

## **Provenance determination of pottery by trace element analysis: Problems, solutions and applications**

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Provenance determinations of pottery by chemical analysis is reviewed and shown to work well. Since pottery is produced from a well homogenized clay paste according to a certain recipe, sharp elemental patterns are expected for a series of products having the same origin. To obtain such patterns when forming compositional groups of pottery, a consideration of experimental errors, a correction for dilution and a choice of only stable elements is necessary. The patterns thus obtained will have low probability of overlap with groups of different origin. Examples for well defined groups of German stonewares and of Mycenaean wares from the Peloponnese are recorded.

### **Introduction**

Scientific examination of pottery is one of the oldest fields in archaeometry. More than 60 years ago SHEPARD<sup>1</sup> published her innovative work about petrographic and chemical analyses on pottery. Her aims have been “first to trace the history of the potter’s craft” and secondly, “to recover the evidence which pottery preserves of cultural development, contacts and influences”. This paper will not cover such a large scope, but be confined to chemical trace element analysis of pottery only. If just provenance is the archaeological question, chemical analysis is the most convenient scientific method according to our experience. Results can be obtained very effectively in most cases, also if manpower, costs and time are considered. However, the general trend during the last years was to recommend a combination of several methods for provenance studies and not to use only chemistry, compare, e.g., JONES,<sup>2</sup> TITE.<sup>3</sup> This might be a consequence of low success rates in several of such studies either due to poorly measured data or due to wrong assumptions and insufficient data evaluation. It will be described here, how the concentration data measured in different pottery vessels should be treated and interpreted and how the chemical compositions can be used to classify and identify different groups of pottery and, in a second step, to locate their places of manufacture. These results, linking find sites to production sites, have then to be interpreted archaeologically and, eventually, will increase our knowledge of cultural contacts, trade and influences and point to economic and social interactions between communities involved.

In general, the interpretation of the compositional variation observed in pottery as a function of the place of origin proved to be very successful as shown by many publications. However, with the increasing number of samples from sites not only geographically far apart, overlapping compositions occurred for some regions making a discrimination of single production places difficult for some cases. For example, ASARO and

PERLMAN<sup>4</sup> wrote about the analytical data of Mycenaean pottery measured in Berkeley: “The question of provenience of the vast quantities of Mycenaean wares has proved perplexing” (p. 213); (the problem of interpreting the data) “is a gigantic puzzle which will require much labor to solve to the degree that is capable of solution” (p. 219). This situation still prevails for Late Bronze Age pottery of some regions of Greece. But first approaches to overcome these problems of discrimination, at least partly, by using “core groups” emerge as will be shown here.

Archaeometric provenance determinations by mineralogical investigations (petrography, X-ray diffraction) use different information stored in the sherds. They can be considered as complementary and are recommended to obtain an additional sub-classification and/or an independent verification of the chemical results. They also might be helpful to explain the reasons for the chemical differences in pottery groups.<sup>5</sup> However, an advantage of elemental analysis is that it produces ‘hard’ quantitative data the interpretation of which does not depend on acquired expertise in recognizing inclusions.

### **Historical review**

Early analytical work on pottery started at a larger scale in the late 50ties/early 60ties<sup>6–8</sup> using already the three methods which, in the following years, produced most of the results, namely neutron activation analysis (NAA), X-ray fluorescence (XRF), and optical emission spectroscopy (OES). With the development of high resolution semiconductor gamma-ray detectors since the mid-60ties NAA reached and surpassed OES and also atomic absorption spectroscopy (AAS) in measurement precision of trace elements.<sup>9</sup> The early provenancing work has been comprehensively summarized by JONES in 1986.<sup>10</sup> Since then, many more analytical methods like inductively coupled plasma emission or mass spectroscopy (ICP-ES, -MS) with or without laser ablation<sup>11</sup> or isotope analysis<sup>12</sup> have been tested and employed and many laboratories have been engaged in pottery analysis.

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### Basic principles

In several review papers and textbooks the main points of provenancing of pottery have been summarized, namely the appropriate selection of sherds, the sampling procedures, the number of specimens to be analyzed, the importance of quantitative and precise measurements, and the data evaluation and interpretation procedure.<sup>3,10,13–16</sup>

The basic idea was, that pottery having the same composition originates from the same production place, since the elemental content of pottery measured by some method of chemical analysis depends mainly on the geochemical composition of the claybed(s) exploited and, therefore, points to its production place. An obvious assumption is that the raw clay was not traded; indeed considering the weight of clay and the limited transport possibilities in old times, this assumption seems well justified. If many elements, about 20 to 25, are measured with sufficiently high experimental precision, the “elemental pattern” obtained has a very high probability of being unique in the world. If now this pattern can be assigned to a production place, e.g., by comparison with reference material, which is selected on archaeological criteria as local, provenance is determined. The chances to encounter a member of an already known production place will increase with the number of measured patterns. As work progresses, a card-file with many patterns can be assembled and provenance determinations will be easier.

But there are several points which complicate this simple procedure. Pottery is made from a well homogenized lump of clay. According to a certain “recipe” the ancient potters purified, mixed or tempered the clays to produce a workable clay paste to form and fire their vessels. For example, coarse parts are taken out by levigation, other non plastic parts may have been added as temper. Also mixing of clays of different geological origin may occur. HANKEY<sup>17</sup> wrote: “Potters, however, are rather like cooks in choosing ingredients”. These refinements performed by a variable human action determine the composition of the clay paste. It is only identical to the raw clay from a claybed, if the clay is found ready to be used. From experience this is rarely encountered, but sometimes lucky and most welcome matches are found like the Motza clay formation near Jerusalem (PERLMAN’s personal communication). The analysis of large numbers of clay samples and a cumbersome geological survey of possible production areas as proposed often in former work to form reference patterns are therefore not needed and not recommendable in pure chemical provenance determination work (compare Reference 18).

During the preparation and refinement procedures of the clay paste described above the ancient potters certainly did not follow always very strict rules.

Therefore, the quantity of non plastic parts will vary in some range. Since the tempering parts consist mainly of quartz, feldspar or calcite which generally have much lower levels of trace elements than clays, dilutions due to these pottery making practices should be considered comparing compositions. A higher proportion of the non plastic tempering parts, e.g.,  $\text{CaCO}_3$  will result in a decrease of the concentration values of all the elements absent in the temper. To correct for such dilutions, elemental concentration ratios should be used. Or, having the same effect, such dilutions should be considered performing a best relative fit of the individual concentration values to the group mean values and correct all values by the fit factor obtained. In such ways only the clay part of the paste which determines chemical provenance is taken into account and the varying parts will be cancelled.

A further assumption to be made is that no changes of the elemental concentrations occurred during the firing and burial. Whereas firing temperature is found to have negligible effects for all of the more than 30 elements measured usually by NAA (except for Br),<sup>19</sup> possible alterations for some elements like Ba, Ca, K, Na, or P<sup>4,20,21</sup> have been reported during burial time which seem to depend partly on the firing temperature of the sherds. We generally find an enlarged scatter of concentrations for these elements (including As; P is not measured by us) in sherds manufactured with certainty in one workshop by the same recipe. These elements should be considered with care during the first data evaluation and classification.

If the ancient potters introduced larger changes in the recipe employed for producing the paste, especially if different clay mixtures were used, then different elemental patterns might be detected in the material from the same workshop. These changes in the recipe might have been done also deliberately to produce different types of pottery with special properties.<sup>22</sup> So, even single production series in a workshop can be distinguished by a chemical analysis, not only the production workshop.<sup>23</sup>

In particular, the scatter or spread (root mean square deviation) of the concentration values of the patterns formed should be inspected. Then, it can be checked if the patterns support the assumptions of production series just described. Although a priori no knowledge exists about the inhomogeneity of a clay paste, in the normal case of homogeneous paste, spread values of less than 5–10% for many elements are expected. Therefore, we look in our data only for “sharp” patterns with small spreads. Large spreads often indicate the existence of possible subgroups which belong to different production series or even to different production sites. In older work, often a large part of the samples taken from one site have been added to a group resulting in unusually large spread values and “diffuse” group patterns difficult

to discriminate from other patterns. If well defined chemical groups are formed, all members will belong with high probability to only one production series.

The next step, the assignment of the group to its manufacturing place, is the most serious problem and depends on the availability of good reference material to form control groups. For each single production series detected a control group is needed. Very well suited are true wasters, which have been thrown to the dump heap of a workshop and certainly have not been traded. They will represent the one or the different production series of the workshop. But sherds looking similar to wasters found in strata of settlements might have been overfired during a destruction event. If pottery workshops and also wasters from dump places are not known, as is often the case, control groups are usually formed by distribution arguments. If a pattern is found mainly in material from one site and only sporadically somewhere else, the assignment of this pattern to this place seems probable. But this means introducing a bias from the beginning. All pieces could be as well imports from elsewhere to the site considered. However, the argument of a local production at a site is strengthened, if the time depth of existence of this pattern is large, i.e., if it is found in early as well as in much later strata. Furthermore, if very different ware types, i.e., coarse, cheap or expensive, rare vessels of high quality, are all members of the group, the probability declines, that all these ware types are imports. Still, to establish good control groups is in many cases a problem difficult to be solved without clear reference material.

### Grouping methods of chemical data and examples

In most cases and under the right conditions groups of samples which have similar compositions can be found without problems and chemical provenance determinations work superbly. With precisely measured concentrations of more than 20 elements and considering measurement errors and possible dilutions, it is normally not difficult to distinguish the differences of chemical patterns of single production series. In practice, because of the large number of values to be sorted, computer aided methods are used for the group formation. But we agree fully with the opinion of PERLMAN (private communication) or SCHNEIDER,<sup>24</sup> that similar or different compositions in pottery sherds should be seen in the data directly and that grouping results obtained only with the help of sophisticated statistical methods are possibly dubious, all the more since some of the assumptions necessary in such calculations (e.g., nature of the probability distribution) might not be fulfilled. As result of the grouping, the pattern of the average concentrations (grouping values) including the

individual elemental spreads should be given, so that concentration differences of groups can directly be seen and good group memberships verified.

As example for this "normal" case, where groups are readily obtainable, we will refer to data of German stoneware from the Rhineland and Saxony. In both regions stoneware vessels of excellent quality have been produced mainly during the end of the medieval period which are difficult to distinguish by archaeological means and which have been exported to the whole of northern Europe. The average concentration values measured in most of the vessels from Siegburg near Bonn<sup>25</sup> and from Waldenburg in Saxony<sup>26</sup> are given as examples in Table 1, column 2, 3 and 4, 5. The two production centers show very different elemental compositions and are easily distinguishable by chemistry. The clear separability is demonstrated in Fig. 1a depicting the result of a discriminant analysis. It is used, after the groups are already formed, to check the group discrimination and to depict the grouping results. The discriminant functions  $W1$ ,  $W2$ , ... are linear combinations of the concentration variables calculated in such a way, that the differences between the presumed groups are displayed as clearly as possible (see, e.g., Reference 15). Besides the two groups from Siegburg and Waldenburg five more groups, all from Saxony, are included to show that even for a geographically limited region and large sample numbers all patterns are very well resolvable. All the groups contain reference material and also single pieces of unknown origin which could be assigned to their production place with the exception of one group representing a production series of unknown provenance.

However, an evaluation of some newly measured Greek data from the Late Bronze Age (LBA) revealed a different situation. Not several well separated groups, but one large group with unusual large concentration ranges was obtained as grouping result using the modified Mahalanobis filter procedure for the statistical grouping.<sup>27,28</sup>

In statistical data analyses a sample is commonly represented by a point in multidimensional space termed also hyperspace. Each dimension of this space corresponds to the concentration values of one chemical element. Samples having not very different compositions will be represented by points not far apart. In order to form groups of vessels of similar composition a "cloud" of points has to be located. Several old and well known statistical methods exist for that exploratory purpose like principal component analysis (PCA) or different kinds of cluster analysis (CA), which all have their problems like the need of a standardization of the whole data set or the treatment of outliers or zero values.<sup>15</sup>

Table 1. Concentrations of 30 elements: averages  $M$  (in  $\mu\text{g/g}$ , ppm), if not indicated otherwise, and spreads  $\sigma$  in % of  $M$ , after a best relative fit of each sample towards the group mean (considering all concentration values except As, Ba, Ca and Na)

Element	Siegburg 41 samples $M$ ( $\sigma$ (%))		Waldenburg 34 samples $M$ ( $\sigma$ (%))		Mycenae/Berbati 252 samples $M$ ( $\sigma$ (%))		Achaia 146 samples $M$ ( $\sigma$ (%))		Aegina 107 samples $M$ ( $\sigma$ (%))	
As	4.14	49	9.72	17	5.61	43	6.68	71	9.10	37
Ba	379.	11	552	8.5	382	26	454	25	307	23
Ca, %	0.30	56	0.42	46	9.80	22	4.95	40	11.0	23
Ce	123	2.9	101	2.7	62.3	2.5	65.8	3.2	46.2	4.2
Co	3.08	20	4.81	14	28.3	6.4	28.5	7.8	20.2	11
Cr	159	4.6	131	12	249	12	265	9.8	449	13
Cs	21.2	6.6	33.1	5.7	8.65	11	7.17	7.8	5.64	12
Eu	1.81	2.3	1.53	4.3	1.14	4.5	1.22	4.7	0.94	3.6
Fe, %	0.53	15	1.05	6.2	5.14	3.8	5.29	5.3	4.14	4.4
Ga	28.1	8.5	57.4	5.6	21.6	26	21.8	18	13.0	35
Hf	8.82	11	6.54	7.3	3.56	9.8	3.90	15	4.06	5.8
K, %	1.93	5.5	1.85	7.5	2.58	10	2.55	6.3	1.99	13
La	62.5	3.0	68.7	3.4	31.3	2.7	31.1	4.3	22.2	3.2
Lu	0.51	3.8	0.43	5.3	0.42	5.3	0.51	23	0.35	5.6
Na, %	0.20	33	0.30	53	0.54	32	0.89	20	1.05	16
Nd	50.8	4.5	40.6	5.2	25.9	5.8	27.7	11	20.0	7.0
Ni	31.6	26	54.2	25	216	13	213	11	329	13
Rb	126	6.2	121	7.8	149	8.5	146	6.4	73.8	9.9
Sb	1.03	11	3.44	7.0	0.57	13	0.58	19	0.76	19
Sc	15.0	3.9	18.7	4.8	21.2	4.3	22.3	4.3	16.0	3.8
Sm	8.34	3.9	7.03	4.9	4.85	4.0	5.20	5.9	3.80	5.5
Ta	1.93	3.1	3.16	3.3	0.80	6.6	0.86	5.2	0.65	6.0
Tb	1.02	5.8	0.84	4.8	0.67	6.9	0.75	8.3	0.57	7.7
Th	14.4	2.4	12.6	4.4	10.9	3.0	11.6	2.9	7.56	5.0
Ti, %	0.85	5.6	0.98	4.2	0.43	20	0.46	9.1	0.38	29
U	3.17	3.7	4.27	4.3	2.26	5.8	2.46	10	2.14	14
W	2.82	10	10.2	4.3	2.16	17	2.21	15	1.43	14
Yb	3.72	5.6	2.97	4.8	2.59	3.3	2.78	4.8	2.17	5.3
Zn	33.7	25	86.7	41	113	10	121	17	89.7	12
Zr	358	14	265	9.9	154	21	190	19	171	22

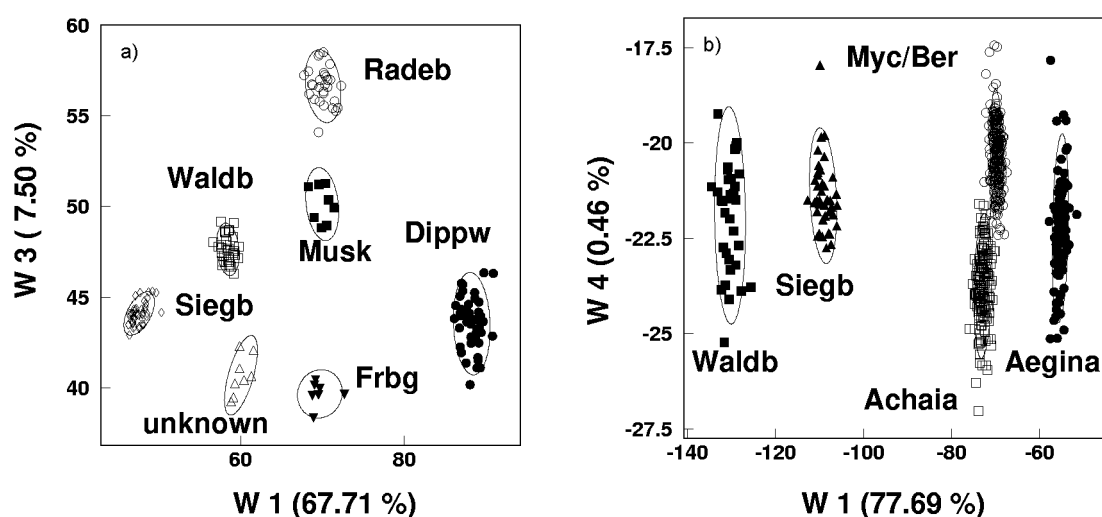


Fig. 1. Discriminant analysis of (a) 172 samples of German earthen- and stoneware sherds from Siegburg, Waldenburg, Dippoldiswalde, Muskau, Freiberg, Radeburg assuming seven groups (one pattern of unknown provenance) and (b) of the 580 samples given in Table 1 assuming 5 groups (Siegburg, Waldenburg, Mycene/Berbati, Achaia, Aegina). Plotted are the discriminant functions W1 and W3(W4) which cover 67.7% (77.7%) and 7.5% (0.5%) of the variance between groups. The ellipses drawn are the  $2\sigma$  boundaries of the groups

To overcome many of these difficulties in forming the groups we developed our own exploratory grouping method which we call the filter method.<sup>27</sup> Assuming normal distributions, our filter examines each sample separately. Generally speaking filter methods are used to test the hypothesis, a data point belongs to a group, and this hypothesis is by statistical methods either accepted or rejected. Considering experimental errors and also a possible constant shift of the data (dilution), a squared modified Mahalanobis distance  $d^2$  to a given group is calculated for each sample which can be regarded as similarity measure. Since the  $d^2$  is normalized according to statistical criteria, it obeys in the ideal case a  $\chi^2$  distribution very closely. Therefore, it can be easily converted into a probability of group membership. Choosing as confidence level of group membership 95% this will correspond to a  $d^2$  cut off value of about 1.6 nearly independent of the number of degrees of freedom for more than 20 elements measured. Therefore, all samples with small  $d^2$  below the chosen limit can be filtered out and have to be added to the starting group. When the procedure is repeated iteratively, employing the new, slightly shifted average concentration values (grouping values) and their spreads, soon all members of a group fulfilling the membership criterion are located.

The large group of LBA Greek pottery vessels mentioned above was formed employing this procedure. During the iteration, more and more sherds of our Greek databank had to be added. In each step, the spreads of the grouping values increased and, therefore, the distances to other samples were lowered, because they are calculated in units of the spreads. Despite the dilution correction, the group pattern showed unusual broad concentration distributions for some elements which contradict the assumption of a well defined, single production series from a homogenized paste. Therefore, a possible subdivision into smaller parts was investigated employing a smaller statistical cut off value of  $d^2$ . Starting again at some volume element, now only samples with smaller  $d^2$  values, e.g.,  $\leq 1.1$  corresponding to a confidence level of about 66%, were added and so "core groups" within the large cloud could be formed. The new procedure reduces the total volume of the large cloud in hyperspace and, therefore, allows a separation of core groups at the cost of higher error probability when rejecting samples as group members. In the hyperspace picture, the large cloud is split into two or several adhering smaller clouds (core groups). Because of the large number of samples – we now have about 3000 samples in our Greek databank – the concentration hyperspace is filled up with points in such a way that it becomes difficult in some cases to separate different clouds according to the standard filter procedure (95% confidence level of group membership). The close chemical similarity of a large part of the Greek LBA pottery has been already reported previously.<sup>29</sup>

As example of a successful separation of samples by forming core groups two Mycenaean patterns assigned to the Peloponnese are presented in Table 1, columns 6, 7 and 8, 9. They are very similar in composition. The first data set belongs to sherds assigned to a production place in the region of Mycenae/Berhati.<sup>30</sup> This assignment is now strengthened, since one waster from the workshop excavated at Berhati<sup>31</sup> and measured in Berkeley is a member of this group. The second pattern separable by core group formation was found to contain samples predominantly from Achaia, in particular from different Mycenaean sites in the neighbourhood of Patras. Inspection of the data reveals, that only the elements Ca and Cs are different and lower for the region of Patras. In a bank of data from the Argolid and Achaia measured in Berkeley<sup>32</sup> core group formation (again without using the knowledge of the finding sites of the samples) resulted in just the same two patterns for these regions only different in Ca and Cs.<sup>33</sup> This supports not only our data for these Peloponnesian regions, but also strengthens the applicability of the core group formation concept for provenancing.

Although core groups differ only very little in composition, an assignment of a sample to such a chemical pattern (and with it to a production place, if the site of the pattern is known) is in most cases possible with sufficiently high probability. This is demonstrated for the two patterns Mycenae/Berhati and Achaia in Fig. 1b depicting again the result of a discriminant analysis. All other chemical patterns can be excluded as in the "normal" case (compare, e.g., Cr, La, Rb or Th of the main pattern assigned to Bronze Age Aegina, given as example also in Table 1 and in Fig. 1b). Only for some vessels a membership to one unique group can not be stated. For these samples which are located in the middle region between the centers of two core groups only assignments to these two core groups with different, non vanishing probabilities are deducible. In these cases, additional provenance determinations by other methods are needed.

## Conclusions

The assumption of a well homogenized clay paste to produce pottery of a certain production series seems in most cases justified, since well defined elemental concentration patterns with small spreads for most of the elements are normally obtained which characterize these single production series. Additionally to the need to measure many elements with high precision, experimental errors, dilution corrections and stable elements have to be considered when forming the groups. These sharp concentration patterns guarantee successful provenance determinations in most cases normally encountered. However, e.g., for some regions of

Mycenaean Greece, vessel compositions are measured which overlap for all elements except for a few. To separate these patterns, core groups have to be constructed at the cost of higher error probability when rejecting some samples as group members. A general disadvantage of this strict grouping procedure is, that (a) a large number of groups results and (b) a large number of control groups is needed to determine the provenance of each pattern. This makes for most projects the analysis of many samples necessary.

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