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MORPHOLOGY OF ANCIENT POTTERIES USING X-RAY DIFFRACTION ANALYSIS AND X-RAY FLUORESCENCE IN SISTAN PLAIN, EASTERN IRAN

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ABSTRACT

Sistan plain, located in the north of Sistan and Baluchestan province, is one of the most significant cultural area in eastern Iran. This region is located between south Asia (Indus valley) and Western Asia (Mesopotamia) and also has been a connector between cultures of Central Asia and South of Persian Gulf area. Sistan was the main area to connecting between west and south Asia. Much of the cultural items found in the site under exploration were huge bulk of diverse pottery. Most pieces of pottery found in the Sistan plain were of the pottery belonging to Shahr-e Sukhteh, and its villages dating back to the third millennium BC, Dahan-e Gholaman of the Achaemenid period 550 BC and a large number of sites belonging to the Islamic period, which vary in term of the colour ranging from buff, gray, black and red and in terms of thickness. This study aims to determine the morphological relations of the pottery of Sistan plain using semi-quantitative X-ray diffraction (XRD) and X-ray fluorescence (XRF) methods. In this regard, 52 pieces of pottery from prehistoric, historic and Islamic eras, which were collected from archaeological surveys, were analysed. The samples were gathered from Gerdi domain, Dahaneh Gholaman, Shahr-e Sukhteh, south of the Hamoun Lake, Rostam castle and around the Shileh River. The instrumentation and cluster analysis of pottery sherds indicated that the prehistoric pottery pieces of Sistan plain have a different composition compared with that of Sistan area. Moreover, the glazed pottery pieces of the Islamic era are different from those of Sistan plain in terms of their chemical and have silica compounds, gypsum and aluminosilicate, which indicates the continuity of local technology, production and trade in Sistan to the Islamic period. In addition, the composition and structure of pottery in this region accounts for the high level of skills and knowledge of potters, who made a variety of pottery pieces with diverse applications in the local communities, which continued from prehistory to the Islamic era in this plain.

KEYWORDS: Sistan plain, Prehistoric pottery, Historical period, Islamic pottery, XRD, XRF, Local manufacturing

1. INTRODUCTION

Sistan area is located in the eastern Iran and in northern Sistan and Baluchestan province. It has an area of 8114 square kilometres. This region from the Bronze Age has had a significant role in the development of human civilization and heritage of this Iranian region (Moradi et al, 2014; Sarhaddi-Dadian et al 2015a; Sarhaddi-dadian et al, 2017a). Shahr-e Sukhteh, Dahaneh Gholaman, Khajeh Mount, the first settlement in the region were dating back to the late fourth millennium BC, which coincided with the history of urbanization in Central Asia. This era prolonged to the second millennium BC, (Sajjadi, 2008; 307, Tosi 1983; 73-125) (many sites have been discovered in Sistan and Share-Sukhteh is only one of significant sites of the region) (Vidale and Tosi 1996; 251-269; Biscione 1987; 394) (each of these regions has a lot of ceramic sherds which are diverse in terms of colour, era, ornaments and technical features. The main feature of the Bronze Age pottery is the application of geometrical, plant ornaments as well as drawing animals inside and outside of the piece with black and ochre colour to the buff pottery or those with decorated with light gray slime (Tosi, 1983; 136-139, Salvatori Andvidale, 1997). There is no evidence of such type of pottery since the Bronze era, about 3200 years BC, to the Achaemenid Empire, 550 AD. The next era which is the historic period of Sistan plain which began from 550 BC and continued to 550 AD included the periods of the Achaemenid, Parthian and Sassanid empires (Chavalas, 1999; 88). The important feature of the pottery of this era was its simplicity and the absence of any type of interior or exterior decoration and the ornamentation is generally carvel lines in the form of reputed grooved Sistani pottery. The other feature of these pottery pieces is the thick or thinner glaze and sometimes in red colour (Mehrafarin and Musavi, 2011; 240-58). Studies indicate that no pottery piece has been found in this region between the late Sassanid era (16th century) and the 13th century AD. Many new sites were discovered following this era in Sistan, from which large pieces of pottery were recovered. Pottery of this period is different than the two previous eras, which is simultaneous with middle Islamic era. This type of pottery pieces is decorated in different colours. The dominant colour glazes of this era are decorated green, gray-blue, milky and black and special decorations like under glaze motif or glaze or black colour glazes (niello) as well as geometrical decoration in the form of wide-mouthed bowls with flat base. The paste colour of pottery is light buff in the Islamic era (Mousavi and Atai 2010). Six pottery variation belonging to the areas of the Shileh River, Gerdi Castle, south of Rostam Castle, Shahre-sukhteh, south of Hamoun Lake, and Dahaneh Gholaman were systematically examined (Sarhadi-Daddian, 2013). Nine hundred pieces of pottery were selected from the region, and most of them belong to the pre- and historic eras. The pottery pieces of the historic period were mainly red and buff in terms of paste colour while the pottery of the prehistoric era generally was buff, gray, red black. Previous studies indicate that the red and gray pottery was less common in the Sistan region: however, the use of this type of pottery was prevalent, especially in Balochestan and marginal areas of India. The samples of this type of pottery were common as the burial pottery among other pottery pieces in the graveyard of Shahre-Sukhteh (Moradi et al, 2013a). The origin of the raw material of pottery — whether it is imported — can be determined by comparing the main and secondary elements according to instrumental analyses. Therefore, this study aims to determine whether all the pottery gathered from different regions of Sistan with a close relation are manufactured in local workshops or are cultural remote communications. The Documentation of the origin of the pottery is very vital for archaeologists and archeometrist as they contribute to investigate local civilization and culture in the manufacture of pottery and commercial activities in connection with other communities cultural activities (See Papageorgiou and Liritzis 2007).

2. MATERIALS AND METHOD

In The pottery of Sharhe-sukhteh found in the site and its surrounding Tape-Aghmari around like hills 3 Km to the south of Shahre-Sukhteh (Tosi, 1934; 34) and the other, which is Biyaban river 25 kilometers south of Shahre-Sukhteh, are the clear sample of ancient pottery workshops in the third millennium BC, are (Tosi, 1983; 42. Vidale & Tosi, 1996; 252). Fifty-eight factors from the wide prehistoric area of Sistan, i.e., Shahreh-sukhteh, and in total 186 images were identified according to the table of Sheppard (Tosi, 1983; 136-9. Salvatori and Vidale, 1997,27). XRF semi-quantitative analysis was conducted to identify the local origin of pottery (Sarhadi-Dadian et. al., 2015b). Scientific analysis conducted on Shahreh-Sukhteh reveals that some segments of red and buff pottery are imported goods and have a large content of lead. (Moradi et al, 2013). Example: Two pieces of the Islamic pottery are covered with plant motif and is decorated glaze. These two samples are decorated with enamels. Sample 1/332 is decorated with plant motifs in blue while Sample 8 / 369ZR are decorated with brown, blue and red plant motifs. The origin of these two pieces of pottery is not clear and is not local which probably belongs to the Safavid era. (Lane, 1947; Lane, 1948). Conducting combined analysis is

one of the most crucial methods to determine the chemical composition of ancient relics such as pottery, mortar, ancinet metals, brick and glass (Bieber et al, 1976. Brokmans et al, 2008. Marghusian et al, 2009;l Wong et al, 2010; Zuliskandar et al, 2013a; 2013b; Bater. 2010). Other historic sites of the plain include Dahaneh Gholaman, Gerdi domain, Shile Rhiver, the south of Hamoun Lake, and Rostam castle, on which little quantitative analysis has been done (Sarhad-daddian et al, 2017b). Given the geographical location of Sistan region which had many impacts on the trade between mentioned zones as well as central Asia and Arabians located south of the Persian Gulf from prehistoric times has been the focus of attention from pre-historic era because of its location among eastern civilizations such as India, Pakistan as well as eastern civilizations such as Elam, Mesopotamia. High traveling in the region has caused the art and civilization to restore; pottery is one of the arts that requires demanding is research. It is an issue that attracts the attention of archeologists and archeometrists

3. RESEARCH METHOD

This study employs an empirical analytic library research method on the basis of field and laboratory studies. After collecting data and samples from six sites, including Shahr-e Sukhteh, Dahaneh Gholaman, the south of Hamoun Lake, Shileh River, Rostam Castle and explorations into Sistan, 52 samples of pottery were collected that includes pottery of gray, red and buff color. To investigate the source of crude pottery, 2 clay samples were also taken from the soil mines in Sistan plain to be tested in the laboratory; one sample was taken from one meter and the other sample was taken from 4 meters below ground level. Since the pottery from various eras was dispersed, first, we put them into three categories according to the prehistoric, historical and Islamic eras, and presented the characteristics and type of decoration in each era. Next, the instrumental analysis of samples was performed using the XRD and XRF elemental analysis. The results and data were investigated using hierarchical cluster analysis (HCA) and Ntsys ver2.1 software based on Nei and Li similarity matrix and Upgma method. Finally, the data shows that the pottery of prehistoric communities to the Islamic era forms the culture in Sistan where the pottery is recognized as a product with specific applications which are found in local areas of the hills or sites discussed here.

4. MATERIALS AND METHODS

This section includes the introduction of samples, sampling and sample preparation process and the equipment used for analysis methods.

4.1 Samples

This study investigates and performs XRF and XRD semi-quantitative analysis on 52 pieces of pottery recovered from six sites in Sistan plain in order to find the connection between these sites and the raw material used in the production of this ancient relic (Table 1).

Table 1: Gross observation of pottery collected from Sistan

Number	Shape	Paste color	Body color	Chamotte	Pattern color	Done	Type	Era
18264-1	Jar	Buff	Buff	Aeolian sand	_____	On a wheel	Standard	II-III
18261-2	Jar	Buff	Buff	Aeolian sand	Dark brown	On a wheel	Standard	II-III
18267-3	Jar	Buff	Buff	Fine gravel	_____	On a wheel	Coarse	II-III
18273-4	Jar	Buff	Buff	Aeolian sand	Brown	On a wheel	Standard	II-III
18262-5	Jar	Brick	Brick	Fine gravel	_____	On a wheel	Coarse	II-III
18263-6	Jar	Buff	Buff	Fine gravel	_____	On a wheel	Standard	II-III
18269-7	Bowl	Buff	Buff	Aeolian sand. Fine gravel	Brown	On a wheel	Standard	II-III
18268-8	Bowl	Buff	Buff	Aeolian sand	Brown	On a wheel	Standard	II-III
18259-9	Bowl	Gray	Gray	Aeolian sand	_____	On a wheel	fine	II-III
18265-10	Bowl	Gray	Gray	Aeolian sand	Black	On a wheel	Fine	II-III
18270-11	Bowl	Buff	Buff	Aeolian sand	Light brown	On a wheel	Standard	II-III
18260-12	Cup	Buff	Buff	Fine sand	Dark brown	On a wheel	Fine	II-III
18266-13	Bowl	Red	Red	Aeolian sand	_____	On a wheel	Fine	II-III
18272-14	Glass shaped	Buff	Buff	Aeolian sand. Fine gravel	_____	On a wheel	Standard	II-III
18271-15	Jar	Red	Red	Aeolian sand	_____	On a wheel	Standard	II-III
QH6-1	Bowl	Red	Red	Aeolian sand	_____	On a wheel	Standard	Historical
QH29-2	Cup	Red	Red	Aeolian sand	_____	On a wheel	Standard	Historical
QH20-3	Bowl	Red	Red	Straw and grass	_____	On a wheel	Coarse	Historical
QH6-1	Bowl	Red	Red	Aeolian sand	_____	On a wheel	Fine	Historical
QH8-15	Jar	Red	Red	Straw and grass	_____	On a wheel	Standard	Historical
QH17-11	Cup	Red	Red	Aeolian sand	_____	On a wheel	Fine	Historical

QH18-13	Bowl	Red	Red	Aeolian sand	_____	On a wheel	Fine	Historical
QH2-14	Bowl	Red	Red	Aeolian sand	_____	On a wheel	Fine	Historical
QH3-8	Bowl	Red	Red	Straw and grass	_____	On a wheel	Fine	Historical
QH23-9	Bowl	Red	Red	Straw and grass	_____	On a wheel	Coarse	Historical
QH26-5	Bowl	Red	Red	Straw and grass	_____	On a wheel	Coarse	Historical
QH29-2	Bowl	Red	Red	Straw and grass	_____	On a wheel	Standard	Historical
QH32-4	Jar	Red	Red	Aeolian sand and fine gravel	_____	On a wheel	Coarse	Historical
QH34-7	Jar	Red	Red	Aeolian sand	_____	On a wheel	Standard	Historical
QH38-12	Bowl	Red	Red	Straw and sand	_____	On a wheel	Standard	Historical
Ghulam-1	Jar	Red	Red	Aeolian sand	_____	On a wheel	Standard	Historical
ZR332/3	Bowl	White	White	_____	White and blue glaze	On a wheel	Standard	Islamic
ZR028/1	Jar	Red	Red	Aeolian sand	Black	On a wheel	Standard	II-III
ZR087/6	Jar	Buff	Buff	Aeolian sand	Engraved decorations	On a wheel	Standard	II-III
ZR077/2	Jar	Buff	Buff	Aeolian sand	_____	On a wheel	Standard	Historical
ZR078/8	Bowl	Buff	Buff	Aeolian sand	_____	On a wheel	Standard	Historical
ZR079/5	Bowl	Buff	Buff	Aeolian sand	_____	On a wheel	Fine	Historical
ZR080/4	Bowl	Buff	Buff	Aeolian sand and fine gravel	_____	On a wheel	Coarse	Historical
ZR081/2	Jar	Buff	Buff	Aeolian sand	_____	On a wheel	Coarse	Historical
ZR083/4	Bowl	Gray	Gray	Aeolian sand	Burnished	On a wheel	Standard	Historical
ZR084/3	Bowl	Buff	Buff	Aeolian sand	_____	On a wheel	Coarse	Historical
ZR086/3	Bowl	Red	Red	Aeolian sand	_____	On a wheel	Standard	Historical
ZR247/4	Bowl	Red	Red	Aeolian sand	_____	On a wheel	Standard	Historical
ZR088/3	Bowl	Buff	Buff	Aeolian sand and fine gravel	Embossed	On a wheel	Coarse	Historical
ZR089/2	Bowl	Buff	Buff	Aeolian sand	_____	On a wheel	Standard	Historical
ZR253/1	Bowl	Gray	Gray	Aeolian sand	Black	On a wheel	Standard	II-III
ZR253/4	Bowl	Red	Red	Aeolian sand	_____	On a wheel	Standard	historical
ZR093/2	Bowl	Buff	Buff	Aeolian sand	_____	On a wheel	Standard	Historical
ZR094/1	Bowl	Red	Red	Aeolian sand and fine gravel	_____	On a wheel	Coarse	Historical
ZR271/5	Jar	Red	Red	Aeolian sand and fine gravel	_____	On a wheel	Coarse	Historical
ZR369/8	Bowl	White	White	_____	White and blue glaze	On a wheel	Fine	Islamic
ZR061/4	Bowl	Buff	Buff	Aeolian sand and fine gravel	_____	On a wheel	Coarse	Historical

We have collected 52 pieces of pottery in Sistan plain belonging to different era. Moreover, to investigate the clay used by potters, two clay samples as raw materials were selected from two different areas to be tested in laboratory, hence comparing the pottery. One sample was taken from a depth of 4 meters in the vicinity of Shahre Sukhteh (about 10 gr) and the other was taken from 3 kilometers to the south of the site of Shahre Sukhteh (about 10 gr) where the people of Sistan still use this clay to make pottery and bricks (Fig.1).

We have divided the pottery into three categories according to the prehistoric, historical and Islamic eras. The 18 pieces of pottery belonging to the prehistoric era were collected from the area of Shahre Sukhteh and the surrounding hills, including the samples with the numbers 253/1, 18259/9, 18265/10, 028/1, 18266/13, 18271/15, 087/6, 18260/12, 18261/2, 18262/6, 18263/6, 18264/1, 18267/3, 18268/8, 18269/7, 18270/11, 18272/14, 18273/4, a collection which statistically includes buff, red and gray pieces of pottery in terms of priority. These pieces have been done on a wheel and in most cases had buff-colored clay paste (Fig.2). Buff pottery is common in Sistan and the paste has different tones ranging from absolutely buff to green. Statistically speaking, the gray pottery has second order dispersion and the red one has third order dispersion (Tosi 1983b; 132). Unfortunately, according to the surveys and data, there is no evidence of Iron Age in Sistan and it is still unknown to us. The evidence may have been covered by sediments that had been carried by the Hirmand River in Sistan plain over thousands of years. The historical era has begun in the Dahaneh Gholaman site. The diversity of the pottery indicates the benefits and usefulness of these relics in the past, and according to the shape, the skilled potter defined a particular usage for it.

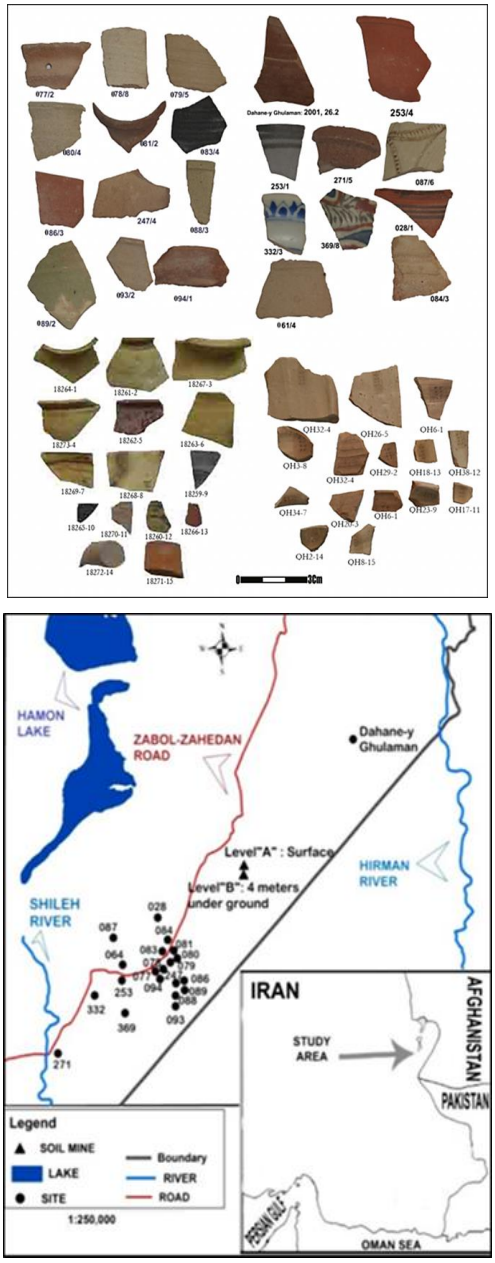

Figure 1: Sistan plain map site locations and representative sherds (Sarhaddi-dadian et al., 2015b ; 2017b)

Since most historical sites that have been investigated in this study belong to the historical era, thirty pieces of pottery selected for chemical studies were taken from the historical sites, the most important of which includes Dahaneh Gholaman, Gerdi domain, Shileh River, the south of Hamoun Lake and Rostam castle. The collection belonging to that era with the numbers: 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 excavated from Shileh River, Rostam castle, the south of Hamoun Lake and Gerdi domain were simple (Fig. 2). The other historical pieces excavated from Dahaneh Gholaman historical site include: Gholaman 2/26 / 2001, QH1/6, QH2/29, QH3/20, QH6/1, QH8/15, QH17/11, QH18/13, QH19/10, QH20/3, QH23/9, QH26/5, QH29/2, QH32/4, QH34/7, QH38/12 which (Figure 2) belong to the Achaemenid empire. (Sarhaddi-daddian, 2013). These pieces are devoid of any decorative motif. They have the paste colors of buff, light red and red and mineral fillers and chopped herbs. It seems that the use of horizontal grooves in decoration started from the Achaemenid period and developed in later periods. The pieces found in Dahaneh Gholaman site have all simple human motifs and these decorative motifs are observed on the outside surface of the cups that have the same size of rim and body (Genito, 1990; 588-601).

The pottery belonging to the Partian era has significant difference in terms of form and style with the Achaemenid era. Most of the pieces are simple and usually covered with buff, bright, red and dark slip. Pottery with burnished decoration (Mehrafarin et al., 2013) is one of the most common motifs in the Partian era in Sistan (Haerinck, 1980; 43-45; Mehrafarin and Mousavi, 2011; 240-258). According to the archaeological studies, there are few Sasanian settlements in southern Sistan and there is not enough evidence related to these parts. The pottery belonging to the Sassanid period has the same properties as the Partian era. This era is also characterized with unglazed ceramics and the dominant color of the pottery is red as before. The pottery is mostly simple, covered with a thick layer of slip. It is decorated with grooves on the surface. The other ones are decorated with stamped patterns, and geometric and plant motifs (Mehrafarin and Mousavi; 2010; 256-272) The Islamic era pottery have been identified in the site 332 with the number 3 and in the site 369 with the number 8. Islamic era pottery is different from other pottery in Sistan. The pottery is glazed like the pottery of other parts of Iran; therefore, the classification includes both the glazed and unglazed patterned pottery (Mousavi and Ataie, 2010; 302-321). The mineral fillers and the patterns of zig-zag lines and small circles are used. Painted and glazed pieces belonging to the Safavid era have been found in sites 332/3 and 369/8 that are decorated with red, brown and blue patterns on a milky background and are transparently glazed (Lane, 1947. Lane, 1948. p 1032) A special type of pottery that was mentioned has patterns under a blue glaze and geometric and plant motifs that are comparable to the pottery belonging to the 6th -13th century AD in southern Sistan. (Golombek, 2003; 53-270. Sconlon, 1948).

4.2 Analysis and Sampling Methods

For the characterization of the shards and clay samples, analytical instruments used included X-Ray Diffraction SIEMENS D5000 Diffractometer and XRF Spectrometer Philips Model PW1480. X-ray fluores-

cence spectrometry (XRF) is employed as a non-destructive analytical method widely used to determine the elemental composition of materials. (Ferretti, 2000; Milazzo, 2004). For the analysis, in order to determine the chemical composition of the pottery, each sample of weight 0.7 g was pulverized, heated up at a temperature of 105°C for one hour and mixed until homogenous with the flux powder, 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 material was molded in a container and cooled gradually into pieces of fused glass with a thickness of 2 mm and a diameter of 32 mm. The samples were of 1:10 dilution. Press pallet samples were prepared by mixing 1.0 g of samples together with 6.0 g of boric acid powder; then, a pressure of 20 psi (137.895 kPa) was applied using hydraulic pressure equipment. The samples of fused pallets and pressed pallets were analyzed by wavelength-dispersive X-Ray Fluorescence (WD-XRF). A Philips PW1480 sequential spectrometer fitted with a rhodium-anode X-Ray tube (3kW 60kV) was used for the analysis of major and trace elements. The spectrometers were controlled using Philips X40 application software package version 3.2 and 4.01 running under the DEC VMS operating system.

5. RESULTS AND DISCUSSION

5.1 X-ray diffraction analysis

X-ray diffraction analysis (XRD) was conducted on 33 pieces of pottery and the two soil samples to determine minerals of the samples (Table 2).

Table 1 a: The results of XRD analyzes on Sistan plain pottery

	ZR 332/3	ZR 028/1	ZR 087/6	ZR 077/2	ZR 078/8	ZR 080/4	ZR 079/5	ZR 081/2	ZR 083/4	ZR 086/3	ZR 084/3
QTZ	+	+	+	+	+	+	+	+	+	+	+
PLQ	+	+	+	+	-	-	+	+	+	+	-
GYP	+	-	-	-	-	-	-	-	-	-	-
DIP	-	-	+	+	+	+	-	+	+	+	+
AND	-	-	-	-	-	-	+	-	-	-	-
LAB	-	-	-	-	-	-	-	-	+	-	-
HEM	-	+	-	-	-	-	-	-	-	-	-
DIK	-	+	-	-	-	-	-	-	-	-	-

Table 1 b: The results of XRD analysis Sistan plain pottery

	LS-1	LS-2	18259-9	18260-12	18270-11	18273-4	ZR 247/4	ZR 088/3	ZR 089/2	ZR 253/2	ZR 093/2
QTZ	+	+	+	+	+	+	+	+	+	+	+
PLQ	+	+	+	+	+	+	+	+	+	+	+
DIP	-	-	-	-	-	-	+	+	+	+	+
GYP	-		+	+	+	+	-	-	-	-	-
DOL	+	+	-	-	-	-	-	-	-	-	-
Ca	+	+	-	-	-	-	-	-	-	-	-
ClF	+	+	-	-	-	-	-	-	-	-	-
MUS	+	+	-	-	-	-	-	-	-	-	-
CHAM	+	+	-	-	-	-	-	-	-	-	-
AND	-	-	-	-	-	-	+	-	-	+	-
TEPH	-	-	-	-	-	-	-	+	-	-	-
PYR	-	-	-	-	-	-	-	-	-	+	-

Table 1c: The results of XRD analysis on Sistan plain pottery

	Gh-1	QH 2-29	QH 8-15	QH 20-3	QH 23-9	QH 26-5	QH 29-2	ZR 094/1	ZR 271/5	ZR 369/8	ZR 061/4
QTZ	+	+	+	+	+	+	+	+	+	+	+
PLQ	+	-	-	+	+	-	+	+	+	+	-
DOL	-	+	+	+	+	+	+	+	-	-	+
DIP	+	-	-	-	-	-	-	-	+	-	-
GYP	-	-	-	-	-	-	-	-	-	+	-
HEM	-	-	-	-	-	-	-	+	-	-	-

PLQ: Plagioklaz
DIP: Diopside Ca(Mg, Al)(Si, Al)2O6
LAB: Labradorite Ca0.65Na0.35(Al1.65Si2.35O8)
TEPH: Tephrite (Mg, Fe,Al, Ti)(Ca, Fe, Na, Mg)(Si,Al)2O6
PYR: Pyrocene (Mg0.998Fe0.002)(Ca0.999Fe0.028)(Si2O6)
CLF: Clinochlore-1MIIb, ferroan, (Mg,Fe)6(Si,Al)4O10(OH)8
CHAM: Chamosite (Mg5.036Fe4.964)Al2.724(Si5.70Al2.30O20)(OH)16
AND: Andesine Na0.622Ca0.368Al1.29Si2.71O8
DIK: DickiteAl2Si2O5(OH)4(HCONH2)

QTZ:Quartz, syn SiO2
GYP:Gypsum CaSO4.2H2O
MUS:Muscovite, KAl2Si3AlO10
Ca:Calcite, CaCO3
DOL:Dolomite CaMg(CO3)2
HEM:Haematite Fe2O3
GH-1: Gholaman
LS: Local Soil

XRD analysis on the pieces of prehistoric pottery indicates that these parts include minerals such as quartz, albite, dickite, hematite and is diopside. Two pottery pieces belong to the Islamic period (ZR 3/332, ZR 8/369) which have color motif and glaze on the surface, and the data is indicative of quartz, gypsum and calcium (sodium Aluminosilicates). Sample ZR3 / 332 according to XRF analyses have a high level of quartz, which as the remained mineral substance suggest the high temperature of firing near the 1000°C. The historic pottery piece also implies the presence of the phases of quartz, diopside, plagioclases in the form of anorthite, albite, andesine, labradorite and lower amounts of minerals such as palladium, gunite and gypsum in these pieces while cuprite has only one of the pieces. Sample of analyzed clay include the phases of quartz, calcite, clinochlore, muscovite, dolomite, Chamosite and plagioclase. Clay samples were supplied from older units that had been used for pottery and bricks.

The diopside only exists in pottery pieces while it is not found in clay samples. Prehistoric pottery has diopside while most of the historic pieces of pottery have diopside. This mineral substance is found in ultramafic igneous rocks (i.e., kimberlite and peridotite), as well as rich diopside agite. Moreover, this is common in mafic rocks such as basalt and andesite. Diopside is also formed in a variety of metamorphic rocks while being in contact with developed metamorphic skarn created from dolomite with high silica. Regarding the remaining diopside in the pottery pieces, it is suggested that diopside mineral substances in the pottery pieces may come from dolomitic present in clay.

5.2 X-Ray fluorescence analysis

Table 3 shows the main elements of the recovered pottery pieces from several ancient sites in Sistan plain, which have a homogeneous composition (Table 3).

Table 3: The main constituent elements of Sistan plain pottery by Dry Weight%

Sample	Major Elements (%)											
	Na2O	MgO	A12O3	SiO2	P2O5	SO3	K2O	CaO	TiO2	MnO	Fe2O3	SrO
QH1-6	1.7	3.8	15.3	64.5	1.2	0.67	1.2	5.6	0.38	0.65	5.4	0.079
QH2-29	1.1	4.6	15.1	60.4	2.1	0.58	1.7	5.8	0.49	0.086	7.9	0.073
QH3-20	0.98	4.4	13.2	63.5	1.6	0.37	1.6	5.9	0.50	0.078	7.8	0.068
QH6-1	1.4	4.2	14.3	61.7	1.0	-	1.1	7.3	0.53	0.087	8.2	0.052
QH8-15	-	4.3	13.4	62.0	1.2	-	1.5	7.2	0.58	0.094	9.5	0.080
QH17-11	-	4.1	13.9	62.4	2.2	-	1.6	5.9	0.63	0.075	8.8	0.075
QH18-13	1.2	4.7	14.2	60.4	1.7	-	1.5	5.6	0.60	0.098	9.8	0.061
QH19-10	2.4	4.9	12.9	66.6	0.84	-	1.4	4.5	0.37	0.058	5.8	0.071
QH20-3	3.1	4.6	10.8	65.4	1.9	0.034	1.7	4.8	0.54	0.060	6.7	0.13
QH23-9	1.4	4.6	15.9	62.1	0.77	-	1.5	4.7	0.50	0.077	8.3	0.072
QH26-5	1.7	6.6	13.8	60.1	0.93	0.026	1.3	7.7	0.46	0.077	7.2	0.058
QH29-2	2.8	6.3	14.3	57.2	1.7	0.52	1.5	5.5	0.58	0.091	9.3	0.073
QH32-4	2.8	5.3	17.6	57.3	1.3	0.49	1.5	4.6	0.50	0.079	8.3	0.081
QH34-7	2.3	5.1	15.7	54.9	5.5	0.62	1.8	5.6	0.47	0.091	7.8	0.095
QH38-12	2.2	6.3	15.5	56.8	1.4	-	1.6	5.7	0.56	0.12	9.3	0.68
18259-9	1.43	4.43	17.84	55.55	0.21	0.42	3.44	5.79	0.81	0.10	8.71	0.06
18260-12	3.04	6.84	12.57	50.88	0.12	0.50	1.80	12.62	0.53	0.10	5.57	0.10
18261-2	2.34	5.46	13.69	51.78	0.13	1.72	2.33	10.99	0.60	0.11	6.01	0.067
18262-5	4.73	5.87	11.25	51.27	1.39	1.05	2.65	10.40	0.44	0.09	4.97	0.11
18263-6	3.86	5.46	11.25	49.30	0.23	0.85	2.47	12.07	0.49	0.09	4.97	0.059
18264-1	2.77	7.67	10.63	49.69	0.17	2.80	2.37	12.71	0.47	0.09	4.81	0.096
18265-10	1.42	4.66	16.58	60.39	0.13	0.15	3.21	2.40	0.82	0.09	8.84	0.016
18266-13	1.82	3.74	16.60	53.14	0.15	0.34	3.42	5.00	0.73	0.09	7.22	0.057
18267-3	2.75	7.03	12.59	52.89	0.16	0.73	1.96	12.75	0.54	0.12	5.56	0.10
18268-8	2.21	5.46	12.36	52.86	0.14	2.00	2.63	11.34	0.52	0.10	5.35	0.092
18269-7	2.77	9.87	10.58	52.30	0.11	1.33	2.14	11.62	0.46	0.09	5.05	0.083
18270-11	2.25	6.00	13.29	51.34	0.30	0.73	2.74	12.36	0.58	0.11	6.10	0.065
18271-15	2.61	3.33	15.26	62.64	0.60	0.17	2.71	1.55	0.75	0.66	6.79	0.063
18272-14	3.23	5.09	10.51	47.36	0.18	2.98	3.09	12.69	0.51	0.08	4.78	0.061
18273-4	2.98	6.07	12.24	49.57	0.27	0.72	2.43	11.85	0.56	0.10	5.64	0.16
ZR 332/3	4.85	0.93	3.13	77.56	0.09	-	1.32	1.52	0.11	0.02	0.48	0.021
ZR 028/11	1.34	4.84	16.73	56.32	0.14	-	3.02	4.10	0.77	0.09	7.64	0.030
ZR 087/6	2.19	5.07	15.08	52.04	0.20	-	2.55	9.78	0.67	0.12	6.52	0.041
ZR 077/2	2.01	6.10	15.43	52.37	0.12	-	2.58	9.07	0.64	0.10	6.31	0.042
ZR 078/8	2.16	5.02	13.16	43.98	0.41	-	2.27	13.64	0.58	0.14	6.04	0.090
ZR 079/5	3.12	7.04	14.59	48.75	0.23	-	1.20	12.28	0.63	0.11	6.00	0.040
ZR 080/4	2.56	6.68	12.55	54.76	0.31	-	2.09	10.04	0.52	0.10	5.12	0.051
ZR 081/2	2.28	4.99	14.76	50.45	0.91	-	2.62	7.90	0.64	0.10	6.60	0.10
ZR 083/4	1.41	4.29	15.34	50.59	0.31	-	3.20	8.77	0.69	0.11	7.38	0.035

ZR 084/3	1.82	5.49	15.34	48.83	0.31	-	2.39	12.81	0.71	0.13	6.42	0.042
ZR 086/3	2.43	4.93	15.22	55.18	0.26	-	3.19	6.68	0.62	0.11	6.10	0.043
ZR 247/4	2.32	5.34	15.95	54.68	0.25	-	2.47	8.71	0.10	0.61	6.35	0.051
ZR 088/3	2.53	5.48	12.79	53.98	0.16	-	1.90	11.78	0.58	0.09	5.16	0.059
ZR 089/2	2.86	5.27	14.33	47.93	0.18	-	1.81	11.57	0.66	0.11	6.32	0.041
ZR 253/1	2.02	5.79	16.75	53.15	0.13	-	2.74	8.53	0.66	0.12	6.87	0.041
ZR 253/4	1.32	4.67	14.96	49.67	0.18	-	2.93	11.22	0.72	0.13	6.76	0.052
ZR 093/2	1.98	5.02	15.79	53.59	0.42	-	2.81	9.18	0.61	0.11	6.28	0.052
ZR 094/11	2.01	6.92	13.46	43.36	0.18	-	2.13	11.20	0.50	0.10	5.20	0.070
ZR 271/5	2.26	4.73	14.60	53.51	0.15	-	2.31	9.43	0.67	0.13	6.21	0.031
ZR 369/8	5.47	1.18	3.88	78.98	0.09	-	1.46	2.23	0.12	0.02	0.79	0.032
ZR 061/4	2.52	7.17	11.66	45.49	0.33	-	1.56	12.92	0.53	0.09	5.06	0.11
Gh-1	2.01	4.90	14.67	57	1.38	-	1.5	4.8	0.75	0.091	7.07	0.032
Clay-LS-1	0.87	4.57	13.14	45.56	0.15	-	2.60	12.94	0.78	0.13	6.95	0.032
Clay-LS-2	0.88	4.74	13.03	45.47	0.14	-	2.57	13.80	0.72	0.13	6.45	0.033

Excluding two pieces of pottery which belong to the Islamic period, the dry weight percent of silica for the layers of prehistoric and historic pottery pieces is approximately from 43 to 65.4 and aluminum content of is 10 to 17. Except for 18271-4, 18256-10, 18259-9, which are probably non-native pottery of the region (Sarhadi-Dadian et. al, 2015b; 2017b) the calcium amount is 4 to 13 calcium and iron ranges approximately from 5 to 5.9. Alkali elements such as magnesium, sodium and potassium percent with dry weight of approximately 4 to 9.87, 1 to 4.73 and 1.20 to 3.25 respectively are shown in 1 the XRF data table.

Two samples of pottery recovered from the Islamic sites show different data, in which the range of dry weight silica is much higher 77-79% and calcium much lower 2.23-2.48%, a very low level of magnesium 0.03-1.185% and high sodium 4.85-5.47%, which is compared with other historic and prehistoric pottery. Sample ZR 8-369% has a small amount of 0.79% iron which has been compared with other samples. The average P2O5 percentage indicates that these containers were not used for organic material. Because of the high percentage of lime in pottery pieces, data shows that Sistan pottery is mainly manufactured by the calcareous clay as the main source for pottery. This data indicated the high level of lead in one of samples of Shahreh-Sukhteh (Sarhaddi-Dadian, 2013). Lead, as one of the dyeing factors, were added by potters to the old clay, and the archaeological studies suggest that lead was used as dyeing element in old social communication in the valley of India (Caleb, 1991). No lead element was detected in the prehistoric and historic pottery pieces. Therefore, there is no lead in historic and prehistoric eras and strengthens our claim that none of these two pieces of pottery belong to Sistan area.

5.3 Dispersion and Hierarchical Cluster Analysis

The Fig. 3 presents the percent values of SiO_2 and CaO in Excel to determine the dispersion of pottery.

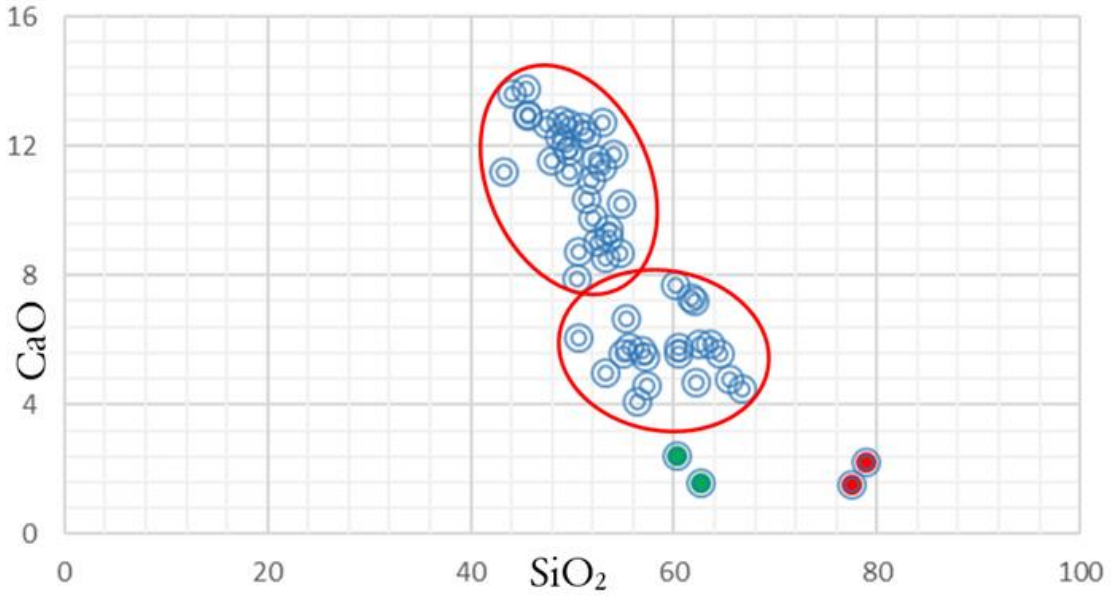

Figure 3: The graph presenting the dispersion of calcium oxide and silicon dioxide

The graph clearly indicates that the two pieces of pottery belonging to the Islamic era with numbers ZR3/332 and ZR 8/369 have different chemical composition that have been compared with the other pottery analyzed and indicated in red color at the bottom right of the graph. Both samples include a very high percentage of silica, sodium, low aluminum, calcium, iron and potassium. Therefore, compared with the other samples, it is suggested that the two samples have been imported from other parts or abroad. Two pieces of pottery marked in green on the graph also have a long association with the two other areas and probably with the imported pottery. The two red spots in the graph indicate the association of the pottery belonging to the prehistoric and historical eras in Sistan plain.

In Fig. 4, a dendrogram of HCA is drawn in terms of SiO_2 and CaO. Here the pottery pieces are classified into three categories A, B and C. In A category, the hierarchical cluster analysis includes the pottery with similar shapes. It is more likely that the pieces in this category are from nearby and related sites compared with the other two categories and it has a coefficient of above 5 which indicates that the pieces are likely to be local. Although group B, which contains the highest rate of the pieces, is believed to be the site for local pieces, it includes a combination of both imported pottery and pottery with its raw material brought from the surrounding areas since the coefficient is significantly below 5. Group C is also presented as containing the imported pottery in the diagram.

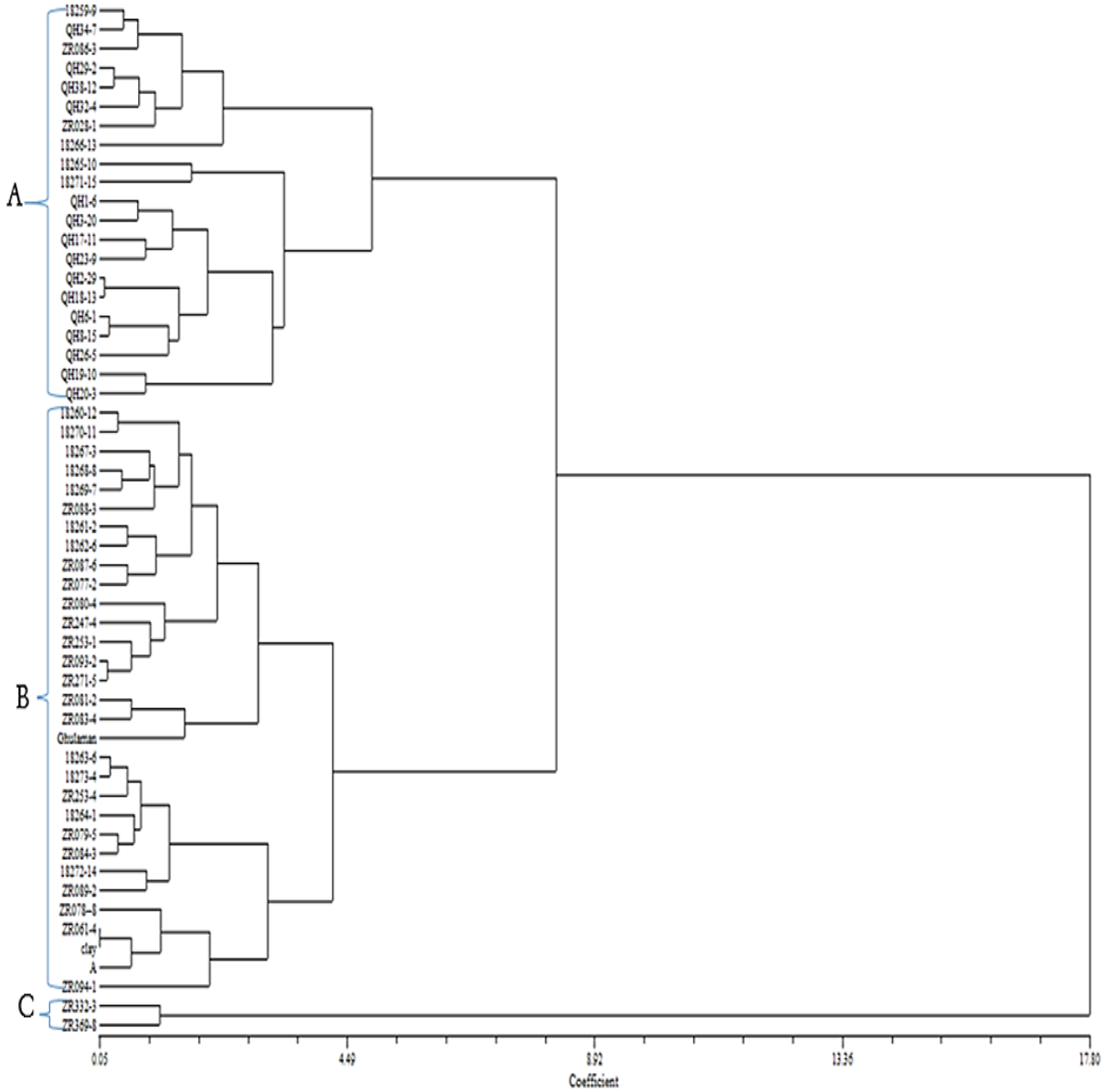

Figure 4: A dendrogram of HCA in terms of SiO_2 and CaO of the tested pieces

5. CONCLUSION

The results of the analysis show that most pieces of pottery found during the archaeological investigations in Sistan are indicative of similarity in the material used suggesting that these pieces are local. The XRD and XRF analysis data show that the two pieces of glazed pottery ZR 332-3 and ZR 369-8 recovered from two different sites have different chemical composition. Both pieces were compared with the other pieces and the high percentage of silicon in these pieces may be due to the glaze used; therefore, regarding the composition material, we recommended that these two pieces do not belong to this category and probably does not belong to Sistan. The piece of pottery entitled ZR 028-1 from the prehistoric era has a different composition compared with the other pieces. ZR 028-1 sample has a lower level of calcium and a higher level of potassium compared with the other pieces belonging to the historical and prehistoric era. Moreover, the XRD analysis determined the mineral substances of the pieces. The raw materials were distanced from the furnaces and they were probably at a distance of 6 to 10 km from the location of the raw materials. The analysis of the pieces found in Sistan shows that there used to be commercial activities in Sistan since the prehistoric era and these activities has continued to the Islamic era. The results also show that the local community had skills and specific knowledge in the field of making pottery from the prehistoric to the historical era in Sistan. The data are applicable in the field of archaeology and archaeometry and the association of this type of cultural item with important sites such as Shahdad and Espidezh can be examined. The pottery morphology of these areas helps us realize the cultural and commercial association of these civilizations.

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REFERENCES

Bater, M. (2010): Structural Studied of Fresco Pigments of Period Parti in Kuh-E Khwaja of Sistan. *Iranian Journal of Crystallography & Mineralogy* 18, no. 3

Bieber, A. M. Jr., D.W. Brooks, G. Harbottle, and E.V. Sayre. (1976): Application of multivariate techniques to analytical data on Aegean ceramics, *Archaeometry*, 18: 59–74.

Biscione, R. (1987): The Elusive Phase 2 of Shahr-I Sokhta Sequence. *South Asian Archaeology* (1987): 391-409.

Broekmans, T., A. Adriaens, and E. Pantos, 2008, Insights into the production technology of north Mesopotamian Bronze Age pottery, *Applied Physics A*, 90: 35–42

Chavalas, M. W. (1999): Ancient Persia from 550 BC to 650 AD. *History: Reviews of New Books* 27, no. 2:88-88.

Ferretti, M. 2000, x-ray fluorescent application for the study and conservation of cultural heritage, in Radiation in Art and Archaeometry(eds D.C. Creagh and D.A. Bradly), Elsevier, Amesterdam, pp 285-296.

Genito, B. (1990): The Most Frequent Tyoes at Dahan-E Gholaman (Sistan) and Their Spatial Variability. *South Asian Archaeology*, 588-601,

Golombek, L. (2003): The Safavid Ceramic Industry at Kirman. *Iran* 41: 253-70.

Haerinck, E. 1980. Twinspouted vessels and their distribution in the Near East from the Achaemenian to the Sasanian periods. *Iran* **18**: 43-54.

Lane, A (1947): Early Islamic Pottery. The Bernard Rackham Collection. Bernard Rackham was Keeper of the Department of Ceramics at the Victoria and Albert Museum from 1914 to 1938. The Bernard Rackham Collection consists of 68 books from his private library, some of which contain loose archive material such as letters and associated personal papers. The collection was given to Cardiff School of Art and Design as a series of donations from Bernard Rackham's family over a six year period from 2002 to 2008.

Lane, A. (1948): Late Islamic Pottey Persian, Syria, Egypt, Turkey. London: Faber and Faber.

Marghussian, A.K., H. Fazeli and H. Sarpoolaky, 2009. Chemical-mineralogical analyses and microstructural studies of prehistoric pottery from Rahmatabad, south-west Iran. Archaeometry, 51(5): 733-747. Piperno, M. and M.

Mehrafarin, R., M.S. Rustaee, and J. Moghadam. (2013): Sofale Daghdar, Sofale Shakhese dore Ashkani dar Sistan Iran" [The Burnishe Pottery, the Distinctive Pottery of Partian period in Sistan, Iran], *Iranian Journal of Archaeological Studies*, 4:6,120-105.

Mehrafarin, R., and S.R.M. Haji (2010): In Search of Ram Shahrestan the Capital of the Sistan Province in the Sassanid Era. *Central Asiatic journal* 54, no. 2: 256-72.

Mehrafarin, R., Musavi Hadji, S.R. and Feyz, S.A (2011): A Study of the Ceramics of East Iran during the Later Parthian Era with Special Reference to Tapa Gowri. *Central Asiatic journal* 55, no. 2: 240-58.

Milazzo, M., Radiation application in art and archaeometry. x-ray fluorescence application to archaeometry. possibility of obtaining non-destructive quantitative analysis, nuclear instruments and methods in physice Research B, 213, 683-692.

Moradi, H., Sarhaddi-Dadian, H. & Rahman, N. H. S. N. A. (2014): Development and Decline of the Bampur Valley, Based on the New Archaeological Evidence from Prehistoric Period. *Iranian Studies*, 47, no 2: 263-287.

Moradi, H., Sarhaddi-Dadian, H., Zuliskandar R., AND Nik Hassan Shuhaimi Nik Abdul Rahman. 2013a: Compositional Analysis of the Pottery Shards of Shahr-I Sokhta, South Eastern Iran. *Research Journal of Applied Sciences, Engineering and Technology* 6(4), 654-659.

Mousavi Haji, S. A., & Atai, M. 2010. The Study of a Collection of Potteries Samples from Sistan Ed. Zahedan: The Organization of Cultural Heritage, Handicrafts and Tourism of Sistan and Baluchesan.

Ramli, Z., N.H.S.N.A. Rahman and A.L. Samian. (2011): X-ray fluorescent analysis on Indo-Pacific glass beads from Sungai Mas archaeological sites, Kedah, Malaysia. *J. Radioanalytical Nuclear Chem*., 227: 741-747

Papageorgiou.I and Liritzis.I (2007) Multivariate mixture of normals with unknown number of components. An application to cluster Neolithic ceramics from the Aegean and Asia Minor. *Archaeometry,* 49, 4, 795-813.

Sajjadi, SMS, M. Casanova, L. Costantini, and KO Lorentz. (2008): Sistan and Baluchistan Project: Short Reports on the Tenth Campaign of Excavations at Shahr-I SokhtaIran: *journal of the British Institute of Persian Studies* 46: 307.

Salvatori, S., and M. Vidale. (1997): Shahr-I Sukhta 1975-1978: Central Quarters Excavations, Preliminary Report. Roma: *Istituto* Italiano per l'Africa e l'Oriente.

Sarhaddi-Dadian, H., Zohre Oveisi-Keikha, Vahid Purzarghan, (2017a) Introducing Troglodyte Architecture at Chabahar City in South-east of Iran, *International Journal of Archaeology*. Vol. 5, No. 1, 1-5. doi: 10.11648/j.ija.20170501.11

Sarhaddi-Dadian, H., Moradi, H,. Zuliskandar, R., and V. Purzarghan. (2017b): X-Ray Fluorescence Analysis of the Pottery Shards from Dahan-e Ghulaman, the Achaemenid Site in Sistan, East of Iran. *Interdisciplinaria Archaeologica* 8(1): Online First

Sarhaddi-Dadian, H., Morad, H., Soltani, M. (2015a): Preliminary Study of Rock Art at Negaran Valley in Baluchistan, Iran. *Rock Art Research* 32, no. 2: 8-11

Sarhaddi-Dadian, H., Zuliskandar, R., Nik Hassan Shuham, N. A. R., Mehrafarin, R. (2015b): X-Ray Diffraction and X-Ray Fluorescence Analysis of Pottery Shards from New Archaeological Survey in South Region of Sistan, *Mediterranean,Archaeology and Archaeometry*, Vol 15(3), 45-56

Sarhaddi-Dadian, H. (2013): Archaeology of Sistan, Iran during the Achaemenian Empire (from 550 BC to 331 BC). National University of Malaysia, Ph.D Thesis.

Scanlon, G.T. (1948): Mamluk Pottery: More Evidence from Fustat, In: Muqarnas Ii: An Animal on Islamic Art and Architecture, Oleg Grabar (Ed). . New Haven Yale University Press.

Sheikhakbari, S. Sarhaddi-Dadian, H., Amirhajloo, S., & Daneshi, A. (2015). Comparative Study of Pottery Industry in Zeh-Klout Historical Period in Roudbar-Kerman with Its Neighbouring Cultures Based on the New Archaeological Evidences. *International Journal of Archaeology*, 3(3), 22-32.

Tosi, M. (1983a): Excavations at Shahr-I Sokhta 1969-70. Prehistoric Sistan 1: 73-125.Tosi, M (1983b) Prehistoric Sistan. IsMEO, Vidale, M., and Tosi. M (1996): The Development of Wheel Throwing at Shahr-I Sokhta Slow and Fast Revolutions Towards Statehood. *East and West* 46, no. 3/4: 251-69.

Wong, E.H, C.A. Petrie, & H. Fazeli. (2010): Cheshmeh Ali ware: a petrographic and geochemical study of a transitional halcolithic period ceramic industry on the northern central plateau of Iran. *Iran XLVII*: 11-26

Vidale, M., & Tosi, M. (1996). The development of wheel throwing at Shahr-i Sokhta slow and fast revolutions towards statehood. *East and West,* 46(3-4):, 251-269.

Zuliskandar R and Nik Hassan Shuhaimi Nik Abdul Rahman. (2013): Composition Analysis of Ancient Bricks, Candi Bukit Kechil, BUjang Valley, Kedah. *Research Journal of Applied Sciences, Engineering and Technology* 6(5): 924-930

Zuliskandar R, Nik Hassan Shuhaimi Nik Abd. Rahman, Abdul Latif S and Mohd Ambar Y. (2013): X-Ray Diffraction and X-Ray Flourescence of Ancient Bricks of Candi Bukit Pendiat (Site 17), Bujang Valley, Kedah. *Research Journal of Applied Sciences, Engineering and Technology*, 6(06): 1094-1100.