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Compositional variability of archaeological ceramics in the eastern Mediterranean and implications for the design of provenance studies

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ABSTRACT

The so-called 'chemical fingerprints' of production sites that are determined in provenance studies of archaeological ceramics comprise not only an estimate of the intrinsic chemical compositions but also an estimate of their variability. The compositional variability of ceramics from a specific production site is affected by the natural variability of the raw materials used, variation in the ceramic production process and potential post-depositional alteration. In order to characterise the production site as whole, average concentrations and their variations are estimated on the basis of a necessarily limited number of samples selected for analysis. The sampling strategy therefore has a significant impact on the results. The compositional variability is interfered from uncertainties introduced during the analysis subject to the analytical method chosen. This paper provides an overview of the sources of variability that influence such analyses. Case studies challenging the classical unbiased provenancing approach are presented using examples from the eastern Mediterranean region.

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1. Introduction

Provenance studies of archaeological ceramics represent one of the most popular subjects in the field of science-based archaeology. During the past few decades, several mineralogical and chemical techniques, and combinations from both, have been developed with the aim of understanding fabric or paste variability, grouping like materials and assigning a provenance to the resulting groups (Day et al., 1999; Maniatis et al., 1984). The first methodology to be investigated and developed for such provenance studies was based on elemental composition, which has been a well-established approach in archaeological science since the end of the1950s (Richards and Hartley, 1960; Sayre and Dodson, 1957), and the eastern Mediterranean has often been the focus of research and provided the basis for new developments in terms of analysis and data interpretation (Jones, 1986).

The basic tenet of provenance studies is that ceramics from a specific production site present a chemical composition distinct from ceramics produced elsewhere, as a result of the use of different raw materials and/or different methods of clay paste modification. This composition can be used to define a local chemical pattern or 'chemical fingerprint' of a production site or even a single workshop. In an unbiased approach, an arbitrary number of patterns can be attributed to a particular site and the assignment of provenance depends on a straightforward comparison of the resulting patterns. In practice, however, a series of constraints

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http://dx.doi.org/10.1016/j.jasrep.2017.03.020 2352-409X/© 2017 Elsevier Ltd. All rights reserved. has to be considered, such as the natural inhomogeneity of raw material sources.

The 'provenience postulate' assumes that chemical differences within a single source of material must be less than the chemical differences between different sources (Weigand et al., 1977). However, analytical studies of clay deposits have shown that, in particular, deposits from the same geological context sometimes present very similar chemical compositions, even though they can be considered different in terms of geographical distance. Furthermore, the natural range of element concentrations appears to be restricted in terms of correlations and in terms of absolute values (Hein et al., 1999). Case studies occasionally report that specific accessory minerals, not homogeneously distributed in the clay, can affect the element composition within a single clay source. The 'human factor' must also be considered a source of variability. It can be assumed that craftspeople within the same cultural context but operating at different sites tried to select raw materials with similar physical properties for the production of similar vessels. In many cases, such raw materials may belong to similar geological contexts and thus decrease the discriminative power of trace element analysis. Furthermore, modification of the clay paste can affect the chemical composition and its variability, for example if different raw materials are mixed together. Finally, the chemical composition of ceramic objects can be altered by environmental factors during burial.

Another type of constraint concerns the sampling strategy and analytical method used in provenance studies. The choice of method affects the subgroup of element concentrations that are determined, the analytical precision and, when the results are compared with reference data, the analytical accuracy. This has become a very important issue,

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as an increasing number of laboratory-based and portable techniques are routinely applied to ceramic studies, with very different performance levels, in terms of detection limit, precision and accuracy, affecting the variability of the measured assemblage.

In summary, the chemical composition of a group of archaeological ceramics is determined by absolute values but also the variability or uncertainty of those values. Without an estimation of this variability, the results cannot be interpreted in terms of similarity or dissimilarity among samples or the formation of a compositional reference group. In this study, the constraints and sources of variability described were examined and discussed using case studies from the eastern Mediterranean region. The ceraDAT database was utilised (Hein and Kilikoglou, 2012), which already holds more than 8000 records of archaeological ceramics from the region, produced from the Neolithic period until the Byzantine period, and more than 200 records of raw materials.

2. Statistical treatment of variability

2.1. Mahalanobis distance and best relative fit

The basic approach to chemical provenance studies is to create reference groups and patterns comprising similar ceramics, which represent specific production sites, and compare the composition of individual samples with the reference patterns. A reference pattern \mathbf{x} is a vector $(x_1, x_2, ..., x_n)$ comprising the average concentrations of n elements that are measured in the samples belonging to the reference group. A straightforward way to evaluate the variability of a reference pattern is to determine the covariance matrix S_x based on the individual sample compositions. The covariance matrix can theoretically be separated according to different sources of variability (Beier and Mommsen, 1994; Bieber et al., 1976):

$$S_x = S_{xN} + S_{xS} + S_{xA}$$

where S_{xN} is the natural variability or the true variability of the ceramic group, S_{xS} is the variability introduced by the sample selection and S_{xA} is the analytical uncertainty. Based on the covariance matrix of a reference pattern, the Mahalanobis distance can be calculated, to assess the similarity or dissimilarity of an individual sample of composition y with this pattern:

$$d_{x,y}^2 = {}^t(y-x)S_x^{-1}(y-x)$$

The Mahalanobis distance can be normalised with n, the number of elements considered, in order to compare different element suites. A similar but simpler distance, more suitable for small groups, is the squared Euclidian distance, weighed by the uncertainties, which can be estimated by the standard deviations of the individual element concentrations. When sample composition y is tested for similarity to the group composition x, the distance can be determined with:

$$d_{x,y}^2 = \sum_i \frac{(x_i - y_i)^2}{\sigma_i^2}$$

This distance corresponds to just the diagonal of the estimated covariance matrix in the Mahalanobis distance.

Additionally, a best relative fit (Beier and Mommsen, 1994; Harbottle, 1976) can be applied to adjust certain differences between two data vectors. If, for example a non-plastic temper, such as quartz or calcite, was added to the ceramics, these are normally poor in trace elements, which will result in a depletion of trace elements. On the other hand, the loss of molecular water or CO_2 at high firing temperatures results in a relative enrichment of trace elements. With a best relative fit, variation in the amount of temper material or firing temperature can be corrected for and the composition of a particular group of ceramics can be more precisely defined.

2.2. Variation matrix and total variation

An alternative approach to estimating the chemical variability of a set of compositional data is to determine their total variation, following the approach of Buxeda i Garrigos and Kilikoglou (2003). First the $n \times n$ variation matrix \mathbf{T} is generated with the matrix elements $\tau_{ij} = var\{\log(x_i/x_j)\}$, which present the variances of the logarithms of the element concentration ratios (Aitchinson, 1986). The total variation of the data is then given by:

$$vt = \sum_{ij} \frac{\tau_{ij}}{2n}$$

The sum τ_s of the variances in a particular column of the variation matrix gives the contribution to the total variation when element *s* is chosen as divisor. Therefore a high ratio of vt/τ_s indicates small variability of the respective element (Buxeda i Garrigos, 1999). On the basis of the variation matrix, an element with a small variability can be selected as a common divisor for a log-ratio transformation of the data, eventually excluding the above-mentioned effects on the absolute amount of trace elements. Because at the same time the concentration values are also normalised by the log-ratio transformation, the data can be studied with standard statistical procedures, such as hierarchical clustering or principal component analysis (Baxter, 1995; Baxter and Freestone, 2006).

3. Geochemical variation of raw materials: the provenience postulate

The basis of the compositional diversity of ceramics, enabling provenance studies by means of their composition, is primarily the variety of raw materials available for their manufacture: the clays of diverse genesis and origin, and temper materials that were potentially added and mixed with the clay paste. The underlying assumption that "differences in chemical composition between different natural sources ... exceed ... the differences observed within a given source" was expressed as the 'provenience postulate' by Weigand et al. (1977). The further assumption that an individual potter or a particular potting community was using a specific raw material source, preferably in the vicinity of the production site, implies that the eventual ceramic composition is characteristic of that workshop. Therefore, when the provenience postulate is applied to clay sources, the differences among pottery produced at different sites using essentially different clays can be expected to exceed the compositional variation within pottery produced from a specific clay. However, some case studies have highlighted difficulties that raise doubts about the general validity of the provenience postulate.

One example is the Minoan pottery from central Crete. Assumptions of a local origin for certain pottery wares produced at major palace sites have been challenged, questioning the validity of chemical reference groups used in provenance studies (Day et al., 1999). Ambiguities in analytical results from the pottery initiated a thorough investigation of raw material variability over the island (Hein et al., 2004). A series of 28 Neogene clay deposits was included as potential raw material sources for Minoan fine wares, and 60 clay samples were collected. Of the sampled deposits, five belonged to the Middle Miocene Viannos formation (Fig. 1). From one of these deposits, Agios Syllas, a series of 14 samples was taken in order to investigate intra-deposit variability. Analysis of the rock samples by neutron activation analysis (NAA) highlighted the difficulties of provenance studies of Minoan ceramics produced in the area from probably exactly those clays. Examination of the total variation of the samples on the basis of 22 element concentrations (Ca, Ce, Co, Cr, Cs, Eu, Fe, Hf, K, La, Lu, Na, Rb, Sb, Sc, Sm, Ta, Tb, Th, U, Yb and Zn) indicated a geochemical similarity among the five deposits of the Viannos formation, which was formed during the Miocene (Fig. 2) (Zachariasse et al., 2011). Following the common criteria of

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