RESEARCH ARTICLE
doi: 10.2306/scienceasia1513-1874.2007.33.013

Fine-fraction Clays from Chiang Muan Mine, Phayao Province, Northern Thailand

Yupa Thasod[a*], Benjavun Ratanasthien[a], Satoshi Tanaka[b], Haruo Saegusa[c] and Hideo Nakaya[d]

[a] Department of Geological Sciences, Faculty of Science, Chiang Mai University, Chiang Mai 50200, Thailand.
[b] Department of Earth Sciences, Kyoto University of Education, Kyoto 612-8522, Japan.
[c] Museum of Nature and Human Activities, Sanda, Hyogo 669-1546, Japan.
[d] Department of Earth and Environmental Sciences, Faculty of Science, Kagoshima University, Kagoshima, 890-0065, Japan.

* Corresponding author, E-mail: thasod@yahoo.com

Received 24 Dec 2004
Accepted 11 Sep 2006

Abstract: Fine-fraction clays from the Chiang Muan mine in Phayao Province, northern Thailand, were studied using the x-ray diffraction method. The analysis determined the parent rocks and depositional environments of clays. Clay minerals in this area are subdivided into three zones of I to III. In zone I, montmorillonite is dominant, followed by kaolinite and illite. These clay minerals were derived from Jurassic rhyolite, tuffaceous shale, and sandstone in the southern part of the Chiang Muan basin. Kaolinite and illite are dominant in zones II and III. These minerals are the alteration products of feldspar and mica and probably had the same origin as those of zone I. Montmorillonite is abundant in the Underburden unit of mine but it is much less in the Lower coal zone unit. This suggests that the climate changed about 13 million years ago. Later the amount of kaolinite and illite increased because of much weathering.

These clay minerals assemblages indicate that the area in which they were deposited had changed from a dry temperate to a tropical climate and a high meteoric water supply. The paleocurrent direction in the area was northward during initial deposition as suggested by clay mineral assemblage. The present current in the area flows from north to south.

Keywords: clay minerals, depositional environments, Chiang Muan mine, x-ray diffraction.

Introduction

Clay minerals are fine-grained and hydrous materials. They occur in every place and are most abundant mineral in sedimentary rocks[1]. The study of clay minerals in the Chiang Muan open-pit coal mine was conducted to determine the parent rocks and depositional environments of the clays. Sediments are composed of interbedded coal, sandstone, siltstone, and mudstone. The coal reserves are approximately 10 million tons and used for electricity generation in the Mae Moh power plants.

The mine is located in Chiang Muan District, Phayao Province, in northern Thailand. It is 300 meters wide and more than 1 kilometer long. The Chiang Muan basin is a small Cenozoic basin (Fig. 1)[2]. The Cenozoic basins in Thailand are mainly north–south trending half grabens and grabens that were initiated in the Oligocene[3]. The collision of the Indian plate with the Eurasian plate 40 to 50 million years ago caused clockwise rotation of Southeast Asia, movement on strike slip faults, and development of Cenozoic basins in this region[3]. The structural style of the Chiang Muan

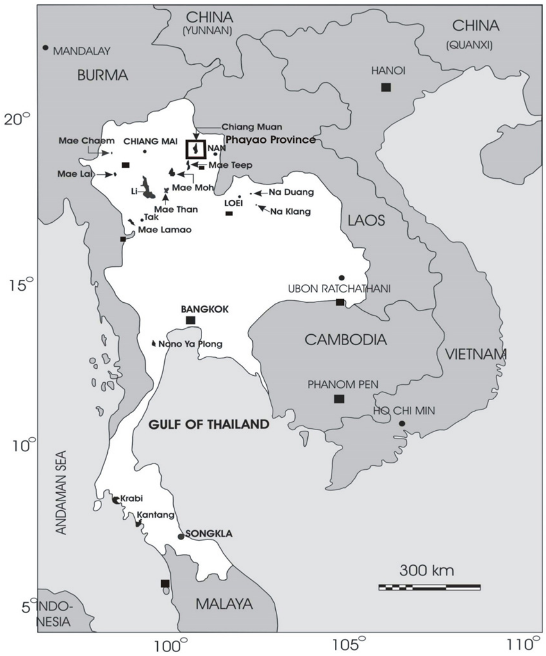

Fig 1. Map of Thailand showing Neogene deposits with coal and oil shale (modified from Ratanasthien, 2002[2]).

mine is simple. Bedding strikes north-south in the southern part of the mine and northeast in the northern part. The oldest strata are exposed on the western margin. The youngest strata are on the eastern margin and dip east to southeast. Minor normal faults are common these show the strike of north-south to east northeast–west southwest[4]. Exposed faults indicate that the north-south faults formed at the first stage. These faults occur in the main coal seams. Superimposed on a few of these faults are weak to well-developed low-angle striations that indicate north northeast-south southwest movement. At the northern end of the mine, an important normal fault oriented east northeast-west southwest has up thrown the youngest overburden and deeply down thrown the main coal seam[4].

An associated mammalian fauna and paleomagnetic analysis indicate that the strata in the Chiang Muan mine are latest Middle Miocene in age, about 13.5 to 10 million years [5,6,7,8].

Materials and Methods

Stratigraphic Sequence

The Chiang Muan basin is bounded by a sequence of Jurassic reddish bed formations that form the north, south, and west flanks of the basin[9] (Figure 2). The lower Jurassic unit is purplish to pale grayish rhyolite, tuffaceous shale, and sandstone facies. The middle Jurassic unit is fine-grained calcareous sandstone interbedded with shale and conglomerate facies. The upper Jurassic unit is sandstone interbedded with micaceous and tuffaceous shale, and conglomerate facies. Quaternary sediments in the basin consist of gravel, sand, silt, clay, mud, and lateritic soil[9].

The Chiang Muan basin contains Tertiary sandstone, claystone, carbonaceous claystone, and coal. These strata are divided into seven units as follows: Underburden, Lower coal zone, Interburden 2, Upper coal zone 2, Interburden 1, Upper coal zone 1, and Overburden units (Fig. 3).

Underburden Unit (UB)

This is the lowest unit and unconformably overlies Jurassic red beds. It consists of sandstone, pebbly sandstone, clayey sandstone, sandy claystone, and claystone, and is moderately reddish brown and light gray to yellowish gray. The unit is generally more than 2 meters in thickness. The claystone will swell and soften under the wet condition.

Lower Coal Zone Unit (L)

The lower part of the unit, which is the lower split coal seam sub-unit, is carbonaceous claystone, coal, and silty claystone. It is brownish black and its thickness

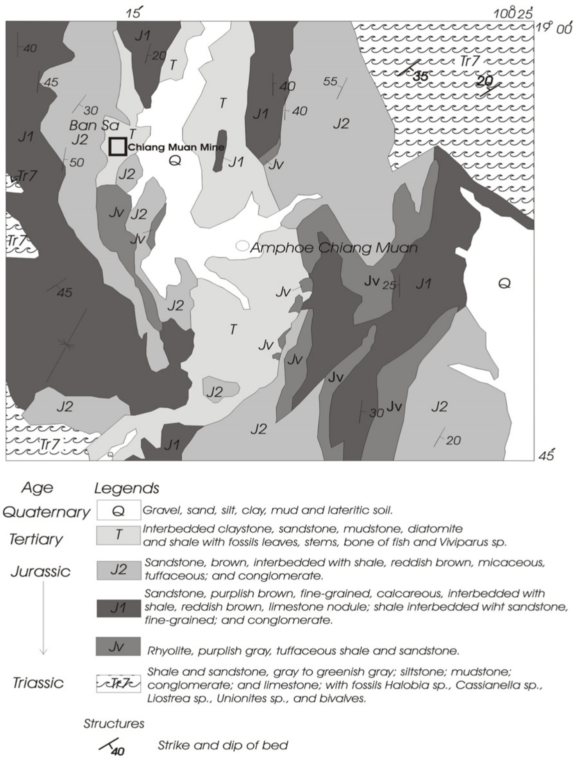

Fig 2. Geological map of the Chiang Muan basin and surrounding rock units (modified from Charoenprawat *et al.*, 1995[9]).

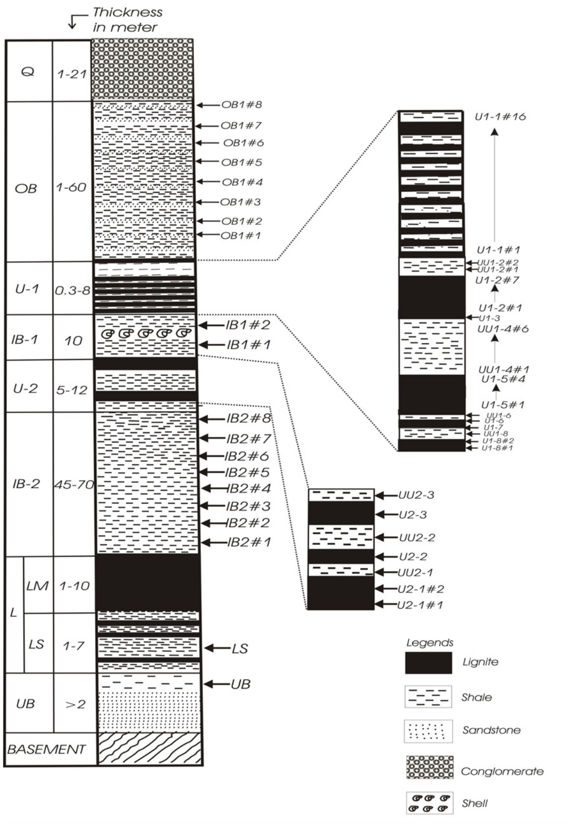

Fig 3. The Chiang Muan mine stratigraphic column and sample locations.

varies from 1 to 7 meters. The upper part, which is the lower massive coal seam sub-unit, is massive, moderately bright, hard and brittle coal. The thickness varies from 1 to 10 meters.

Interburden 2 Unit (IB2)

This unit is a very hard, thick sequence of reddish brown to light gray clayey sandstone and sandy claystone. The sandstone is fine- to coarse-grained. The thickness of the unit varies from 45 to 70 meters. The unit is a prominent oxidized zone and it will swell under the wet conditions.

Upper Coal Zone 2 Unit (U2)

The unit includes coal, carbonaceous claystone, claystone, and silty claystone. It is brownish black and light gray, and its thickness varies from 5 to 12 meters. Vertebrate remains and fossil seeds are common. The unit has a high sulfur content in the form of pyrite lenses and pyrite replacing plant structure in coal. Some of the pyrite is oxidized to sulphate that forms gypsum.

Interburden 1 Unit (IB1)

This unit is interbedded light gray sandy claystone, silty claystone and clayey sandstone. The sandstone is fine- to coarse-grained. The thickness of this unit is approximately 10 meters. Fossil leaves and fruit remnants are abundant, and molluscan fossils are observed in some horizons.

Upper Coal Zone 1 Unit (U1)

This unit is a carbonaceous claystone and coal. There are three main layers of carbonaceous claystone that are intercalated with light gray claystone. However, in the northern part of the mine, there are nine layers. The thickness is 0.3 to 8 meters. A high sulfur content occurs in some places in the form of pyrite and gypsum.

Overburden Unit (OB)

This unit is a series of claystone, sandy claystone, and silty claystone intercalations. Its lower part is light to deep greenish gray and the upper part is yellowish brown to gray. Its thickness varies 10 to 60 meters. This zone has the features for paleosols like as soil color horizons, root traces and fossil burrows.

The Tertiary sequence is capped by Quaternary deposits that are mainly brownish red to orange gravel, sand and some clay.

Sample Collection and Preparation

Seventy-one samples were collected by the channel sampling method. Their color and texture were determined. Sample descriptions are listed in Table 1 and sample locations are shown in Fig. 3.

Each sample was oven-dried overnight at 40°C, crushed to less than 5 millimeters, and then quartered by riffling. One sample quarter was ground into powder. The bulk composition was analyzed. The oriented samples were prepared by suspending each sample in a 1000-ml cylinder for 3 hours and 36 minutes. The particles in all samples were less than 2 micrometers in diameter[10]. 40 ml of each sample was pipetted at 5 centimeter depth from the tip of pipette, transferred to a beaker containing a glass plate, and left to air-dry. The oriented samples were ready for x-ray diffraction analysis. Those oriented samples were later treated by ethylene glycol by placing the samples in a desiccator containing 1-cm-deep of ethylene glycol at the base of

Table 1. Description and thickness of samples collected from Chiang Muan mine.

Sample no.	Description	Thickness (m)
UB	Mudstone with coal fragment, brownish gray.	1
LS	Coal accumulated from leaves, grayish black.	1
IB2#1	Gravelly sandstone, yellowish gray; gravel composed of feldspar, chert, quartz and rock fragments, angular to sub-angular; sandstone fine- to coarse-grained, poorly sorted, mottered by red spots.	5
IB2#2	Gravelly sandstone, yellowish gray, poorly sorted, very fine- to coarse- grained, mottered by olive.	5
IB2#3	Sandy siltstone, yellowish gray, moderate sorted, mottered by moderate yellowish brown.	5
IB2#4	Sandstone, pale olive, well sorted, angular to sub-angular, quartz, feldspar, rock fragment, mottered by dark yellowish orange.	5
IB2#5	Sandy siltstone, yellowish gray, fine to medium grained, moderate sorted, Fe- concretion, rooted, mottered by moderate brown.	5
IB2#6	Gravelly siltstone, yellowish gray to light olive brown; gravel: sub-angular, rock fragment, root traces.	5
IB2#7	Gravelly sandstone, moderate sorted, yellowish gray; gravel: angular, quartz, rock fragment; sandstone: quartz, rock fragments, feldspar, well sorted, sub-angular to sub-round.	5
IB2#8	Sandy siltstone, yellowish gray, root traces, mottered by yellowish orange.	5
U2-1#1	Coal, grayish black, clay parting, pyrite.	0.5
U2-1#2	Clay to carbonaceous clay, dark yellowish brown, root traces, Fe-concretion, coal fragments.	1
UU2-1	Siltstone, grayish orange to dark yellowish orange, mottered by yellowish orange, Fe-concretion, coal fragments.	1
U2-2	Coal, clay parting, grayish black, leaves accumulation, soft.	0.5

Table 1. Cont'd.

Sample no.	Description	Thickness (m)
UU2-2	Carbonaceous claystone, olive gray.	0.5
U2-3	Coal, grayish black, soft, brittle.	0.5
UU2-3	Mudstone, light olive gray, root traces, coal fragments, mottered by dark yellowish orange.	1
IB1#1	Mudstone, greenish gray, mottered by dark yellowish orange, root traces.	3
IB1#2	Sandstone, fine-grained, greenish gray, well sorted, quartz, fledspar and rock fragments.	3
U1-9	Coal, black to brownish black, dense, accumulated from tree trunks and leaves.	0.3
U1-8#1	Coal, grayish black, brittle.	0.3
U1-8#2	Coal, grayish black, accumulated from leaves.	0.2
UU1-8	Mudstone, greenish gray to yellowish gray, mottered by brown, Fe-concretion, rooted.	0.2
U1-7	Coal, grayish black, dense.	0.2
U1-6	Coal, grayish black, soft, accumulated from leaves.	0.2
UU1-6	Sandy mudstone, mottered by pale olive/moderate yellow, poorly sorted, feldspar, quartz, rock fragments, rooted, Fe-concretion.	0.2
U1-5#1	Coal, grayish black to black, accumulated from leaves.	0.2
U1-5#2	Carbonaceous clay to coal, brownish black.	0.3
U1-5#3	Carbonaceous clay and clay, dark gray.	0.2
U1-5#4	Coal, grayish black, clay parting.	0.3
UU1-5	Carbonaceous claystone, brownish gray.	0.2
UU1-4#1	Mudstone, greenish gray.	0.3
UU1-4#2	Silty sandstone with coal fragment, grayish orange/grayish black, mottered by moderate brown.	0.3
UU1-4#3	Sandstone, fine-grained, quartz, feldspar, yellowish gray/dark yellowish orange, mottered, Fe-concretion, root traces.	0.3
UU1-4#4	Mudstone, pale olive, mottered by light brown/dark yellowish orange, root traces.	0.3
UU1-4#5	Mudstone, greenish gray, rooted.	0.2
UU1-4#6	Siltstone, light yellowish gray, mottered, roots traces.	0.3
U1-3	Coal, parting with clay, dark gray, sheet fissures.	0.2
U1-2#1	Carbonaceous claystone, brownish gray.	0.2
U1-2#2	Mudstone, moderate yellowish brown to greenish gray, mottered by dark yellowish orange.	0.3
U1-2#3	Siltstone, dusky yellow, roots traces.	0.3
U1-2#4	Siltstone, moderate yellowish brown, root traces, Fe-concretion.	0.3
U1-2#5	Silty sandstone, pale olive, mottered by light olive brown, Fe-concretion.	0.3
U1-2#6	Mudstone, inside is pale olive and surface is dark yellowish orange/ moderate yellowish brown.	0.3
U1-2#7	Carbonaceous claystone, dark yellowish brown/grayish yellow.	0.2
UU1-2#1	Siltstone, pale olive, roots traces.	0.3
UU1-2#2	Siltstone, moderate greenish yellow, pyrite, mottered by yellow and brownish yellow.	0.3
U1-1#1	Carbonaceous claystone, dark yellowish brown.	0.2
U1-1#2	Coal with parting clay, clay is yellowish gray, coal is grayish black.	0.2
U1-1#3	Claystone, coal fragment, brownish black to light olive gray, mottered by yellowish orange.	0.2
U1-1#4	Coal parting by yellowish gray clay, brittle, sheet.	0.2
U1-1#5	Mudstone, yellowish gray, small spots by dark yellowish orange.	0.3
U1-1#6	Claystone, pale yellowish brown, mottered by grayish yellow.	0.2
U1-1#7	Claystone, dark yellowish brown, coal fragment, mottered by grayish yellow.	0.3
U1-1#8	Carbonaceous mudstone, olive gray.	0.2
U1-1#9	Carbonaceous claystone, olive gray, sheet, more coal content than the lower part, coal penetrated in the fissures.	0.2
U1-1#10	Siltstone, pale olive, Fe-concretion, mottered by dark yellowish orange.	0.3
U1-1#11	Siltstone, yellowish gray, mottered by dark yellowish orange, small coal fragments.	0.3
U1-1#12	Sandy siltstone, yellowish gray, Fe-concretion, rooted, quartz, feldspar.	0.3
U1-1#13	Siltstone, greenish gray, mottered by reddish brown, little spots, Fe-concretion.	0.3
U1-1#14	Siltstone, yellowish gray, mottered by reddish brown, Fe-concretion.	0.3
U1-1#15	Sitlstone, yellowish gray, high Fe-concretion, rooted.	0.3
U1-1#16	Mudstone, yellowish gray to moderate yellow, roots traces, some coal fragment.	0.3
OB1#1	Mudstone, pale olive, root traces, mottered by moderate yellow/dark yellowish orange.	3
OB1#2	Siltstone, greenish gray, low mottered.	1
OB1#3	Siltstone, yellowish gray.	3
OB1#4	Mudstone, yellowish gray mottered by moderate olive brown.	2
OB1#5	Brownish black coal parting by brownish gray clay.	1
OB1#6	Claystone, very light gray, mottered by pale yellowish orange, roots traces.	2
OB1#7	Carbonaceous claystone and claystone, dark yellowish brown.	0.5
OB1#8	Mudstone, pale reddish brown/moderate yellow.	5

the desiccator and keeping in an oven at 60°C for about 4 hours or longer until they were ready for the x-ray diffraction analysis[11].

X-ray Diffraction

X-ray diffraction is the most widely used method for identification of clay minerals and their crystalline characteristics. The x-ray diffractograms were generated using MAC-M18XHF, at the Department of Earth Science, Kyoto University of Education, Japan. The lamp of this instrument generated CuKα radiation, at 18 kilowatts, 40 kilovolts, and 50 milliamperes. The scanning speeds were 4° per minute. Additional x-ray diffraction analysis was carried out by a Bruker x-ray diffractometer (model D8 Advance) at Department of Geological Sciences, Chiang Mai University, which generated CuKα radiation at 18 kilowatts, 40 kilovolts and 30 milliamperes. Bulk analysis used the following scanning process: 2-theta start at 2°, stop at 60°, step 0.04°. For fine-fraction clays, the scanning process was 2-theta start at 2°, stop at 40°, step 0.04°. Semi-quantitative estimation was determined. The highest intensity of *d*-spacing reflection obtained from each mineral was selected to represent the proportion of the mineral in a sample. The estimation was based on the composition of the half peak area calculated from the height of the peak multiplied by the half width at the midpoint of the peak height. The above method of calculating the half peak areas overcomes problems associated with inclined base lines and overlapping peaks. The calculated half peak area of every mineral found in a sample was summed and recalculated to 100 percent (Fig. 4). The proportion of each mineral in a sample was calculated and presented as a percentage value that could be an under- or overestimate of the true proportion present. Thus, the most effective comparisons can be made only between samples containing similar constituents[12].

Clay Mineral Identification

The presence of an integral series of *d*-spacing reflections is based on the (001) reflection of the x-ray diffraction pattern of the oriented mounts (Table 2). Kaolinite reflected as 7.13 to 7.19 Å = (001); 4.18 to 4.48 Å = (020, 110, 111), 3.50 to 3.57 Å = (002) and unaffected by glycolation. Disordered kaolinite can be distinguished from the kaolinite by a stronger intensity of x-ray diffraction pattern in the vicinity of 4.48 Å in unoriented sample mounts and broad (001 and 002) *d*- spacing reflection at the same position of those in ordered kaolinite in the oriented mounts. Chlorite is distinguished from kaolinite by the basal spacing of chlorite of about 14.2 Å = (001); 7.12 Å = (002); 4.75 Å = (003) and 3.56 Å = (004). Illite can be identified with the first order at about 10 Å; 4.97 to 5.1 Å = (004); 3.33 Å = (024, 006). Montmorillonite reflects at 13.4 to 14.48 Å = (001); 4.75 to 5.1 Å = (003) and about 3 Å = (005) (Table 2). After glycolation, the (001) reflection of montmorillonite extends to 17 to 19 Å and 21 to 26 Å (Fig. 5).

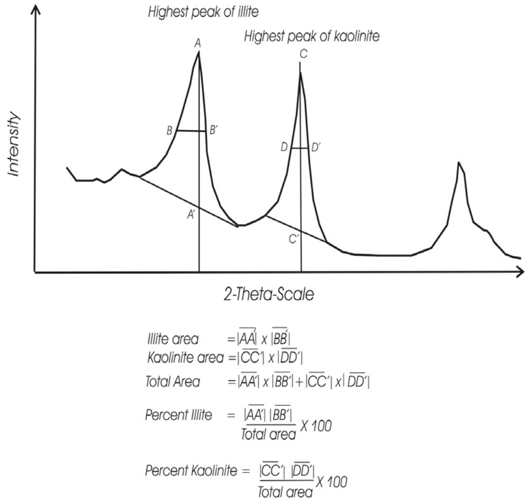

Fig 4. The half peak area technique for calculating percent of clay minerals (after Ratanasthiens, 1975[12]).

Results

Bulk quantities of mineral percentages are shown in Table 3. The percentages of minerals in fine-fraction clays from each layer are shown in Table 4. In the fine-fraction clays, the most abundant minerals are kaolinite (56 layers and a maximum of 72 percent), illite (53

Table 2. *d*-Spacing of dominant minerals using in x-ray diffraction identification of minerals in the Chiang Muan mine[1,13,14].

Mineral	d-spacing 1	hkl	d-spacing 2	hkl	d-spacing 3	hkl
Quartz	4.26	100	3.34	101	2.45	110
Kaolinite	7.13 to 7.19	001	4.18 to 4.48	020	3.57	002
Illite	9.98 to 10.01	002	4.97 to 5.01	004	3.33	024, 006
Montmorillonite	13.8 to 14.48	001	4.73	003	~3	005
Chlorite	14.2	001	7.12	002	4.75	003
Gypsum	7.56 – 7.68	020	4.27	121	3.06	141

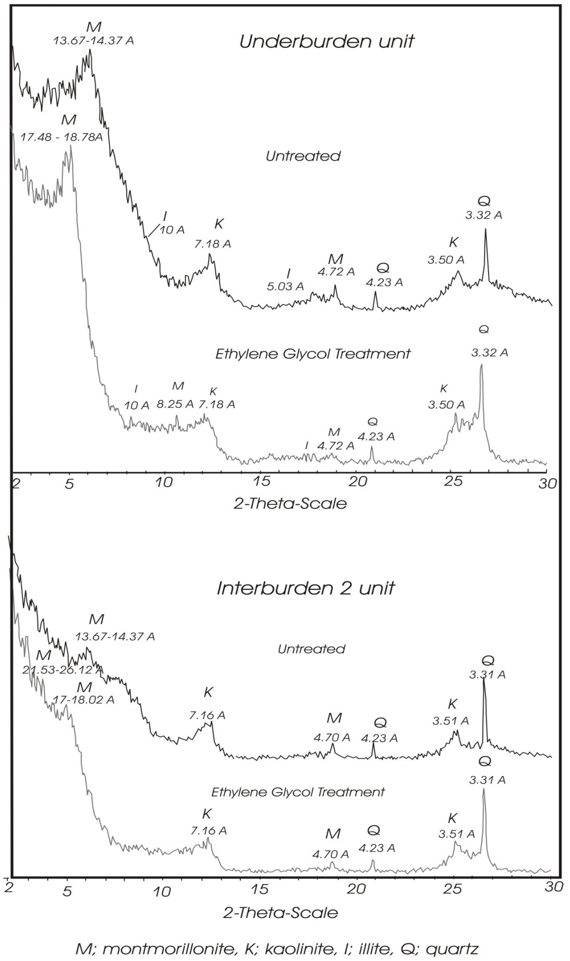

Fig 5. X-ray patterns of clay samples with different treatments.

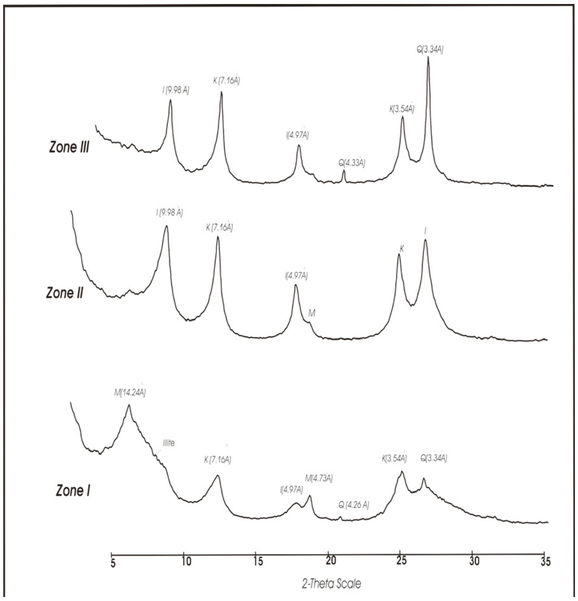

Fig 6. X-ray diffraction patterns of representative samples os each zone. M, montmorillonite; K, Kaolinite; I, Illite; Q, Quartz.

layers and a maximum of 71 percent), quartz (51 layers and a maximum of 97 percent), and montmorillonite (31 layers and a maximum 69 percent). Trace minerals are chlorite, disordered kaolinite and mixed-layers clay minerals. Other minerals are gypsum, calcite, anhydrite and pyrite. Gypsum and pyrite contents are high in coal-bearing layers.

The mineral assemblages in the Chiang Muan mine occur in three zones from bottom to top as illustrated in Fig. 6.

Table 3. Bulk composition of minerals from the Chiang Muan mine[15].

Unit / Mineral	Quartz (%)	Kaolinite (%)	Illite (%)	Montmorillonite (%)
OB	54 - 83	10 - 22	0 - 24	0 - 6
IB1	56	21	23	-
IB2	80 - 97	0 - 4	0	1 - 17
UB	25	11	27	36

Zone I: montmorillonite, kaolinite, and quartz in the Underburden unit, Lower coal zone unit, and Interburden 2 unit.

Zone II: quartz, illite, and kaolinite in the Upper coal 2 unit and the Interburden 1 unit.

Zone III: quartz, illite, kaolinite, and montmorillonite in the Upper coal zone 1 unit and the Overburden unit.

The fine-fraction clays of the Underburden unit have abundant montmorillonite, moderate amounts of kaolinite and chlorite, and only small amounts of quartz (Table 4). Montmorillonite has high intensity and a very broad peak. Kaolinite and quartz also have broad peaks reflecting poor crystallinity.

The fine-fraction clays of the lower coal zone sub-unit contain a high content of disordered kaolinite and moderate amounts of quartz and montmorillonite (Table 4). Quartz shows the sharpest peaks reflecting

Table 4. Percentages of minerals in fine-fraction clays from the Chiang Muan mine.

Sample	Quartz	Kaolinite	Illite	Mont	Gypsum	Others
UB	5	16	10	69	0	0
LS	37	0	0	12	0	51 dk
IB2#1	12	53	20	15	0	0
IB2#2	14	50	22	14	0	0
IB2#3	9	57	0	34	0	0
IB2#4	46	23	4	12	0	15 ch
IB2#5	18	72	0	10	0	0
IB2#6	12	61	20	7	0	0

Table 4. Cont'd.

Sample	Quartz	Kaolinite	Illite	Mont	Gypsum	Others
IB2#7	10	67	0	23	0	0
IB2#8	25	51	0	24	0	0
U2-1#1	28	0	8	0	55	10 dk,cal
U2-1#2	76	14	11	0	0	0
UU2-1	34	34	30	3	0	0
UU2-2	24	40	34	2	0	0
UU2-3	45	24	31	0	0	0
IB1#1	0	38	59	3	0	0
IB1#2	31	31	36	1	0	0
U1-8#1	33	0	0	0	67	0
U1-8#2	39	0	0	0	61	0
UU1-8	64	18	16	2	0	0
U1-6	58	0	0	0	0	42 dk
UU1-6	63	16	21	0	0	0
U1-5#1	0	0	0	0	0	100 py,mix
U1-5#2	51	21	28	0	0	0
U1-5#3	67	5	28	0	0	0
U1-5#4	81	0	4	0	15	0
UU1-5	42	30	25	1	0	3 mix
UU1-4#1	29	48	23	0	0	0
UU1-4#2	75	6	9	0	0	10 ch
UU1-4#3	0	27	22	52	0	0
UU1-4#4	48	30	22	trace	0	0
UU1-4#5	0	53	43	4	0	0
UU1-4#6	0	59	39	3	0	0
U1-3	49	0	4	15	13	19 dk
U1-2#1	63	0	11	0	0	25 dk
U1-2#2	0	29	71	0	0	0
U1-2#3	0	49	51	0	0	0
U1-2#4	0	49	51	0	0	0
U1-2#5	81	13	6	0	0	0
U1-2#6	97	3	0	0	0	0
U1-2#7	58	6	17	0	12	7 cal
UU1-2#1	0	32	64	4	0	0
UU1-2#2	90	7	3	0	0	0
U1-1#1	63	9	29	0	0	0
U1-1#2	19	0	3	15	45	18 dk, cal
U1-1#3	49	27	24	0	0	0
U1-1#4	17	0	0	52	30	0
U1-1#5	72	17	12	0	0	0
U1-1#6	67	18	15	0	0	0
U1-1#7	57	19	24	0	0	0
U1-1#8	36	18	45	0	0	0
U1-1#9	22	39	39	0	0	0
U1-1#10	24	27	48	trace	0	0
U1-1#11	78	9	10	0	3	0
U1-1#12	0	39	61	0	0	0
U1-1#13	0	41	55	4	0	0
U1-1#14	0	33	67	0	0	0
U1-1#15	61	25	14	0	0	0
U1-1#16	70	14	16	0	0	0
OB1#1	0	54	46	trace	0	0
OB1#2	0	50	40	0	0	10 anh
OB1#3	37	33	28	2	0	0
OB1#4	53	25	19	trace	0	4 mix
OB1#5	33	7	24	0	0	36 ch, mix
OB1#6	0	62	35	3	0	0
OB1#7	19	33	44	3	0	0
OB1#8	0	44	56	0	0	0

Other minerals: ch = chlorite; dk = disordered-kaolinite; cal = calcite; py = pyrite; mix = mixed-layer clays; anh = anhydrite

good crystallinity. The disorderd kaolinite also shows sharp peaks. It is characterized by the peak at 4.48 Å (020) which is higher in intensity than those of 7.16 Å (001) and 3.57 Å (002)[16]. Montmorillonite appears as a small broad peak.

The Interburden 2 unit is located between the Upper coal zone 2 unit and the Lower massive coal seam sub-unit. The fine-fraction clays of this unit contain more kaolinite concentrations and have moderate amounts of quartz and montmorillonite (Table 4). Chlorite and illite occur as trace minerals. Quartz and kaolinite peaks in this unit have low intensity and are slightly broad. Montmorillonite occurs in every layer as small and broad peaks at 13.6 Å to 14.4 Å.

The Upper coal zone 2 unit consists of coal-bearing sediments. The minerals of the fine clay fraction are heterogeneous (Table 4), including gypsum, quartz, illite, disordered kaolinite and kaolinite. The peaks are not smooth. However, these clastic sediments contain nearly the same quantity of quartz, kaolinite and illite. Montmorillonite occurs as a trace mineral. Quartz peaks are sharp to moderately sharp. Illite and kaolinite peaks are also moderately sharp. Montmorillonite is only slightly reflected.

The fine-fraction clays of the Interburden 1 unit are mainly illite, kaolinite and quartz (Table 4). A small amount of montmorillonite is found. The peaks of illite and kaolinite are moderately sharp, though quartz gives the sharpest peaks.

The fine-fraction clays of the Upper coal zone 1 unit have a high content of quartz but lesser amounts of illite and kaolinite than the lower units (Table 4). Montmorillonite occurs as a trace mineral, though it is plentiful in samples UU1-4#3, U1-1#2, U1-1#4, U1-3 and UU1-4#3. Quartz peaks are sharpest when compared with those of other minerals. This indicates that quartz is very well crystalline. Kaolinite and illite peaks are moderately sharp to slightly broad. The peaks of montmorillonite and gypsum in samples U1-1#2, U1-1#4 and U1-3 are very sharp. Nevertheless, the broad peaks of montmorillonite also reflected at 13.67 Å to 14.37 Å.

The fine-fraction clays of the Overburden unit contain a slightly higher kaolinite quantity than illite, with a smaller amount of quartz (Table 4). Trace minerals are montmorillonite, chlorite and mixed-layer clays. Anhydrite occurs only in the sample OB1#2 which may be dehydrated from gypsum. This anhydrite or gypsum could be an oxidation product of pyrite which is highly abundant in the Upper coal zone 2 unit and/or it could be a product of marine incursion during the coal unit deposits[16]. The peaks of illite and kaolinite are very similar; both are slightly broad to slightly sharp. The peak shape indicates that the minerals are poorly to moderately crystalline. Quartz peaks are moderately sharp; the crystals could be moderately crystalline.

Discussion

The assemblage and abundance of clay minerals depend on the degree of weathering, the type of parent rocks, and the associated environments[1,17,18]. In the Chiang Muan coal mine, the abundant minerals in clay fractions are kaolinite, illite, montmorillonite and quartz (Table 4), which are subdivided in to three zones.

Zone I is dominated by montmorillonite, kaolinite and quartz (Fig. 6). These minerals are interpreted as the weathering products from volcanic rocks that are exposed in the southern flank of the Chiang Muan basin (Fig. 2). Broad x-ray graphs indicate that montmorillonite crystals are thin and impure[19, 20]. The highest montmorillonite content is in the Underburden unit. This strongly contrasts with that in the upper part of deposition. Montmorillonite is typically formed in soils developed under warm to temperate climates characterized by alternating humid and dry seasons[21]. Kaolinite is typically formed on land and is characteristic of tropical and warm humid climatic zones with well drained soils, accelerated leaching of the bedrock[22], and a minimum temperature of 15°C[23]. Kaolinite can crystallize in permeable sandstone[24]. The decrease of montmorillonite content, along with an increase in kaolinite and illite in the upper units, suggests that the climate changed and that there was a greater sediment supply from the land[25].The magnetostratigraphy suggests the age of the lower lignite between 12.4 to 13.0 million years ago[6,7]. Thus, the climate changed at about 13 million years ago.

Zone II is dominated by quartz, illite, and kaolinite. These minerals are common materials. The strata in this zone are interbedded coal, mudstone, and fine-grained sandstone. The clay fractions are interpreted as the weathering product from feldspartic rocks in the surrounding areas. They also can develop between grains of sandstone and siltstone[24]. The beds in this zone are considered to be lacustrine and floodplain deposits[7]. Palynological study suggested this zone was deposited in a moist environment, probably along the coastal area of a Miocene lake[26]. Sulfur isotopic study, however, suggested that the upper coal seam 2 unit was associated with a marine environment, but there is no evidence from clay minerals to support this.

Zone III is dominated by quartz, illite, kaolinite and montmorillonite. The clay fraction in this zone is similar to that of zone II. However, there is much montmorillonite in some layers (Table 4). Popcorn structure, which is a character of swelling clays, occurs in outcrops that correspond to the occurrence of montmorillonite. This zone is an alternation of thin coal layers, mudstone and sandstone. It was interpreted as lacustrine and floodplain deposits[7]. The high content of montmorillonite in some layers may be from re-worked, uplifted beds of the lower part or from another source. The ancient environment of this zone was fresh water and is suggested by palynology[26].

Acknowledgements

Financial support from the Thailand Research Fund through the Royal Golden Jubilee Ph.D. Program (Grant No.PHD/0075/2544) to Y. Thasod and B. Ratanasthien is acknowledged. We thank the Graduate School, Chiang Mai University, for financial support for this research. We also thank the Department of Earth Science, Kyoto University of Education, Japan, for conducting the experiments and the Department of Geological Sciences, Faculty of Science, Chiang Mai University for providing the instrument used in this research. We especially thank the Chiang Muan mine officers and everyone who assisted.

References

1. Grim E (1968) Clay mineralogy (second edition), 596 p. McGraw-Hill Co. New York.
2. Ratanasthien B (2002) Problems of Neogene biostratigraphic correlation in Thailand and surrounding areas. *Revista Mexicana de Ciencias Geol'gicas* **19 : 3**, 235-41.
3. Polachan S, Pradidtan S, Tongtaow C, Janmaha S, Intarawijitr K and Sangsuwan C (1991) Development of Cenozoic basins in Thailand. In: *Marine and Petroleum Geology* **8**, 84-97.
4. Morley CK, Woganan N, Sankumarn N, Hoon TB, Alief A and Simmons M (2001) Late Oligocene-Recent stress evolution in rift basins of northern and central Thailand: implications for escape tectonics. *Tectonophysics* **334**, 115-50.
5. Nakaya H, Saegusa H, Kunimatsu Y and Ratanasthien B (2002) Miocene mammalian faunas of northern Thailand and their geological age. *Primate Res* **18**, 131-41 (in Japanese with English abstract).
6. Suganuma Y, Okada M and Nagaoka S (2002) Preliminary magnetostratigraphy of the Tertiary sedimentary rocks, Mae Moh Group from Chiang Muan in Northern Thailand. *Primate Res* **18**, 165-73 (in Japanese with English abstract).
7. Suganuma Y, Hamada T, Tanaka S, Okada M, Nakaya H, Kunimatsu Y, Saegusa H, Nagaoka S and Ratanasthien B (2006) Magnetostratigraphy of the Miocene Chiang Muan Formation, northern Thailand: Implication for revised chronology of the earliest Miocene hominoid in Southeast Asia. *Palaeogeography, Palaeoclimatology, Palaeoecology* **239**, 75-86.
8. Benammi M, Urrutia-Fucugauchi J, Chaimanee Y and Jaeger JJ (2003) Preliminary paleomagnetic study of the Chiang Muan basin in northern Thailand. In: *Proceeding of the 8th International Congress on Pacific Neogene Stratigraphy: Pacific Neogene Paleoenvironments and Their Evolution* (Edited by B. Ratanasthien, S.L. Rieb, and S. Chantraprasert), pp 352 (abstract).
9. Charoenprawat A, Chuaviroj S, Hinthong C and Chonglakmani C (1995) Geological map of Thailand, Sheet Lampang (NE47-7), scale 1: 250,000, Geological Survey

Division, Department of Mineral Resources, Bangkok, Thailand.
10. Hardy R and Tucker M (1988) X-ray powder diffraction of sediments. In: *Techniques in Sedimentology* (Edited by Tucker M), pp 191-228. Blackwell Scientific Publications, Great Britain.
11. U.S. Geological Survey Open-File Report 01-041 (2001) A Laboratory manual for x-ray powder diffraction; Ethylene glycol treatment. *http:// pubs.usgs.gov/of/of01-041/htmldocs/ methods.htm#eglycol*.
12. Ratanasthien B (1975) The geochemistry of some recent argillaceous sediment. A Thesis submitted for the degree of Doctor of Philosophy, The University of Aston in Birmingham, 348 p.
13. Brown G (1972) The x-ray identification and crystal structures of clay minerals, 544 p. Mineralogical society (Clay mineral group), London.
14. Carroll D (1970) Clay Minerals: A guide to their x-ray identification. *The Geological Society of America, special paper* **126**, 80 p.
15. Chantarakamthorn K (2004) Mineral types and size distribution of clay in Chiang Muan Coal mine, Phayoa province. Independent study (205499), Department of Geology, Faculty of Science, Chiang Mai University.
16. Silaratana T, Ratanasthien B, Takayasu K, Fyfe WS, Asnachinda P Kandharosa W and Kusakabe M (2004) Sulfur isotopic implication of Middle Miocene marine incursion in northern Thailand. *Science Asia* **30**. 43-58.
17. Millot G (1970) Alkaline chemical sedimentation with attapulgite, sepiolite and montmorillonite. In: *Geology of Clays*, pp 199-203.
18. Weaver EC and Pollard DL (1975) The chemistry of clay mineral, 213 p. Elsevire Scientific Publishing Company.
19. Moore M D, Robert C R (1997) X-ray diffraction and the identification and analysis of clay minerals, 378 p. Oxford University Press.
20. Weizong X, Johnston C T, Parker P and Agnew S F (2000) Infrared study of water sorption on Na-, Li-, Ca-, and Mg-exchanged (Swy-1 and Saz-1) montmorillonite. *Clays and Clay minerals* **48**, 120-31.
21. Chamley H (1998) Clay mineral sedimentation in the ocean. In: *Soils and sediments (mineralogy and geochemistry)* (Edited by Paquet H and Clauer N), pp 269-302. Springer Verlag, Berlin.
22. Robert C and Chamley H (1991) Development of early Eocene warm climates, as inferred from clay mineral variation in oceanic sediments. *Global and Planetary Change* **89**, 315-32.
23. Gaucher G (1981) Les facteurs de la Pédogenèse. Dison, 730 p. Belgium.
24. Weaver CE (1989) Clays, muds, and shales: Developments in sedimentology (44), 819 p. Elsevier, New York.
25. John MC, Adatte T and Mutti M (2006) Regional trends in clay mineral fluxes to the Queensland margin and ties to middle Miocene global cooling. *Palaeogeography, Palaeoclimatology, Palaeoecology* **233**, 204-24.
26. Songtham W, Ratanasthien B and Mildenhall DC (2003) New species of algae *Actinastrum Lagerheim* and *Closterium* Nitzsch ex Ralfs from Milddle Miocene sediments of Chiang Muan basin, Phayao, Thailand, and their related tropical pollen composition. *Mahasarakham University Journal, Proceeding of 1st International Conference on Palaeontology of Southeast Asia* **22**, 104-22.