

Major, Trace and Rare Earth Elements Geochemistry of Isan Clay Southwestern Nigeria: *Implications for Provenance*

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Abstract

Isan clay occurs within the Precambrian Basement rocks in the southwestern Sector of the Nigerian Basement Complex. This present study focuses on the determination of the provenance of Isan clay using major, trace and rare earth elements geochemistry. Representative clay samples were collected at different horizons and subjected to Inductively Coupled Plasma-Mass Spectrometry (ICP-MS) analysis. On the TiO_2 vs Al_2O_3 diagram, Isan clays plot in the granite/rhyolite field. This indicates that the provenance of Isan clay is predominantly from felsic rocks. The provenance of Isan clay could thus be attributed to the granitic rocks from Isan area. On the La versus Th plot, Isan clays data fall below the range of UC with all the samples having a significant felsic component. This suggests that Isan clays have a uniform provenance of felsic component and exhibited high ratios of La/Co, Th/Co, La/Cr, Th/Cr and Ba/Co which indicate that the clays were derived mainly from felsic rocks. The clays have fractionated REE patterns, the LREE are enriched and the HREE patterns are almost depleted. The $(\text{La/Yb})_N$ ratio of Isan clay ranges from 4.96 to 15.96 and $(\text{Gd/Yb})_N$ ratio ranges from 1.20 to 1.68 with a negative Eu anomaly of 0.50 to 0.67. These characteristics attest that the provenance of Isan clay was felsic and the negative Eu anomaly is regarded as evidence of a differentiated source, similar to granite.

Keywords: *Felsic, provenance, anomaly, differentiation and patterns*

Introduction

Isan clay is located within the Precambrian Basement rocks in the southwestern sector of Nigerian Basement Complex as shown in Fig. 1. Clays are fine particles emanating from the decomposition of rocks under the influence of climate over time (Ekosse, 2011). Clays are composed of mixtures of finer grained clay minerals and clay-sized crystals of other minerals such as quartz, carbonate and metal oxides (Murray, 2007).

Clay minerals are secondary minerals derived from chemical alteration of mainly feldspars and micas. Clay minerals constitute an integral part of clay deposits and consist of layered silicates arranged in tetrahedral and octahedral sheets (Heckrodt, 1991). Clays and clay minerals are generally plastic (except for flint clay) and hardens on drying or firing; both are natural consisting mainly of phyllosilicates (Bergaya and Lagaly, 2006; Ekosse, 2011).

Kaolin (hydrated aluminum silicate, $\text{Al}_2\text{Si}_2\text{O}_5(\text{OH})_4$) is an important industrial clay for economic benefit. Properties of fine particle size, platy shape, inertness, non-toxicity, as well as high brightness and whiteness make it a more versatile mineral, with applications in a wide variety of industries. Commercial kaolin resources are found as sedimentary deposits and as weathering or hydrothermal alteration product of rocks containing a

high proportion of aluminosilicate minerals (Hughes and Brown, 1979).

Isan clay was estimated to be 7.5 million tons and the assessments of industrial suitability based on physical properties indicate that the clay bodies possess adequate mouldability and reddish tones when fired. Thermal characteristics such as loss of ignition, linear shrinkage and water absorption capacity values are within the ranges of various commercial specifications for ceramics, building bricks and refractory (Bolarinwa, 1992; Elueze and Bolarinwa, 1995). There are limited reports on the provenance of Isan clay. The present study focuses on the application of major, trace and rare earth elements geochemistry in the determination of Provenance of Isan clay.

Study Location

The study area is located on longitudes $5^\circ 08'$ to $5^\circ 25'$ E and latitudes $7^\circ 55'$ to $8^\circ 24'$ N (Fig. 2) while Isan clay deposit is accessible through Ikun-Ayede-Oye Ekiti road. Isan area is underlain by crystalline rocks and five groups of rocks have been recognized (Fig. 2) and they include: Migmatite-Gneiss Complex (early gneiss and granite gneiss); Slightly migmatized to non-migmatized metasedimentary rocks (quartzite and marble); Charnockitic rocks (Coarse grained Charnockites); Members of the Older Granite Suite (porphyritic

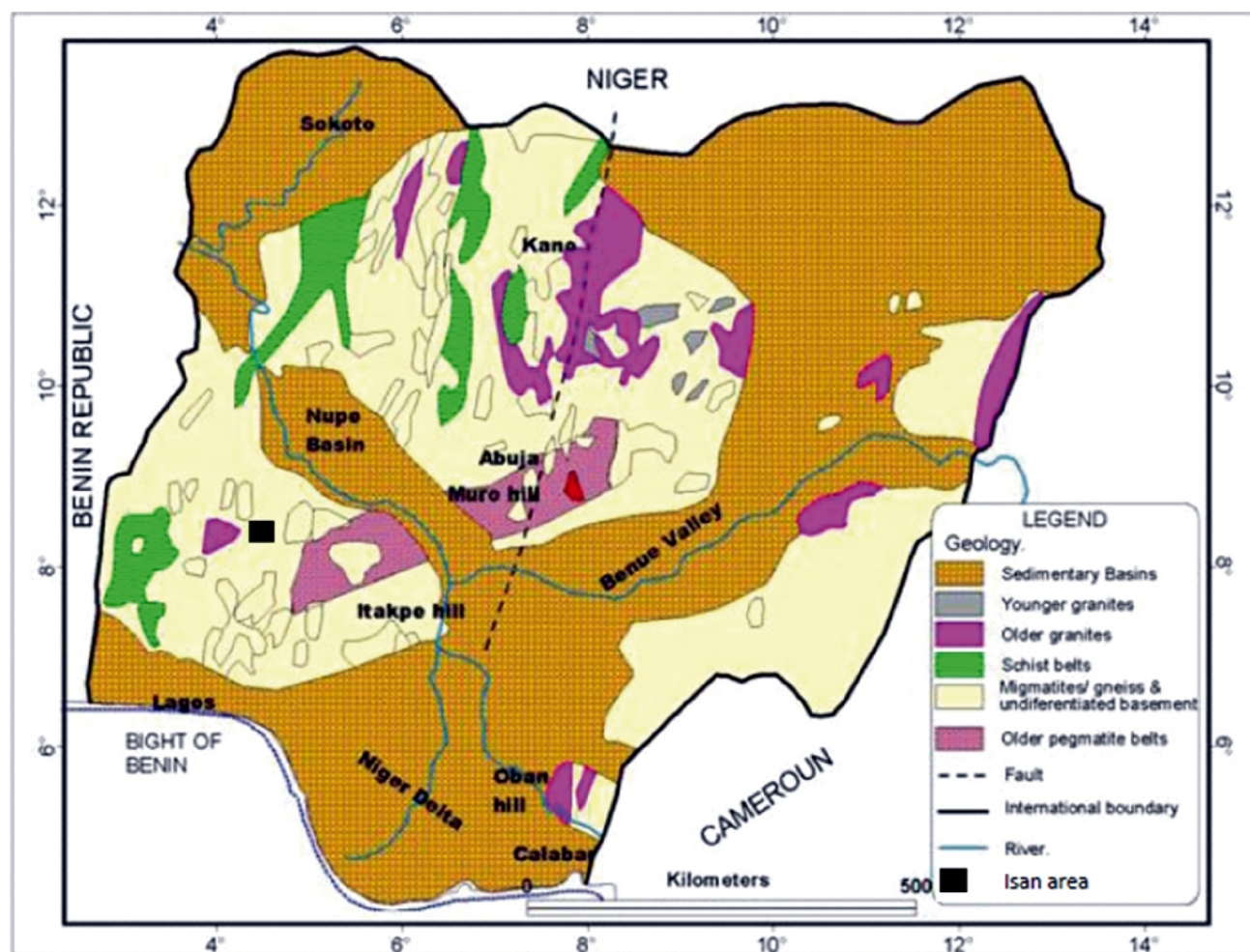


Fig. 1: Geological map of Nigeria showing location of the Isan area (Geological Survey of Nigeria, 2005)

granite, fine to medium-grained granite, coarse grained granite, pegmatites and vein quartz) and Unmetamorphosed dolerite dyke (Odewumi, 2016).

Materials and Methods

The representative clay samples were collected using chisel at different depths and horizons from the deepest to the topmost part of each of the clays, corresponding to macroscopic features. The thickness of each horizon was measured using measuring tape and the clay samples were stored in air-tight plastic bags labeled with a sample number.

The clay samples from Isan area were air-dried at the Department of Geology Laboratory, University of Ilorin, Ilorin, Nigeria. These clay samples were also subjected to Inductively Coupled Plasma-Mass Spectrometry (ICP-MS) analysis comprising of major, trace and rare earth elements at Acme Laboratory Ltd, Vancouver, Canada. The samples were analyzed for

major oxides (SiO_2 , TiO_2 and Al_2O_3); trace elements (Ag, As, Au, Ba, Be, Bi, Cd, Co, Cu, Cs, Ga, Hf, Hg, Mo, Nb, Pb, Rb, Sb, Se, Sn, Sr, Ta, Th, U, V, W, Zn, Zr and Y) and rare earth elements (REE) (La, Ce, Pr, Nd, Sm, Eu, Gd, Tb, Dy, Ho, Er, Tm, Yb and Lu) using a Lithium metaborate/tetraborate fusion and nitric acid digestion of 0.2g of each sample. The provenance of the clays was inferred from TiO_2 versus Al_2O_3 segregation diagram for granite, rhyolite and basalt (after Ekosse, 2001) and La versus Th diagram for felsic and mafic sources (after Nyakairu and Koeberl, 2001). Thus, information on provenance of clays could provide clues on type of parent materials from which they were derived. The Eu anomaly (Eu/Eu^*) was computed using normalized ratio of $\text{Eu}_N/[(\text{Sm}_N \cdot (\text{Gd}_N)^{1/2})]$.

Results

Field Characteristics of Isan Clay

Isan clay is mainly underlain by granite gneiss while the

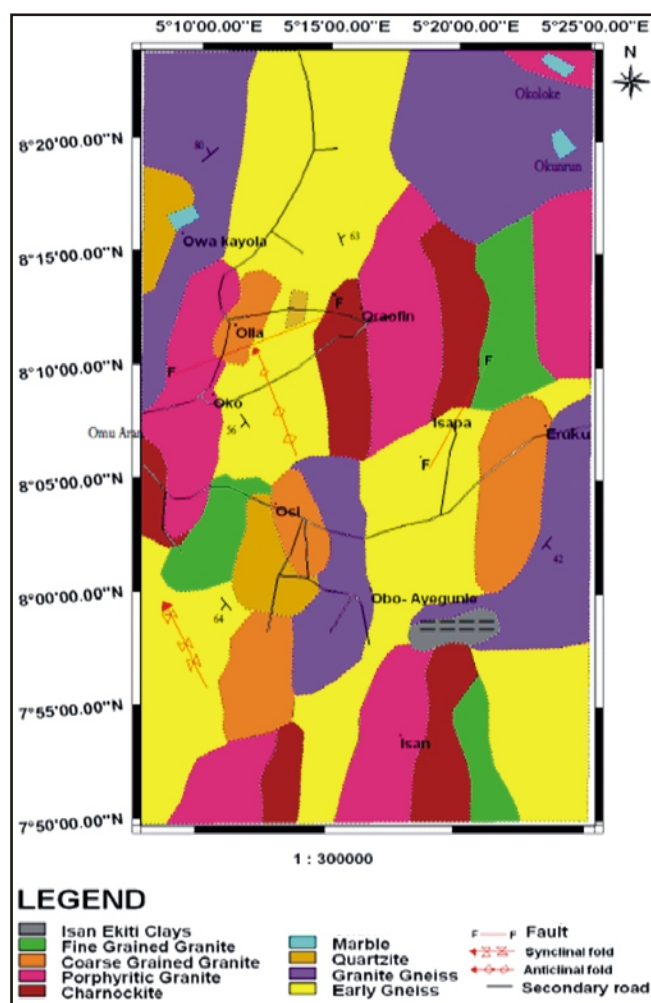


Fig. 2: Geological map of Isan area (Odewumi, 2016)

other Precambrian Basement rocks identified in the area include early gneiss, charnockites, fine to medium-grained granite, porphyritic granite and pegmatite (Fig. 2) which constitutes part of the southwestern sector of Nigerian Basement Complex.

Sampling was carried out at mining pit of Isan clays (Fig. 3) and the pit has a depth of about 17 m, which in some places extends to about 20 m. The clay deposit can be divided into four horizons: the thickness of reddish clay horizon varies from 1.5 to 1.8 m which overlies the kaolinized granite horizon with thickness ranging from 3.0 to 4.0 m. Kaolinized granite horizon overlies the variegated clay horizon with thickness ranging from 5.0 to 7.6 m which in turn overlying the greyish kaolin clay horizon with thickness varying from 5.0 m onwards until weathered Basement is reached. The overburden encountered was lateralized ironstone with an average thickness of 1.8 m.

Geochemical Compositions of Isan Clays

The major oxides and trace elements compositions of Isan clay (SA) are presented in Table 1 while rare earth elements composition of Isan clay is presented in Table 2. The SiO_2 content of Isan clay ranges from 55.49 to 62.05 wt%, Al_2O_3 value ranges from 17.77 to 20.67 wt% and TiO_2 content ranges from 0.02 to 0.2 wt% respectively (Table 1). This indicates that Isan clay is aluminosilicate.

Discussion

The primary source of the clay materials were inferred from TiO_2 vs Al_2O_3 segregation diagram for granite, rhyolite and basalt (Ekosse, 2001). The information on provenance of clays could provide clues on type of parent materials from which they were derived. The TiO_2 vs Al_2O_3 diagram (after Ekosse, 2001) of Isan clay (Fig. 4) indicates that the provenance of Isan clay is predominantly from felsic rocks, as they plot in the granite/rhyolite field. Isan clay provenance could thus be attributed to the granitic rocks from Isan area which constitutes part of the Precambrian Basement rocks in southwestern sector of Nigerian Basement Complex (Odewumi, 2011).

Isan clays have low contents of Cr and Co, with high concentrations of the REE, Th and Hf (Tables 1 and 2). This indicates that the clays were derived from predominantly felsic sources, rather than from basic rocks. High field strength elements (HFSE, including Y, Zr, Ti, Nb, Ta), Th, Hf and Co are the most suitable ones for provenance determination, because of their relatively low mobility during weathering, transport, diagenesis and metamorphism (Nyakairu and Koeberl, 2001).

The ratios of both incompatible and compatible elements are useful for differentiating between felsic and mafic components (Nyakairu and Koeberl, 2001). Isan clays have high ratios of La/Co, Th/Co, La/Cr, Th/Cr and Ba/Co (Table 1) which indicate that the clays were derived mainly from felsic rocks (Cullers et al., 1987, 1988; Cullers, 1988). The immobile elements La and Th are more abundant in felsic than in basic rocks, whereas Co is more concentrated in basic rocks than in felsic rocks (Taylor and McLennan, 1985; Wronkiewicz and Condie, 1987). It has been shown that ratios such as La/Co, Th/Co, Co/Th and Eu/Eu* in siliciclastic sediments, allow to place constraints on the average provenance composition (Cullers et al., 1988; Cullers, 1994, 1995; Wronkiewicz and Condie, 1987, 1989,

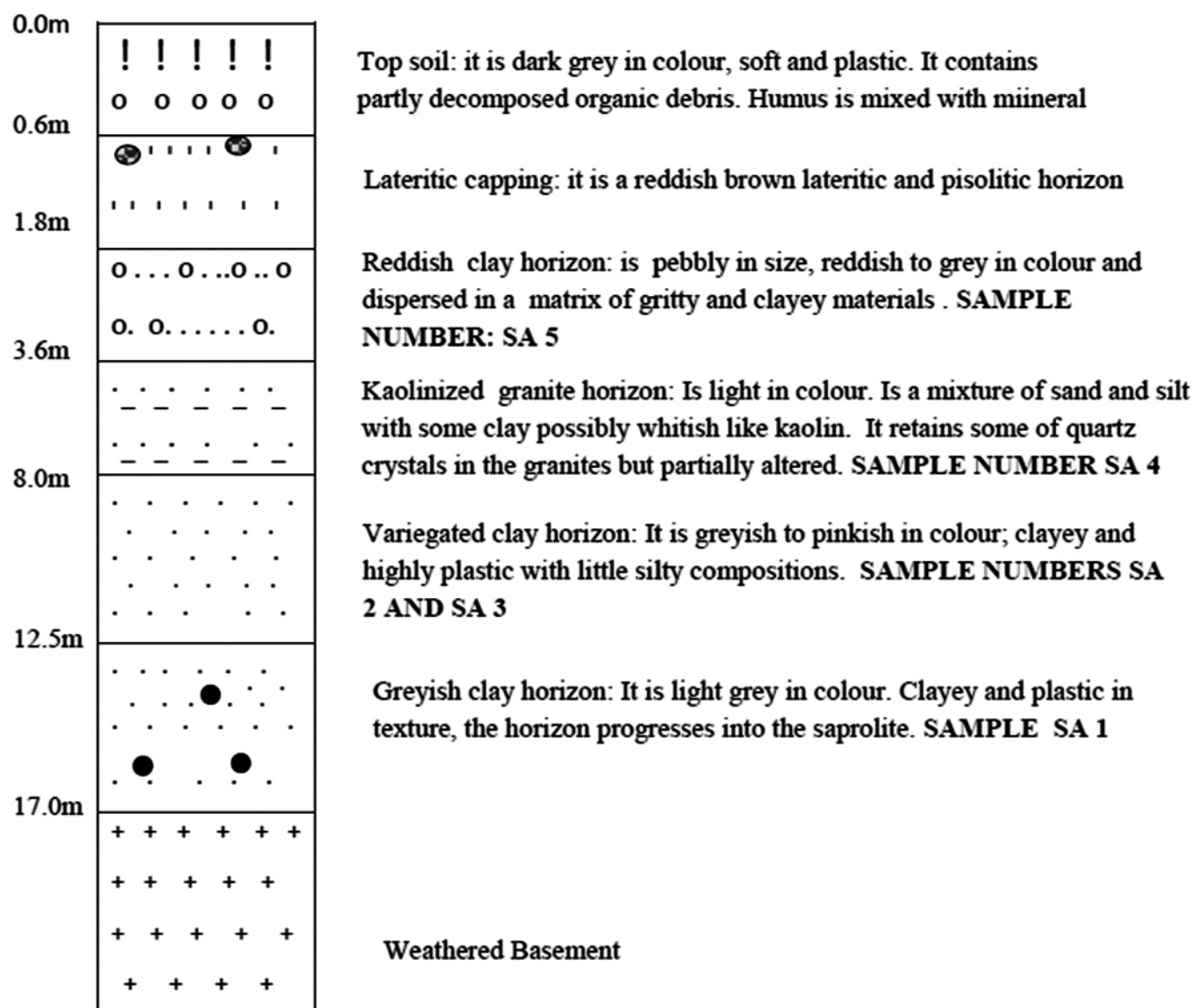


Fig. 3: Profile Section of Isan Clay

1990; Cox et al., 1995).

The geochemical differences between elements such as Th and La (indicative of a felsic source) and Co and Cr (indicative of a mafic source) have been exploited to distinguish between felsic and mafic provenance (McLennan, 1989; McLennan and Taylor, 1991; McLennan et al., 1980; Wronkiewicz and Condie, 1990). The relative enrichments of (normally) incompatible elements (e.g. LREE, Th) over compatible elements (e.g. Co) in the sediments, indicate relatively felsic average provenance compositions and a relatively severe weathering regime (McLennan et al., 1993).

On the La versus Th plot for Isan clay (Fig. 5), Isan clays data fall below the range of UC with all the samples

having a significant felsic component. This indicates that these clays have a uniform provenance (after Nyakairu and Koeberl, 2001). This is similar to the report of Odewumi et al. (2015) on Nahuta clay in northcentral Nigeria where the data showed a significant felsic component.

On the La versus Th plot (Fig. 5; after Nyakairu and Koeberl, 2001), The behavior of trace elements during soil formation is complex due to factors including weathering, adsorption, provenance and metamorphism (Garrels and Mackenzie, 1971; Kronberg et al., 1979; Nesbitt et al., 1980; Taylor and McLennan, 1985; Wronkiewicz and Condie, 1987). The trace element data show large variations compared to Post Archean Australia Shale (PAAS), North American Shale

Table 1: Major oxides (wt%) and trace element compositions (ppm) of Isan clays

Major oxides and Trace Elements	SA 1	SA 2	SA 3	SA 4
SiO ₂	55.49	57.32	62.05	60.47
Al ₂ O ₃	18.63	18.05	20.67	17.77
TiO ₂	0.05	0.2	0.2	0.02
Ba	24	65	236	210
Be	1	2	4	1
Co	1	6	9	9.6
Cs	0.4	0.3	3.9	1.6
Ga	5	6	47.8	21
Hf	5.3	7.5	11.2	10.8
Nb	26.4	34.9	63.1	54.2
Rb	3.7	10.8	105.9	76.78
Sn	608	14	37	11
Sr	7.8	14.5	51.2	68.9
Ta	5.2	2.8	6.4	3.9
Th	7.3	20.8	60.2	74.6
U	2.1	3.9	7.8	5.5
V	<8	41	88	56
W	4.6	3.2	6.4	1.3
Zr	178	230.4	373.7	410.7
Y	8.7	7.6	49.7	33.0
Mo	1.8	10.3	1.2	4.8
Cu	4.1	0.8	17.6	0.3
Pb	6.3	55.2	43.2	67.4
Zn	19	23	39	14
Ni	1.7	0.4	9.5	3.0
As	1.3	0.8	1.7	0.7
Cd	<0.1	<0.1	<0.1	<0.1
Sb	<0.1	<0.1	<0.1	<0.1
Bi	<0.1	0.2	0.7	0.1
Ag	<0.1	<0.1	<0.1	<0.1
Au	1.8	0.6	0.9	0.7
Hg	0.01	0.02	0.03	0.02
Tl	<0.1	<0.1	0.3	<0.1
Se	<0.5	<0.5	<0.5	<0.5
La/Th	1.80	1.42	1.77	1.37
La/Co	13.1	4.93	11.82	10.62
Th/Co	7.3	3.47	6.69	7.70
Th/Cr	5214.3	30588.2	28666.7	109705.9

anomaly in clays is interpreted as being inherited from igneous sources (McLennan and Taylor, 1991; Taylor and McLennan, 1985; Awwiller, 1994).

In addition, the REE patterns have been also used to infer the sources of clays since basic rocks contain low LREE/HREE ratios and no Eu anomalies whereas more silicic rocks usually contain higher LREE/HREE ratios and negative Eu anomalies (Cullers and Graf, 1983). Therefore, the REE patterns of the source rocks may be

Table 2: Rare earth element compositions (ppm) of Isan clays

REE	SA 1	SA 2	SA3	SA 4
La	13.1	29.6	106.4	101.9
Ce	20.9	45.8	202.7	230.6
Pr	2.55	10.3	19.48	22.10
Nd	10	38.2	66.8	59.3
Sm	1.84	5.90	11.17	13.4
Eu	0.27	1.10	1.81	2.25
Gd	1.52	6.33	9.37	7.90
Tb	0.22	0.78	1.31	1.48
Dy	1.46	3.57	8.01	9.30
Ho	0.25	1.24	1.58	1.64
Er	0.86	3.82	4.47	4.77
Tm	0.21	1.43	0.95	0.55
Yb	0.88	3.99	4.46	5.23
Lu	0.15	0.42	0.64	0.69
(Gd/Yb) _N	1.38	1.27	1.68	1.20
(La/Yb) _N	9.96	4.96	15.96	13.03
(Sm/Gd) _N	1.65	1.27	1.62	2.31
Eu/Eu*	0.50	0.55	0.54	0.67

Composite (NASC) and Upper Continental Crust (UC). Isan clays have similar contents in trace elements with Kuba and Nahuta clays (Odewumi et al., 2015) and are enriched in Y, Zr, Nb, Hf, Ta, Th and U

These elements are associated with heavy minerals such as zircon, which is resistant to weathering. They are preferentially partitioned into melts during crystallization and anatexis (Feng and Kerrich, 1990), and as a result, these elements are enriched in felsic rather than mafic rocks. Thorium and uranium behave differently during weathering, as U unlike Th is chemically mobile as U⁶⁺ and there is a tendency of the Th/U ratio to be elevated above Upper crustal values of 3.5 to 4.0 (McLennan et al., 1993). The La/Th ratio of Isan clay ranges from 1.37 to 1.80, this indicates a single source of the clays from Isan area (after Nyakairu and Koeberl, 2001).

The rare earth elements (REE) are suitable ones for provenance determination, because of their relatively low mobility during weathering and metamorphism. The results of the REE analyses of the Isan clay deposits are summarized in Table 2. All analyzed clay samples have similar concentrations of the REEs as shown in Figure 6.

Chondrite-normalized patterns (after Haskin et al., 1968) for Isan clay (Fig. 6) show enrichments of the LREEs. All samples have pronounced negative Eu anomaly ranging from 0.41 to 0.71. The Eu anomaly parallels the depletion in Na₂O and CaO suggesting that it developed at least partially in response to plagioclase weathering, where most of the Eu is hosted. The Eu

preserved in the clays (Taylor and McLennan, 1985; Wronkiewicz and Condie, 1987, 1989).

Isan clays have fractionated REE patterns. The LREE are enriched and the HREE patterns are almost depleted. The $(La/Yb)_N$ ratio ranges from 4.96 to 15.96 with $(Gd/Yb)_N$ ratio ranges from 1.20 to 1.68 in Isan clay and a negative Eu anomaly (0.50 to 0.67). These characteristics indicate that the source material was felsic and the negative Eu anomaly is regarded as evidence of a differentiated source, similar to granite (McLennan, 1989; McLennan et al., 1993; Taylor and McLennan, 1985, 1995).

Chondrite-normalized REE patterns of clays from Ville Marie area, Quebec, Canada (Panahi et al., 2000) is characterized by LREE enrichment and depleted, slightly "curved" HREE which is similar to the patterns of clays from Isan area. The negative Eu anomalies obtained from Isan clay (Fig. 6) attest to the provenance of the clays as felsic sources (probably from granites).

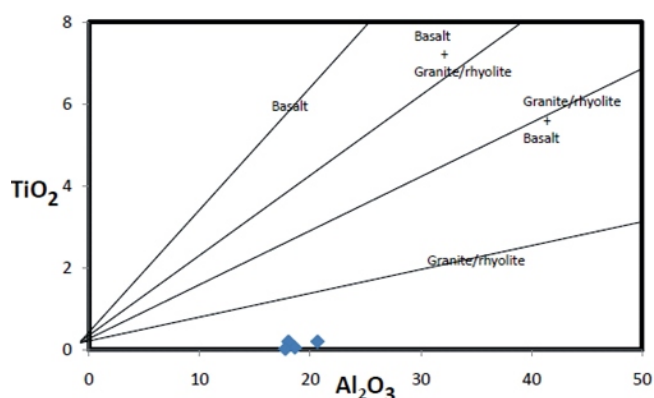


Fig. 4: TiO_2 (wt%) versus Al_2O_3 (wt%) binary plot of Isan clay (after Ekosse, 2001)

Conclusion

On the TiO_2 vs Al_2O_3 diagram, Isan clays plot in the granite/rhyolite field and on the La versus Th plot, Isan clays show a significant felsic component. This indicates that Isan clays have a uniform provenance and is predominantly from felsic rocks.

Isan clays have high ratios of La/Co, Th/Co, La/Cr, Th/Cr and Ba/Co; this indicates that Isan clays were derived mainly from felsic rocks. The relative enrichments of (normally) incompatible elements (LREE and Th) over compatible elements (Co and Cr) in Isan clays are suggestive of felsic provenance and a relatively severe weathering regime

Isan clays have fractionated REE patterns with LREE enrichment and HREE patterns are almost depleted. This further attests that the provenance was felsic and the negative Eu anomaly is regarded as evidence of a differentiated source, similar to granite.

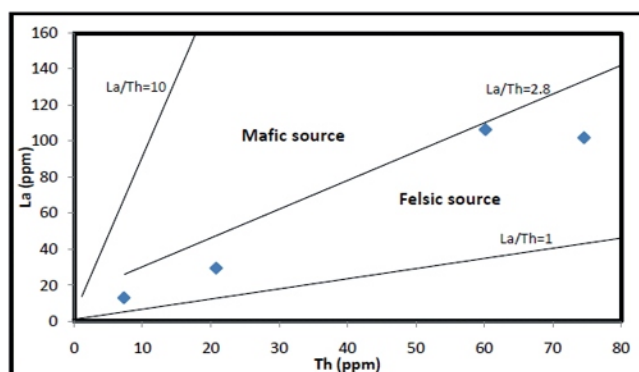


Fig. 5: La versus Th plot for Isan clay (after Nyakairu and Koeberl, 2001)

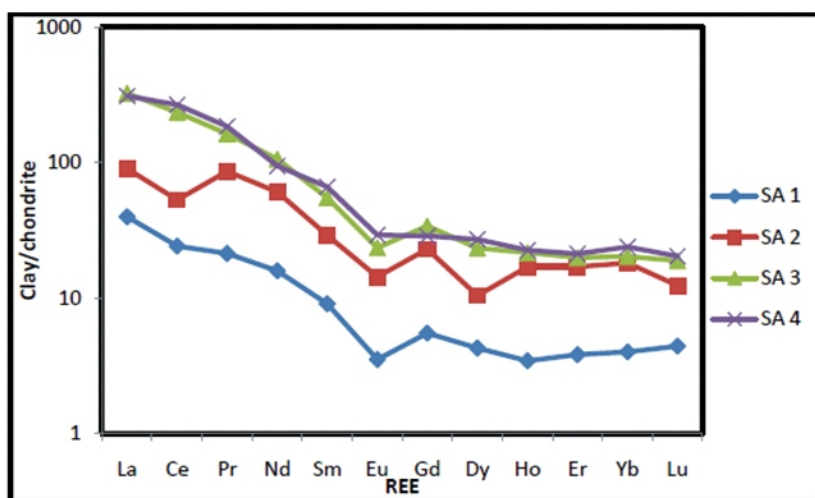


Fig. 6: Chondrite normalized plot of Rare Earth Elements of Isan clay

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