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A review that brick can act as geochemical indicator of soil. Reference: Elemental and trace elements characterization

Amit Kumar Mishra *

Archaeological Survey of India, GPO Complex, 'D' Block, Puratattva Bhawan, Science Branch, 6th Floor, INA, New Delhi-110023, India.

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Abstract

The elemental and trace element characterization of the fired clay bricks of monuments and soil of the surrounding area showed the high wt. % of SiO_2 (~ 71 % $_{Brick}$-78 % $_{Soil}$), Al_2O_3 (~ 14 % $_{Brick}$ - 10 % $_{Soil}$), and Fe_2O_3 (~ 4 % $_{Brick}$ - 6% $_{Soil}$) with low wt. % of Na_2O (~ 1%), K_2O (~ 1%), MgO (~ 2 % $_{Brick}$ - 1 % $_{Soil}$), CaO (~ 1%), etc. The raw materials of brick are siliceous and Ca-poor; the same nature is shown by the soils of surrounding area. In both brick and soil samples, the high percentage of non-metal and transition metals among different families of trace elements indicates geogenic similarity among their sources. The result indicates that brick can act as a geochemical indicator of the soil. The results also indicate that the local community was involved in the production of the bricks and the construction of the monumental structures.

Keywords: Brick; Soil; Elemental oxide; Trace element; Geochemical

1. Introduction

Brick is one of the oldest man-made fundamental masonry materials in the history of human civilizations. Generally, bricks are recommended for construction activities because of their ease of production, easy accessibility of raw materials, excellent engineering properties, fire resistance, durability, molding tendency, light weight, low maintenance cost, and serviceability. The earliest civilizations (ancient Egypt and Babylonia) also used bricks, clay blocks, as masonry materials (Elert et al., 2003). Clay is the essential ingredient of soils, and it is chiefly made up of silica, alumina, and water. It has low amounts of iron, alkalis, and alkali earth metals and has plasticity, which is required for molding clay bricks. The presence of mineral oxides (Fe_2O_3, MgO, and CaO) in clay determines its areas of application, such as in bricks, whereas the alkali metal oxides (Na_2O, K_2O, and CaO) determine their suitability for making ceramic products (Nnuka and Enejor, 2001). The properties of clay bricks vary at regional levels because of their dependency on the characteristics of the soils used and production conditions (Shrestha, 2017). Thus, bricks can serve as geochemical indicators for any region (Shrivastav et al., 1995). The firing processes, which depend on firing temperature, raw materials, and atmospheric conditions, produce a series of mineralogical, textural, and physical changes and influence the properties of bricks (Lopez-Arce et al., 2003). However, the firing process does not disturb the elemental oxide and trace elemental composition of the clay to a large extent; therefore, it can be used to identify the source (soil) of clay (Cogswell et al., 1996; Meloni et al., 2000). In most cases, when bricks deteriorated, restorers replaced them with similar bricks. Here, care must be taken to ensure compatibility between the raw material and firing technique of new bricks. Incompatible bricks can accelerate the deterioration of historic materials and cause irreversible damage to structures. The current study focused on the elemental and trace element characterization of the fired clay bricks and soils of the surrounding area of four Mughal era monuments (Figure 1, Table 1) with reference to their geochemical similarity for conservation of these monumental structures.

* Corresponding author: Amit Kumar Mishra

Table 1 Monumental structures and surrounding area of Haryana region of India**.**

Monumental Structures & Surrounding area	**Year of construction**	**Brick-Assigned code**	**Soil-Assigned code**
Tomb of Sheikh Chilli's, Thanesar (Kurukshetra)	CE 1650	BSCT	SSCT
Mughal Sarai gateway, Gharaunda, (Karnal)	CE 1637	BMSG	SMSG
Kabuli Bagh Mosque, Panipat	CE 1526	BKBM	SKBM
Khwaza Khizr's Tomb, Sonepat	CE 1522-24	BKKT	SKKT

Figure 1 Mughal period monumental structures of Haryana region of India

1.1. Study Area

The Haryana region is the northwestern part of India and is geographically situated between 27^0 39' to 30^0 35'N latitudes and 74^0 28' to 77^0 36'E longitudes (Figure 1). The total geographical area of the state is 44,212 km^2. The overall climate of Haryana is subtropical, semi-arid to sub-humid, continental, and monsoon type, with hot summers (30^0C - 48^0C), cold winters (5^0C - 25^0C) (IMD 2025). The Haryana region was the land for the effervescence of Mughal rule (Grover, 1981). During their period of regime (1526 CE-1761 CE) the Mughals built many huge monumental structures for their social, economic, political, and cultural developments and brought about changes in indigenous architecture and manufacturing technologies of masonry materials (Gajrani, 2004). The Haryana region's monumental structures are built of bricks, and it was a general practice at that time to produce bricks using soil from the surrounding areas of the monument (Chandra, 2003).

2. Materials and Methods

2.1. Brick and soil sampling

To perform the study, brick and soil samples were collected from each monumental structure (Figure 2). A total of 12 Nos. brick samples were collected from selected monuments, i.e., 03 Nos. from each monument, by dividing the samples' stack into 03 Nos. sections and drawing 01 No. sample from each section (BIS, 1978a).The soil samples were collected from each monumental structure's surrounding areas by digging to a depth of 50 cm below the natural ground surface in two segments, i.e., 0-25 cm (n=2) and a further 25 cm -50 cm (n=2) (BIS, 1978b). A total of 16 Nos. soil samples were collected from the surrounding area of selected monuments, i.e., 04 Nos. from each monument.

Figure 2 Brick and soil samples from monumental structures

2.2. Test Method

The analysis of soil and bricks using XRF techniques can provide basic information about geochemical similarity and production technologies adopted for bricks (Pérez-Monserrat et al., 2024). To estimate elemental oxides and trace elements, powder forms of brick and soil samples were prepared. The collected brick samples were cleaned of undesirable material and ground manually using agate mortar and passed through 300 mesh screens (53 micron-sized openings). The soil samples were dried in an oven at 105 °C, ground manually using an agate mortar, and passed through a 2 mm sieve. The obtained powders were also dried in an oven at 500°C -550°C for 21 h to remove the organic compounds. Brick and soil samples were analyzed with X-ray fluorescence (Model: Epsilon 5, PANalytical B.V.). The parameters used for the X-ray lamp were a current of 30 mA and voltage of 40 kV. The analysis was performed by mounting a compressed boric acid pellet of the samples (Singh et al., 2015). The pellets of the samples were prepared by homogeneously mixing 1.0 g of the samples with 6.0 g of boric acid powder at a pressure of 20 psi using hydraulic pressure equipment (Dadiana et al., 2017).

3. Results and Discussions

3.1. Element Oxide

Table 2 shows the mean values (dry wt. %) of elemental oxides and their Pearson correlation in brick samples. The results indicate that the bricks have a high wt. % of SiO_2 (~ 71 %), Al_2O_3 (~ 14 %), and Fe_2O_3 (~ 6.0%) with low wt. % of Na_2O (~ 1 %), K_2O (~ 3 %), MgO (~ 2 %), CaO (~ 11 %), TiO_2(~ <1 %), and MnO (~ <1 %). High wt. % of SiO_2 (~ 71%) and low wt. % of CaO (~ 0.85 %) in the bricks' composition indicate that Ca-poor clay with high silica content was used in the manufacture of bricks (Gulzar et al., 2013; Rai and Dhanapal, 2013). Table 3 shows the mean values (dry wt. %) of elemental oxides and their Pearson correlations in soils of the surrounding area. The results showed that the soils of the surrounding area have a high wt. % of SiO_2 (~ 78%), Al_2O_3 (~ 10 %), and Fe_2O_3 (~ 4 %), and a considerably low wt. % of Na_2O (~ 1 %), K_2O (~ 1 %), MgO (~ 1 %), MnO (~ <1 %), P_2O_5 (~ <1 %), and CaO (~ 1 %). High SiO_2 content indicates that the soils are siliceous in nature, whereas low CaO (~ 1%) indicates a Ca-poor soil in the surrounding area, which is in agreement with the elemental oxide composition of monumental bricks.

TiO_2 was found in much lower wt. % (~ 0.66 %), which is commonly used in refractory bricks to provide better performance under high-temperature conditions and greater resistance to cracking and spalling (Rai and Dhanapal, 2013). The main elemental oxides are Al_2O_3 and SiO_2. An increase in Al_2O_3 increases the refractoriness and mechanical strength, whereas SiO_2 decreases the shrinkage and refractoriness of the clay bricks (Katsuki et al., 2016). The presence of Na_2O, K_2O, MgO, CaO, TiO_2, and MnO are reliable fluxing components that decrease the viscosity of the liquid phase of the clay brick products (Njoya et al., 2017).

Table 2 Mean value (n=3) of elemental oxides (wt. %) and Pearson correlation in masonry bricks

Study sites	**Al_2O_3**	**SiO_2**	**Fe_2O_3**	**K_2O**	**MgO**	**CaO**	**Na_2O**	**TiO_2**	**MnO**
BSCT	15.56	67.31	6.38	3.36	2.11	1.05	1.37	0.67	0.15
BMSG	13.47	72.58	5.85	2.92	1.51	0.76	1.26	0.66	0.12
BKBM	13.46	72.72	5.73	2.92	1.49	0.79	1.21	0.65	0.13
BKKT	14.92	70.46	5.97	3.07	2.04	0.78	0.81	0.66	0.14
Pearson correlation ($p < 0.05$)									
Parameter(s)	Al_2O_3	SiO_2	Fe_2O_3	K_2O	MgO	CaO	Na_2O	TiO_2	MnO
Al_2O_3	1.00								
SiO_2	-0.96	1.00							
Fe_2O_3	0.91	-0.98	1.00						
K_2O	0.94	-1.00	0.98	1.00					
MgO	0.99	-0.90	0.84	0.87	1.00				
CaO	0.77	-0.91	0.92	0.94	0.65	1.00			
Na_2O	-0.10	-0.18	0.30	0.25	-0.26	0.54	1.00		
TiO_2	0.81	-0.87	0.94	0.87	0.76	0.77	0.27	1.00	
MnO	0.94	-0.92	0.84	0.91	0.91	0.81	-0.04	0.63	1.00

Table 3 Mean value (n=4) of elemental oxides (wt. %) and Pearson correlation in surrounding soils

Study sites	**Depth (Cm)**	**Na_2O**	**MgO**	**Al_2O_3**	**SiO_2**	**K_2O**	**CaO**	**P_2O_5**	**Fe_2O_3**	**MnO**
SSCT	0-25	0.86	1.14	10.12	70.23	1.34	1.08	0.10	3.99	0.17
	25-50	0.91	1.01	9.78	74.26	1.23	1.29	0.14	4.01	0.19
Mean		0.89	1.08	9.95	72.25	1.29	1.19	0.12	4.00	0.18
SMSG	0-25	1.01	1.11	10.23	77.89	1.19	1.13	0.15	4.67	0.18
	25-50	0.95	0.98	9.45	81.23	1.21	1.16	0.18	4.86	0.19
Mean		0.98	1.05	9.84	79.56	1.2	1.15	0.17	4.77	0.19
SKBM	0-25	1.05	1.34	10.67	78.12	1.23	1.18	0.17	4.80	0.17
	25-50	0.99	1.21	9.12	82.67	1.18	1.21	0.16	4.69	0.19
Mean		1.02	1.28	9.90	80.40	1.21	1.20	0.17	4.75	0.18
SKKT	0-25	0.59	0.95	10.87	79.89	1.31	1.38	0.09	3.89	0.13
	25-50	0.61	0.89	8.98	83.23	1.36	1.27	0.07	4.05	0.17
Mean		0.60	0.92	9.93	81.56	1.34	1.33	0.08	3.97	0.15

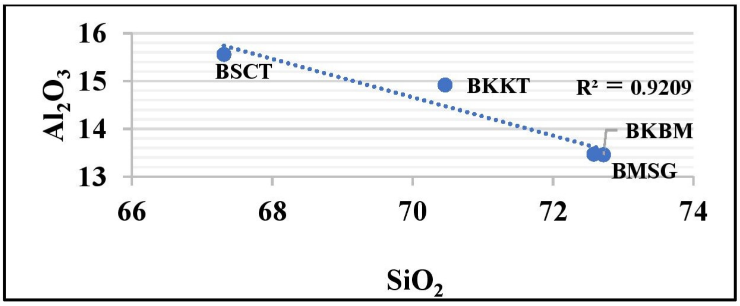
Pearson correlation ($p<0.05$)									
Parameter(s)	Na_2O	MgO	Al_2O_3	SiO_2	K_2O	CaO	P_2O_5	Fe_2O_3	MnO
Na_2O	1								
MgO	0.83	1							
Al_2O_3	-0.50	-0.14	1						
SiO_2	-0.23	-0.07	-0.47	1					
K_2O	-0.92	-0.70	0.78	-0.12	1				
CaO	-0.92	-0.56	0.54	0.42	0.84	1			
P_2O_5	0.95	0.76	-0.72	0.05	-1.00	-0.86	1		
Fe_2O_3	0.79	0.64	-0.85	0.40	-0.96	-0.65	0.94	1	
MnO	0.94	0.59	-0.56	-0.38	-0.86	-1.00	0.88	0.69	1

The fluxing oxides (especially Na_2O and K_2O) react with the silica and alumina of clay minerals to promote liquid phase formation, which facilitates the densification of brick bodies at high temperatures (Sokolar et al., 2017). Apart from the fluxing function of Fe_2O_3, it is also a major colorant agent along with others and the furnace atmosphere, and is responsible for providing red color to bricks after firing (Domínguez et al., 2016). The Pearson correlation indicates a significant correlation, indicating that the same raw materials are used for the production of bricks. SiO_2 exhibited a negative correlation with other elemental oxide. The negative correlation between SiO_2 and Al_2O_3 may be because both are from two different mineral phases, while negative correlations of SiO_2 with other oxides reveal the fractionation of aluminous and ferromagnesian phases during clay firing (Moreno-Tovar et al., 2017).Among other constituents that are commonly important is MgO, showed a positive correlation with CaO, TiO_2, and MnO, indicating similar geological sources of raw materials (Rao et al., 2011).The regression graphs (Figure 3) between elemental oxides (Al_2O_3 versus SiO_2), (Al_2O_3 versus Fe_2O_3), and (Al_2O_3 versus K_2O) showed coefficients of regression (R^2) values of 0.92, 0.82, and 0.87, respectively, indicating good correlation among elemental oxides and similarity with its primary source of raw materials (clay) (Arsenovic et al., 2014).

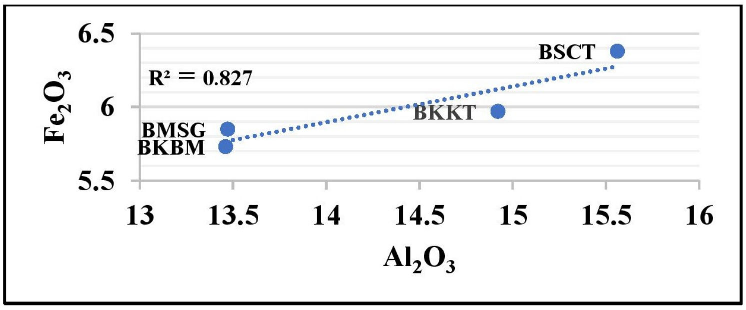
The siliceous nature of soils may be due to the higher amount of quartz present in the parent material (Khan et al., 1997; Walia and Rao, 1997). The CaO and MgO indicate the lithological discontinuity and minerals deficient in calcium and magnesium (Sireesha and Naidu, 2015). Low Na_2O and K_2O levels suggest sodium and potassium weathering of feldspar minerals, whereas K_2O indicates the occurrence of K-bearing clay minerals such as mica and feldspars (Raina et al., 2006). The presence of CaO, Fe_2O_3, MgO, Na_2O, and K_2O in all soil types is acceptable for the production of fired-clay bricks because these oxides act as fluxes, which decrease the temperature needed to produce glassy material during brick manufacturing. The values of oxides also indicate that the geochemical weathering mechanisms were not strong enough to cause significant changes in the distribution pattern. The Pearson correlation between elemental oxides of soil indicates geogenic similarity in soil composition. Figure 4, shows a comparative analysis between SiO_2 and $Al_2O_3$of the monumental brick samples and surrounding soils, the overlap between the contents (SiO_2 and Al_2O_3) signifies that the raw material used to make bricks was picked from the surrounding localities (Ali and Ramli, 2022). This finding also suggests that local communities used raw materials from their common geological source to make bricks for monumental structures.

3.2. Trace element

The compositions of trace elements are recognized as unique features of geographical locations and raw materials sources (Cardiano et al., 2004). Table 4 shows the mean value of trace elements in the brick samples and their Pearson correlation. The trace elements in the brick samples were the non-metal elements (As, Cl), alkali metal (Rb), alkaline-earth metals (Ba, Sr), transition metals (V, Cu, Ni, Zn, Y, Nb, W, and Zr), and metals (Ga, Pb). The results showed that the content was >100 ppm for Ba, Rb, Zr, W, and Cl (except for BSCT, which may be due to some contamination) in all the brick samples and was almost consistent. The other trace elements (V, Ni, Cu, Zn, Ga, As, Sr, Y, Nb, and Pb) were < 100 ppm and consistent with their abundance order. Table 5 shows the mean value of trace elements in the soil samples and their Pearson correlation. The trace elements in the soil samples show the presence of non-metals such (As, Se), transition metals (Cu, Ni, Zn, Cd, Co, and Cr), lanthanides (Hg), and metals (Pb). The results showed that the concentrations of Ni, Cu, Zn, As, Pb, Cd, Co, Cr, and Hg, are quite low (< 50 ppm), except in Se (> 100 ppm).



(a)



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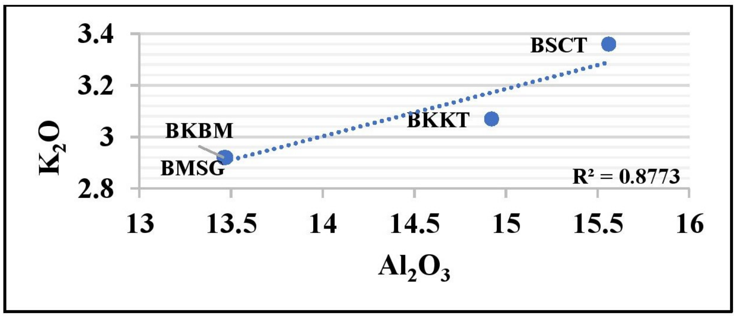

(c)

Figure 3 Regression graphs (a) Al_2O_3 and SiO_2 (b) Al_2O_3 and Fe_2O_3 and(c) Al_2O_3 and K_2O

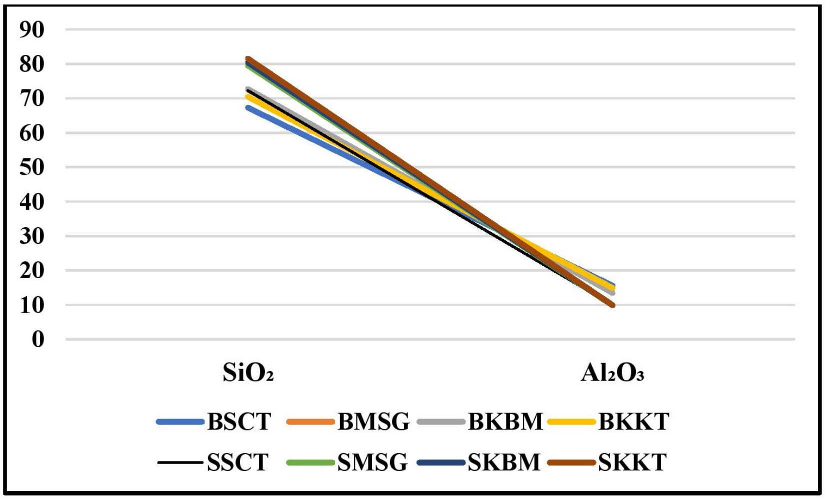

Figure 4 Dry weight % of SiO_2 and Al_2O_3 in brick samples and soil samples

Table 4 Mean value (n=3) of trace elements (ppm) and Pearson correlation in masonry bricks

Study sites	**Cu**	**Zn**	**Ni**	**Pb**	**V**	**Ga**	**As**	**Rb**	**Sr**	**Zr**	**Y**	**Nb**	**Ba**	**W**	**Cl**
BSCT	39.81	90.24	63.30	39.86	15.98	18.59	5.00	162.20	105.59	240.11	28.51	16.41	467.02	141.12	0.41
BMSG	41.92	96.28	53.76	62.46	60.37	17.80	0.99	170.67	90.54	239.05	32.66	16.44	490.03	149.18	288.75
BKBM	37.56	86.93	48.26	48.00	11.44	15.83	5.82	158.67	94.82	283.87	29.61	17.24	478.52	145.20	1493.92
BKKT	42.70	93.61	59.13	36.61	8.33	16.35	4.09	152.67	87.16	246.32	31.17	15.80	523.89	135.53	1588.00
Pearson correlation ($p < 0.05$)															
Parameter (s)	Cu	Zn	Ni	Pb	V	Ga	As	Rb	Sr	Zr	Y	Nb	Ba	W	Cl
Cu	1.00														
Zn	0.92	1.00													
Ni	0.47	0.28	1.00												
Pb	0.01	0.40	-0.56	1.00											
V	0.35	0.70	-0.19	0.91	1.00										
Ga	0.23	0.36	0.67	0.16	0.44	1.00									
As	-0.69	-0.92	0.01	-0.72	-0.92	-0.37	1.00								
Rb	0.03	0.41	-0.14	0.87	0.91	0.62	-0.69	1.00							
Sr	-0.55	-0.51	0.41	-0.23	-0.21	0.61	0.45	0.20	1.00						
Zr	-0.79	-0.79	-0.77	-0.03	-0.44	-0.77	0.63	-0.35	-0.06	1.00					
Y	0.68	0.83	-0.29	0.63	0.72	-0.12	-0.87	0.37	-0.82	-0.32	1.00				
Nb	-0.91	-0.70	-0.73	0.38	0.00	-0.26	0.37	0.26	0.32	0.78	-0.34	1.00			
Ba	0.73	0.51	0.06	-0.23	-0.11	-0.48	-0.26	-0.51	-0.86	-0.18	0.59	-0.69	1.00		
W	-0.32	0.09	-0.59	0.93	0.77	0.20	-0.46	0.87	0.07	0.15	0.32	0.64	-0.55	1.00	
Cl	-0.09	-0.30	-0.48	-0.34	-0.56	-0.97	0.40	-0.77	-0.63	0.65	0.09	0.05	0.62	-0.42	1.00

Table 5 Mean value (n=4) of trace elements (ppm) and Pearson correlation in surrounding soils

Study sites	Depth (Cm)	Ni	Cu	Zn	Se	As	Pb	Cd	Co	Cr	Hg
SSCT	0-25	31.34	36.98	43.84	301.11	0.10	41.87	0.08	0.01	0.10	0.09
	25-50	39.88	34.79	39.08	288.32	0.08	39.09	0.08	0.01	0.07	0.17
Mean		35.61	35.89	41.46	294.72	0.09	40.48	0.08	0.01	0.09	0.13
SMSG	0-25	41.75	41.45	43.13	279.65	0.09	49.23	0.06	0.06	0.09	0.10
	25-50	40.21	39.92	38.99	293.32	0.10	42.24	0.07	0.07	0.06	0.19
Mean		40.98	40.69	41.06	286.49	0.09	45.74	0.06	0.06	0.07	0.15
SKBM	0-25	43.99	39.97	42.88	281.63	0.10	41.24	0.08	0.08	0.08	0.17
	25-50	41.47	34.30	43.26	297.32	1.01	39.19	0.08	0.08	0.08	0.14
Mean		42.73	37.13	43.07	289.48	0.55	40.22	0.08	0.08	0.08	0.16
SKKT	0-25	28.99	31.47	31.74	269.65	0.08	43.08	0.09	0.05	0.04	0.19
	25-50	29.88	31.23	33.22	287.62	0.09	39.24	0.08	0.04	0.04	0.08
Mean		29.43	31.35	32.48	278.64	0.08	41.16	0.09	0.04	0.04	0.13
Pearson correlation ($p < 0.05$)											
Parameter (s)		Ni	Cu	Zn	Se	As	Pb	Cd	Co	Cr	Hg
Ni		1.00									
Cu		0.87	1.00								
Zn		0.90	0.79	1.00							
Se		0.53	0.49	0.85	1.00						
As		0.63	0.17	0.51	0.22	1.00					
Pb		0.28	0.66	0.06	-0.21	-0.42	1.00				
Cd		-0.61	-0.90	-0.46	-0.16	0.18	-0.92	1.00			
Co		0.61	0.37	0.22	-0.31	0.70	0.25	-0.29	1.00		
Cr		0.78	0.71	0.98	0.95	0.39	-0.03	-0.37	0.00	1.00	
Hg		0.84	0.55	0.56	0.07	0.84	0.14	-0.34	0.93	0.37	1.00

The Pearson correlation between the mean values of trace elements in brick samples indicates significant correlation and reveals geogenic similarity in the composition of raw materials, while soil samples indicated similarities in the compositional nature of the parent materials. The behavior of trace elements of brick samples is similar to that of major elemental oxides, and they are thought to reflect their origin composition because of their immobile behavior. Among trace elements, Pb, Zn, and Cu are indicators of changes in primary mineral phases due to hydrolysis and alteration during brick and mortar firing (Moreno-Tovar et al., 2017). Differences in the concentration of trace metals in soils may be due to the inherited properties of the parent materials from which the soil was derived or may be due to anthropogenic factors. Weathering and pedogenic processes (clay migration, gley formation, and podzolization) influence the consistency of phases produced during soil growth, as well as the distribution and action of trace elements. Figure 5 shows the distribution of the copper element against lead for the brick and soil samples. The results indicate that there is one major source of raw material. The use of surrounding soils (raw materials) also revealed that the local community was involved in the production of the bricks and the construction of these monumental structures.

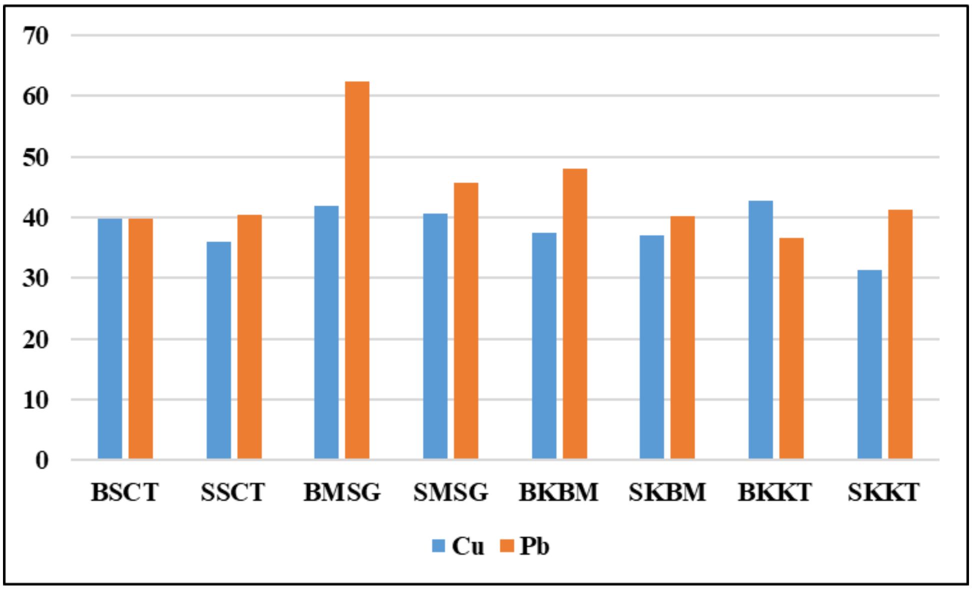

Figure 5 Cu and Pb (in ppm) in brick samples and soil samples

Highlights

- Brick can serve as geochemical indicator for any region.
- Geogenic similarity among sources of raw materials used for manufacturing bricks and surrounding soil
- The local community was involved in the production of the bricks and the construction of the monumental structures.

4. Conclusions

The elemental oxides and trace elements compositions of monumental bricks and the soil of the surrounding areas show geogenic similarity among their origins. The characterisation also indicates the same geochemical nature of raw materials used to produce bricks, and they were from the surrounding area of the monumental structures. The results reveal that the local community was involved in the production of the bricks and the construction of the monumental structures. The study also emphasizes the need for more research to fully understand the other geochemical indicator.

Compliance with ethical standards

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Disclosure of conflict of interest

No conflict of interest to be disclosed

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