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A COMPLETE CHEMICAL GROUPING OF THE PERLMAN/ASARO NEUTRON ACTIVATION ANALYSIS DATABANK ON MYCENAEAN AND MINOAN POTTERY

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The Perlman/Asaro databank contains the analytical data of about 900 samples of Mycenaean and Minoan ceramics, which were measured by neutron activation analysis at Berkeley in the early 1970's. Several studies on parts of these data were carried out and published until now, each focussed on pottery from particular regions. We will present part of the results of a statistical grouping of the whole databank, according to the statistical procedures utilized in Bonn. The established new chemical reference patterns agree in all applicable cases with the corresponding groups in the Bonn databank and mutually confirm the NAA results.

KEYWORDS : NAA, CERAMICS, PROVENANCING, BRONZE AGE, MYCENAEAN, MINOAN

INTRODUCTION

About 30 years ago the archaeometry work group in Berkeley established Neutron Activation Analysis (NAA) as a method for provenance studies of archaeological ceramics (Perlman and Asaro 1969). A first large set of samples, which were selected for an examination, consisted of Mycenaean ceramics (mainly LH III A–C, but LH I-II as well) from Greece and Late Minoan ceramics (LM III A-C) from Crete. Included in this set were a variety of different vessel types, forms and shapes. Finally the elemental concentration data of about 900 samples had been assembled. But problems emerged with the interpretation of the analytical results. Among other things, the concentration data partly seemed to be rather similar for ceramics, which most probably was produced in different regions. The Berkeley group published only parts of the data (Asaro and Perlman 1973). In 1985 Perlman handed over lists of the concentration data to the Manchester archaeometry group (V. Robinson and A. Hoffmann) and to the British School at Athens (E. French). The Manchester group transferred the data in electronic form and made it available to the Bonn archaeometry group in 1990, where they were used mainly for comparison with own data. Since the acquisition of the analytical data several studies were made based on the Perlman/Asaro databank (PADB), but each concerned only parts of the whole data set, focussed on the origin of the samples from different regions (e.g.: Karageorghis *et al*. 1972, Hoffmann *et al*. 1992, French 1993, Tomlinson 1997).

In this paper we will present results of a statistical evaluation of the whole PADB according to the statistical procedures which are utilized in Bonn (Beier and Mommsen 1994). This approach was motivated by the already mentioned close similarity of the chemical compositions of ceramics from particular regions. Though this similarity is rather unusual, considering the large number of elements measured by NAA, the narrow ranges of elemental concentration values in Greek Bronze Age ceramics from different regions also emerges in our own measurements (Mommsen *et al*. 1997, Hein *et al*. 1999). In order to verify a possible differentiation between the regions it is necessary to regard the data set as a whole. A major issue of our approach is to form first stable groups of samples with similar chemical composition. The aim is a pure chemical classification not depending on archaeological assumptions. These can be taken in account afterwards but should not give a bias for the chemical classification from the beginning. After the completed chemical grouping the archaeological classification can help to interpret the chemical classification, for example to identify local groups. If pottery from a particular find site is conspicuously prevalent in a chemical group the probability for a local origin from this site is high. The probability is even higher, if pottery associated to kiln sites or even kiln wasters were selected for the analysis.

Finally the groups, which we formed from the analytical data in the PADB, will be compared with our own data on Mycenaean ceramics that we measured in Bonn. Since the Bonn ceramics standard, which is

used for the calibration of NAA data in Bonn, is based on the Berkeley pottery standard and the data evaluation in both laboratories was quite similar, the direct comparison of NAA data is possible without extensive intercalibration.

Due to the large number of samples and of pottery groups formed the main focus of this paper will be on ceramics from the Northern Peloponnese on the one hand and from Boeotia and Crete on the other hand. In both cases close ranges of element concentrations and thus problems with separating the regional production centres are reported (Asaro and Perlman 1973, Catling *et al*. 1980, Jones 1986, Tomlinson 2000). We will show that a suitable statistical approach, like the statistics procedures in Bonn, can help to solve these problems. A more detailed report on the PADB project is presented elsewhere, including all established chemical groups, a comparison with other studies on the Berkeley material and detailed assignments of each PADB sample to the established chemical reference groups (Mommsen *et al*. forthcoming).

STATISTICAL GROUPING METHOD

At the starting point we have a database with N samples, in our case 885. For each sample there is a set of m concentration values x_k with corresponding uncertainties σ_{xk}, according to the statistical experimental error. The number of measured elements m is 27 in the PADB. From a statistical point of view there are N points in an m-dimensional hyperspace. The problem is to subdivide the points into different groups, or to find conglomerates of points of contiguous or nearly similar position. Groups of points representing similar composition can be treated again as one point, which corresponds to the average concentration values of the group members. In this case the spreads (root mean square deviations) of the concentration values can be taken as uncertainties.

For this reason the modified Mahalanobis filter procedure was used. The modified squared Mahalanobis distance was developed in Bonn as a measure of dissimilarity between points or samples (Beier and Mommsen 1994). In a simplified version without regarding correlation it is:

$$d^2 = \frac{1}{m-1}\sum_{k=1}^{m}\frac{(fx_k - y_k)^2}{\left(f^2\sigma_{x_k}{}^2 - \sigma_{y_k}{}^2\right)}$$

Here x_k and y_k are the concentration values of element k in the samples or groups **x** and **y** and σ_{xk} and σ_{yk} are the corresponding uncertainties. The factor f is a best relative fit factor, also called ‘dilution’ factor, which is applied to the data set of each sample. The dilution effect can have various reasons. The clay paste can have more or less amounts of coarse ingredients, which are usually low in trace elements and thus dilute the trace element concentrations of the clay fraction. Variations of the amount of coarse ingredients in the ceramics can be due to variations in the production process, to deviations in the raw materials or even to post burial alteration (Buxeda 1999). Other reasons for constant shifts are for example errors during the measurement procedures, e.g. weighing errors or, in case of NAA, inhomogeneous neutron flux. Since the d^2 are normalized according to statistical criteria considering the uncertainties of the concentration values they can be easily converted into a probability of group membership. Thus, in case of a chemical group pattern a cut-off value of $d^2 < 1.6$ corresponds to a 95% confidence interval.

The complete formula including the covariance matrices and basics and details of our mathematical methods can be found in Beier and Mommsen 1994.

The actual grouping procedure is a kind of a ‘bottom to top’ approach in contrast to other statistical methods. Starting from one sample, which should be selected suitably but can be arbitrary, the database is searched for similar samples. These samples form a preliminary group, which serves as new starting point in hyperspace to search for similar samples. The process is repeated iteratively. Even though the spreads increase with the number of group members and thus the distances of the other samples to the group pattern decrease, the group forming process normally results in a definitive chemical group. The other samples in the database should be clearly separated according to their distances to the group pattern. After this, another, until then ungrouped, sample is selected in order to start to build the next group. The whole procedure ends when the remaining ungrouped samples can not be used to form new groups and have to be classified as chemical singles. In the case that further samples are added to the data set, the grouping procedure can be continued.

Since the grouping procedure is semi-automatic the user has at every step the opportunity to check the data set for conspicuous samples or particular concentration data and to take this into consideration. Also the cut-off value for d^2 can be chosen suitable to the particular data set, if it is reasonable.

RESULTS OF THE STATISTICAL GROUPING

As mentioned above we disregarded the available archaeological information when we started with the grouping. Each sample of the data set was treated without consideration of find site or ware type. In a first attempt we used our usual acceptance level (95 % confidence level) for similarity of chemical patterns. Besides others a relatively large group of 491 samples was formed, mainly with ceramics from the Peloponnese, especially from the Argolid and Achaia. In a next step, focussed on this large group, we used sharper conditions for similarity by decreasing the cut-off value for d^2. In this way substructures could be found in the large Peloponnese group. A motivation for splitting up the group were unusually high spreads of elements like Ca, Co, Cr and Cs compared to the average statistical measuring error and to typical spreads, which we know from other groups. Furthermore, as mentioned above, we expected rather similar chemical patterns especially for these regions due to experiences with our own data. The element concentration data for the large Peloponnese group and the four subgroups are shown in Table 1 together with the corresponding average measurement errors. A disadvantage of the sharper similarity conditions was that several samples remained ungrouped or without definitive group assignment respectively. Thus, the number of all samples in the four subgroups is smaller than the number of the original large Peloponnese group. If the spreads are taken into account the concentration values for most of the elements look still similar in the four subgroups. But as the discriminant analysis in Figure 1 shows, the four groups can be distinguished clearly by considering all 27 concentration values.

Besides the four groups from the Argolid and Achaia, which were chemically rather similar, a number of other chemical groups were found. Most of them were discriminating very clearly. There was one other conglomerate of groups, which could be subdivided only by using sharper similarity conditions. The samples in this structure came from Boetia and Central Crete. A chemical similarity of the ceramics of these regions was reported before as well (Asaro and Perlman 1973, Catling *et al*. 1980, Jones 1986, Tomlinson 2000). The chemical patterns of the six subgroups are shown in Table 2. And a discriminant analysis of these groups is shown in Figure 2. Even though the groups PHAP and PHBP are not perfectly separated, according to the first two discriminant functions, the groups THEP, THFP, PHCB and PHDP can be clearly distinguished.

The other chemical patterns, which were found in the PADB, belonged mainly to ceramics from the Southern Peloponnese, Attica and Western Crete and will not be treated in detail in this paper. However, the chemical discrimination of these patterns is distinct.

ARCHAEOLOGICAL ASSIGNMENT OF THE CHEMICAL GROUPS – COMPARISON WITH DATA FROM BONN

If at this point the group assignments of the samples are compared with their find sites a regional significance of the found patterns becomes obvious. This comparison for the case of the four Northern Peloponnese groups is shown in Table 3. Though samples belonging to the large group MBP can be found at almost every sampled site, the main part corresponds to ceramics from the Argolid, especially to ceramics from Mycenae, Berbati and Zygouries. Besides, in the PADB there are two wasters from Berbati. One of them fits to MBP, whereas the other deviates only in Cr from MBP and finally was assigned to EMBP. Thus, the origin of this pottery from the region Mycenae/Berbati is rather probable. Furthermore the rare appearance of this chemical pattern in the material of other find sites gives a hint to a limited material exchange between different regions of Mycenaean Greece (the main part of the MBP pottery assigned to ‘other sites’ was found in Abu Hawam). A comparison with the Bonn databank shows that MBP is identical with the pattern MB, which is also assigned to the region Mycenae/Berbati (MB, Mommsen *et al*. 1988).

The samples of the group TAP came mainly from Tiryns and Asine. This pattern too is well known from our own data (TA, Mommsen *et al*. 1988). Within the Bronze Age pottery from Greece, which was analysed in Bonn, mainly samples from Tiryns and Asine showed this pattern as well, including a number of wasters from Tiryns. Thus an origin of this ware from the Southern Argolid is rather probable. Looking at the whole

data set it is obvious that the TA/TAP ware was not as widespread as the MB/MBP ware. In the PADB there are only three samples in the material from Tell Abu Hawam and one sample from Korakou, which fit to the TAP group. In our own material for example we have several samples from Qantir in Egypt that belong to this ware type (Mommsen *et al*. 1996).

The third Peloponnese group ACHP corresponds to the Bonn pattern achaia-a (Mommsen *et al*. 1997), which is formed by ceramics from the Western Peloponnese. Also in the PADB mainly samples from Achaia and Elia can be found in this chemical group. The pattern is chemically very close to MB/MBP. The main difference is in Ca and Cs. But the regional allocation of the chemical groups emphasizes the chemical distinction (Hein *et al*. in press).

Finally the fourth Peloponnese group EMBP is also located in the Argolid. As mentioned before, one waster from Berbati is assigned to this group. In the Bonn databank this pattern is known from material from Aigina, which was archaeologically classified as imports from the Argolid (Mommsen and Maran 1999, Mommsen *et al*. 2001). Most of the ceramics, which is assigned to EMBP, is classified as rather early material (LH I –II). Due to the chemical similarity to MBP and the archaeological classification we assume a close vicinity of the production place or even a temporal variation of the production technique in the same workshop or production place.

In Table 4 an assignment of chemical groups to the find sites in Boeotia and Central Crete is shown. As mentioned above, the chemical differentiation between the ceramics from Boeotia and from Central Crete required a similar treatment as the subdivision of the large group of Northern Peloponnes ceramics. Though the chemical patterns look partly similar the archaeological assignment of the groups shows that the differentiation is reasonable. Furthermore the two groups THEP and THFP correspond to the groups a-theb and b-theb (Mommsen *et al*.1998) in the material measured in Bonn. These groups were formed by samples from Boeotia as well. In the PADB there are also conspicuous numerous samples from Thebes, which belong to MBP. This could be due to the selection of the samples for analysis, because the Theban ceramics in the Bonn databank seems to be mainly local. The ceramics from Crete forms several groups. The pottery from Western Crete can be distinguished clearly from the reference groups from Central Crete (Knossos and Phaistos). The ceramics from Central Crete analysed in Bonn forms similar groups (Mommsen *et al*. 1995), even though with the Bonn data set it was not possible to distinguish between ceramics from Knossos and Phaistos, like in the PADB the groups PHAP and PHDP. They are differing only in Eu and Dy. Because the ceramics from Knossos and Phaistos was measured separately this could also be due to a batch difference.

CONCLUSIONS

The groups, which were established from the NAA data in the PADB, correspond in all applicable cases to those, which were formed based on concentration data of Mycenaean and Minoan pottery measured in Bonn. Thus, the measurements, which were carried out in Berkeley and in Bonn independently and with a time interval of partly 30 years, have been precise and correct. This emphasizes the importance and necessity of precise and comprehensible analytical work. With reliable data and considering 'dilutions' different pottery production series - defined by the use of a clay refinement procedure leading to a chemical similar clay paste - can be distinguished easily even within the same area of production and, hence, provenance can be established, if reference samples exist. In the present case the purely chemical approach provides sufficient information in terms of provenance or assignment of single vessels to particular production series respectively.

The conclusions, which were drawn out from our own data until now especially concerning the Mycenaean ceramics, are confirmed by similar findings in the PADB. During the whole Mycenaean period regionally different pottery groups prevail, at least in the periods and regions covered by our own data and by the Berkeley material. Despite the typological and stylistical homogeneity of the Mycenaean pottery, it can be assumed that the production took place in regional workshops or production centres, and that there was no need for an extensive trade of pottery. In each region only a few main patterns can be detected, which seem to correspond to particular production series. The production covered obviously in all workshops a full spectrum of ware types, from coarse ‘every day use’ ceramics to high quality ceramics like pictorial jars. The presence of many of these patterns in ceramics of the whole Mycenaean period indicates a continuity of the production technology during this time.

In the Bonn laboratory there are still about 1000 samples of Mycenaean ceramics from other regions of Mainland Greece, which have to be analysed yet. Thus, our results and conclusions remain to be verified by this material.

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Table 2 *Element concentrations of chemical groups in the PADB: the average values M are in μg/g (ppm), if not indicated otherwise, and the spreads σ in % of M. For each sample a best relative fit was applied to the groups mean, considering all elements except for Ca and Na.*

Table 3 *Assignment of pottery from sites in the Northern Peloponnese to the four chemical groups from this area: EMBP – early (LH I-II) Mycenae/Berbati, MBP – Mycenae/Berbati, ACHP – Achaia/Elia, TAP – Tyrins/Asine, single – chemical singles, other – samples assigned to other groups; numbers in parentheses correspond to associated group members*

Table 4 *Assignment of PADB pottery from sites in Boeotia and Central Crete to the six chemical groups from these areas; numbers in parentheses correspond to associated group members*

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Figure 1 *Discriminant analysis of 376 samples from the Northern Peloponnese assuming 4 groups: Plotted are the discriminant functions W1 versus W2, which cover 66 % and 29% of the between group variance respectively. The ellipses drawn are the 2σ boundaries of the groups.*

Figure 2 *Discriminant analysis of 100 samples from Boeotia and Central Crete assuming 6 groups: Plotted are the discriminant functions W1 versus W2, which cover 75 % and 13% of the between group variance respectively. The ellipses drawn are the 2σ boundaries of the groups.*

	Peloponnese		EMBP		MBP		ACHP		TAP		ave. meas.
	491 samples		42 samples		275 samples		32 samples		25 samples		error (%)
	M	s (%)	M	s (%)	M	s (%)	M	s (%)	M	s (%)	
Al %	8.11	5.6	7.91	4.2	8.31	4.3	8.03	4.1	7.95	2.6	2.4
Ca %	9.18	24	8.35	24	9.53	14	5.55	32	8.73	14	5.9
Ce	62	3.4	63.5	3.7	62.2	2.7	61	4.3	64.8	2.2	1.6
Co	28.9	9.6	34.9	9.2	28.8	5.2	28.3	5.4	23.9	4.2	1.3
Cr	250	13	280	11	243	8.2	274	6.8	203	9.6	1.8
Cs	8.6	25	12	23	9.06	7.6	6.48	9.6	4.98	5.6	3.3
Dy	4.53	5.3	4.64	3.7	4.47	4.1	4.78	6.9	5	4.6	3.1
Eu	1.21	7.8	1.24	5	1.19	7.5	1.26	6.3	1.36	5.2	4.6
Fe %	5.26	5.2	5.03	5.9	5.33	3.7	5.41	3.4	5.22	2.8	1.4
Hf	3.67	14	3.43	9.8	3.62	13	4.15	10	4.24	9.6	8.3
K %	2.45	21	2.73	20	2.57	15	2.44	13	2.1	19	11
La	31.5	4.1	32.1	3.2	31.9	3.3	30	3.2	31.3	2.6	2
Lu	0.39	5.5	0.41	4.4	0.39	5.1	0.42	5.1	0.42	4.6	4.5
Mn	921	12	886	20	925	8.2	971	17	975	6.7	1.1
Na %	0.66	39	0.42	24	0.59	27	0.82	18	1.3	22	2
Nd	28.2	6.7	28.9	6.6	28.3	6.4	28.1	7.2	28.9	8.9	6.4
Ni	223	20	314	14	212	12	220	12	165	11	8.7
Rb	147	20	156	18	156	12	139	16	112	14	12
Sc	21.2	5.1	21.4	2.7	21.5	4	21.4	4	20.1	2.6	0.3
Sm	5.18	3.7	5.29	2.2	5.12	2.7	5.31	3.7	5.71	2.4	0.7
Ta	0.83	6.3	0.85	7.6	0.82	4.6	0.85	5.2	0.88	2.9	0.9
Tb	0.79	8.2	0.8	7.4	0.78	8.1	0.81	8.1	0.9	5.8	6.3
Th	10.8	3.8	10.7	4.4	11	3	11	3.1	11	2.3	1.4
Ti %	0.44	6.2	0.43	4.9	0.45	6	0.47	5.3	0.46	4.7	4
U	2.36	7.9	2.49	5.9	2.35	4.6	2.3	5.8	2.13	3.4	1.4
Yb	2.71	5.2	2.78	4.3	2.7	5.1	2.84	4.5	2.88	4.1	3.8
Zn	120	13	117	15	123	9.9	113	14	104	11	6.8

Table 1

	THEP		THFP		PHAP		PHBP		PHCP		PHDP	
	24 samples		9 samples		35 samples		5 samples		16 samples		11 samples	
	M	s (%)	M	s (%)	M	s (%)	M	s (%)	M	s (%)	M	s (%)
Al%	6.87	5	7.92	3.4	7.25	6.7	7.33	6.7	8.1	3.5	6.94	3.2
Ca%	9.21	25	9.29	15	8.13	20	10.8	9.3	7.64	25	8.97	23
Ce	52.4	4.2	62.1	2.4	55	2.7	57.4	3.3	54	3.5	53.8	3.2
Co	39.9	11	33.7	5.5	33.3	6	28.8	1.8	39.8	5.2	30.7	7
Cr	499	14	498	17	432	12	333	5.9	459	8.1	390	5.4
Cs	5.02	16	6.45	9.4	6.82	19	7.89	8.8	8.61	9.3	6.44	15
Dy	4.01	5.6	3.92	5.4	4.17	4.6	4.11	4.1	4.13	5.2	4.47	3.7
Eu	1.18	7.2	1.12	9.7	1.1	7.3	1.06	8.4	1.08	8.3	1.26	3
Fe%	5.53	4.8	5.28	3.8	5.49	3.8	5.19	2.2	6.28	3.6	5.23	1.8
Hf	3.26	8.7	3.26	7.5	3.78	8.9	3.99	7.4	3.6	8.1	3.67	8.8
K %	2.18	14	2.66	16	2.39	17	2.54	12	2.5	22	2.07	22
La	24.8	4.4	29.5	3.7	27.6	3	28.7	2.2	27.6	2.7	26.9	4.3
Lu	0.34	6	0.35	7.2	0.36	6.1	0.37	5.1	0.38	4.4	0.35	5.8
Mn	887	16	770	9.8	920	15	824	4.5	853	12	700	14
Na%	0.54	17	0.6	11	0.78	25	0.61	3.2	0.67	37	0.67	26
Nd	23.3	7.1	26.3	6.6	23.5	7.1	25.3	9.4	23.7	7.6	24	6.9
Ni	544	13	426	5.3	382	11	291	6.8	494	7.5	397	8.8
Rb	111	14	147	10	117	14	137	13	143	18	94.8	13
Sc	20.7	2.5	21	1.3	20.6	5.4	20.1	2.7	24.2	4.6	20.1	3
Sm	4.43	3.5	4.66	2.4	4.62	2.4	4.71	1.9	4.49	3	4.66	1.7
Ta	0.76	9.6	0.9	12	0.91	5.7	0.95	3.3	0.92	8	0.88	6.8
Tb	0.71	11	0.68	6.3	0.74	8.8	0.77	6.1	0.72	8.4	0.67	4.9
Th	8.41	5.8	9.87	2.4	9.45	2.9	9.56	3.2	10	3.4	9.04	3.6
Ti%	0.44	5.5	0.48	4.5	0.43	5.4	0.43	7.4	0.43	6	0.41	6.5
U	1.97	14	2.58	9.8	2.35	11	2.88	4.7	2.33	3.9	2.87	16
Yb	2.28	4.4	2.45	6.9	2.43	4	2.47	5.6	2.46	3.3	2.46	4.5
Zn	107	11	99.9	7.4	114	13	125	6.1	125	0	90.8	9.8

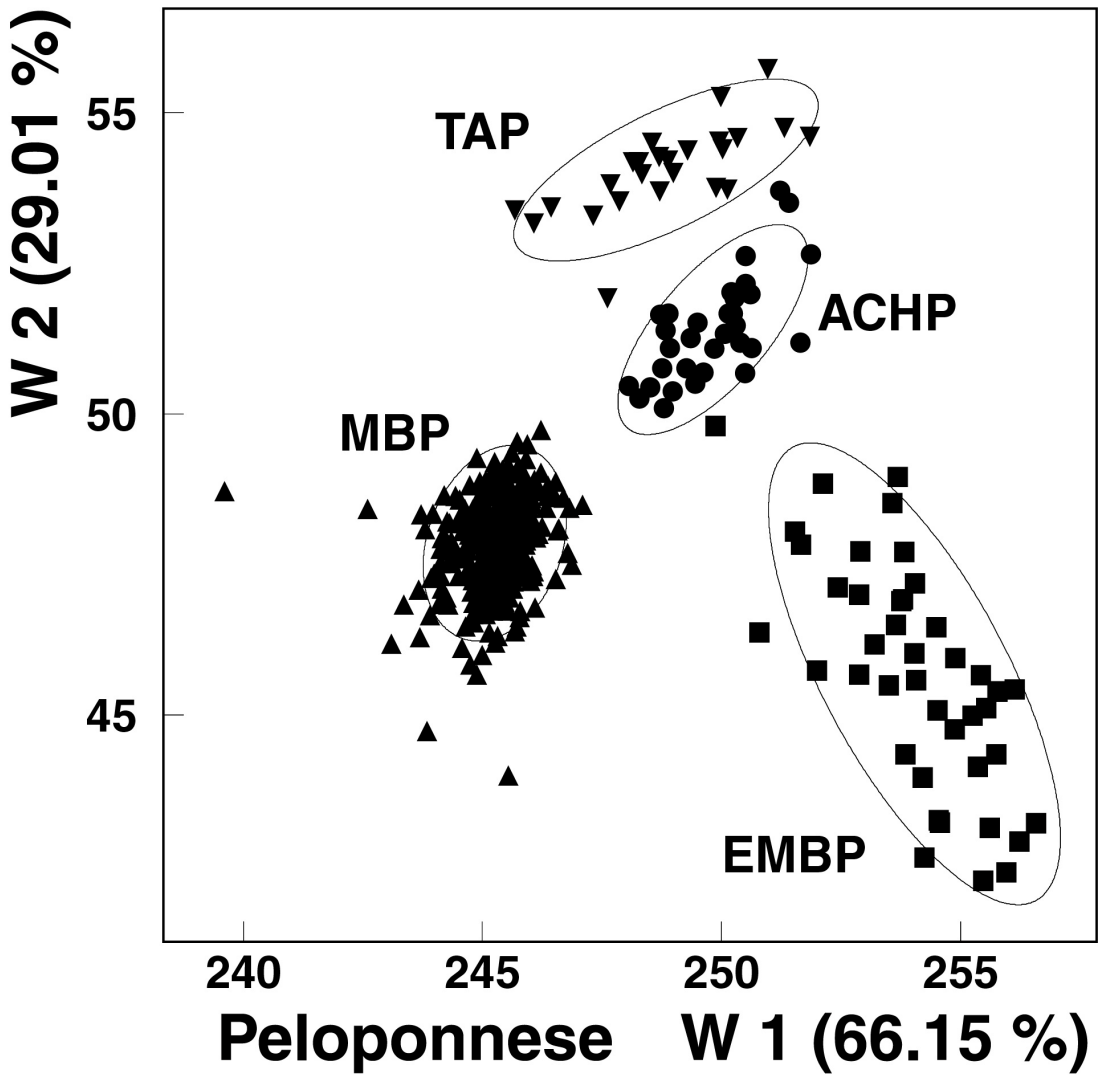
Table 2

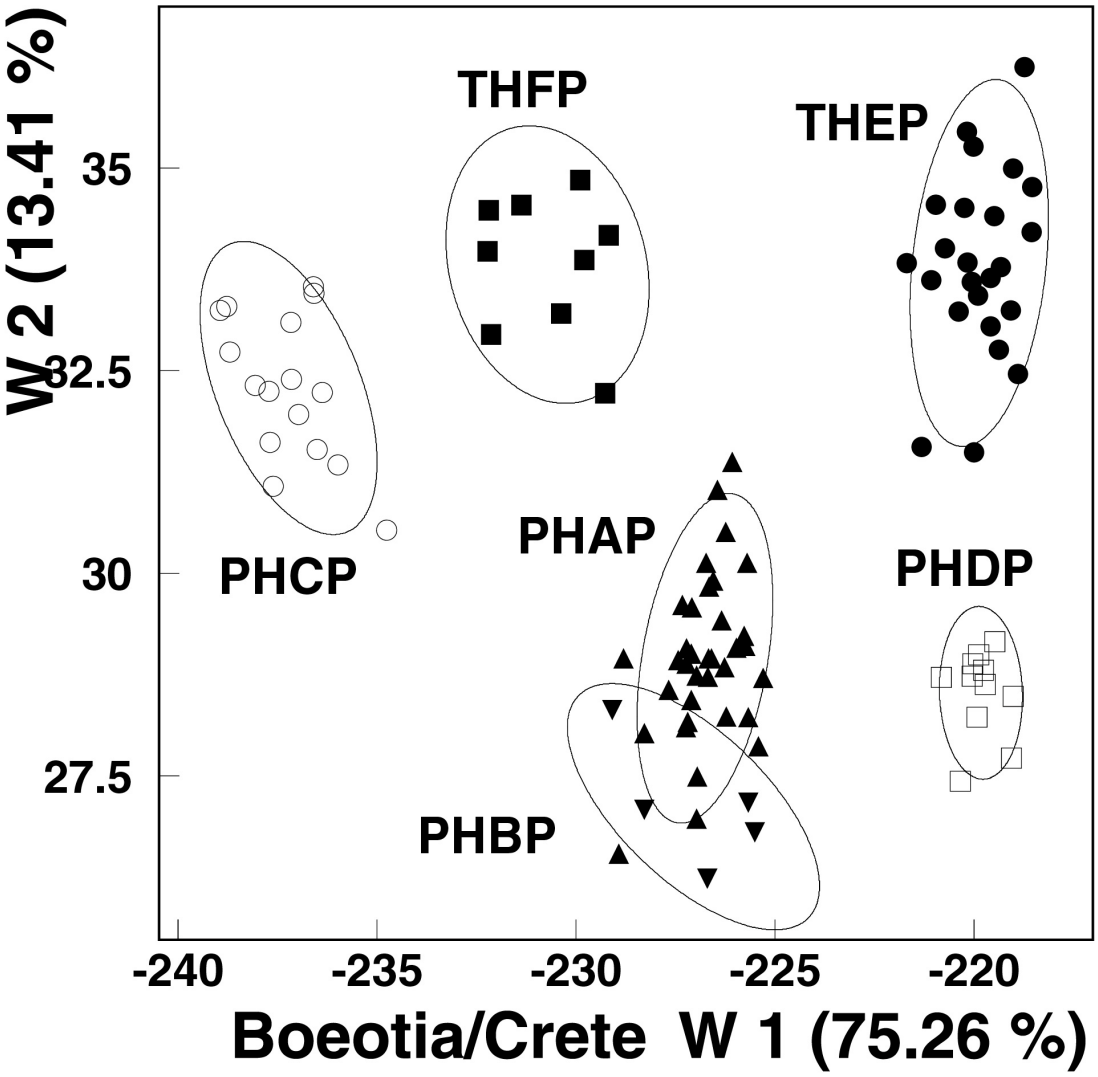
	EMBP	MBP	ACHP	TAP	single	other	sum
Argolid							
Mycenae	13(3)	54(3)		2	1	2	78
Berbati	1(4)	29	(2)		1	1	38
Zygouries	(1)	32(3)			3	1	40
Tiryns	1(3)	26		12(1)	2		45
Asine	5(1)	33(2)	(1)	8	6	5	61
Corinthia							
Korakou	1(6)	16(9)		(1)	7	5	45
Achaia							
var. sites			10(6)		3	1	20
Elia							
Olympia		(1)	11(5)		1	2	20
Platanos	1	1	10(1)		3	4	20
other sites	20(9)	86(7)	1(6)	3	73	313	518
sums	42(27)	277(25)	32(21)	25(2)	100	334	885

Table 3

	THEP	THFP	PHAP	PHBP	PHCP	PHDP	sums
Boeotia							
Eutresis	1						1
Tanagra		4					4
Thebes	21	5	(2)				28
Crete							
Knossos	2		2(1)		1(2)	11	19
Phaistos			33(3)	4	15(1)		56
sums	24	9	35(6)	4	16(3)	11	108

Table 4


W 2 (29.01 %)
55
50
45
TAP
ACHP
MBP
EMBP
240
245
250
255
Peloponnese W 1 (66.15 %)


THFP
THEP
PHCP
PHAP
PHDP
PHBP
35
32.5
30
27.5
-240
-235
-230
-225
-220
W 2 (13.41 %)
Boeotia/Crete W 1 (75.26 %)