



DOI: 10.5281/zenodo.18359

## ISLAND OF ANDROS POTTERY IN ARGILOS? ARCHAEOMETRIC STUDY USING $\mu$ -XRF AND MULTIVARIATE STATISTICAL ANALYSIS

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Received: 02/03/2015

Accepted: 28/05/2015

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### ABSTRACT

The present work constitutes the first two phases of the archaeometric study of the so-called "Siphnian" ware found in two different geographical regions in Greece, Argilos, in eastern Macedonia, and its metropolis in southern Greece, Andros island. Identification of the provenance of the Siphnian-type pottery found in the above regions is the principal question that has to be investigated and is of major importance, since socio-economic interactions between Argilos and Siphnos island are not verified, while there are only few published data about "Siphnian" ware.

For this purpose, non-destructive multi-element micro X-ray fluorescence spectroscopy was employed for the determination of the elemental composition not only of the above Siphnian-type ceramics, but of several other reference groups, which were recruited for comparison purposes. Furthermore, the analytical data were statistically treated using multivariate exploratory techniques (Principal Component Analysis, PCA and Cluster Analysis). As a result, useful provenance associations were derived from the statistical combination of the studied groups. More specifically, results indicate that the Siphnian-type pottery found in Argilos and Andros most probably have common provenance. However, it was proved that neither Argilos nor Andros is the origin of these ceramics, making the determination of their provenance a complicated task.

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**KEYWORDS:** *Provenance, Andros, Argilos, Siphnian pottery, Geometric period,  $\mu$ -XFR, multivariate statistics*

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## 1. INTRODUCTION

In most cases, the archaeological observation solely is not sufficient to determine the exact provenance of the finds. For this purpose, archaeometry plays a vital role, which, by means of the thorough physico-chemical analysis and characterization of the finds, complements the archaeologists' observations and allows the determination of the origin of the under-study materials or the distinction between excavated items of different sources (e.g. Peacock, 1970; Rice, 1987; Tykot, 2004; Papageorgiou and Liritzis, 2007).

To this respect, the present study constitutes the first two phases of the archaeometric investigation of the so-called "*Siphnian*" ware found at two different geographical sources in Greece, Argilos, in eastern Macedonia, and its metropolis in southern Greece, Andros island, identified to have occurred at the end of the third quarter of the 6<sup>th</sup> century b.c. (Bonias *et al.*, 2011). Compared to the major categories of ancient Greek pottery (e.g. the Attics, the Corinthian etc.) with historically known production workshops, Siphnian ware is less well known and more difficult to define (Butt, 1977).

The first Siphnian-type pottery brought to light was published by Brock and Mackworth Young (1949) who found a considerable number of vessels inside a sanctuary in Siphnos. Due to their abundance and location, the excavators attributed their origin to a local pottery workshop, while they gave detailed descriptions of their stylistic features.

Few years later, Boardman and Hayes (1966; 1973) published the excavation results at Tocra, a town in the Cyrenaica region of north-eastern Libya. Among the imported pottery, several finds were considered to originate from Siphnos due to their stylistic resemblance to those described by Brock and Mackworth Young (1949). The above were vases and potsherds, mainly from vessels intended to contain liquids (e.g. skyphoi, cups, lekanes and kotyles) (Bonias *et al.*, 2011). However, the results of the chemical analysis and the provenance study of the above ceramics were not conclusive, since, although the studied objects composed a homogeneous group, their Siphnian provenance could not be verified (Jones, 1986).

From a historical point of view, the presence of Siphnian pottery at sites of the ancient Near East is peculiar, since there is no information indicating that Siphnos had established any colonies in these sites (Bonias *et al.*, 2011). As already discussed, an impressive quantity of vases and fragments were unearthed recently on the ancient city of Argilos and Andros island, metropolis of Argilos (Bonias *et al.*, 2011), making the determination of their (common?) provenance a complicated mystery.

According to the above, the principal question concerns the identification of the source for the samples of Siphnian-type pottery found in the regions of Argilos and Andros. For this purpose, elemental analysis of samples from the pottery currently being studied took place by means of a non-destructive technique. A  $\mu$ -XRF spectrometer was employed for the chemical composition determination of the ceramic samples, which is considered quite advantageous compared to other techniques due to the coexistence of three significant features; it is a non-destructive multi-element technique, which makes it a very valuable and useful tool for the chemical analysis of archaeological finds, since their historical and cultural value is indisputable (Sakalis *et al.*, 2013), and the analysis requires less time and has a relatively lower cost than other similar techniques (Rice, 1987).

Finally, the acquired chemical data were further evaluated employing simple elemental biplots and by means of multivariate statistical analysis, namely principal component analysis (PCA) and cluster analysis. The above analysis allowed the classification of the samples into distinctive groups and provided feedback for further discussion about their provenance.

## 2. ARCHAEOLOGICAL CONTEXT

The archaeological site of the ancient city of Argilos is set on a hill known as Palaiokastro, on the edge of the Northern Aegean Sea about four kilometers west of the Strymona river delta (Figure 1). Excavations on the site were initiated in 1992 jointly by Greek and Canadian archaeologists. Argilos was founded in 655/654 B.C. and shortly became a prosperous city thanks to its trading activities in a region rich in gold and silver mines. Excavations have brought to light numerous finds demonstrating Argilos' interaction with the Thracian tribes, the Macedonians, the Persians, and other colonial Greeks in the region (e.g. Liampi, 2005; Perrault and Bonias, 2007).

Argilos city was established by Andros settlers, a Cycladic island in which well-organized towns (e.g., Zagora and Hypsele) were gradually developed during the Geometric period (towards the end of the eighth century B.C.). During the Archaic period, island "poleis" were created by the unification of communities which peacefully abandoned their previous settlements to found a new city. Such an example is Palaiopolis which probably received an influx of population from Zagora and Hypsele (Coldstream, 2003) (Figure 1).

Coldstream (2003) also notes that Andros is one of the islands which were less well explored and as a

result archaeologists were led to misinterpretations due to the incomplete data. For example, Zagora was considered to be the only major settlement on Geometric Andros, but this picture of the settlement pattern of Andros during the Geometric period was completely changed when the settlement at Hypsele was excavated in the last decades.

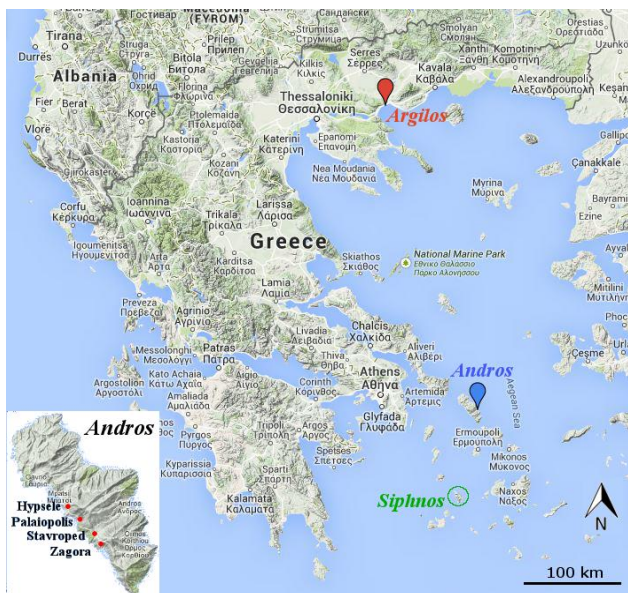


Figure 1. Geographical location of the studied areas

Since Argilos composes the oldest colony of Andros in the area, commercial transactions between the two regions cannot be doubted. On the other hand, no similar relationship between Argilos and Siphnos is evident. The above constitute the most important reason which puzzles the archaeologists and lead them to dismiss the theory that the Siphnian-type pottery found in Argilos and Andros has actually been manufactured in Siphnos (Bonias et al., 2011). In parallel, it seemed impossible to regard the Argilos finds as of local production, since then their presence in distant regions, such as Tocra, could not be explained (Bonias et al., 2011).

The archaeometric study of this specific material with advanced statistical techniques is expected to shed light on the provenance of the Siphnian-type pottery, for which published data are negligible. At the same time the network development among the studied areas constitutes a key issue waiting to be unveiled.

### 3. EXPERIMENTAL

#### 3.1 Instruments and methods

A portable and compact  $\mu$ -XRF spectrometer (SPECTRO, COPRA model) was employed for the  $\mu$ -XRF measurements. Detailed specifications of the instrument, its detection limits and how potential heterogeneity issues in the studied ceramics are

overcome can be found elsewhere (e.g. Papadopoulou et al., 2007; Sakalis et al., 2013). The operating conditions during the  $\mu$ -XRF measurements were high voltage 40 kV, current 0.75 mA and measurement time 5.0 min. The current and voltage values were chosen based on the results of a preliminary investigation regarding the presence and the intensity of the characteristic peaks.

One hundred and five (105) archaeological ceramic samples were analyzed in total by  $\mu$ -XRF. Three independent  $\mu$ -XRF measurements were performed in a point scan mode on different points on the clay body for each sample in order to achieve a statistically representative elemental characterization. The ceramic standard reference material SARM69 (MINTEK South Africa and Department of Geology, University of Free State) was used in the form of pellet for the calibration of the  $\mu$ -XRF spectrometer and the quantification of the detected elements in each sample. It must also be noted that SARM69 was also measured as a sample. A specialized software (WinAxil, Canberra, BENELUX) was used for spectra interpretation and data quantification. Finally, the mean value of the three concentrations for each element was calculated and considered as representative of the clay body chemical profile of each sample.

The chemical data were then statistically processed in order to determine similarities, differences and possible classification of the samples according to their provenance. For this purpose, biplots as well as multivariate statistics such as Principal Component Analysis (PCA) and Cluster Analysis were employed using suitable statistical software.

#### 3.2 Sample preparation

All samples were thoroughly cleaned and preserved through a suitable procedure during the archaeological treatment. Prior to the analysis of the clay body, a few  $\mu$ m of the external layer were carefully removed from an area of 0.5-1.0 mm (diameter) from the surface using a drill with a tungsten carbide cutter (Dremel®, mod. 395, type 5, code 32, The Netherlands), with a rotation speed of 10.000-33.000 rotations/min. The sample surface was then washed with double distilled water and allowed to dry before conducting the measurement. The above procedure was followed to achieve a flat, smooth and free of contamination measuring area in order to minimize interferences and undesired effects from surface anomalies and roughness.

### 4. RESULTS AND DISCUSSION

#### Phase I

The present study was conducted in two consecutive phases. The scope of the first phase (Phase I) was to investigate the potential connection between

ceramic samples found in Argilos and Andros. In Phase I, sixty-eight (68) samples were measured and statistically analyzed, consisted of forty-two (42) ceramic samples stylistically similar to the known as *Siphnian* pottery and twenty-six (26) reference samples certified as local production of Argilos area according to the archaeologists.

According to the archaeological observation, the aforementioned ceramic samples were classified as follows:

- **Group A:** Siphnian-type pottery found in Argilos
- **Group B:** Siphnian-type pottery found in Andros
- **Group C:** Corinthian pottery found in Argilos
- **Group E:** Local pottery from Argilos (reference group)

Samples of Group B have been excavated from three distinct regions in Andros, Hypsele (sample No 1-21), Palaiopolis (sample No 22-26) and Zagora (sample No 27-32). It should be noted that all ceramic sherds chosen for the archaeometric study are of the same archaeological period.

Group C consisted of thirteen (13) *Corinthian* ceramics found in Argilos and it was also analyzed for comparison purposes during the statistical analysis.

In Table 1, all measurements of Phase I (element concentrations) concerning the chemical composition of the clay bodies are presented for all samples. Thus, a multi-dimensional data set is created (81 measurements x 10 elements), demanding a multi-variation statistical interpretation in order to extract the maximum useful archaeometric information.

Using the chemical data of Table 1, Figures 2a-c are plotted, which are biplot graphs representing concentrations of the major and minor elements, namely Ca-Si, Ca-Fe and Mn-Ti respectively, for the clay body of all Phase I studied samples.

From Figure 2, some initial conclusions can be derived regarding the raw materials employed for the production of the studied samples, since they are directly linked to the chemical composition of the clay. These conclusions can also provide some first evidence about the potential connection between the studied Groups.

A first observation from Figures 2a and 2b is that all Corinthian samples found in Argilos (Group C) exhibit much higher concentration of Ca (>10%) compared to the rest of the samples, indicating the use of rich calcareous materials (Ca-rich clays) for their manufacture.

On the other hand, in the rest of the samples (Groups A, B and E) raw materials of relatively low Ca may have been used for their manufacture. It must be accentuated that in the case of the Siphnian-type samples found in Argilos (Group A) all ceramics present very similar Ca concentration indicating the use of one recipe for their manufacture. The above does not seem to be valid for Groups B and E, since relatively high dispersion in the Ca concentration is observed in both Groups.

In the same respect, from Figure 2a, it can be deduced that in almost all samples of Groups A, B and E, the concentration of Si is above 46% indicating that silicate raw materials have been used in all cases (Rice, 1987). However, it should be pointed that some of the samples of Group B have lower Si concentration than the rest of the samples of the same group, indicating different manufacturing methods or existence of various workshops.

As regards the Fe concentration in the clay paste of the samples, it is obvious from Figure 2b, that clay of all Siphnian-type pottery found in Argilos (Group A) exhibit almost the same value (~7%). On the other hand, samples of Siphnian-type pottery found in Andros (Group B) present variations in the Fe concentration, similar to the samples of Group E, which can constitute further evidence that different clay sources may be used for these ceramics.

Interesting are the findings when other less important major element concentrations such as Mn and Ti are also taken into consideration. From Figure 2c, one can readily observe that in the case of Group A samples, a significant homogeneity in concentration of both Ti and Mn exists. On the other hand, for the samples of Group B, considerable variations are observed in the concentrations of both elements, which further enhances the above statement of different clay sources used.

**Table 1. Clay body elemental chemical composition of all samples considered in Phase I of the present study by means of portable  $\mu$ -XRF analysis**

	Sample number	CaO %w/w	TiO <sub>2</sub> %w/w	Cr ppm	MnO %w/w	Fe <sub>2</sub> O <sub>3</sub> %w/w	Ni ppm	Zn ppm	K <sub>2</sub> O %w/w	Cu ppm	SiO <sub>2</sub> %w/w
Group A	4516	5.37	0.64	566.67	0.24	8.30	493.33	286.67	3.43	44.00	65.00
	8767	4.43	0.65	533.33	0.25	7.70	520.00	240.00	3.83	56.67	61.67
	9356	4.87	0.55	433.33	0.18	7.27	533.33	210.00	3.10	40.00	52.33
	10082	3.70	0.62	500.00	0.21	7.63	460.00	266.67	3.47	40.00	56.00

	10086	3.40	0.57	500.00	0.20	6.90	460.00	213.33	3.07	50.00	50.33
	11068	3.47	0.54	486.67	0.20	6.83	490.00	230.00	3.33	50.00	52.00
	11070	4.33	0.57	533.33	0.18	7.30	533.33	256.67	3.30	46.67	53.33
	11593	3.00	0.59	466.67	0.18	7.23	433.33	213.33	3.23	36.33	50.67
	14528	3.93	0.59	500.00	0.22	7.17	376.67	240.00	3.30	43.33	53.00
	14741	4.43	0.62	500.00	0.24	7.60	350.00	253.33	3.47	36.67	55.33
Group B	1	7.97	1.09	200.00	0.20	9.87	206.67	93.33	3.97	32.00	56.00
	2	5.40	0.98	193.33	0.18	9.13	150.00	100.00	3.32	40.67	47.33
	3	7.23	1.10	226.67	0.19	10.43	230.00	160.00	4.00	33.00	51.00
	4	9.87	1.11	783.33	0.15	10.47	766.67	86.67	1.60	55.00	42.33
	5	3.43	0.83	873.33	0.35	8.97	666.67	303.33	3.28	33.00	60.00
	6	4.23	0.77	833.33	0.25	9.00	766.67	370.00	2.76	41.00	52.00
	7	3.77	0.76	843.33	0.28	9.57	766.67	333.33	3.57	39.00	59.33
	8	2.76	0.69	716.67	0.23	8.10	600.00	290.00	3.30	34.33	54.67
	9	3.70	0.70	776.67	0.24	8.87	700.00	300.00	3.29	35.33	51.67
	10	1.15	1.00	190.00	0.07	10.40	600.00	110.00	4.03	63.33	50.00
	11	4.40	0.67	500.00	0.19	9.07	1366.67	366.67	2.25	50.00	49.00
	12	3.43	0.67	466.67	0.27	8.90	1366.67	343.33	2.65	56.67	52.33
	13	4.27	0.59	433.33	0.26	8.80	1566.67	346.67	3.00	66.67	53.00
	14	2.78	0.67	500.00	0.20	9.83	1633.33	400.00	1.46	60.00	43.33
	15	5.57	0.56	433.33	0.23	7.93	1233.33	346.67	2.39	43.33	49.00
	16	5.50	0.83	180.00	0.17	10.63	533.33	120.00	3.00	43.33	44.00
	17	3.43	0.63	500.00	0.21	8.80	1400.00	350.00	3.27	53.33	53.33
	18	4.43	0.73	500.00	0.20	8.90	1433.33	363.33	2.97	43.33	56.67
	19	3.70	0.70	533.33	0.27	10.50	1666.67	406.67	2.55	60.00	51.67
	20	3.93	0.77	533.33	0.18	10.63	1333.33	430.00	1.61	56.67	50.33
	21	2.00	0.58	400.00	0.27	7.53	1200.00	296.67	2.73	46.67	50.00
	22	3.67	0.80	933.33	0.18	9.90	233.33	333.33	1.92	42.67	48.33
	23	1.50	0.63	733.33	0.18	8.77	200.00	343.33	3.00	42.33	43.00
	24	3.77	0.79	866.67	0.21	8.77	200.00	336.67	2.70	41.00	53.00
	25	5.40	0.56	700.00	0.26	7.50	233.33	306.67	2.90	46.33	50.67
	26	8.00	0.72	866.67	0.32	10.27	300.00	336.67	1.98	52.00	52.00
	27	4.23	0.85	1000.00	0.28	11.20	200.00	466.67	1.50	51.33	47.67
	28	3.43	0.73	733.33	0.22	8.47	200.00	306.67	2.97	44.67	51.67
	29	4.00	0.75	700.00	0.27	9.07	200.00	400.00	3.37	46.67	58.33
	30	4.13	0.73	866.67	0.31	10.20	266.67	400.00	2.57	53.00	54.33
	31	4.33	0.84	833.33	0.27	9.47	300.00	400.00	2.38	52.00	49.00
	32	3.37	0.76	866.67	0.16	10.30	300.00	433.33	1.73	51.00	44.33
Group C	18b	10.33	0.93	246.67	0.11	8.50	200.00	100.00	2.80	56.67	43.00
	235	15.33	0.98	273.33	0.15	9.53	266.67	73.33	2.40	83.33	48.67
	2259	11.67	0.76	200.00	0.12	6.96	233.33	73.33	2.45	53.33	41.67
	2566	17.00	0.91	230.00	0.15	8.77	200.00	100.00	2.42	76.67	49.00
	2567	16.67	0.88	240.00	0.13	8.17	213.33	96.67	1.96	63.33	49.33
	7101	17.33	0.82	200.00	0.18	7.77	266.67	90.00	2.40	60.00	43.67
	8248	18.33	1.04	233.33	0.14	9.10	200.00	103.33	3.06	60.00	55.33
	9071	14.33	0.99	266.67	0.14	9.23	200.00	110.00	3.20	60.00	48.00
	9225	21.00	0.78	226.67	0.12	7.33	233.33	100.00	0.98	73.33	41.33
	11166	19.33	1.05	300.00	0.18	10.70	333.33	126.67	3.23	43.33	51.00
	11323	15.67	0.91	333.33	0.12	9.23	266.67	110.00	0.73	76.67	41.33

	11712	15.67	0.93	196.67	0.13	8.33	200.00	96.67	2.93	53.33	44.00
	15234	15.67	1.00	333.33	0.16	9.37	300.00	113.33	3.20	70.00	54.00
Group E	04R	2.08	1.30	75.00	0.07	8.00	50.00	130.00	3.53	60.00	62.50
	05R	1.58	1.22	63.33	0.09	7.80	90.00	146.00	3.52	47.50	62.20
	06R	9.80	1.00	96.00	0.10	8.40	70.00	90.00	2.20	67.50	70.00
	07R	2.10	1.10	65.00	0.08	8.00	200.00	160.00	3.44	55.00	72.00
	08R	2.26	1.28	65.00	0.07	11.20	90.00	126.00	3.52	74.00	54.60
	09R	1.68	1.08	52.50	0.07	7.60	48.00	92.00	3.16	55.00	49.80
	10R	1.34	1.24	220.00	0.11	5.76	150.00	132.00	2.64	48.00	80.00
	11R	7.14	1.16	160.00	0.11	9.60	192.00	122.00	3.18	80.00	49.80
	12R	8.00	1.24	114.00	0.17	9.20	86.67	150.00	2.62	68.00	50.00
	13R	6.02	1.42	168.00	0.19	11.20	85.00	162.00	3.56	72.00	68.00
	14R	6.12	1.44	140.00	0.12	10.00	70.00	108.00	3.46	92.50	68.00
	15R	2.36	1.04	74.00	0.08	8.20	51.00	96.00	3.48	60.00	60.20
	16R	3.40	0.78	72.50	0.07	6.18	56.00	80.00	2.80	62.50	55.20
	17R	1.28	0.88	50.00	0.03	6.20	49.00	92.00	3.00	56.00	55.80
	18R	1.72	1.10	70.00	0.04	8.20	70.00	82.00	4.30	60.00	72.00
	19R	10.60	0.58	156.00	0.16	8.40	172.50	96.00	2.76	86.00	58.00
	20R	3.38	1.50	138.00	0.13	12.20	93.33	128.00	2.76	122.00	68.00
	21R	3.52	1.26	77.50	0.09	10.40	47.00	134.00	3.76	82.00	70.00
	22R	6.04	0.84	40.00	0.07	6.42	110.00	112.00	2.50	102.00	50.00
	23R	1.66	1.12	77.50	0.05	9.60	50.00	130.00	3.08	76.00	62.00
24R	4.18	1.16	108.00	0.04	9.20	85.00	124.00	3.50	80.00	72.00	
25R	3.72	1.22	80.00	0.06	9.20	70.00	158.00	3.64	60.00	70.00	
27R	1.45	1.13	82.50	0.06	9.25	52.00	142.50	3.80	110.00	65.00	
28bisR	1.77	0.91	103.33	0.07	6.90	200.00	96.00	2.78	37.50	57.20	
29R	5.50	1.06	105.00	0.13	7.80	160.00	118.00	2.68	95.00	68.00	
30bisR	1.96	0.94	287.50	0.04	6.12	300.00	80.00	2.92	36.00	67.40	

From the same Figure (Figure 2c), it is worth noticing that taking into account the Mn and Ti concentrations of the clay paste, Siphnian-type pottery found both in Argilos and Andros is differentiated from the reference group from Argilos (Group E), which was not apparent in Figures 2a and 2b. In the case of Group E samples, materials of high Ti and low Mn concentration have been used for their manufacture.

The PCA and cluster analysis of the clay bodies of the whole material of Phase I are presented in Figures 3 and 4 respectively. Results in both PCA and cluster analysis are similar. More specifically, samples are classified into four (4) distinct chemical groups based on the chemical composition of their clay body, which are in agreement with the archaeological observation. However, it is also apparent (mainly in Figure 4) that similarities between few samples of Groups C and E exist, while Siphnian-type samples of Group B could be further divided into two subgroups.

Of course, the above classification of the samples in these four chemical groups cannot constitute conclusive evidence about their different provenance. Yet, it is indicative of different manufacturing processes used or different workshops even from the same geographical area, the latter however demanding additional evidence to support it.

The above chemical group classification as deduced by the PCA and cluster analysis shown in Figures 3 and 4 can be explained by the chemical data of the clay body of the samples.

From Figure 2 and Table 1, it is apparent that the clay body of the samples in Group E presents relatively higher Si concentration than the rest of the samples, while the main attribute of their chemical composition is the much higher Ti and lower Mn concentration compared to the samples of the other groups.

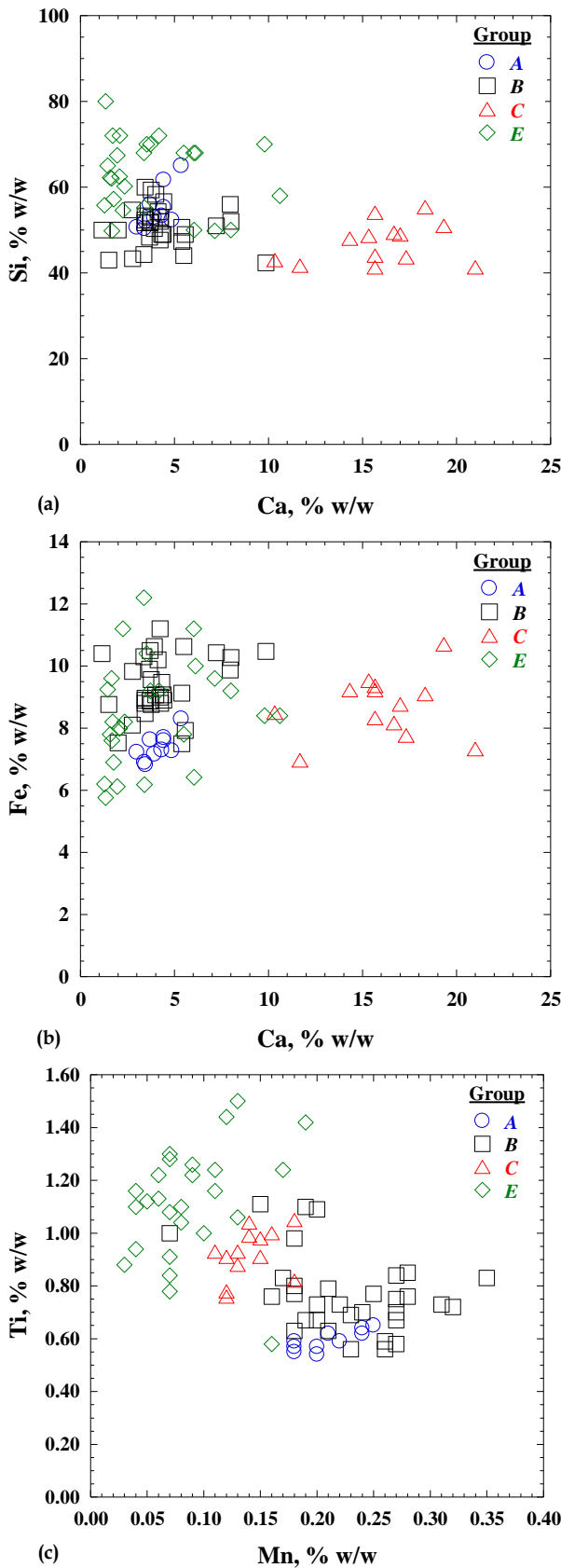


Figure 2. Binary plots for the clay pastes of samples studied in Phase I: (a) Ca-Si, (b) Ca-Fe and (c) Mn-Ti.

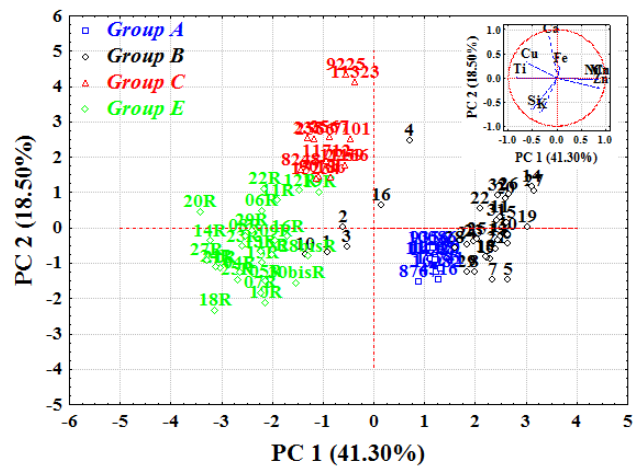


Figure 3. Plot of the two first principal components obtained through PCA of the chemical data from the clay bodies of the ceramic samples of Phase I and the relative loadings (inset).

In this respect, the Corinthian samples found in Argilos (Group C) is completely differentiated from the rest of the samples due to its unique elemental composition in most of the elements detected, as already discussed (Figure 2 and Table 1), rendering them a clearly distinct group. The homogeneity of Group C (Figure 3) should also be highlighted at this point to amplify its uniqueness, since it composes ceramics of one of the major ancient Greek pottery categories (i.e., the Corinthian) of distinctive style and clay quality and with historically known production workshops.

Another significant observation from Figures 3 and 4 is the homogeneity of the other separate chemical groups as well. Group A is characterized by a remarkable homogeneity among its samples based on the chemical composition of their clay paste. The above can serve as evidence that all ceramics of Group A are probably manufactured from the same workshop.

In this regard, the relatively high homogeneity observed among samples of Group E, which are considered to be local production in Argilos area, may support the statement that they were also manufactured in the same workshop.

On the other hand, although the homogeneity of Group B cannot be doubted, several exceptions are observed (Figures 3 and 4), namely samples 1, 2, 3, 4, 10 and 16 which are separated from the rest of the samples of this Group. This could mean that samples of this group may have been manufactured in different workshops of the same geographical region, which however demands additional evidence to justify it (e.g. more reference samples). It should also be accentuated that the above samples (1, 2, 3, 4, 10 and 16) were all found in the Hypsele area.



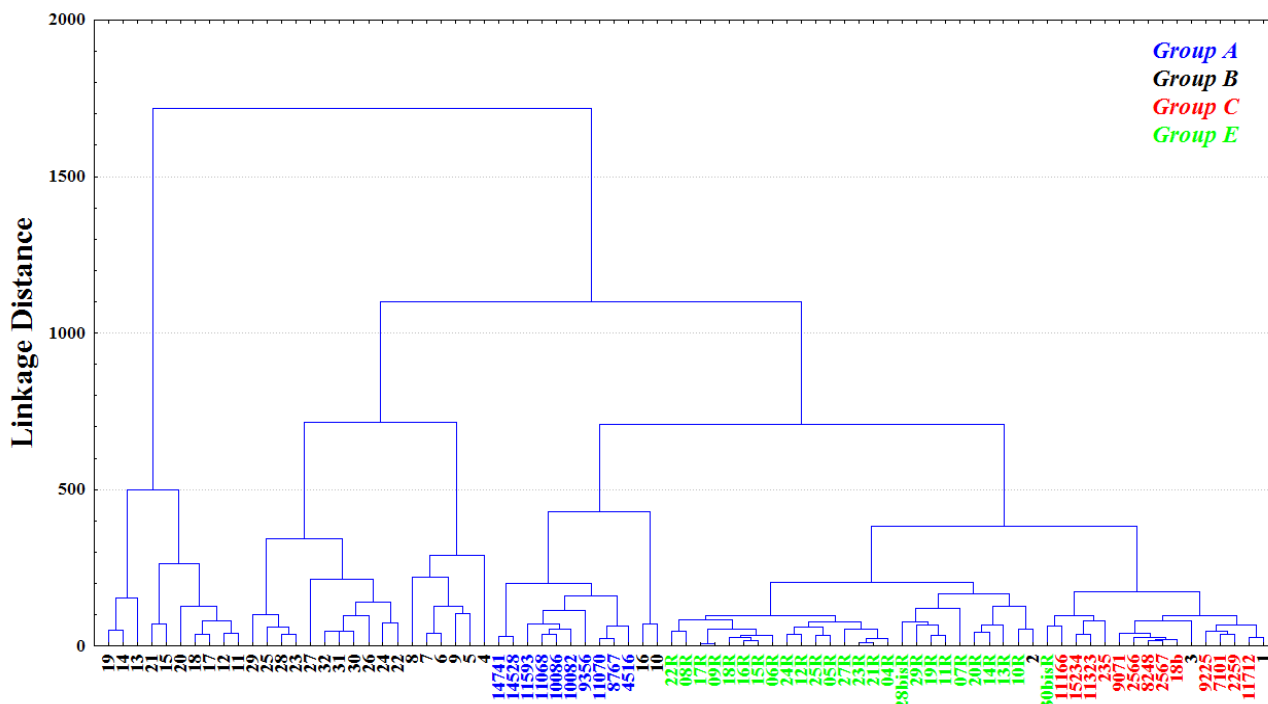


Figure 4. Comparison between pottery found in Argilos (Groups A and C), in Andros (Group B) and reference from Argilos (Group E) by means of Cluster analysis (Complete Linkage, Euclidian Distances)

In addition, from Figure 3, interesting is the observation that the chemical composition of Group A shows more similarities with that of Group B rather than with that of the Argilos local pottery (reference Group E), from which it is utterly differentiated. Group E constitutes an independent chemical group exhibiting no similarities with the rest of the samples. Moreover, it seems that the Siphnian-type pottery (Groups A and B) constitute a wider group based on the chemical composition of the clay, although they were found in completely different regions; Group A in Argilos and Group B in Andros, serving as an indication of the common provenance of these ceramic samples.

According to the above, provenance of samples found in Argilos (Group A) is questioned, while it seems that they have not been manufactured in Argilos, but they could be a product of Andros due to their elemental composition similarities with Group B.

The above scenario constituted the case study of Phase II, i.e. to investigate using chemical and statistical analysis whether Siphnian-type pottery found in Argilos (Group A) were actually manufactured in Andros.

## Phase II

During Phase II, in order to further investigate the provenance of the Siphnian-type pottery found in Argilos, a new reference group was taken into account in the analysis, namely twenty-four (24) ce-

ramic samples verified as local production of Andros by the archaeologists (e.g. Liritzis, 2007), which was compared with Groups A and B of the previous Phase.

It should be mentioned that these samples are situated in the late Geometric period, the late Hellenistic and Roman period, according to the archaeological observation, meaning that they are not contemporary to the ceramics of Groups A and B. The above samples were further classified into seven (7) archaeological groups, based on where they were found on Andros:

- **Group G1:** Andros local pottery found in *Hypsele*
- **Group G2:** Andros local pottery found in *Hypsele*
- **Group Z:** Andros local pottery found in *Palaiopolis*
- **Group H:** Andros local pottery found in *Palaiopolis*
- **Group J:** Andros local pottery found in *Palaiopolis*
- **Group I:** Andros local pottery found in *Stavropeda*
- **Group K:** Andros local pottery found in *Aprovatou (north of Hypsele)*

It should be noted that ceramics of Groups G1 and G2 and Groups Z, H and J, were found in different sections of the same excavation site (G1 and G2 in *Hypsele* and Z, H and J in *Palaiopolis*).



The same procedure with Phase I was followed and the chemical composition of the clay paste of the above samples are presented in Table 2.

Using the chemical data of Table 2, the PCA and the cluster analysis of the groups are plotted in Figures 5 and 6 respectively, in order to examine whether the archaeological classification of the above groups complies with the chemical composition of the clay body as well and to extract more information about their provenance.

From Figure 5, it is apparent that the studied samples exhibit similarities concerning the elemental composition of their clay paste and it could be regarded that they all compose one chemical group, although they could be further divided into several sub-groups. However, several exceptions are observed, such as FOL1\_A (Group G1), FOL2\_D

(Group G2), PAL1\_B (Group H), ST3 (Group I) and AP1 (Group K), which are only slightly correlated with the rest of the material.

It should also be noted that dispersion to a great extent among samples of the same archaeological group is observed in all Groups.

In a similar way, from Figure 6, one can readily observe that two major chemical groups are present, but with relatively small difference between them concerning the chemical composition of their clay body. Several sub-clusters with high correlation among them are also observed. According to the above, it could be concluded that all samples have common provenance, consistent with the archaeological observation, manufactured in more than one workshops of Andros though.

**Table 2. Clay body elemental chemical composition of Andros local production samples considered in Phase II of the present study by means of portable  $\mu$ -XRF analysis**

	Sample number	CaO %w/w	TiO <sub>2</sub> %w/w	Cr ppm	MnO %w/w	Fe <sub>2</sub> O <sub>3</sub> %w/w	Ni ppm	Zn ppm	K <sub>2</sub> O %w/w	Cu ppm	SiO <sub>2</sub> %w/w
G1	FOL1_A	2.67	2.50	266.67	0.12	18.53	235.00	95.00	0.86	50.00	51.00
	FOL1_B	1.22	1.53	266.67	0.04	15.40	336.67	123.33	4.30	83.33	66.33
G2	FOL2_A	1.40	1.17	333.33	0.21	12.60	180.00	100.00	4.03	50.00	64.33
	FOL2_B	1.20	1.01	466.67	0.06	16.70	340.00	146.67	2.28	60.00	63.33
	FOL2_C	6.13	1.37	200.00	0.22	13.07	245.00	136.67	3.37	50.00	60.67
	FOL2_D	9.20	1.40	766.67	0.16	12.83	896.67	140.00	1.88	93.33	52.33
Z	PAL1_A	2.07	1.30	176.67	0.17	11.67	235.00	126.67	3.47	56.67	67.67
	PAL1_B	8.93	1.13	900.00	0.31	14.07	250.00	110.00	1.95	60.00	55.33
	PAL1_C	3.50	1.13	333.33	0.30	12.50	506.67	146.67	3.21	55.00	60.00
H	PAL2_A	0.65	1.50	190.00	0.11	10.63	205.00	90.00	2.32	50.00	55.00
	PAL2_B	2.10	1.33	400.00	0.13	13.20	436.67	133.33	1.94	66.67	55.33
	PAL2_C	1.67	1.47	400.00	0.16	16.73	376.67	150.00	2.16	76.67	51.00
J	PAL3_A	2.60	1.33	186.67	0.13	9.53	253.33	163.33	4.87	60.00	64.33
	PAL3_B	2.33	1.67	366.67	0.27	14.70	306.67	210.00	3.39	60.00	60.00
	PAL3_C	2.67	1.23	240.00	0.08	14.50	205.00	130.00	3.37	63.33	56.67
	PAL3_D	1.40	1.80	266.67	0.17	14.03	330.00	140.00	4.90	43.33	69.33
I	ST1	0.62	1.23	260.00	0.05	13.07	180.00	100.00	2.04	60.00	45.67
	ST2	2.73	1.00	75.00	0.24	9.10	165.67	100.00	1.67	40.00	53.67
	ST3	10.03	1.30	300.00	0.21	12.23	713.33	93.33	2.11	53.33	65.00
	ST4	5.53	1.23	366.67	0.14	11.90	793.33	123.33	3.20	35.00	56.67
K	AP1	4.77	1.07	666.67	0.10	10.10	500.00	100.00	3.97	30.00	57.67
	AP2	3.00	1.50	163.33	0.13	12.57	290.00	123.33	3.87	36.67	58.33
	AP3	1.09	1.07	270.00	0.12	15.07	255.00	100.00	4.43	63.33	59.00
	AP4	2.47	1.93	466.67	0.33	19.50	550.00	160.00	2.42	60.00	56.33

The next step of Phase II was to compare the Siphnian-type pottery found in Andros (Group B) with the reference samples of Andros (Groups G-K

and Z). For this purpose the corresponding PCA and cluster analysis are constructed and presented in Figures 7 and 8 respectively.

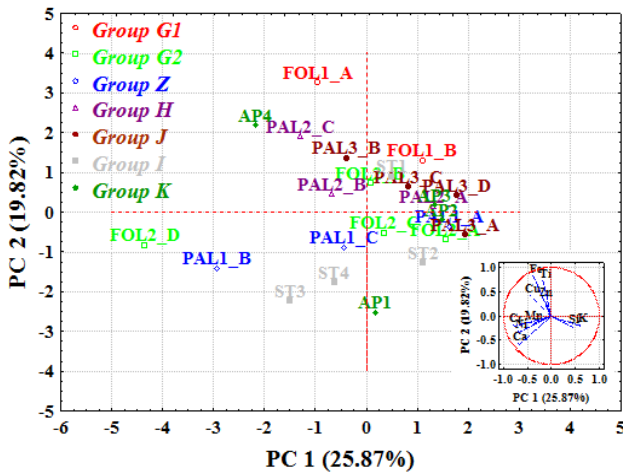


Figure 5. Plot of the two first principal components obtained through PCA of the chemical data from the clay bodies of the ceramic samples of Andros local pottery (Groups G-K and Z) and the relative loadings (inset).

An observation of major importance from Figure 7 is that samples tend to be classified into two distinctive chemical groups; one consisting of the samples of Group B (on the right side of the PCA, with few exceptions) and the other composing the samples of the rest of the groups (on the left side of the PCA, Groups G-K and Z).

Homogeneity of Group B is high with only exceptions samples 1, 2, 3, 4, 10 and 16, which are differentiated from the rest of the samples of the same group and seem to be correlated, based on the chemical composition of the clay paste, with the reference samples of Andros (Groups G-K and Z). In the same respect, samples FOL2\_D (Group G2) and PAL1\_B (Group H) are slightly separated from the rest of the reference pottery of Andros.

In the same respect, the cluster analysis (Figure 8) demonstrates that the reference samples of Andros are differentiated from the Siphnian-type pottery found there (Group B), the latter being divided into two sub-clusters. However, as also observed in the corresponding PCA (Figure 7) few samples of Group B exhibit more similarities with the reference ceramics rather than with the rest of the samples of the same Group. In a similar way, samples FOL2\_D (Group G2) and PAL1\_B (Group H) tend to cluster with samples of the Group B.

The above can be regarded as evidence of the different provenance of the Siphnian-type pottery found in Andros, since they are chemically completely differentiated from the Andros local pottery.

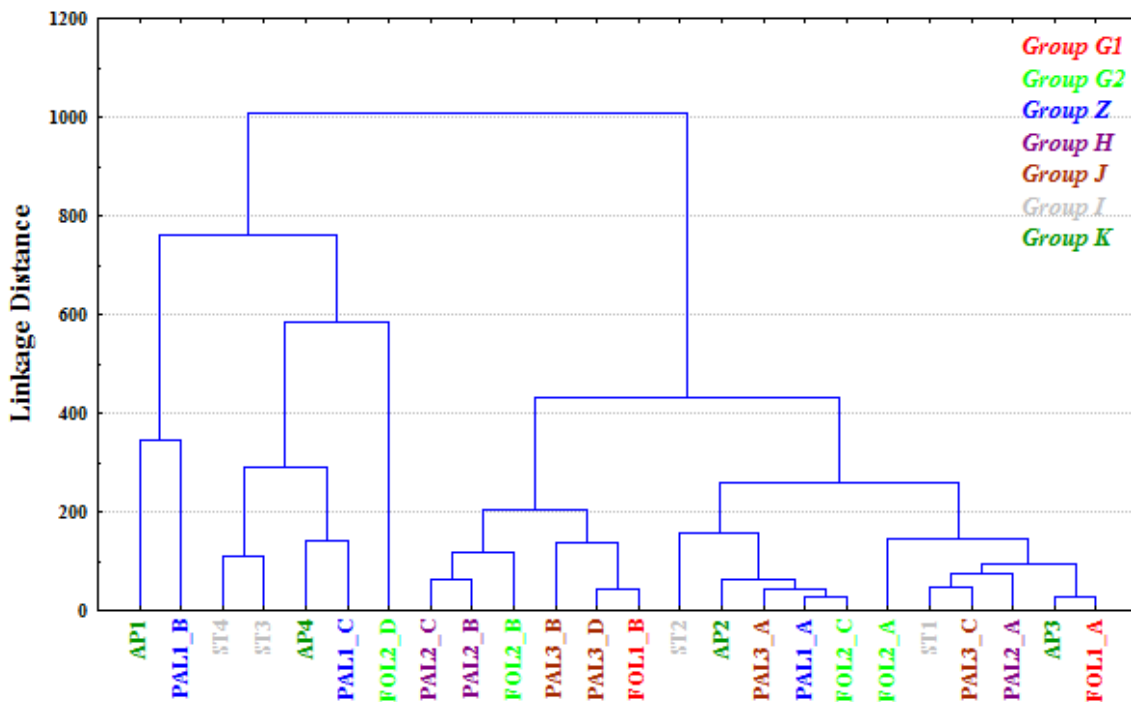


Figure 6. Comparison between samples considered as local production of Andros (Groups G-K and Z) by means of Cluster analysis (Complete Linkage, Euclidian Distances)

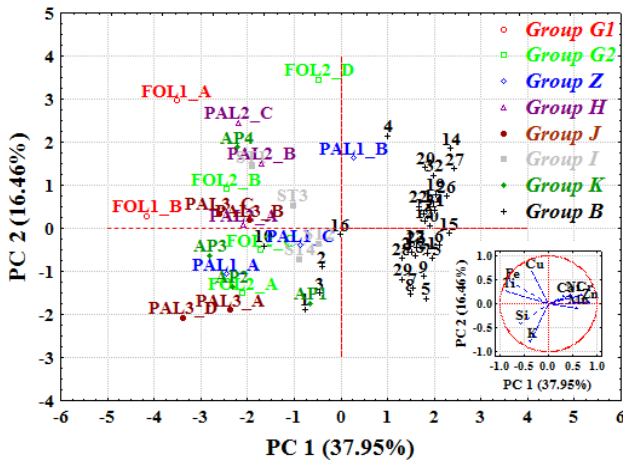


Figure 7. Plot of the two first principal components obtained through PCA of the chemical data from the clay bodies of the ceramic samples of Andros local pottery (Groups G-K and Z) and the Siphnian-type pottery found in Andros (Group B) and the relative loadings (inset).

In the final step of Phase II, the chemical data of Group A samples (Siphnian-type pottery found in Argilos) are also taken into consideration in the statistical analysis. For this purpose, biplot graphs representing concentrations of Ca-Fe and Mn-Ti respectively for the clay body of all Phase II studied samples are plotted (Figures 9a-b).

From Figure 9, several conclusions can be extracted regarding the raw materials employed and the recipes used for the manufacture of the studied samples in Phase II, which will allow to justify differentiations observed among the Siphnian-type pottery from Argilos (Group A) and Andros (Group B) and the local pottery from Andros (Groups G-K, Z).

A first observation from Figure 9a is that the majority of the samples of the Andros local production are characterized by significantly low calcareous clay, since in most of them the concentration of Ca is lower than 3%. However, the high dispersion of the samples based on the Ca concentration of the clay paste is indicative of the use of different clay sources in Andros. On the other hand, as already discussed, Ca concentration in the clay body of the Siphnian-type pottery of Argilos and Andros is definitely more than 3%, but still falls into the low calcareous category (Rice, 1987).

Figure 8. Comparison between samples considered as local production of Andros (Groups G-K and Z) and the Siphnian pottery found in Andros (Group B) by means of Cluster analysis (Complete Linkage, Euclidian Distances)

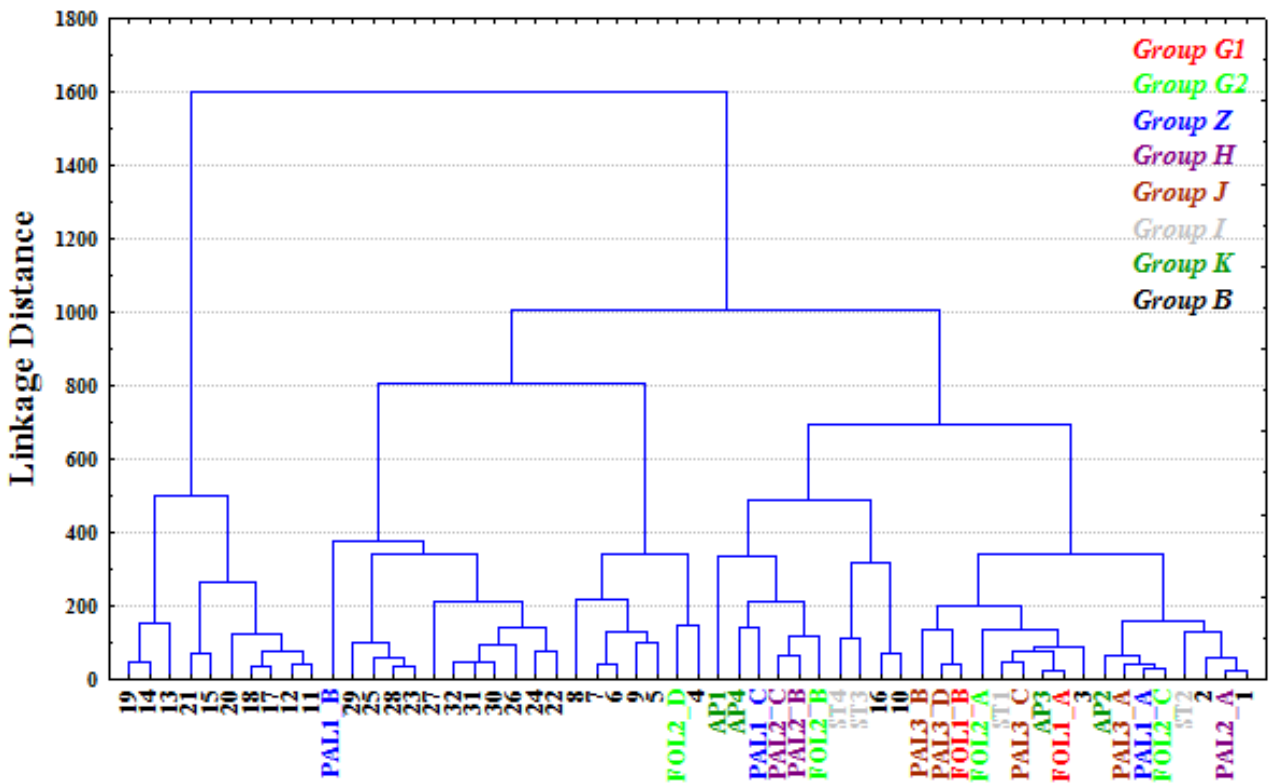


Figure 8. Comparison between samples considered as local production of Andros (Groups G-K and Z) and the Siphnian pottery found in Andros (Group B) by means of Cluster analysis (Complete Linkage, Euclidian Distances)

In the same respect, from Figure 9a one can readily notice the difference in the Fe concentration of the clay body between the Siphnian-type pottery (Groups A and B) and the Andros local production

(Groups G-K, Z). With few exceptions, the high Fe concentration of the samples from Andros is worth mentioning, since it is above 10% and in some cases approaches even ~20%, while the Siphnian-type pot-

tery have been manufactured with raw materials containing Fe below 10%. The heterogeneity of the samples of Andros local pottery concerning the Fe concentration should be highlighted, contrary to the relatively high homogeneity of the samples of both Group A and Group B.

When Mn and Ti concentrations of the clay paste are also taken into consideration (Figure 9b), the above differentiation of Groups A and B with the samples from Andros is further enhanced. The main attribute of the clay body of all Andros samples (Groups G-K, Z) is the relatively high Ti concentration (above ~1%) while the corresponding concentration in the majority of the Siphnian-type samples lies in the range 0.5-0.8%.

As regards the Mn concentration, it seems that at least two different recipes might have been used for the production of the Andros local pottery, i.e., those with clay paste Mn concentration lower than 0.15% and those higher than that. Once again, the high dispersion of the Andros samples (Groups G-K, Z) observed in Figure 9b, provides further evidence of different recipes or workshops used for their manufacture.

The PCA of the Siphnian-type pottery found in Andros and Argilos and the reference samples from Andros is also produced in Figure 10.

From Figure 10, one can readily observe that samples tend to be classified into three major chemical groups; one composing the Groups G-K and Z (Andros local production) and the other two consisting of the Siphnian-type pottery Groups A (from Argilos) and B (from Andros) respectively. This chemical group classification as deduced by the PCA can be justified by the chemical data of the clay body of the samples. As already discussed, the main attributes of the reference samples clay paste are the higher Fe and Ti concentrations (Figure 9) compared to the rest of the materials presented in the PCA. As also observed in Figure 7, samples FOL2\_D (Group G2) and PAL1\_B (Group H) are slightly differentiated from the rest of the reference pottery of Andros.

The most significant finding is that the correlation between Groups A and B found in Figure 3 of Phase I of the present study is also found in Figure 10 of Phase II. Albeit these two Groups are chemically differentiated when compared with Groups G-K, Z, a close relationship between their clay paste chemical composition is also a fact. On the one hand, their provenance is called into question since they are chemically completely differentiated from local pottery of both Andros and Argilos. It seems that the Siphnian-type pottery studied in this work does not originate from Argilos nor from Andros, but from somewhere else. On the other hand, the similar clay composition of the samples of these Groups could

provide further evidence of their potential common provenance as imports in both cases (Argilos and Andros). In the same time, it seems that samples of Group A should indeed originate from a common workshop, due to its notable homogeneity.

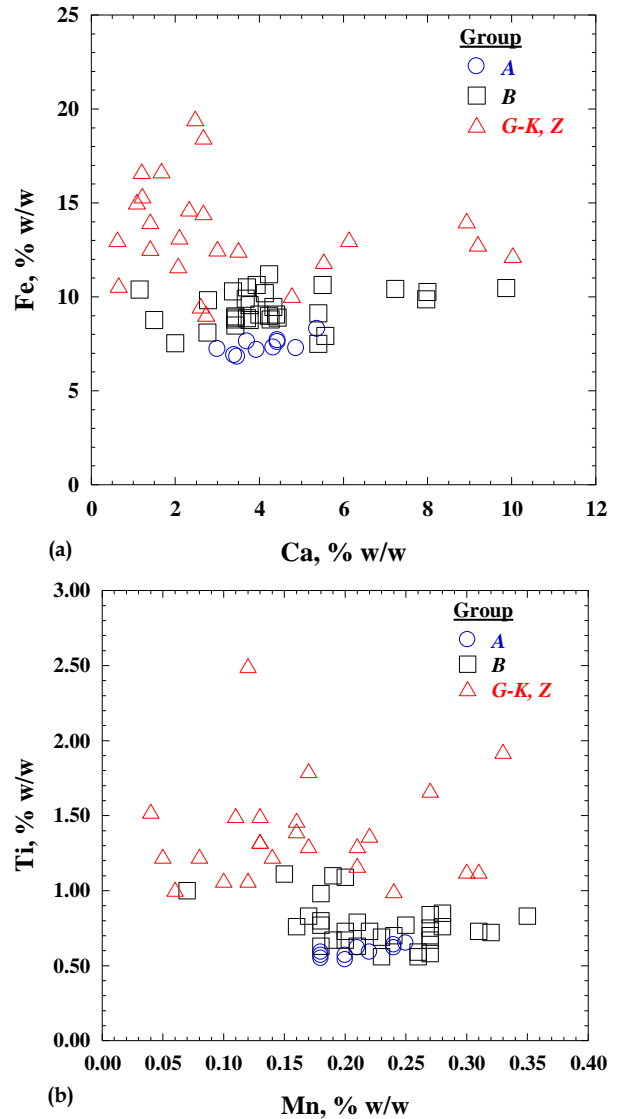


Figure 9. Binary plots for the clay pastes of samples studied in Phase II: (a) Ca-Fe and (b) Mn-Ti.

From Figure 10 it is also apparent that several samples of Group B tend to be differentiated from the rest of the ceramics of the same group, as previously discussed (Figures 3-4 and Figures 7-8). These samples are 1, 2, 3, 4, 10 and 16 and they tend to be merged into the chemical group composed by Groups G-K and Z (Andros local pottery). Thus, they could be attributed to the reference groups (G-K and Z) and be considered as possible case of Siphnian-type pottery originated from Andros.

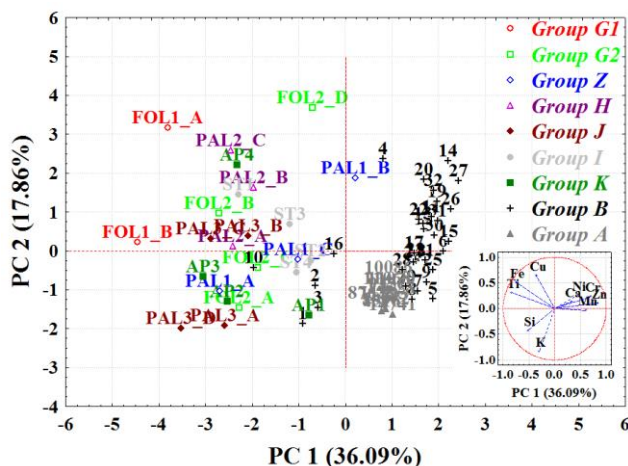


Figure 10. Plot of the two first principal components obtained through PCA of the chemical data from the clay bodies of the ceramic samples of Andros local pottery (Groups G-K and Z) and the Siphnian-type pottery found in Andros (Group B) and Argilos (Group A) and the relative loadings (inset).

## 5. CONCLUSIONS

In the present work an archaeometric study of samples of Siphnian-type pottery found in Argilos and its metropolis, Andros, took place in order to extract information about their provenance. For this purpose, several reference groups, namely ceramics of local production from Argilos and Andros and Corinthian samples found in Argilos were also studied for comparison, while the study was materialized in two Phases.

Chemical analysis of the clay bodies by means of  $\mu$ -XRF combined with statistical analysis of the chemical data revealed important information concerning the potential provenance and the technological characteristics of the studied pottery.

According to the chemical composition of the samples, the Siphnian-type pottery Groups (A and B) are chemically differentiated, but in close relationship to each other, an indication of possible common provenance. Nevertheless, according to the statistical

analysis of the chemical data, it is evident that samples of both groups have not been manufactured in Argilos.

The remarkable homogeneity of Group A samples should also be mentioned, indicative of the use of the same recipe or their manufacture from the same workshop.

In addition, results show that samples of Group E, which is considered to be local production in Argilos area, were probably manufactured in the same workshop, since they exhibit relatively high homogeneity as regards the clay chemical composition.

The above seems not to be valid for samples of Andros local pottery (Groups G-K, Z). Chemical data revealed their common provenance, but the high dispersion among the samples indicate that they are products of several workshops.

Finally, according to the statistical analysis, it seems that the Siphnian-type pottery found in Argilos and Andros has not been manufactured in Andros either, since their clay chemical composition exhibits significant variations from that of the Andros reference group as well. However, the different time period between the under study samples (Groups A and B) and the reference group (Groups G-K, Z) should also be taken into consideration before drawing concrete conclusions, since chemical composition of clay even of the same area can vary with time.

Future work, which is indeed in progress, will focus on comparison of the Siphnian-type pottery currently being studied with contemporary local pottery from Andros and with Siphnian-type pottery from other regions of Cyclades, and particularly Siphnos island, to shed light into their provenance. Further thorough archaeological and archaeometric study of specific exceptions, such as samples 1, 2, 3, 4, 10 and 16 of Group B, which at present can be regarded as Siphnian-type pottery originated from Andros, is also scheduled for the near future.

## ACKNOWLEDGEMENTS

The Authors wish to thank the archaeologists Dr. Zisis Bonias, Prof. Jacques Perreault, Dr. Stavros Paspalas and Dr. Christina Televantou for providing the samples of the present study.

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