoic extrusive rocks, and Neogene-Quaternary basaltic rocks along with the weak tectonic activity and the tropical East Asian monsoon climatic condition in the Ba River drainage basin are responsible for the prevailing smectite and kaolinite with low illite and chlorite in the clay mineral assemblage of Miocene sedimentary rocks and modern river sediments.

3. The elemental mobility and weathering trends of Miocene sedimentary rocks and modern river sediments demonstrate that Miocene sedimentary rocks could be formed mainly from felsic igneous rocks, meanwhile modern river sediments can be derived by materials of both mafic and felsic igneous rocks. The dominant appearances of smectite and kaolinite are associated with leaching of Ca, Na and Mg first and of K and Si thereafter during chemical weathering. In the Ba River drainage basin, the good correlations of CIA with kaolinite/illite and Al/Rb indicate moderate chemical weathering during the Miocene and in the present time.
Declarations

Author contribution statement

Pham Nhu Sang: Performed the experiments; Analyzed and interpreted the data; Contributed reagents, materials, analysis tools or data; Wrote the paper.

Zhifei Liu: Conceived and designed the experiments; Analyzed and interpreted the data; Contributed reagents, materials, analysis tools or data; Wrote the paper.

Yulong Zhao, Xixi Zhao, Phan Dong Pha, Hoang Van Long: Contributed reagents, materials, analysis tools or data.

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Competing interest statement

The authors declare no conflict of interest.

Additional information

No additional information is available for this paper.

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