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X-RAY DIFFRACTION AND X-RAY FLUORESCENCE ANALYSIS OF POTTERY SHARDS FROM NEW ARCHAEOLOGICAL SURVEY IN SOUTH REGION OF SISTAN, IRAN

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ABSTRACT

The aim of this study is to determine whether pottery shards from new archaeological survey in south region of Sistan are locally made or imported. Many artefacts especially pottery shards have been found during the archaeological survey. These pottery shards are variable in color; from buff, grey, black, and red. The analytical techniques involved X-Ray Fluorescence (XRF) and X-Ray Diffraction (XRD), that were applied to determine the major and trace elements and also the mineral content of the pottery shards. The results show that most of the pottery shards taken from archaeological survey in Sistan are locally made. Two of the samples from Islamic Period however shows a different content of chemical composition compared to other shards suggesting a different that Sistan origin. A prehistoric shard also shows a different in chemical composition and not originated from Sistan area. The analysis of pottery shards indicate that since prehistoric period, there has been a local production and trade activity in Sistan that continues until the Islamic Period. Result of the analysis also shows that local community at Sistan since prehistoric period are very skilful and keep a tradition in pottery making until Islamic Period.

KEYWORDS: *Sistan, X-Ray Fluorescence (XRF) and X-Ray Diffraction (XRD), pottery, Shahr-I Sokhta, Iran*

1. INTRODUCTION

Sistan is located in the east of Iran and in the north of the vast province of Sistan and Baluchestan (see Fig. 1) with the area of 8.117 square kilometers (Lashkari et al, 2012). Sistan with its rich culture has many unspoken words at the bottom of its fluid geomancy and heavy alluviums (Moradi et al, 2014). Continuous attempts by archaeologists and comprehensive studies by researchers have shown a new face of the history of this region, which today is only a desert. Sistan since the Bronze Age until now has had an effective and salient role in the creation of human culture and civilization in Iran (Sarhaddi Dadian et al., 2012: 312; Moradi et al 2013b; Moradi et al 2014; Sarhaddi-Dadian et al 2015,). *Shahr-I Sokhta* (Sajjadi, 2008: 307; Tosi, 1983a: 73-125) *Dahaneh Ghulaman* (Scerrato, 1977: 709-735; Latere and Genito, 2010: 77), and *Kuhe Khawaja* (Bater, 2010; Ghanimati, 2000: 137-150), are not counted as the only historical places of Sistan that have been identified. The first settlement in this area was dated to the late fourth millennium BC i.e the Bronze Age (Sajjadi and Casanova, 2006: 347-357).

It coincides with the beginning of urbanization in the Middle East (Alden, 1982: 613-640). This period lasted until the early second millennium BC, many sites have been identified in the southern Sistan that the *Share Sokhta* is one of the hundreds site from this period. (Vidale and Tosi 1996: 251-269; Biscione 1987: 394). Each of the site have abundant of pottery shards, which are different from those of other periods in terms of type, shape, decoration, and technical characteristics. The main features of potteries are using the decoration of the geometrical, plant, and animal pattern in inner and outer of the containers, which have been decorated by the black or ochre colours on the buff or light grey slips (Tosi 1983: 136-139, Salvatori and Vidale, 1997). From the Bronze Age until the Achaemenian Empire (550 BC), there was no any evidences of earthenware, and other cultural materials in Sistan. Therefore, we have a big gap from 1800 BC to 550 BC in Sistan Region.

The second period of the potteries in Sistan belonged to the Iranian historical periods (550 BC-550 AD) that the dynasties of Achaemenid, Parthian, and Sassanid had under control the large parts of the Middle East (Chavalas, 1999: 88).

One of the important features of potteries is without any decoration in internal and external surface of them. The decoration of the potteries are simple such as the excised decoration at outer, Moreover, the others major characteristics are such as dense slip, smooth and burnished, and inside surface colours were in red (Mehrafarin and Musavi, 2011: 240-58).

In Sistan from the late Sassanid period (sixth century AD) to 13th century AD, the survey shows that there is no earthenware was found. After this period many new sites were detected in Sistan, which have many potsherds. The potteries of this period are different from the previous two periods in terms of shape and decoration, which coincided with an Islamic period middle in Sistan. The Islamic potteries have been decorated with the various colours. The most important glaze colours are including green, gray bluish, milky and black that have been decorated with the specific decorative such as the excised motifs under glaze or glaze with black colour (black pen), and also the geometric and plants decorative under light glaze. The dominate form is the open mouth bowl with the flat base. The dough's colour for these includes light buff in the Islamic Period (Mousavi & Atai, 2010). Four parts were selected for field works in southern Sistan Basin that include Shileh River, Gerdi Castle, South of Rostam Castle, and south of Hamon Lake zones. They have been surveyed systematically. The areas of those parts were 2223 square kilometres. During the survey 95 sites have been detected including 20 sites from Shileh River, 3 sites from the South of Hamon lake, 47 sites from the Gerdi Castle district, and 25 sites from south of Rostam Castle. About 626 pottery shards have been selected from the surface of the sites.

Most of the pottery shards found from survey in several sites from Sistan belong to historical period. Three of the samples belong to prehistory period and two of the samples belong to Islamic period. Pottery shards from prehistoric and historic period were buff, grey, red and black in colour. Previous study showed that grey and red wares, unlike the buff wares of *Shahr-I Sokhta*, are prevalent in south-eastern Iran, especially in Baluchistan and in the Indo-Iranian borderland, even though some of them have been found in the graves of *Shahr i Sokhta* (Moradi et al. 2013a). Scientific analysis on pottery from *Shahr-I-Sokhta* showed that some of the buff and red shards are imported item and the high content of lead indicated that the shards are originated from Indus Valley (Hosseini et al. 2013). Two of the Islamic potteries were decorated by flora motif and the surface was covered by glaze. Sample ZR332/1 decorated with flora motif which in blue colour while sample ZR369/8 decorated with flora motifs which in brown, blue and red colour. The origin of these two shards is unknown and the possibility of these two shards are not local origin is high and the shard is dated to the Safavid period (Lane, 1947; Lane, 1948, Pl. 32A).

Compositional analysis is one of the best method used to determine the chemistry of the ancient arte-

facts such as pottery, bricks and glass beads (Bieber et al. 1976; Broekmans et al. 2008; Marghussian et al. 2009; Wong et al. 2010; Ramli et al. 2011a, 2011b, 2012; Zuliskandar et al. 2011a, 2011b, 2013a, 2013b). Comparing the major and trace elements of the pottery shard, we can determine whether the potteries came from the same sources or raw materials or were imported. Therefore the objective of this study is to determine whether all the pottery shards collected in different sites of Sistan are locally made or not. For archaeologists, the data of the origins of the pottery is very important for them because from the data, they can interpret the culture and trade activity of the community that they are studying.

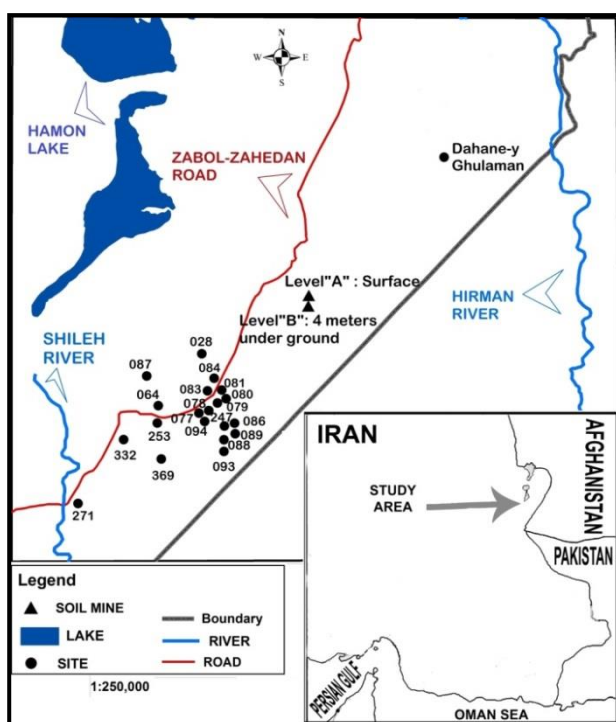


Figure 1. Location of the sites where pottery shards have been taken (Map drawn by Maral Maleki)

2. MATERIALS AND METHOD

In total 22 pottery shards have been chosen from different periods of Sistan region for chemical analysis, and also one soil sample of Sistan has been selected as the raw material for the Laboratory methods from two levels; surface level and 4 meters underground, to compare them with ancient potsherds of Sistan Region for provenance study. An amount of 30g of soil has been taken as a sample for comparison which located 3 kilometers south of the Shahre Sokhta Site, that today Sistani people also take advantage of that soil in making pottery and brick. Shahre Sokhta Site is also situated in 56 kilometers south of the modern city of Zabol. As we mentioned earlier the potshards are divided in to three categories such as prehistoric period, historical period and

Islamic period. We selected 3 prehistoric pottery shards, which are 028/1, 087/6, and 253/1. The pottery was wheel made and in the majority of the cases, the color of the paste is buff. Buff ware is indeed the prevalent pottery at Sistan Region and it ranges from an absolute buff to green. From the statistical point of view gray ware is the second in diffusion and red ware is the third. We also gathered two samples of gray, one red pottery, and one buff pottery shards to include all classes of pottery produced in prehistoric potteries, which are comparable to the Shahre Sokhta site, form the shape and the motifs, dated to the third millennium BC in Sistan region (Tosi 1983b: 132). There is a cultural gap after prehistoric period in Sistan Region.

The historical period was started from Dahane y Ghulaman Site. Unfortunately, there were no evidences of Iron Age in Sistan Region after the archaeological survey in the whole plain of the Sistan. Still the Iron Age is unknown in this region for us; probably the evidence of this period has been covered by the flood sediments during the thousand years, which has been carried out by Hrimand River into the Sistan region. The variety of potteries shows that they have been important and useful products during the history of the Sistan. From the historical sites have been chosen 17 pottery shards for the chemical studies, because most of the historical sites, which have been discovered in this research belonging to the historical periods. The potshards are 061/4, 077/2, 078/8, 079/5, 080/4, 081/2, 083/4, 084/3, 086/3, 247/4, 088/3, 089/2, 093/2, 094/1, 253/4, 271/5 and Dahane-y Gulaman 2001/26/2. A large group of potteries, which have been detected from the districts of Shileh River, Rostam Castle, South of Hamon Lake, and Gerdi were simple and they belonged to the historical periods, especially, the Achaemenian Empire (Sarhaddi Dadian, 2013). In total we were able to figure out the settlements of Achaemenid period in Sistan Region, thus potteries do not have any decorative motifs. The colours of pottery shards are buff, light red (brick colour), and red colours. The tempers were consisted of minerals and a little vegetation. It is very hard to figure out the forms of those potteries from the potshards. We can mention several shapes such as bowls and vats with the open mouth, flat rim, and the bowl with inverted and averted rims. We can also mention the cups with smooth edge and flat body. In total, the decorative motifs of those pottery includes excised decoration, added motifs, from excised decoration, we can point out the grooved designs. It seems that the creation of grooves horizontal lines started from the Achaemenian Period and the later period has been developed. The human stylized motif is the design that has been found on the potteries of Dahan-

ne-y Ghulaman Site, and usually these motifs have been decorated on the outer surface of the cups, which have the same size on mouth and body (Genito, 1990: 588-601).

The pottery of Parthian Period has considerable differences from the Achaemenian Period in terms of form and style. Most of potteries are simple, and usually they have been covered by different slips such as buff, light and dark red. The pottery with the excised decoration is still one of the common motifs in the Parthian Period in Sistan Region (Haerincq, 1980: 43-54; Mehrafarin and Musavi, 2011: 240-258)

According to archaeological survey the settlements of Sassanid in south of Sistan were not many, and there are not enough evidence about this period in those parts of Sistan unlike other parts. The pottery features of the Sassanid Period continue from the Parthian Period. In this period also the ceramics without glaze, and the dominant colour is red as previous stage. Most of the potteries are quite simple that are covered with thick layer and in some cases their surfaces are covered with thick mud slurry. The dominant colour of the pottery are the Sistani grooved pottery style common in this period. The new sample of the pottery that we can mention is mould motifs (stamp). The decorative motifs are often geometric and have plants designs. (Mehrafarin and musavi, 2010: 256-272)

The evidences of Islamic Period have been detected in the sites Nos. 332 sample No. 3; and site No.369, sample No. 8. The potteries of this period is very different from other periods in Sistan Region. Those potteries are as most other parts of Iran have glaze, moreover, they divided painted pottery with glaze and non-glaze, simple pottery without slip, simple pottery with excised decoration. (Mousavi and Ataie, 2010: 302-321). The tempers of potteries are mineral. Their designs include zigzag lines, small circles. The painted pottery with glaze is a type of potshards that we have been found in the sites 332/3 and 369/8. The plants motifs in red, brown and blue on milky background under clear glaze, which are dated to the Safavid period (Lane, 1947; Lane, 1948, Pl. 32A). We can mentioned special type of pottery such as painting under the blue glaze, which include geometric and plants motifs. They are comparable with the potteries of 6-13 A.D in south of Sistan. (Golombek, 2003: 253-270; Scanlon, 1984).

3. SAMPLE PREPARATION

For the analysis, in order to determine the chemical composition of the potteries, each sample weighing 0.4g was refined and heated up for one hour at a temperature of 105°C and mixed until homogenous with the flux powder of a type of Spectroflux 110

(product of Johnson & Mathey). These mixtures were baked for one hour in a furnace with a temperature of 1100°C. The homogenous molten was moulded in a container and cooled gradually into pieces of fused glass with a thickness of 2mm and a diameter of 32mm. The samples were of 1:10 dilution. Press pallet samples were prepared by mixing 1.0g of samples together with 6.0g of boric acid powder and then pressure of 20 psi was applied by using hydraulic pressure equipment. The samples of fused pallets and pressed pallets were then analysed using a Philips PW 1480 equipment for analysis of major and trace elements.



Figure 1. Pottery shards from several archaeological sites in Sistan.

Scatter plot diagrams of SiO₂ versus CaO, and strontium versus rubidium were then performed to demonstrate the differences among the groups and were analysed using Microsoft Excel software. The main purpose is to see the distribution of the samples in the group and subsequently to compare with the clay elements. Hierarchical cluster analysis (HCA) was applied to the chemical data from the four components, namely silica (SiO₂) and calcium (CaO), strontium and rubidium, of all 22 pottery shards samples in order to verify the presence of compositional groups of brick fragments differentiated by their probable major element sources. The measurement of distance used in the assignment rule was based on Ward's Linkage and Squared Euclidean Distance algorithm. The results are presented in the form of a dendrogram (Fig. 4 & 6) showing in the graphical form the distance between the pottery

samples on the basis of their SiO₂ and CaO percentage and strontium and rubidium concentration. The applicability of the analytical methods for the multi elemental analysis by XRF of the pottery shards is evaluated using the analysis of certified reference material, 315 Fire Brick (Calibration: G_FBVac28 mm) for major elements and certified reference material, SY-2 (Calibration: Trace Element P_20) for trace elements. The CRM was also used as the quality control material of the analytical procedure.



Figure 2. Pottery shards from several archaeological sites in Sistan.

4. RESULTS AND DISCUSSION

4.1 XRD and XRF

X-Ray Diffraction analysis (XRD) on 22 pottery shards and two clay samples is conducted to determine the mineral content in the shards and also clay samples (see Table 1). Analysis on shards from prehistoric period (ZR028/1, ZR087/6, ZR253/1) shows that the shards contain mineral such as quartz, albite, dickite, haematite, anorthite and diopside. Two shards (ZR332/3, ZR369/8) which are from Islamic Period that had motif and colour glaze on the surface contain mineral such as quartz, gypsum and calcium sodium aluminium silicate. Sample ZR332/3 only has quartz as a remaining mineral and it shows that the sample fired at very high temperature exceeding 1000°C. The rest of the pottery shards are from historical period contain mineral such as quartz, diopside, anorthite, albite, andesine, labradorite and minor mineral such as palladium, cuprite, giniite and gypsum. Quartz, calcite, clinocllore, muscovite, dolomite, chamosite and albite are mineral present in clay samples taken from ancient oasis

in Sistan. Diopside mineral only present in the pottery shards but not present in the clay samples. Two of the shards (ZR087/1, ZR253/1) from prehistoric period had diopside while most of the shards from history period contain diopside mineral.

Diopside is found in ultramafic (kimberlite and peridotite) igneous rocks, and diopside-rich augite is common in mafic rock, such as olvine basalt and andesite. Diopside is also found in a variety of metamorphic rocks, such as in contact metamorphic skarns developed from high silica dolomites. We suggest that diopside mineral in the shards probably came from dolomite present in clay samples.

Table 2 shows the content of major element of the pottery shards taken from several potential archaeological sites in Sistan. The pottery samples show quite homogeneous composition except for two samples from Islamic Period. The range of the silica dry weight percentage for shards categories as prehistoric and historical period is from ~43 to 56%. Content of aluminium is from ~11 to ~17%. Content of calcium and iron are from 4 to 14% and 5 to 8%, respectively. Alkaline element such as magnesium, sodium and potassium shows content of dry weight percentage from 4.30 to 7, 1 to 3 and 1 to 3%, respectively. Two samples from Islamic Period shows a different data which the range of silica dry weight percentage is much higher (77-79%), very low calcium (2.23-2.48%), a lower content of magnesium (0.03-1.185) and potassium (1.32-1.46) and much higher sodium (4.85-5.47%) compare to other shards from prehistoric and historical period. Sample ZR369/8 also has a very low content of iron (0.79%) compare to other samples. The percentage of P₂O₅ which is average in every shard indicates that none of the shards have been used as a container for some organic materials. The high percentage of CaO in the shards shows that potters in Sistan used calcareous clays as their main resources.

The content of trace elements is shown in Table 3. The result clearly shows that two shards from Islamic period have a very different chemical content. The shards have very high content of lead (1500ppm, 3700ppm), cobalt (100ppm, 300ppm) and barium (3000ppm, 4500ppm) and lower rubidium (15ppm, 24ppm). Analysis on Shahr-I-Sokhta pottery shards also showed that one of the pottery shards has a high content of lead (Hosseini 2013). Lead is usually added as a colouring agent into the pottery by ancient potters and archaeological research confirm that the ancient community in Indus Valley always used lead as a colorant (Caleb 1991). No lead was detected in prehistory and historical shards and therefore our suggestion that both shards from Islamic Period are not originated from this region is strongly supported.

4.2 Scatter Plot and Cluster Analysis

Figure 1 show a scatter plot of SiO₂ and CaO and from the figure it's clearly shows that two shards from Islamic Period namely ZR332/3 and ZR369/8 have different chemical composition compare to other shards. These two samples show much higher of silica and sodium and lesser of aluminium, calcium, iron, and potassium content and therefore, we suggest that these two samples came from other region probably from outside of Iran.

Figure 2 show a scatter plot of strontium and rubidium and from the figure shows that most of the pottery shards have the same trace elements concentration. Most of the shards also have a similar trace element concentration with raw resources taken from Sistan region not far from the surveyed sites. Two shards from Islamic Period namely ZR332/3 and ZR369/8 also have different trace elements concentration compare to other shards.

Hierarchical agglomerative clustering of SiO₂ and CaO percentage shows that there are three component of groups which are group A that is considered as local production of pottery shards and has significant value below 5; group B is also considered as a local production pottery shards which has significant value below 10 and group C is considered as a imported pottery shards (see Fig. 3).

Fig. 4 shows scatter plot of strontium versus rubidium; it shows that most of the prehistoric and historical pottery shards and also the clay samples are in one group. The Islamic Period shards are belonging to other group which is non local pottery product. Some of the shards (ZR078/8, 081/2,/061/4) contain much higher strontium compare to other shards. Hierarchical agglomerative clustering of strontium and rubidium percentage

shows that there are three component of groups which are group A that is considered as local production of pottery shards and has significant value below 5; group B is also considered as a mixture of local production and imported pottery shards which has significant value below 10 and group C is considered as a pottery shards which have a much higher content of strontium (see Figure 4).

5. CONCLUSION

The compositional analysis showed that most of the pottery shards taken from archaeological survey in Sistan, Iran are locally made. Two of the samples from Islamic Period which are ZR332/3 and ZR369/8 shows a different content of chemical composition compared to other shards. We strongly suggest that these two samples are not originated from Sistan. A shard from prehistoric period, the ZR028/1, also shows a different composition. Sample ZR028/1 has a lower content of calcium and a higher content of potassium compare to other shards from prehistoric and historical period. The mineral content also differ with other shards. We suggest that sample ZR028/1 is not originated from Sistan. Usually potters took their raw material not far from their kiln; 6 to 10 kilometres from their kiln location. The analysis of pottery shards from Sistan shows that since prehistoric period, there has been a trade activity in Sistan and then the activity continues until the Islamic Period. Result of the analysis also shows that local community from prehistoric period to historical period in Sistan was very skilful and knowledgeable in making the pottery.

Table 1. Content of mineral in pottery shards and clay samples

Sample	Mineral Content
ZR332/3	Quartz, syn SiO ₂ Calcium Sodium Aluminum Silicate Ca _{0.8} Na _{0.2} Al _{1.8} Si _{2.2} O ₈ Gypsum CaSO ₄ .2H ₂ O
ZR028/1	Quartz, syn SiO ₂ Albite, calcian, ordered (Na,Ca)Al(Si,Al) ₃ O ₈ Dickite Al ₂ Si ₂ O ₅ (OH) ₄ (HCONH ₂) Haematite Fe ₂ O ₃
ZR087/6	Quartz, syn SiO ₂ Diopside Ca(Mg, Al)(Si, Al) ₂ O ₆ Anorthite, sodian, ordered (Ca,Na)(AlSi) ₂ Si ₂ O ₈ Albite, disordered Na(Si ₃ Al)O ₈
ZR077/2	Quartz, syn SiO ₂ Diopside Ca(Mg, Al)(Si, Al) ₂ O ₆ Anorthite, sodian, ordered (Ca,Na)(AlSi) ₂ Si ₂ O ₈ Albite, disordered Na(Si ₃ Al)O ₈
ZR078/8	Quartz, syn SiO ₂ Diopside Ca(Mg, Al)(Si, Al) ₂ O ₆
ZR079/5	Anorthite CaAl ₂ Si ₂ O ₈ Andesine Na _{0.622} Ca _{0.368} Al _{1.29} Si _{2.71} O ₈
ZR080/4	Quartz, syn SiO ₂ Quartz, syn SiO ₂

ZR081/2	Diopside $\text{Ca}(\text{Mg}, \text{Al})(\text{Si}, \text{Al})_2\text{O}_6$ Quartz, syn SiO_2 Diopside $\text{Ca}(\text{Mg}, \text{Al})(\text{Si}, \text{Al})_2\text{O}_6$
ZR083/4	Albite, calcian, ordered $(\text{Na}, \text{Ca})\text{Al}(\text{Si}, \text{Al})_3\text{O}_8$ Quartz, syn SiO_2 Anorthite, ordered $\text{CaAl}_2\text{Si}_2\text{O}_8$ Labradorite $\text{Ca}_{0.65}\text{Na}_{0.35}(\text{Al}_{1.65}\text{Si}_{2.35}\text{O}_8)$ Diopside $\text{Ca}(\text{Mg}, \text{Al})(\text{Si}, \text{Al})_2\text{O}_6$
ZR084/3	Quartz, syn SiO_2 Diopside $\text{Ca}(\text{Mg}, \text{Al})(\text{Si}, \text{Al})_2\text{O}_6$
ZR086/3	Quartz, syn SiO_2 Albite, calcian, ordered $(\text{Na}, \text{Ca})\text{Al}(\text{Si}, \text{Al})_3\text{O}_8$ Anorthite $\text{CaAl}_2\text{Si}_2\text{O}_8$ Diopside $\text{Ca}(\text{Mg}, \text{Al})(\text{Si}, \text{Al})_2\text{O}_6$
ZR247/4	Quartz, syn SiO_2 Albite, calcian, ordered $(\text{Na}, \text{Ca})\text{Al}(\text{Si}, \text{Al})_3\text{O}_8$ Anorthite $\text{CaAl}_2\text{Si}_2\text{O}_8$ Diopside $\text{Ca}(\text{Mg}, \text{Al})(\text{Si}, \text{Al})_2\text{O}_6$
ZR088/3	Andesine $\text{Na}_{0.622}\text{Ca}_{0.368}\text{Al}_{1.29}\text{Si}_{2.71}\text{O}_8$ Quartz, syn SiO_2 Diopside $\text{Ca}(\text{Mg}, \text{Al})(\text{Si}, \text{Al})_2\text{O}_6$ Tephrite $(\text{Mg}, \text{Fe}, \text{Al}, \text{Ti})(\text{Ca}, \text{Fe}, \text{Na}, \text{Mg})(\text{Si}, \text{Al})_2\text{O}_6$ Albite calcian low $(\text{Na}_{0.75}\text{Ca}_{0.25})(\text{Al}_{1.26}\text{Si}_{2.74}\text{O}_8)$
ZR089/2	Quartz, syn SiO_2 Diopside $\text{Ca}(\text{Mg}, \text{Al})(\text{Si}, \text{Al})_2\text{O}_6$ Anorthite, ordered $\text{CaAl}_2\text{Si}_2\text{O}_8$
ZR253/4	Quartz, syn SiO_2 Albite, calcian, ordered $(\text{Na}, \text{Ca})\text{Al}(\text{Si}, \text{Al})_3\text{O}_8$ Diopside $\text{Ca}(\text{Mg}, \text{Al})(\text{Si}, \text{Al})_2\text{O}_6$ Pyrocene $(\text{Mg}_{0.998}\text{Fe}_{0.002})(\text{Ca}_{0.999}\text{Fe}_{0.028})(\text{Si}_2\text{O}_6)$ Andesine $\text{Na}_{0.622}\text{Ca}_{0.368}\text{Al}_{1.29}\text{Si}_{2.71}\text{O}_8$
ZR253/2	Quartz, syn SiO_2 Palladium (H-Loaded), syn Pd Cuprite Cu_2O Giniite, ferric, syn $\text{Fe}_5(\text{PO}_4)_4(\text{OH})_3 \cdot 2\text{H}_2\text{O}$
ZR093/2	Gypsum $\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$ Quartz, syn SiO_2 Diopside $\text{Ca}(\text{Mg}, \text{Al})(\text{Si}, \text{Al})_2\text{O}_6$
ZR094/1	Anorthite, ordered $\text{CaAl}_2\text{Si}_2\text{O}_8$ Quartz, syn SiO_2 Calcite CaCO_3 Albite high $(\text{K}_{0.02}, \text{Na}_{0.78})(\text{AlSi}_3\text{O}_8)$ Diopside $\text{Ca}(\text{Mg}, \text{Al})(\text{Si}, \text{Al})_2\text{O}_6$
ZR271/5	Quartz, syn SiO_2 Diopside $\text{Ca}(\text{Mg}, \text{Al})(\text{Si}, \text{Al})_2\text{O}_6$ Anorthite, sodian, ordered $(\text{Ca}, \text{Na})(\text{AlSi})_2\text{Si}_2\text{O}_8$ Albite, disordered $\text{Na}(\text{Si}_3\text{Al})\text{O}_8$
ZR369/8	Quartz, syn SiO_2 Calcium Sodium Aluminum Silicate $\text{Ca}_{0.8}\text{Na}_{0.2}\text{Al}_{1.8}\text{Si}_{2.2}\text{O}_8$
ZR061/4	Gypsum $\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$ Quartz, syn SiO_2 Diopside $\text{Ca}(\text{Mg}, \text{Al})(\text{Si}, \text{Al})_2\text{O}_6$
Ghulaman	Quartz, syn SiO_2 Diopside $\text{Ca}(\text{Mg}, \text{Al})(\text{Si}, \text{Al})_2\text{O}_6$ Albite, calcian, ordered $(\text{Na}, \text{Ca})\text{Al}(\text{Si}, \text{Al})_3\text{O}_8$
Clay A	Quartz, syn SiO_2 Calcite, CaCO_3 Clinochlore-1MIIB, ferroan, $(\text{Mg}, \text{Fe})_6(\text{Si}, \text{Al})_4\text{O}_{10}(\text{OH})_8$ Muscovite, $\text{KAl}_2\text{Si}_3\text{AlO}_{10}$ Dolomite $\text{CaMg}(\text{CO}_3)_2$ Chamosite $(\text{Mg}_{5.036}\text{Fe}_{4.964})\text{Al}_{2.724}(\text{Si}_{5.70}\text{Al}_{2.30}\text{O}_{20})(\text{OH})_{16}$ Albite, calcian, ordered, $(\text{Na}, \text{Ca})\text{Al}(\text{Si}, \text{Al})_3\text{O}_8$
Clay B	Quartz, syn SiO_2 Calcite, CaCO_3 Clinochlore-1MIIB, ferroan, $(\text{Mg}, \text{Fe})_6(\text{Si}, \text{Al})_4\text{O}_{10}(\text{OH})_8$ Muscovite, $\text{KAl}_2\text{Si}_3\text{AlO}_{10}$ Dolomite $\text{CaMg}(\text{CO}_3)_2$ Chamosite $(\text{Mg}_{5.036}\text{Fe}_{4.964})\text{Al}_{2.724}(\text{Si}_{5.70}\text{Al}_{2.30}\text{O}_{20})(\text{OH})_{16}$ Albite, calcian, ordered, $(\text{Na}, \text{Ca})\text{Al}(\text{Si}, \text{Al})_3\text{O}_8$

Table 2. Content of major elements in pottery shards and clay samples

Sample	Dry Weight (%)									
	Si	Al	Ca	Fe	Mg	Mn	Na	K	Ti	P ₂ O ₅
ZR332/3	77.56	3.13	1.52	0.48	0.93	0.02	4.85	1.32	0.11	0.09
ZR028/1	56.32	16.73	4.10	7.64	4.84	0.09	1.34	3.02	0.77	0.14
ZR087/6	52.04	15.08	9.78	6.52	5.07	0.12	2.19	2.55	0.67	0.20
ZR077/2	52.37	15.43	9.07	6.31	6.10	0.10	2.01	2.58	0.64	0.12
ZR078/8	43.98	13.16	13.64	6.04	5.02	0.14	2.16	2.27	0.58	0.41
ZR079/5	48.75	14.59	12.28	6.00	7.04	0.11	3.12	1.20	0.63	0.23
ZR080/4	54.76	12.55	10.21	5.12	6.68	0.10	2.56	2.09	0.52	0.31
ZR081/2	50.45	14.76	7.90	6.60	4.99	0.10	2.28	2.62	0.64	0.91
ZR083/4	50.59	15.34	8.77	7.38	4.29	0.11	1.41	3.20	0.69	0.31
ZR084/3	48.83	14.57	12.81	6.42	5.49	0.13	1.82	2.39	0.71	0.31
ZR086/3	55.18	15.22	6.68	6.10	4.93	0.11	2.43	3.19	0.62	0.26
ZR247/4	54.68	15.95	8.71	6.35	5.34	0.10	2.32	2.47	0.61	0.25
ZR088/3	53.98	12.79	11.78	5.16	5.48	0.09	2.53	1.90	0.58	0.16
ZR089/2	47.93	14.33	11.57	6.32	5.27	0.11	2.86	1.81	0.66	0.18
ZR253/1	53.15	16.75	8.53	6.87	5.79	0.12	2.02	2.74	0.66	0.13
ZR253/4	49.67	14.96	11.22	6.76	4.67	0.13	1.32	2.93	0.72	0.18
ZR093/2	53.59	15.79	9.18	6.28	5.02	0.11	1.98	2.81	0.61	0.42
ZR094/1	43.36	13.46	11.20	5.20	6.92	0.10	2.01	2.13	0.50	0.18
ZR271/5	53.51	14.60	9.43	6.21	4.73	0.13	2.26	2.31	0.67	0.15
ZR369/8	78.98	3.88	2.23	0.79	1.18	0.02	5.47	1.46	0.12	0.09
ZR061/4	45.49	11.66	12.92	5.06	7.17	0.09	2.52	1.56	0.53	0.33
Ghulaman	50.51	16.67	6.11	7.82	4.90	0.12	2.10	3.25	0.75	0.38
Clay A	45.56	13.14	12.94	6.95	4.57	0.13	0.87	2.60	0.78	0.15
Clay B	45.47	13.03	13.80	6.45	4.74	0.13	0.88	2.57	0.72	0.14

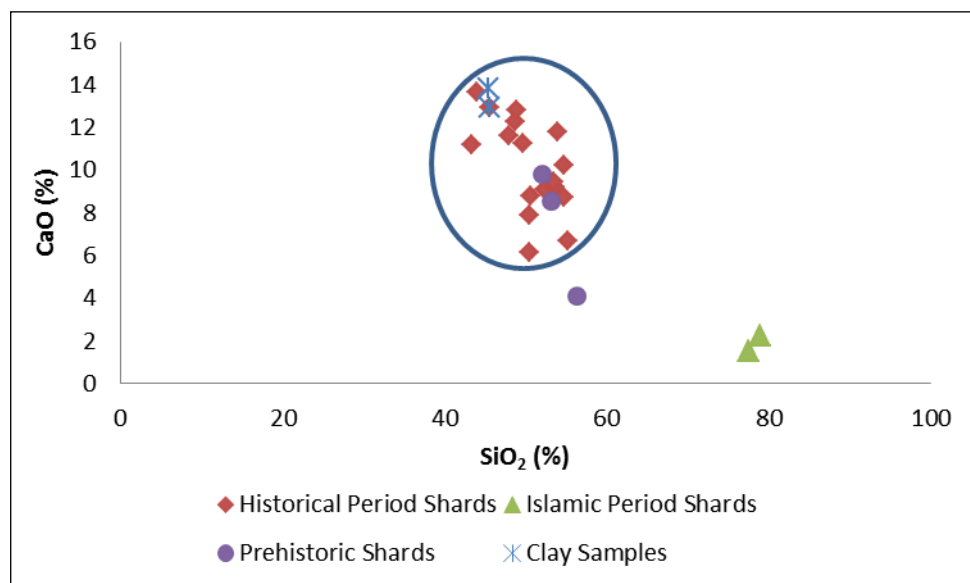
Figure 1. Scatter plot of SiO₂ and CaO percentage of shards and clay samples from Sistan Region

Table 3. Content of trace elements in pottery shards and clay samples

Sample	ppm ($\mu\text{g/g}$)									
	Pb	Cu	Ba	Sr	Zr	Rb	Zn	SO ₃	Cl	Co
ZR332/3	1500	200	3000	216	100	15	58	2400	2300	100
ZR028/1	Nd	89	400	306	200	104	200	1100	400	nd
ZR087/6	Nd	68	400	415	100	90	100	1300	600	nd
ZR077/2	Nd	88	500	425	100	108	100	2700	800	14
ZR078/8	Nd	61	600	900	400	118	nd	22700	1200	nd
ZR079/5	Nd	66	Nd	406	200	38	100	900	800	24
ZR080/4	Nd	56	Nd	510	100	66	200	2200	800	28
ZR081/2	Nd	79	500	1000	400	123	100	7000	2500	34
ZR083/4	Nd	70	500	305	100	125	300	500	200	nd
ZR084/3	nd	81	500	424	200	105	100	1600	400	nd
ZR086/3	nd	62	500	432	100	107	100	3100	4000	29
ZR247/4	nd	46	600	514	100	104	100	1000	500	nd
ZR088/3	nd	51	600	519	200	69	100	700	500	nd
ZR089/2	nd	63	Nd	411	100	87	100	2300	1600	nd
ZR253/1	nd	72	600	419	100	113	200	700	400	nd
ZR253/4	nd	84	500	526	200	115	100	1400	600	nd
ZR093/2	nd	68	600	529	100	118	100	1500	900	nd
ZR094/1	nd	70	800	700	500	63	200	8000	1300	21
ZR271/5	nd	67	400	312	100	104	100	600	800	32
ZR369/8	3700	57	4500	323	200	24	nd	3200	4100	300
ZR061/4	nd	94	400	1100	400	52	nd	16000	2200	nd
Ghulaman	nd	90	600	320	100	107	nd	1800	2000	40
Clay A	nd	83	Nd	325	96	98	110	2000	100	4
Clay B	nd	80	Nd	330	100	109	100	500	nd	nd

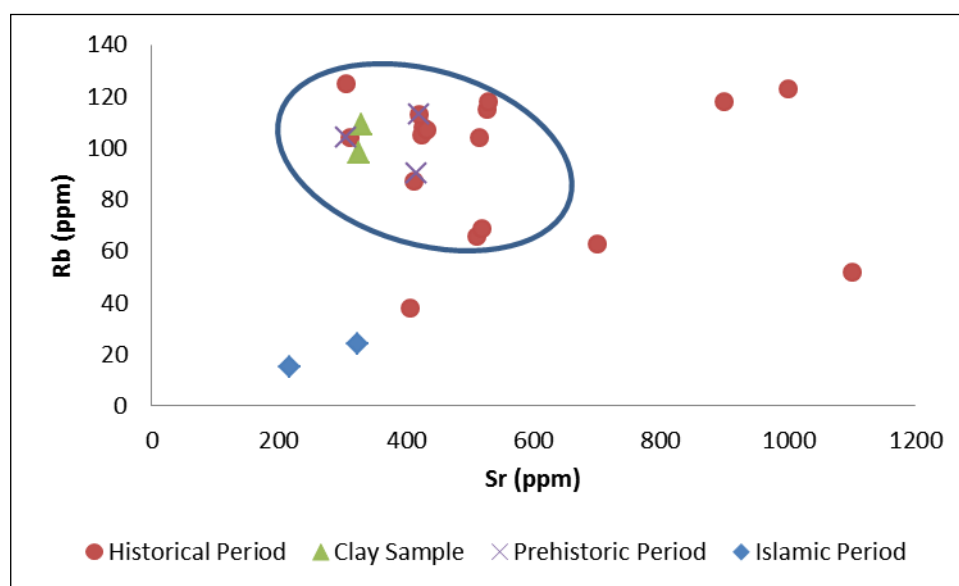


Figure 2. Scatter plot of Strontium and Rubidium concentration in pottery shards and clays sample from Sistan Region

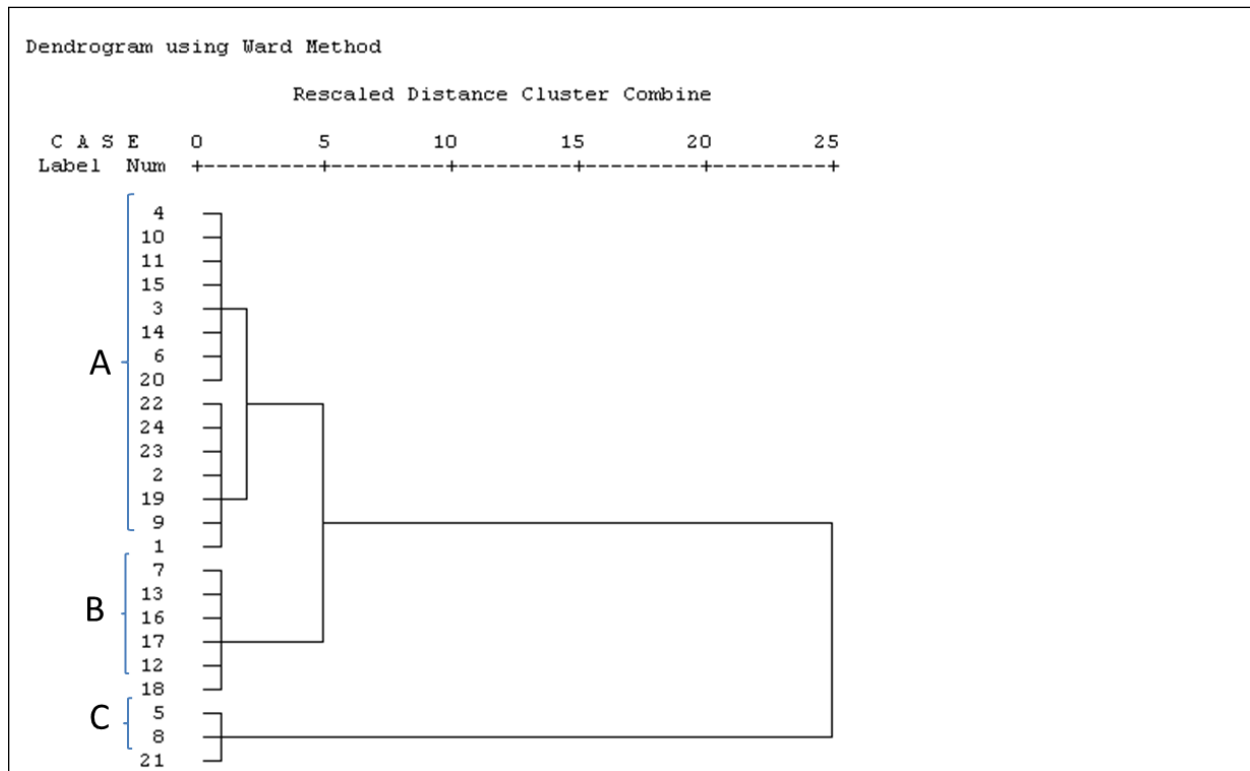


Figure 3. Hierarchical agglomerative clustering of the SiO_2 and CaO percentage of the pottery shards from several sites at Sistan, Iran

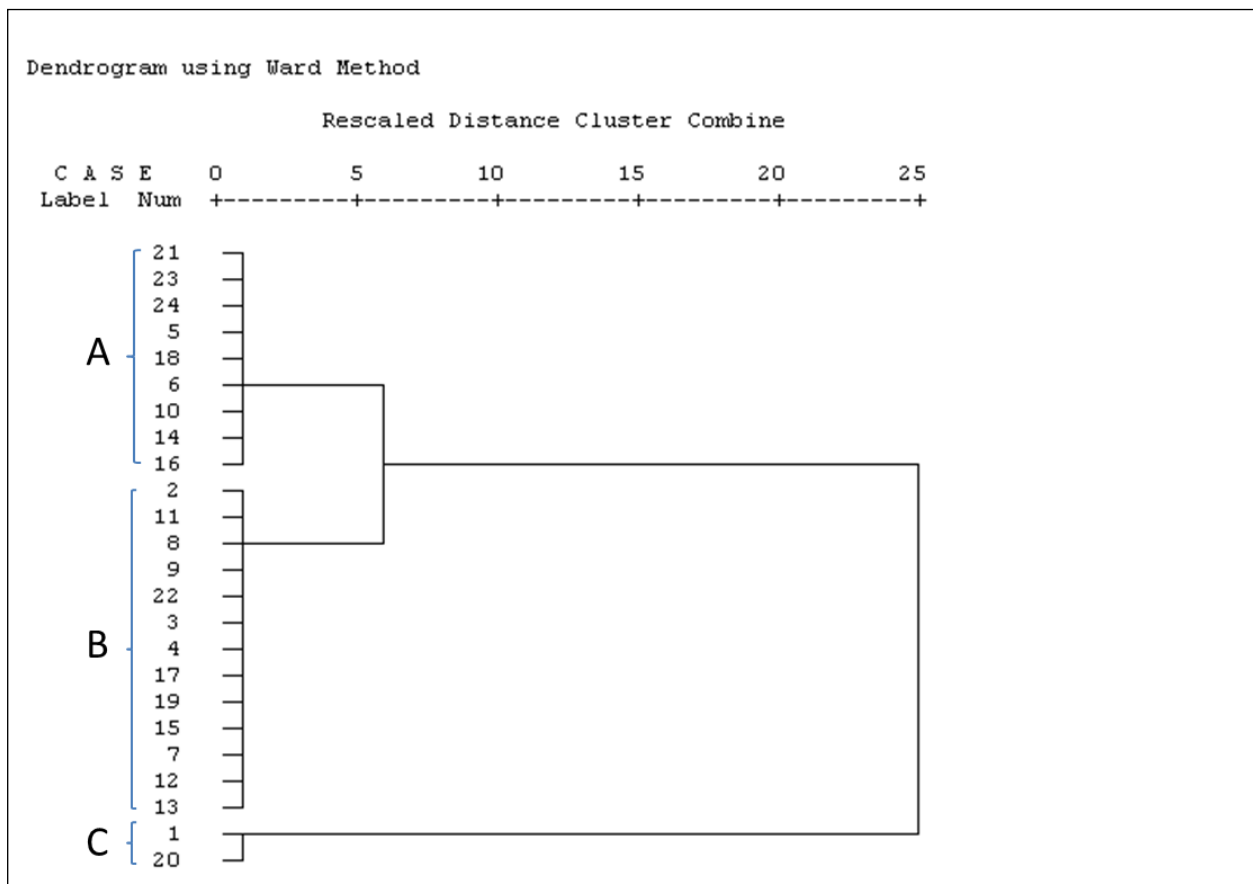


Figure 4. Hierarchical agglomerative clustering of the Strontium and Rubidium concentration of the pottery shards from several sites at Sistan, Iran

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