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INVESTIGATING THE GEOCHEMICAL CONTENT OF ANCIENT LATERITE BRICKS FROM BUKIT CHORAS ARCHAEOLOGICAL SITE USING X-RAY DIFFRACTION (XRD) AND X-RAY FLUORESCENCE (XRF)

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Abstract: The Bukit Choras Archaeological Complex is located at the summit of Bukit Choras, approximately 14 km north of Gunung Jerai in the state of Kedah, Malaysia. The Bukit Choras complex consists of a main structure believed to be a stupa, surrounded by mounds and water tanks. Surface remains suggest that the structures were constructed mainly of laterite bricks. However, the origin of the raw laterite used to make the laterite bricks is not yet known. An investigation needs to be carried out to determine the possible locations from where the raw laterites were mined. This research is focused on the geochemical composition of building materials used to construct the site, which may be determined using X-Ray Diffraction (XRD) and X-Ray Fluorescence (XRF). Nine raw material samples collected from various points around Bukit Choras were compared with two laterite bricks taken from the main structure in the complex. The XRD and XRF analyses detected minerals, major elements and trace elements that showed similar composition between the laterite bricks and several raw material samples. The findings of this study suggest that the raw materials used to construct the complex structures were likely taken from the south-eastern outcrop of Bukit Choras.

Keywords: X-Ray Diffraction, X-Ray Fluorescence, laterite bricks

Introduction

The Bujang Valley is a historical region where the port-polity of Ancient Kedah was located, spanning from Bukit Choras in the northwest, to Gunung Jerai and the Merbok-Muda River valley in the south. Some traces of the settlement have also been found further south in the town of Cherok Tokkun in the neighbouring state of Penang. Known for the existence of many cultural remains, the valley is among the richest archaeological site in Malaysia. Among the artefacts discovered include tradeware, iron smelting sites, potteries as well as Hindu-Buddhist sculptures, inscriptions and shrines (Khaw, 2011).

By studying the building materials of structural ruins, archaeologists can gain a better understanding on the history and culture of Ancient Kedah. To date, more than 88 structural ruins have been documented in the area, comprising Hindu, Buddhist and secular structures (Allen, 1988; Khaw, 2011; Saidin, 2016). Observation on these archaeological sites in the context of their geomorphological environment suggests that the structures were constructed using materials that are accessible from the immediate surroundings. In the Bujang Valley, the common building materials found in the ancient ruins include clay, laterite, granite and, to lesser extent, river pebbles.

The Bukit Choras complex is one of several sites in the Bujang Valley that is believed to host a Buddhist temple (Figures 1, 2 and 3). Surface remains suggest that the structures are almost entirely made of laterite bricks, which are among the raw materials commonly used in the area (Rahman & Yatim, 1992). Although Bukit Choras and its surrounding areas is abundant with iron rich materials, the exact location from where the ancient builders obtained the raw laterite and brought it to the site to build the structures remains a mystery.

Information on the possible locations from where the raw laterite is mined may provide an important insight into the material culture of Ancient Kedah's society, their mode of transportation, as well as their form of manipulation of local resources. The questions regarding the origin of raw laterite used to construct the structures in Bukit Choras may be ascertained by identifying the mineral content, as well as the major and trace elements in the laterite bricks. These elements constituted the chemical fingerprint of the structures, which may be compared with the contents of naturally-occurring laterites found around the area.

The geochemical composition may be determined through X-Ray Diffraction (XRD) and X-Ray Fluorescence (XRF), which have been widely applied to study the origin of raw materials from other sites of the Bujang Valley. The XRD and XRF methods have been used to study ancient bricks at Candi Bukit Pendiat (Ramli *et al.*, 2011; Ramli *et al.*, 2013a), Candi Sungai Mas (Ramli *et al.*, 2012), Candi Pengkalan Bujang (Ramli *et al.*, 2011; Ramli & Rahman, 2012; Ramli *et.al.*, 2013b), Candi Kampung Baru (Ramli *et al.*, 2018) as well as the brick ruins in Sungai Batu (Rapi *et al.*, 2020). These 7th to 13th century C.E. Hindu and Buddhist sites are mostly located near the banks of Sungai Bujang, Sungai Merbok and Sungai Muda, which are rich in clay.

In the studies, raw clay samples were randomly collected from sources located nearest to the sites and brought to the laboratory for XRD and XRF analyses (Ramli *et al.*, 2011; Ramli & Rahman, 2012; Ramli *et al.*, 2012; Ramli *et al.*, 2013a; Ramli *et al.*, 2013b; Ramli *et al.*, 2018; Rapi *et al.*, 2020). Comparison between XRD and XRF analysis results of the ancient bricks and raw clay revealed similar chemical properties. It has thus been demonstrated that the materials used to construct all the ruins had been obtained from the nearest localities where the raw materials were available. The use of XRD and XRF in the present study aims to investigate the geochemical composition of the laterite bricks at the archaeological site and origin provide possibilities on the origins of the building materials of Bukit Choras archaeological structures.

Figure 1: Location of Bukit Choras in Bujang Valley, Kedah, Peninsular of Malaysia

Figure 2: Remnants of a main archaeological structure at Bukit Choras in Kedah, Malaysia

Figure 3: Remnants of a subsidiary structure at Bukit Choras in Kedah, Malaysia

Research Area

The archaeological site is located at the south-western tip of a crescent-shaped ridge (N5.964121° E100.418753°) extending from north to south. The ridge is surrounded by paddy fields, a Chinese cemetery, a bauxite quarry, a religious school, villages and rubber plantations on the slopes. The nearest main town is the state capital of Alor Setar, which is about 26 km away. Located in the northernmost limit of the Bujang Valley, Bukit Choras is the only known archaeological site that is isolated from the rest, which are mostly concentrated in the Merbok-Muda Valley south of Gunung Jerai.

Preliminary excavation by Quaritch-Wales (1940) has led to the discovery of a laterite basement measuring 6.87 m long, 7.16 m in width and 0.91 m in height, besides a low platform measuring at 6.4 m long and 6.7 m wide, as well as other artefacts like Buddhist inscriptions, iron nails and broken pottery. A survey by Kamaruddin (1989) identified two large trenches to the east of the structures, which he believed to be ancient reservoirs. These structural ruins are relatively well-preserved, where the lower portion of the ancient buildings may be distinguished from the surface (Khaw *et al.,* 2020; Muztaza *et al.,* 2020). The Bukit Choras complex can be dated back to the 6th or 7th century C.E. Geophysical mapping at the site has detected many more structures that are buried (Khaw *et al.,* 2020; Muztaza *et al.,* 2020).

The area around Bukit Choras is mainly flat terrain of approximately 7 m above sea level, although there are several smaller hills with similar elevation and geological setting. The slopes of Bukit Choras are extremely steep, especially in the north-western part of the ridge. However, the south-eastern slope has more gradient and thus, providing easier access to the hilltop complex. The northern part of Bukit Choras is near the banks of Sungai Sala, which flows westward into the Straits of Malacca.

The geological composition of north-western Kedah, where Bukit Choras is located, consists of the Kubang Pasu Formation (Carboniferous) that overlies the Mahang Formation (Ordovician-Devon), with intervals of chert, mudstone inter-bedded with sandstone, and thick sandstone sequence representing the upper layer of the formation as shown in Figure 4 (Harun & Jasin, 2000). Quaternary sediments in the area consist of two sequences: The upper part is the Sugar Formation (marine alluvium) and the lower part is the Beruas Formation (terrestrial alluvium). The flat terrain around the hill is made up of unconsolidated quaternary marine and continental deposits, which are clay, silt, sand, peat and minor gravel (Jones, 1981). Based on the topography map in Figure 5, Bukit Choras is the highest point of elevation in the area at the height of approximately 50 m. Figure 6 shows a picture of the hill taken from the southern access point.

There are several hills within the area which share similar geological characteristics. These hills are Bukit Besar, Bukit Chepat, Bukit Jambul, Bukit Pinang, Bukit Raya Dalam, Bukit Seni and Bukit Kobah. Sungai Sala to the north of Bukit Choras has the potential features of an ancient route for ships to navigate inland. The unique position of Bukit Choras complex has raised much curiosity regarding its form of architecture and role in the history of Ancient Kedah.

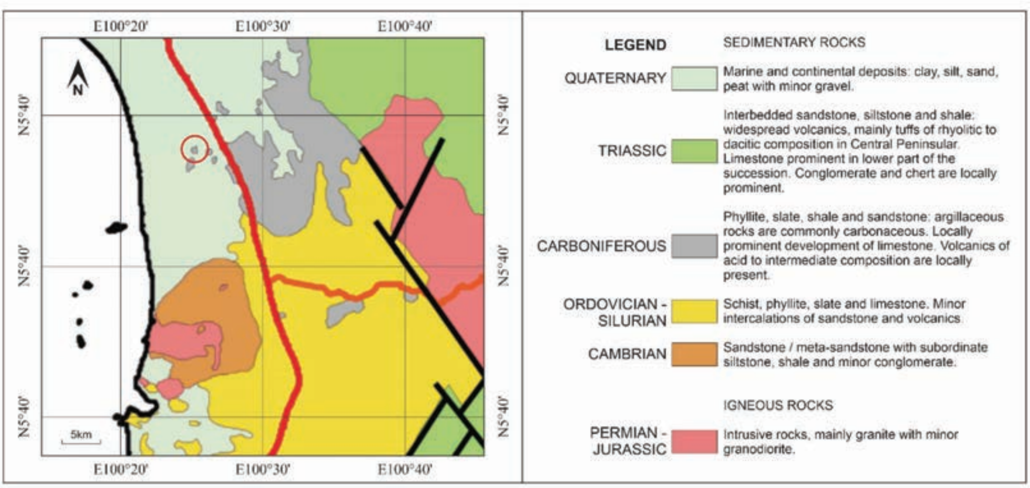

Figure 4: The geological setting of the study area (red circle)

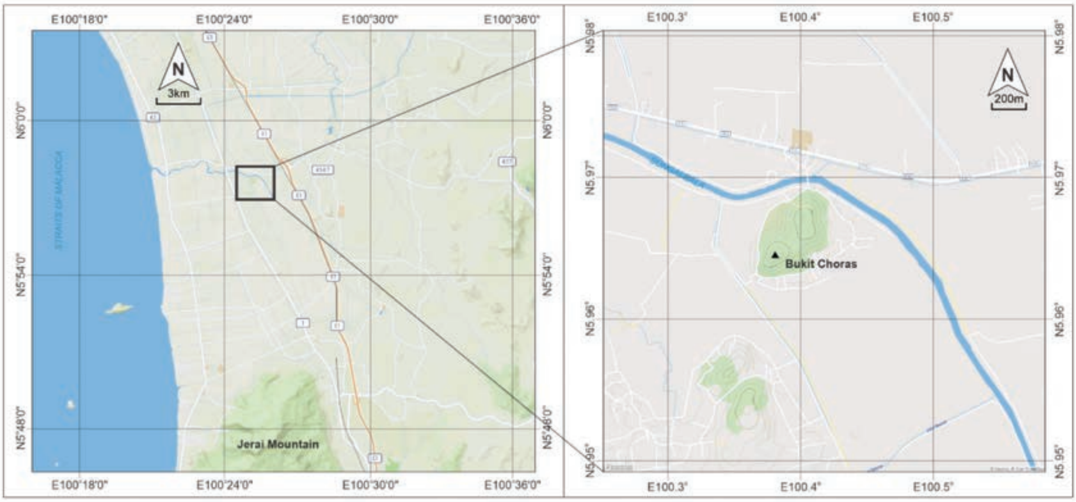

Figure 5: Topography map of Bukit Choras showing its elevation and the crescent ridge where the archaeological complex is located

Figure 6: A picture of Bukit Choras taken from the southern access point

Materials and Methods

Material and Sample Collection

Previous works had shown that the structures in Bujang Valley were mostly built using materials available in their surroundings (Ramli *et al*., 2011; Ramli & Rahman, 2012; Ramli *et al*., 2012; Ramli *et al*., 2013a; Ramli *et al*., 2013b; Ramli *et al*., 2018; Rapi *et al*., 2020). Similarly, the building materials of Bukit Choras complex could have also been extracted from nearby areas. The raw laterite could have been taken from the hill slopes or foothills, wherever the building materials might be easily extracted and conveniently transported to the site.

To determine the possible locations from where raw laterite could have been mined, focus was given to laterite rich areas around the archaeological site. Thus, a foot survey from the summit of Bukit Choras down to the foothill was carried out as shown in Figure 7. Samples were randomly collected at every locality where there was a laterite stone outcrop, and scattered laterite rocks or soil were visible. This survey had identified several localities with the presence of red mudstone, red shale and most importantly laterite soil. The south-eastern and western foothills of Bukit Choras were found to have the most abundant sources of laterite. Samples of other raw materials, such as red mudstones and laterite soil were also collected.

During the fieldwork, samples were collected using either hand pick or hand auger techniques. Some samples were found fully exposed on the surface or partially buried. These samples were extracted manually using rudimentary equipment such as a geological hammer and pickaxe, while some could just simply be picked by hand. As for buried samples, they were extracted via the auger method, which involved inserting a steel rod into the soil using a hammer. Unlike samples which were collected from the surface, the auguring method provided geological samples which were not exposed to the elements, as well as information regarding stratigraphy. A total of 11 samples were obtained from the slopes and foothills surrounding the archaeological complex. The coordinates, location and type of samples are shown in Table S1 and S2. The collected samples were properly wrapped, and their locations and physical descriptions were recorded. The samples were then brought to the Earth and Material Characterization Laboratory, Centre for Global Archaeological Research at Universiti Sains Malaysia (USM) for XRD and XRF analyses. The colour of the samples was described according to the Munsell colour chart of the Washable Edition, Munsell Colour (firm), 2010.

Samples BCL1, BCL2, BCBawah, BCBM2, BCBM3, BCBK2a, BCBK2b, and BCBK(U) were collected manually by hand. Table S1 summarizes the colour, weathering profile and laterite profile of the hand-picked samples. BCL1 and BCL2 were two laterite bricks taken from the main structure at the archaeological site. Sample BCL1 was olive yellow 5Y 6/8, reddish yellow 7.5YR 6/8 and Yellow 10YR 7/8. Sample BCL2 was Reddish Yellow 5YR 7/8, Red 2.5YR 4/8 and Dark reddish Gray 2.5Y 4/1. BCBawah was a laterite sample taken from a trench at a paddy field near the western foothill of Bukit Choras.

Figure 7: A map showing the locations (red and blue circle) where laterite samples were collected around Bukit Choras

Scattered laterite boulders could be seen around the area. The colour was 10YR 8.8 Yellow 5YR 4.1 Dark Grey 7.5R 4/6 Red and 5YR 6/4 Light Reddish Brown. BCBM2 and BCBM3 were laterite samples collected from the south eastern outcrop of Bukit Choras, where a large amount of raw laterite could be observed. The colour of BCBM2 was Red 2.5YR 5/8, Weak red 2.5YR 5/2 and Yellow 10YR 8/8. Meanwhile, BCBM3 was Olive 5Y 5/4 and Reddish Yellow 7.5YR 7/8 and 5YR 6/8. Sample BCBK(U) comprised red mudstones obtained from a bauxite quarry at the northern slope of the hill, where iron-rich mineral ores could be observed. The colour was 7.5R 5/4 Weak Red 5R 5/2 Weak Red 2.5YR 5/4 Reddish Brown. BCBK2a and BCBK2b were two red mudstone samples collected from Water Tank II. Geophysical survey at the area suggested the presence of iron-rich minerals (Nasha *et al.,* 2020; Nordiana *et al.* 2020). The colours were 7.5YR 6/8 reddish yellow 10YR 8/8 yellow.

Samples BCP1A, BCP2 and BCP2B were extracted using the hand auger method. At these three locations, the samples were augured at various depths between 0 to 129 cm. At these three points, the hand auger is driven into the soil as far down as possible, until the soil composition becomes too hard and compact for the steel rod to penetrate any further. Table S2 summarizes the colour, weathering zone profile and laterite profile of the hand augured samples by depth. After the samples were pulled out, iron-rich minerals like laterite and red mudstones were selected for analysis. Sample BCP1A was taken from the south eastern part of the main structure, where the soil could be observed on the surface. Sample BCP2 was taken from an elevated point 23 m above sea level northwest of Bukit Choras, where laterite could also be observed on the surface. As for sample BCP2B, it was extracted at the base of water tank II at the eastern part of the complex.

The auguring data shows that the composition from the hilltop to the hillside of Bukit Choras has a uniform weathering profile with the presence of topsoil, ferricrete/duricrust layer and mottled zone (Table S2). Surveys at the foothill of Bukit Choras (12 m above sea level) revealed lateritic zone patterns namely (1) humus zone, (2) duricrust, (3) mottled zone, (4) palid zone (5) saprolite (partially altered parent rock – horizon C) (6) parent rock–argillaceous Mahang Formation (Figure 8). Regolith of laterite profile at Bukit Choras is not very thick, which is only between 1.5 -

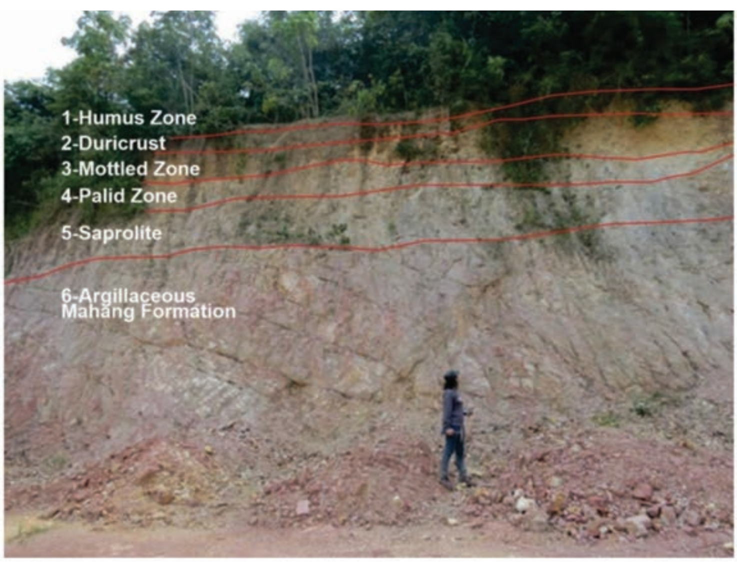

Figure 8: Field photograph of southern laterite profile area showing different zones from argillaceous Mahang Formation to top humus zone

2.0 m. Flat landscape surrounding Bukit Choras consisted of a thick layer of alluvium (5 - 10 m), which is the deposition of quaternary alluvial sediments. Therefore, facies changes between the hilltop area and the surrounding flats were not identified, and the weathering profile can only be observed at the area between the hilltop and hillside of Bukit Choras.

Methods

After the samples were collected from the site and brought to the laboratory, they were prepared for XRD and XRF analysis. The raw materials and laterite brick samples were cleaned and dried before being crushed and pulverized using the PM100 milling machine (Retch GmbH, Haan, Germany) to standardise the grain size at 20 - 30 µm. The crushed samples were dried for two hours in an oven (Memmert GmbH, Schwabach, Germany) at 105°C at half fresh airflow.

The XRD analysis was used to detect mineral composition in the samples. Powdered samples were transferred using a spatula to a 25 mm PMMA sample holder and flattened by placing a piece of glass on top before being analysed with a Bruker D8 Advance XRD machine (Bruker AXS, Karlsruhe, Germany). Scanning parameters were set at 10 - 70° (2theta) ranges, 0.04°/step with Copper (Cu) X-Ray tube (Rapi *et al.,* 2020). Diffractograms were analysed using the DIFFRAC.EVA Ver 3.0 software for qualitative identification based on more than 200,000 reference materials, such as organic, inorganic, mineral, hydrated materials, forensic materials and pigments included in the Powder Diffraction File provided by the International Centre for Diffraction Data (ICDD). However, according to Bunaciu *et al.* (2015), there might be some limitations in performing XRD, such as the sample should be homogeneous for best identification of unknown elements. For mixed material/minerals, the detection limit was approximately 2% of the total sample and peak overlay might occur in high angle reflections.

The XRF analysis was used to detect major and trace elements in the samples. In this study, all samples were also analysed with the Axios Max Panalytical XRF instrument (Panalytical B.V., Almelo, the Netherlands) to get detailed insight into the geochemical properties of the samples. The XRF analysis was performed by preparing fuse beads for major element analysis, while pressed pellets embedded in boric acid were used for trace element analysis. A total of 1.5 g of dried powdered samples was placed on a petri dish using a spatula. Each sample was then transferred into a mould and mixed with 5.5 g of boric acid before being compressed with a plastic rod into a pellet. Another 0.5 g of sample was added with 5 g of flux and smelted with Fluxana to produce a glass pellet. All samples were also burnt in a furnace at 1,200°C and weight changes were reported as *Loss on Ignition* (L.O. I) values (Abdullah *et al.,* 2018). In XRF analysis, a detection limit called the Lower Limit of Detection (LLD) was taken into consideration while interpreting the data. It was assumed that the smallest amount of analyte in the specimen would be related to the lowest net peak intensity of the analyte present in the XRF spectrum (Kadachi & Al-Eshaikh, 2012). Some trace elements might fall below the detection limit due to detector sensitivity and homogeneity of the samples.

Results and Discussion

Laterite is a red to reddish brown clayey material, formed by the weathering of iron rich rocks under tropical or sub-tropical environment, and is usually found compacted into a matrix (Little, 1969; Nichol, 2000). Most importantly, it could easily be cut and chiselled into blocks that harden when exposed to the sun, which made it a suitable construction material (Watsantachad, 2005). Laterites were usually extracted from a natural outcrop before being cut and trimmed into intended shapes and measurements. This material had been used in eastern India in the Neolithic period, as well as in Southeast Asia since the 5^{th} century C.E (Smith & Mohanty, 2017; Watsantachad, 2005).

The XRD analysis on the 11 samples collected from Bukit Choras generally showed the presence of several minerals, such as quartz (SiO_2), kaolinite ($Al_4(OH)_8(C_4O_{10})$), kaolinite-1A ($Al_2Si_2O_5(OH)_4$), muscovite-2Ml ($KAl_2(Si,Al)_4O_{10}(OH)_2$), hematite ($Fe_{1.957}O_3$), microline ($KAlSi_3O_8$) and goethite ($FeO(OH)_2$) (Table 1). The minerals that characterised the building samples taken from the ruins in the Bukit Choras complex were quartz, kaolinite, goethite and hematite, all showing similar composition with those reported by Watsantachad (2005) (Table 1).

Samples taken from various sites around the hill that showed similar mineral content with the structural ruin samples were BCBM2, BCBM3 and BCBK(U), which contained quartz, kaolinite, and hematite (Figure 9). Goethite was detected only in the laterite brick samples, and not in any other natural soil and rock samples analysed in this study. This mineral is formed as a result of weathering of iron rich minerals through the oxidation of Fe^{2+} to Fe^{3+}, and composed of Fe, O and OH, as well as other impurities like Al_2O_3, MnO, CaO and SiO_2 (Mohapatra *et al.,* 2008). As for BCBawah, the sample showed similarities with BCL1 and BCL2 in terms of the presence of quartz and hematite. However, the presence of kaolinite-1A, instead of kaolinite, showed that the mineral composition was slightly different.

The quantitative value of major elements obtained using XRF analysis is shown in Table 2. In all 11 samples, Silica (Si) was recorded as the highest dry weight percentage in the range of 25.97% to 56.55%. The second highest is Ferum (Fe) in the range of 5.36% to 45.72%, followed by Aluminium (Al) in the range of 17.79% to 27.08%. Other elements showed significantly smaller percentage as compared to the first three elements.

Figure 10 shows the two-dimensional scatter plot of Si vs Al to visualise the correlation between laterite brick BCL1 and BCL2 samples

Table 1: Mineral content of building materials and soil samples based on XRD analysis

SAMPLE	LOCATION	QUARTZ	KAOLINITE	KAOLINITE-1A	HEMATITE	GOETHITE	MUSCOVITE-2M1	MICROCLINE
BCL1	Main structure	X	X		X	X		
BCL2	Main structure	X	X		X	X		
BCBawah	Paddy field trench at western foothill	X		X	X			
BCBM2	Laterite outcrop southeast of the hill	X	X		X		X	
BCBM3	Laterite outcrop southeast of the hill	X	X		X			
BCBK(U)	Bauxite quarry at northern slope	X	X		X		X	
BCBK2a	Water tank II	X	X				X	
BCBK2b	Water tank II	X	X				X	
BCP1A	Southeast of the main structure	X		X			X	X
BCP2	Northwestern slope	X		X			X	
BCP2B	Water tank II	X	X				X	X

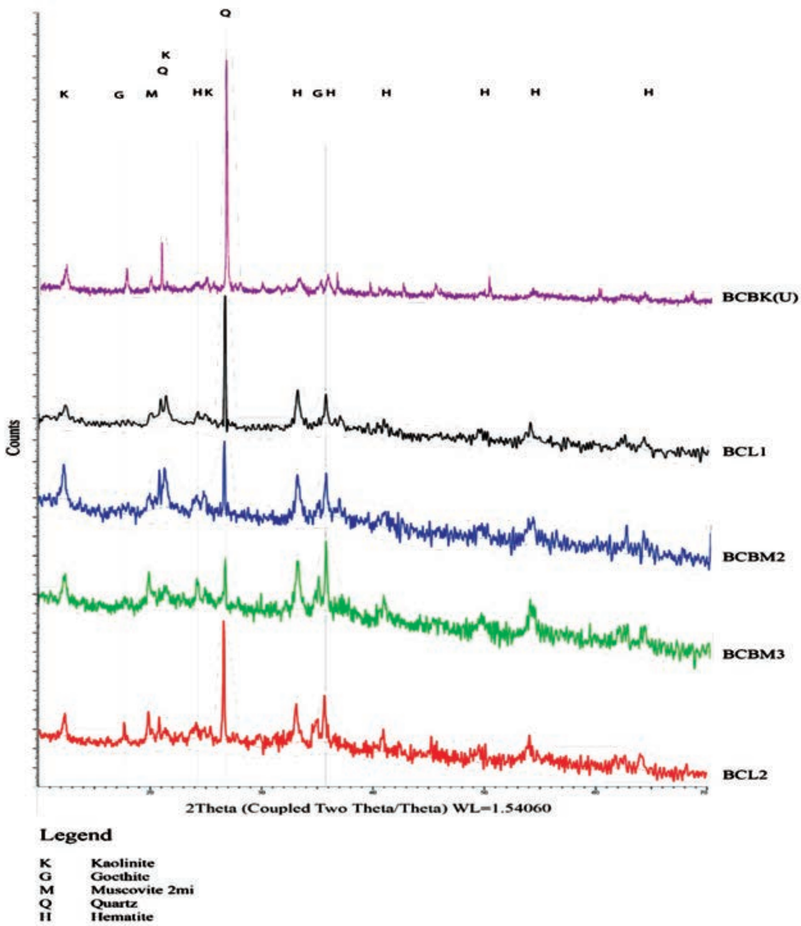

Figure 9: XRD diffractorgramme for BCL1, BCL2, BCBK(U), BCBM2 and BCBM3 samples

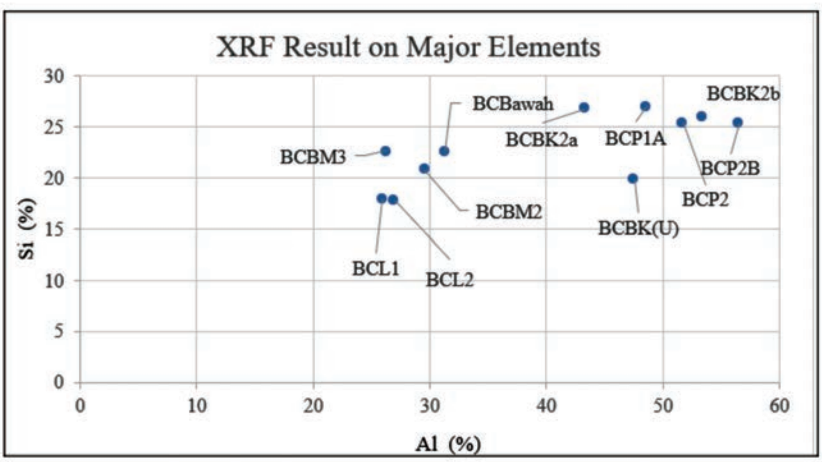

Figure 10: Two-dimensional scatter plot for the percentage of major elements (Si vs Al)

with other raw soil and rock samples. Si and Al were selected because they had the highest percentage and were among the most common elements found in laterite (Watsantachad, 2005). Both BCL1 and BCL12 showed miniscule variation in terms of the composition of most major elements. However, in comparison with other samples, a pattern could be observed in terms of the dry weight percentage between Si and Al.

In the plot, samples BCL1, BCL12, BCBM2 and BCBM3 were clustered together within the 25% to 30% range for Si and 17% to 23% range for Al. The BCBawah sample, on the other hand, was plotted near the cluster. As for other samples, they were located farther away, which were within approximately 20% to 27% range for Al and 43% to 57% range for Si. The XRF results clearly demonstrated the close correlation of major element composition between BCL1, BCL12, BCBM2 and BCBM3, while the BCBawah sample showed slight variation of the percentage.

Aside from detecting major elements, the XRF analysis was also used to determine the concentration of trace elements in the samples (Table 3). There were 22 trace of elements that have been recorded using the XRF Omnion standardless analysis. The elements' concentration in every sample was recorded in parts per million (ppm) due to the trace amount present in the samples. The elements such as Cl, Cr, Nb, Rb, S, Y, Ga, Zr and Ba were detected in the samples. Ba was recorded at higher concentrations in BCBK2a, BCBK(U), BCBM3, BCBawah, BCP1A, BCP2 and BCP2B. Figure 11 shows the two-dimensional scatter plot for the concentration of Zr versus Cr. Zr and Cr were chosen because they were among the most stable elements in soil (Yamasaki *et al.,* 2016). Samples BCL1 and BCL2 showed close reading for Cr at 684 ppm and 648 ppm, respectively, while there was a gap for Zr concentration (321 ppm and 452 ppm, respectively). In comparison with other samples, BCL1, BCL2, BCBM2, BCBM3 and BCBK2a were clustered together within the range of 608 ppm to 684 ppm for Cr. As for Zr, these samples were scattered across a wider range, which was from 151 ppm to 452 ppm. Sample BCL2 was detected to have the same concentration of Cr with BCBK2a and BCBK2b (648 ppm), while Zr concentration for BCL1 (321 ppm) was extremely close to the concentration in BCBK(U) (319 ppm).

The XRF analysis detected several immobile elements such as Fe, Al, Ti, Zr, Y, and Nb. According to Budihar & Pujar (2018), these geochemical elements remained immobile during incipient to moderate stages of

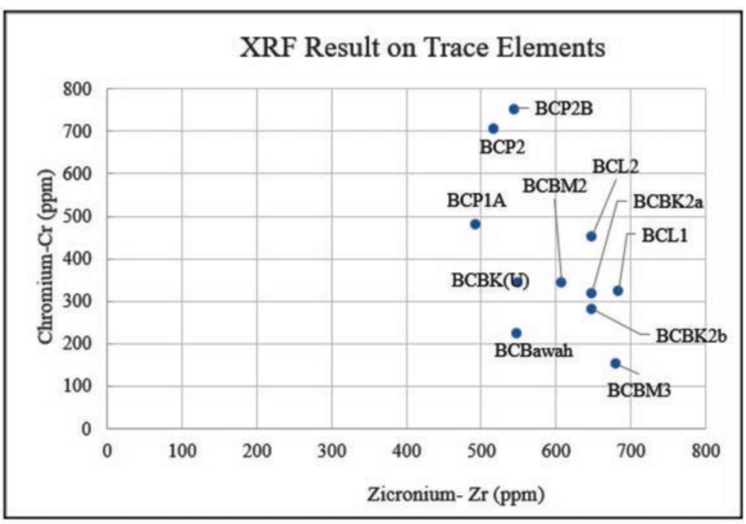

Figure 11: Two-dimensional scatter plot for concentration of trace elements (Cr vs Zr)

weathering. The two-dimensional scatter plots on the ratios for the immobile elements of Zr vs Y (Figure S1), Al vs Nb (Figure S2), Fe vs Al (Figure S3) and Ti vs Al (Figure S4) generally exhibit lesser variations between BCL1, BCL2, BCBM2 and BCBM3. The two-dimensional scatter plots of immobile elements ratios seem to show similar trends with those of Si vs Al and Cr vs Zr (Figures 10 and 11). The results therefore suggest that these four samples share similar geochemical properties.

The laterite bricks from BCL1 and BCL2 constituted the main building materials for the structural ruins in Bukit Choras. Our study suggested that raw materials used to produce the laterite blocks might have been taken from the south-eastern outcrop of Bukit Choras, which is directly east of the archaeological complex. This was supported by XRD analysis, where a similar mineral composition was observed between both laterite brick samples and samples extracted from the south-eastern outcrop (i.e., BCBM2 and BCBM3). The mineral composition shared by all four samples was quartz, kaolinite and hematite. The percentage of major and trace elements detected by XRF analysis had also shown clear correlation between the two brick samples with BCBM2 and BCBM3. These included the compositions of Si, Al, Zr, Cr as well as other immobile elements which were clustered together as compared to other soil and laterite rock samples (Figures 10, 11, S1, S2, S3 and S4).

According to Watsantachad (2005), the chemical and mineral composition of laterite mostly comprised sesquioxides of Al and Fe, possibly with minerals such as quartz, kaolinite, goethite, hematite and gibbsite. Other elements commonly detected in laterite included Ti, Ca, Mg, Mn, K and Na (Young, 1976; Watsantachad, 2005). Most of these minerals and elements had been detected in XRD and XRF analyses. The laterite brick samples from the main structural ruins of Bukit Choras complex contained signature minerals and elements which could be compared with other samples of raw materials such as quartz, goethite, hematite and kaolinite, Zr, Cr, Al and Si. Comparison of mineral content and chemical composition with other samples found similarities with samples BCBM2 and BCBM3, taken from the south-eastern outcrop of Bukit Choras foothill (Figure 12), which

Table 2: Quantification of major elements in samples using XRF analysis.

Sample	Dry Weight (%)										
	Si	**Ti**	**Al**	**Fe**	**Mn**	**Mg**	**Ca**	**Na**	**K**	**P**	**L.O.I**
BCL1	25.97	0.78	17.95	45.72	0.03	0.23	0.06	0.11	0.86	0.14	9.28
BCL2	26.98	0.83	17.79	43.28	0.02	0.20	0.07	0.03	0.77	0.14	-
BCBawah	31.32	0.70	22.50	33.44	0.01	0.22	0.05	0.16	3.27	0.03	-
BCBM2	29.55	0.79	20.75	36.70	0.03	0.44	0.14	0.02	1.72	011	-
BCBM3	26.37	0.65	22.64	37.75	0.01	0.17	0.06	0.15	2.89	0.07	-
BCBK(U)	47.55	0.74	19.92	23.22	bdl	0.13	0.05	0.48	3.21	0.03	5.02
BCBK2a	43.32	0.84	26.78	17.70	bdl	0.12	0.05	0.70	4.01	0.16	7.89
BCBK2b	53.45	0.85	25.96	9.41	bdl	0.13	0.06	0.63	4.42	0.08	5.67
BCP1A	48.62	0.95	27.08	11.12	bdl	0.12	0.05	0.81	4.25	0.06	6.11
BCP2	51.70	0.95	25.40	9.96	bdl	0.15	0.05	0.63	4.19	0.06	6.51
BCP2B	56.55	0.92	25.35	5.36	bdl	0.14	0.05	0.71	3.95	0.04	5.88

*bdl: Below Detection Limit

Table 3: List of trace elements with quantitative value in samples collected from Bukit Choras

Sample	Elements (ppm)																				
	As	**Ba**	**Ce**	**Cl**	**Cr**	**Cu**	**Co**	**F**	**Ga**	**Hg**	**Nb**	**Ni**	**Pb**	**Rb**	**S**	**Sr**	**Th**	**V**	**Y**	**Yb**	**Zn**	**Zr**
BCL1	439	bdl	Bdl	137	648	55	bdl	bdl	bdl	bdl	22	bdl	59	68	1070	35	bdl	701	30	bdl	49	321
BCL2	528	bdl	Bdl	190	648	66	bdl	bdl	289	bdl	47	bdl	108	76	813	27	bdl	824	29	193	39	452
BCBK2a	bdl	4017	Bdl	195	648	bdl	bdl	bdl	83	bdl	84	314	bdl	733	1496	285	108	Bdl	128	bdl	84	319
BCBK2b	bdl	bdl	Bdl	235	648	bdl	bdl	bdl	49	bdl	66	101	bdl	546	627	169	bdl	Bdl	89	bdl	29	282
BCBK(U)	bdl	4782	Bdl	232	550	161	bdl	bdl	47	bdl	58	bdl	100	544	751	222	bdl	Bdl	126	bdl	184	343
BCBM3	787	1083	Bdl	221	682	bdl	bdl	bdl	35	bdl	48	bdl	140	288	971	46	bdl	408	54	bdl	bdl	151
BCBM2	174	bdl	Bdl	164	608	bdl	bdl	bdl	249	bdl	40	89	74	154	698	bdl	97	608	38	bdl	43	345
BCBawah	534	1227	Bdl	193	548	92	bdl	bdl	36	bdl	38	133	115	368	741	52	bdl	Bdl	58	bdl	60	223
BCP1A	bdl	3980	Bdl	231	494	bdl	1998	679	93	bdl	113	214	bdl	1053	46	549	139	Bdl	148	bdl	75	481
BCP2	215	4840	953	205	518	bdl	bdl	bdl	133	bdl	201	bdl	167	1184	185	471	110	Bdl	219	bdl	99	705
BCP2B	bdl	6417	1638	160	545	128	bdl	398	187	93	154	322	131	1366	137	622	bdl	Bdl	213	bdl	bdl	750

*bdl: Below Detection Limit

could be reached from the site through the most convenient and gradient route.

Conclusion

Previous studies on structural ruins in the Kampung Sungai Mas, Kampung Pengkalan Bujang, Kampung Baru and Sungai Batu Archaeological Complexes had shown that the building materials used by ancient builders mostly came from their surrounding areas. In this study, two laterite brick samples were taken from the main structure at Bukit Choras. Survey around Bukit Choras had identified two areas abundant in raw laterite, one of them being a natural laterite outcrop, which was at the south-eastern foothills. Several other samples were also taken at potential areas where iron rich minerals could be spotted. Samples BCBM2 and BCBM3, which were collected from the south-eastern natural laterite outcrop, showed close resemblance to the mineral contents of BCL1 and BCL2. Samples BCL1, BCL2, BCBM2 and BCBM3 showed relatively similar composition for elements such as Si, Al, Zr and Cr. These results backed the assumption that the raw laterite used to make the ancient structures was possibly extracted from the southeast outcrop, which is around 400 m east of the complex.

There is a track connecting the laterite outcrop at the foothills to the archaeological complex. The track is a gradually inclined slope compared to other routes, which were steeper and more difficult to trek. This route was probably the easiest to collect and transport the laterite to the archaeological site. This study backed the hypothesis that the raw materials used to construct the ancient structures on Bukit Choras were likely obtained nearby, just like other sites in the Bujang Valley. The abundance of raw laterite in Bukit Choras was also possibly one of the reasons why the area was selected as the site for this religious complex. The usage of construction materials from the immediate surroundings of Bukit Choras suggested that the builders might be locals who knew the terrain well. Like other sites in the Bujang Valley, the finding in Bukit Choras archaeological complex

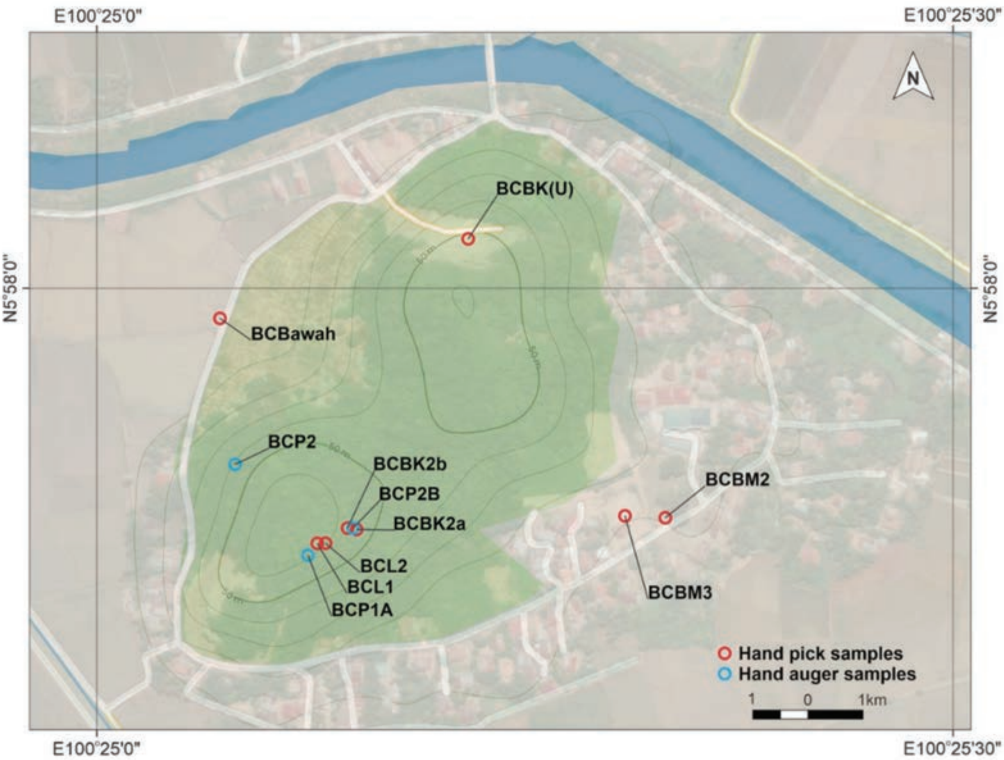

Figure 12: Map showing the contour of Bukit Choras

demonstrates the ability of the society of Bukit Choras in manipulating their local resources to construct religious monuments.

The ancient society living in Bukit Choras and the adjacent areas was part of the Ancient Kedah's network of feeder-points, consisting of several riverine and coastal settlements. These feeder-points which also included the sites of Kuala Selinsing, Perlis, Cherok Tokkun and Takuapa contributed to the economic development of the main entrepot of Ancient Kedah. The position of Bukit Choras to the north of the Gunung Jerai possibly made it an important relay point for travelers coming from the north, before continuing their journey to the main port of Ancient Kedah, located at the Merbok-Muda River valley. The area of Bukit Choras also had favourable geomorphological configuration with coastal hills and a river, which acted as natural lighthouse with continuous supply of freshwater. The ancient settlement could have composed mostly of houses built on tall stilts located along the ancient shorelines and Sungai Sala estuary, similar to other coastal and riverine settlements of the Malay Peninsula.

The society there was most possibly actively involved in the mining of the raw material as well as the construction of the sites. They also seem to be well-aware of the presence and location of suitable raw materials needed for the construction. The structures at the top of Bukit Choras appear to have been quite a large complex, with well-planned building layout and water reservoirs. This suggests that the local workforce was not only able to mine and transport the raw laterite to the construction site, but were also skilled in trimming the laterite stones into right shapes and measurements, as well as assembling them to form the edifices. The findings in Bukit Choras therefore suggest that the population of the area and its surroundings at that time could have been considerable, and while the workforce was well-trained and had considerable knowledge in building technology. Such cultural development in Bukit Choras was synchronous with the period of economic and political growth of Ancient Kedah from the 6th to the 13th Century C.E.

Our findings had thus formed new narratives regarding the cultural matrix of the ancient society in Bukit Choras, and their place in the historical framework of Ancient Kedah. This could contribute towards future management, conservation and restoration of this archaeological complex not only as a Malaysian cultural heritage site, but also as potential spot to be developed as a geotourism or archaeotourism site.

Acknowledgements

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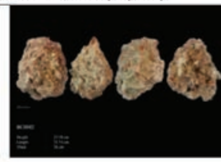
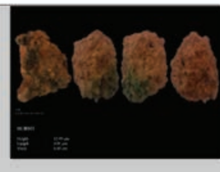
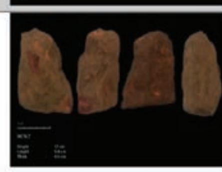
SUPLEMENTARY TABLES

Table S1: Summary of sample taken by hand pick at Bukit Choras

SAMPLE	LOCATION	COLOUR / WEATHERING PROFILE / (height x length x thickness) (cm)		LATERITE PROFILE
BCBM2	South-eastern outcrop N 5.964536° E 100.422199° Ele = 12 m	2.5YR 5/8 red 2.5YR 5/2 weak red 10YR 8/8 yellow B horizon 23.6 x 21.5 x 20.0		Ferricrete / duricrust *
BCBM3	South-eastern outcrop N 5.964498° E 100.421823° Ele = 12 m	5Y 5/4 olive 7.5YR 7/8 reddish yellow 5YR 6/8 reddish yellow B horizon 13.0 x 9.9 x 8.5		Ferricrete / duricrust *
BCBK(U)	Bauxite quarry (northern hill) N 5.966986° E 100.420702° Ele = 52 m	7.5R 5/4 weak red 5R 5/2 weak red 2.5YR 5/4 reddish brown C horizon 11.4 x 9.7 x 3.2		Fragment of argilite *
BCBK2a	Water Tank II N 5.964197° E 100.418980° Ele = 56 m	7.5YR 6/8 reddish yellow 10YR 8/8 yellow C horizon 13.0 x 9.9 x 8.5		Fragment of argilite *
BCBK2b	Water Tank II N 5.964137° E 100.419037° Ele = 57 m	7.5YR 6/8 reddish yellow 10YR 8/8 yellow C horizon 13.0 x 9.9 x 8.5		Fragment of argilite *

*sample analysis XRD & XRF

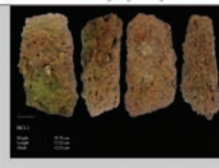
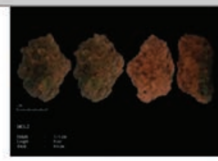
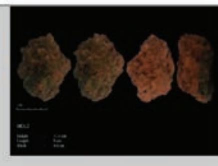
SAMPLE	LOCATION	COLOUR / WEATHERING PROFILE / (height x length x thickness) (cm)		LATERITE PROFILE
BCL1	Main structure N 5.964121° E 100.418753° Ele = 64 m	5Y 6/8 olive yellow 7.5YR 6/8 reddish yellow 10YR 7/8 yellow B horizon 29.7 x 17.2 x 12.2		Ferricrete / duricrust *
BCL2	Main structure N 5.964106° E 100.418795° Ele = 62 m	5YR 7/8 reddish yellow 2.5YR 4/8 red 2.5Y 4/1 dark reddish gray B horizon 11.4 x 9.0 x 4.6		Ferricrete / duricrust *
BCBawah	Western foothill N 5.966289° E 100.417879° Ele = 7 m	10YR 8/8 yellow 5YR 4/1 dark gray 7.5R 4/6 red 5YR 6/4 light reddish brown B horizon 11.4 x 9.0 x 4.6		Ferricrete / duricrust *

Table S2: Summary of sample taken by hand auger at Bukit Choras

SAMPLE	LOCATION	LAYER (cm)		COLOUR / WEATHERING ZONE PROFILE	TYPE / LATERITE PROFILE
BCP1A	South-eastern part of the main structure N 5.964014° E 100.418624° Ele = 61 m	Layer 1 (0-18.3)		7.5YR 4/2 brown 10R 6/4 pale red O Horizon Humid Layer	Humus and loose soil mix with laterite Soil (organic)
		Layer 2 (18.3-36.6)		7.5YR 4/4 brown 10R 5/6 red A Horizon Humid Layer	Loose soil, red clay and laterite Soil (ferricrete / duricrust)
		Layer 3 * (36.6-54.9)		7.5YR 5/3 brown 10R 5/6 red B Horizon Subsoil	Hard red clay and laterite Soil (ferricrete / duricrust)
		Layer 4 (54.9-73.2)		5YR 6/2 pinkish gray 10R 5/6 red 2.5YR 6/6 light red B Horizon Subsoil	Compacted red clay and laterite Mottled zone
BCP2	Northwest hill N 5.965025° E 100.418572° Ele = 41 m	Layer 1 (0-18.3)		7.5YR 4/4 brown 2.5YR 4/6 red O Horizon Topsoil, humid layer	Humus and loose sandy soil with red and brown clay Soil (organic)
		Layer 2 (18.3-36.6)		2.5YR 4/6 red 5YR 5/4 reddish brown 2.5YR 4/4 reddish brown A Horizon	Brown and red clay and laterite Soil (ferricrete / duricrust)
		Layer 3 * (36.6-54.9)		10R 4/3 weak red 5YR 6/6 reddish yellow 2.5YR 4/4 reddish yellow A Horizon	Laterite mixed with red and brown clay Soil (ferricrete / duricrust)

SAMPLE	LOCATION	LAYER (cm)		COLOUR / WEATHERING ZONE PROFILE	TYPE / LATERITE PROFILE
		Layer 4 (54.9- 73.2)		7.5YR 6/4 light brown 2.5YR 4/4 reddish brown 10R 4/3 weak red B Horizon Subsoil	Laterite with compacted clay Mottled zone
BCP2B	Water Tank II N 5.964159° E 100.419003° Ele = 56 m	Layer 1 (0-43)		7.5YR 5/4 brown 7.5YR 5/6 strong brown 7.5YR 6/6 reddish yellow O-A Horizon Topsoil Humid layer	Loose clay with red mudstone Soil (Top)
		Layer 2 (43-64)		7.5YR 6/6 reddish yellow 10R 5/4 weak red A Horizon Topsoil Humid layer	Loose clay with red mudstone Soil (Top)
		Layer 3 (64-99)		7.5YR 6/4 light brown 10YR 6/4 light yellowish brown 7.5YR 7/1 light gray 5R 5/8 red A Horizon Topsoil Humid layer	Loose clay with red mudstone Soil (ferricrete / duricrust)
		Layer 4 (99-126)		5YR 6/2 pinkish gray 2.5YR 5/8 red 5YR 5/8 yellowish red 5YR 5/6 yellowish red 5YR 6/1 gray 7.5R 5/4 weak red 2.5YR 5/4 reddish brown A-B Horizon Subsoil	Loose clay with red mudstone and laterite Soil (ferricrete / duricrust)

SAMPLE	LOCATION	LAYER (cm)	COLOUR / WEATHERING ZONE PROFILE	TYPE / LATERITE PROFILE
		Layer 5 * (126-129) 	10R 4/6 red 10YR 6/6 brownish yellow B Horizon Subsoil	Laterite soil, loose clay with red mudstone and groundwater level Soil (ferricrete / duricrust)

*sample analysis by XRD & XRF

SUPPLEMENTARY FIGURES

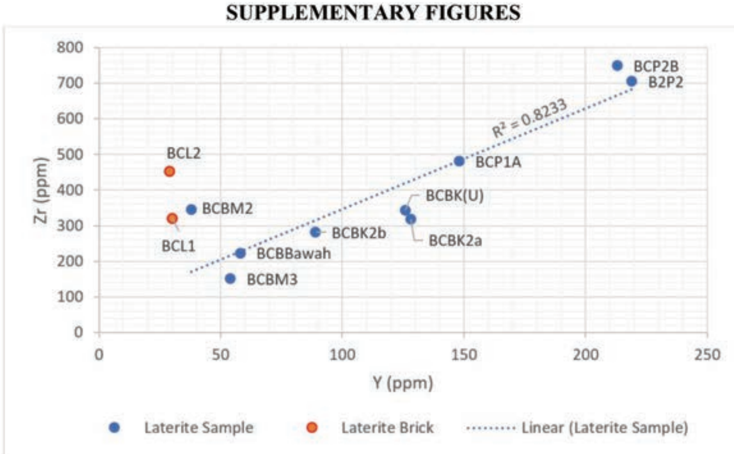


Figure S1: Two-dimensional scatter plot for concentration of immobile elements (Zr vs Y)

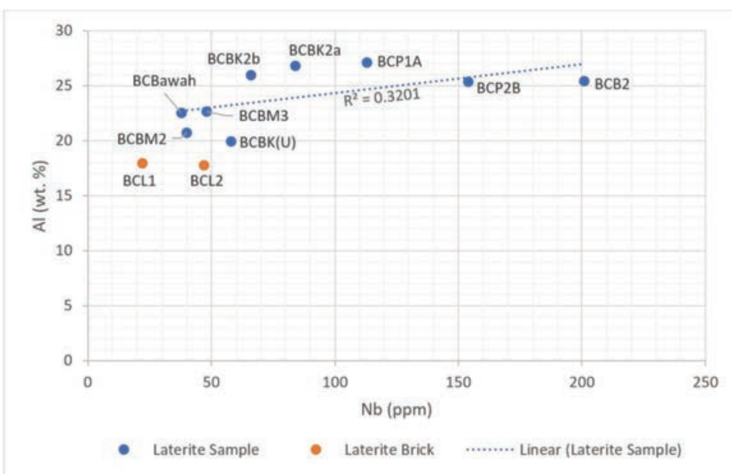


Figure S2: Two-dimensional scatter plot for concentration of immobile elements (Al vs Nb)

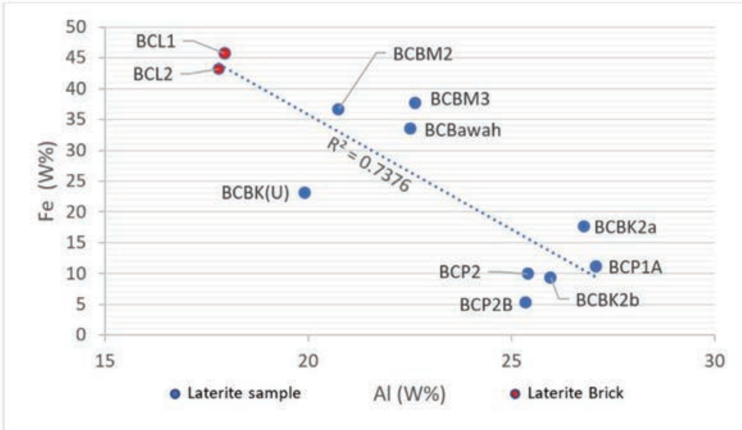


Figure S3: Two-dimensional scatter plot for concentration of immobile elements (Fe vs Al)

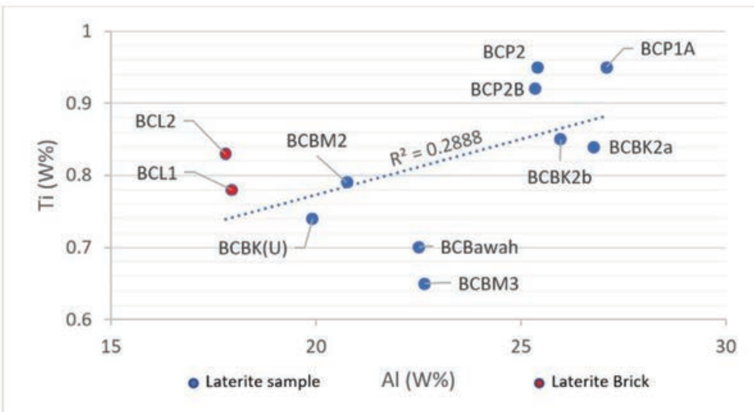


Figure S4: Two-dimensional scatter plot for concentration of immobile elements (Ti vs Al)