Aeginetan Ware (AW) Provenancing Report: Characterization to Source Interpretation

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1.0 - Introduction

This report provides a brief summary of our provenancing results for the site of Kolonna itself (Kolonna Sample Set #1: Permit # YPPO/SYNT/F44/3235/52385, and Kolonna Sample Set #2: Permit # YPPO/SYNT/F44/2518/70941) and sample sets from three archaeological sites (Tsoungiza: Permit #YPPO/SYNT/F44/2881/60620, Halieis: Permit # YPPO/SYNT/F44/2055/37912, and Athens: Permit # YPPO/SYNT/F44/2225/37518).

On the basis of stylistic criteria, the samples from our Kolonna sample sets #1 and #2 carry an Aeginetan provenance. Likewise, the samples from Tsoungiza, Halieis and Athens are presumed to have an Aeginetan provenance. Based on our geochemical results, it does not appear that all of the sherds are from Aegina. Brophy has discerned a K-Na ratio in amphibole compositions for the South Aegean Volcanic Arc (SAVA), especially for Aegina. All provenance interpretations in this report are based on a comparison between sherd and raw materials and allowed a high probability provenance for ceramic material (on a single sherd basis) from the Aeginetan volcanic center. The results are summarized in Tables 1-5. The individual Na_2O-K_2O diagrams for every sherd are provided, as well as the geochemical data files used for source interpretation (see plots, tables and data files under supplementary materials).

2.0 - Critical Observations

2.1 Accepted Analysis of What AW represents

Based on stylistic and limited petrographic analysis, archaeologists have identified a distinctive group of ceramics known as "Aeginetan Ware". Ceramic production encompassed a wide range of products including table, storage and presumably cooking wares. These ceramics are believed to have been produced at a single production site, Kolonna, on the island of Aegina in the South Aegean Volcanic Arc during the Middle and Late Helladic periods of the Aegean Bronze Age (2000-1065 B.C.).

2.2 Our Analysis of What AW represents

Early on in our research it became apparent that "Aeginetan Ware" was not a monolithic ceramic product of Aegina; rather, it was a series of volcanic and metamorphic fabrics which together reflected specific ceramic functions and production locations. Thus, AW represents a functional ware of multiple origins and time periods. These observations are evident in the permitted sample sets.

Our research has not only further characterized AW (fine and coarse), but set the stage for a new interpretation of long-standing ideas about cultural change, i.e. artifact distribution, craft specialization, and local technological influence. We now need to conceive of the idea that specific fabrics were exploited in the Aegean area for their physical, mineralogical and chemical characteristics.

2.3 Fabric Issue

Based on our extensive fieldwork, the AW sherds that have metamorphic fabrics have no clay source counterpart on Aegina. One must conclude that these fabrics, although similar to AW in physical appearance, were not produced on Aegina.

There are two other potential clay sources for AW on Aegina Island: (1) an Early Pliocene marl, and (2) a Pleistocene terra rossa soil. On the basis of textural and mineralogical grounds these two sources were discounted. We are confident that the volcanic ash deposit, located in the northwestern part of Aegina, represents the dominant, if not the sole clay source material, used in the production of local AW.

The small number of AW samples with a fine-grained volcanic fabric is somewhat enigmatic. They are dominated by a very fine-grained matrix reminiscent of either the marine marl or the terra rossa. But, they also contain very small fragments of multiple volcanic minerals (e.g. quartz, feldspar and hornblende) and small volcanic rock fragments. These latter features indicate that they are best grouped with the local volcanic fabric and represent a volcanic ash clay source.

3.0 - Methods: Developing a Quantitative Methodology for Ceramic Provenance

3.1 Previous Comparative Provenance Research

Shriner has previously sought to develop a quantitative method for the use of source reference materials for comparative provenance for Early Bronze Age ceramic artifacts produced at the site of Lerna. The result (Shriner, 1999; Shriner and Dorais, 1999) was a provenance study that employed the Electron Microprobe (EMP) to characterize the chemical composition of different types of mineral grains in the various source materials, thus providing a reference standard against which one could compare the chemical compositions of the same type of minerals grains in any given artifact. Because the source materials were derived from the weathering of *metamorphic* rocks, the different minerals used in this study were of metamorphic origin (e.g. muscovite, biotite, chloritoid, etc.)

Building on this initial provenance work, Shriner extended the EMP approach to potential sources of *volcanic* origin in the South Aegean Volcanic Arc (SAVA). The goal was to use the compositions of volcanic minerals (e.g., hornblende, biotite, feldspar) to characterize *potential* source materials on the different volcanic centers of SAVA (e.g. Aegina, Melos, Poros) and use this as a basis for determining the origin (i.e. provenance) of specific artifacts known to have been produced from clay material of volcanic origin. The initial research of Dorais and Shriner (2002a, 2002b) resulted in a characterization that did, indeed, permit one to provenance specific artifacts to specific South Aegean volcanic islands.

The goal of the present work has been to build upon and possibly simplify the work of Dorais and Shriner. In so doing, we have collected significant amounts of additional mineral composition data from *potential* source materials (both rock and clay-rich sediments) on the islands of Aegina, Melos, Poros, Santorini, and the peninsula of Methana. The minerals in these reference materials were then analyzed by electron microprobe techniques to develop a mineral composition databank against which mineral compositions in individual ceramic artifacts can then be compared.

The bulk of our efforts have been devoted to identifying and characterizing the *actual* clay source for ceramics on Aegina Island. Brophy, Christidis and Shriner conducted field work for potential clay sources on Aegina in the summers of 2002, 2003 and 2005. With the data from our Integrated Petrologic Approach (i.e. integration of data sets from Petrography (PE), X-ray diffraction (XRD), Rare Earth Elements (REE), and electron microprobe analysis (EMPA), we identified an extensive archaeological clay deposit on the island (Shriner and Brophy, 2003; Shriner et al., 2003, 2005, 2007, 2008; Christidis et al., 2008). Bulk chemical analysis allowed us to link this deposit to published chemical data (Mommsen et al., 2001) for Kolonna ceramics.

3.2 Hornblende Composition as a Provenancing Tool - Rationale

Many ceramic artifacts in the Aegean are produced from clay-rich material created by the alteration of felsic[1] volcanic lava and/or ash. Typically, such lavas or ashes contain fragments of minerals that were growing in the molten magma before it erupted onto the surface as lava or ash. During the alteration process, these mineral fragments are usually preserved intact. As discussed below, these minerals provide a potential means of determining provenance of a given ceramic artifact.

The original study of Dorais and Shriner (2002a, 2002b) and Dorais, Lindblom and Shiner (2004) utilized the compositional variations of a single volcanic mineral, hornblende, for ceramic artifact provenancing. The current investigation follows their lead. An important mineral property of hornblende is that it displays a feature known as solid solution. The mineral hornblende, with the general chemical formula shown below,

$$K_{0\text{-}1}(Ca, Na)_{2\text{-}3}(Mg, Fe^{+2},Fe^{+3},Ti,Al)_5Si_6(Si, Al)_2O_{22}(OH)_2$$

displays this characteristic in a complex fashion. Ultimately, a given hornblende crystal can have varying amounts of K, Na, Ca, Mg, Fe^{+2}, Fe^{+3},Si, Al and Ti!

Why is solid solution of potential importance when it comes to provenancing ceramic artifacts? A fundamental property of minerals that crystallize in a magma is that their chemical composition *reflects the chemical composition of the magma itself* (e.g. Carmichael et al., 1975). For example, a magma with a high ratio of Na_2O/K_2O would yield hornblende crystals that themselves had a high ratio of Na_2O/K_2O. A magma with a low ratio of Na_2O/K_2O would yield crystals with a low ratio of Na_2O/K_2O. A magma with a high ratio of MgO/FeO would yield a hornblende crystal with a high ratio of MgO/FeO and vice versa. In short, since hornblende displays the property of solid solution, the compositions of any given hornblende crystal will be a direct reflection of the composition of the magma from which it originated. Moreover, if potential clay sources for ceramic artifacts were derived from lavas and/or ash deposits that came from different composition magmas, then the unaltered minerals in the different clay sources would also have different compositions. Thus, the composition of hornblende has the

[1] Igneous rocks are classified on the basis of how much silica (SiO_2) they contain. Rocks with <53% SiO_2 (by weight) are called mafic. Intermediate rocks contain 53 50 63 % SiO_2 and felsic rocks contain >63% SiO_2.

potential for identifying the clay source material from which a given ceramic artifact was produced (i.e. its provenance).

In hornblende, two elemental ratios that are particularly sensitive to small differences in magma composition are Na_2O/K_2O (e.g. Sisson and Grove, 1993) and TiO_2/Al_2O_3 (e.g. Dietrich et al., 1988). In principle, either one of these element ratios in hornblende could be used for provenancing purposes. However, it is also known that Al in hornblende is very sensitive to crystallization pressure (e.g. Hollister et al., 1987); thus, making the TiO_2/Al_2O_3 in hornblende sensitive to both magma composition *and* pressure. This makes it a less reliable indicator of magma composition alone. For this reason we have chosen to concentrate on the Na_2O/K_2O ratio in hornblende as a potential method for provenancing ceramic artifacts derived from volcanic sources.

4.0 - Geological Reference Material Sample Collection, Preparation and Analysis

For any given island or volcanic center, multiple samples were collected from dacitic lava flows and/or altered volcanic ash horizons. The sampling was guided by published geologic maps (Aegina: Dietrich et al., 1991; Methana: Dietrich et al., 1995 and Fytikas et al., 1972; Poros: Dietrich et al., 1991; Melos: Fytikas, 1977; Santorini: Druitt et al., 1999). All attempts were made to collect at least one sample from each of the volcanic units identified on the maps. Sample collection on Aegina is discussed in further detail below.

For every dacite lava sample, the following procedure was employed. Fist-sized samples of rock were cut into 0.5" slabs using conventional water-cooled diamond rock cutting saws. The location and orientation of the slab was always randomly chosen. Each slab was then further reduced to 1.5"x .75"x 0.5" rectangular billets which were then sent to a commercial outfit for final hi-polished thin section preparation.

For selected samples of the volcanic ash horizon on Aegina Island (see Figure 4), the following procedure was employed. The sample was first gently disaggregated by hand using a large mortar and pestle. The sample was then passed through multiple sieves of progressively smaller sieve size, thus separating the sample into different size fractions. The different size fractions were then studied with a binocular dissecting scope to identify both fragments of hornblende phenocryst-bearing dacite rock fragments as well as mineral fragments of hornblende. Individual rock-fragments and mineral fragments were hand-picked and sent off for commercial hi-polished thin-section preparation as grain mounts.

The major element compositions of individual hornblende phenocrysts or mineral fragments were determined by wavelength-dispersive X-ray analysis at 15 kV, using a CAMECA SX50 electron microprobe, located in the Department of Geological Sciences, Indiana University. Major elements were analyzed using a 2μm beam with a beam current of 20nA and a peak counting time of 20 seconds. Analytical reproducibility was within 2%. The accuracy of the analyses was monitored using reference materials of known compositions. Elements analyzed included: Si, Ti, Al, Fe, Mn, Mg, Ca, Na and K. For a given mineral analysis wt. % abundances were determined for the following oxides: SiO_2, TiO_2, Al_2O_3, Fe_2O_3, FeO, MnO, MgO, CaO, Na_2O and K_2O. Raw counts were converted to oxide concentrations using an on-line PAP correction scheme of Pouchou and Pichoir (1985).

If available, 8 individual hornblende phenocrysts were analyzed for each dacite lava sample. Typically, three individual analyses were performed on each phenocryst including one in the core, one at the rim and one in the middle. This approach yielded a maximum of 24 hornblende analyses per sample. Though no statistical tests were performed, it was assumed that this large number of analyses was sufficient to fully characterize the range of hornblende compositions in any given sample. For the grain mounts, the number of hornblende analyses of both phenocrysts in dacitic rock fragments as well as individual mineral fragments was dictated by the number of hornblende crystals present. Again, three analyses (core, middle, and rim) were typically conducted on each crystal.

Throughout the course of this investigation, multiple persons were responsible for the actual EMP analysis but only one person (Brophy) was responsible for a quality control check before a given analysis was incorporated into the reference databank. The first step was to check for the quality of the analysis**.** Any analysis that had total oxide abundances (including H_2O) less than 95% was excluded from further consideration (see protocols for data manipulation file under supplementary materials). The second step was to determine whether or not the mineral that was analyzed was indeed hornblende. If it turned out to be some other mineral (typically augite or biotite) it was excluded from further consideration. The third step was to check for internal stoichiometric consistency (i.e. the relative proportions of oxide abundances had to fall within certain parameters dictated by the crystalline structure of hornblende). Only those analyses that passed this final test were incorporated into the reference data set. Finally, every analysis was "normalized" via the following procedure: (1) wt. % Fe_2O_3 was converted to wt. % FeO and then summed with the existing FeO to yield a "total iron" called FeO*; (2) wt. % H_2O was set equal to zero; (3) the abundances of the remaining oxides were summed to yield a total oxide abundance; and (4) each oxide abundance was divided by the total oxide abundance and then multiplied by 100 to yield a "normalized" oxide abundance on an "anhydrous" basis. The purpose of this normalization procedure is to permit more direct comparison of individual mineral analyses.

5.0 - Ceramic Sample Collection, Preparation and Analysis

In 2003 the Greek government and Drs. Felten and Hiller permitted us to re-section 81 samples from Mommsen et al.'s (2001) original Kolonna collection. In the summer of 2003 Shriner (with the permission of the Greek government and Drs. Camp, Papadopoulos, Pullen, Rotroff, Rutter, and Wright) developed AW sample sets for Tsoungiza, Halieis, and Athens. These three sample sets were cut and exported to Indiana University in September, 2004. Drs. Berger and Gauß developed a pivotal new 78-piece technological sample set for Kolonna. The sample set was cut and exported to Indiana University in September, 2005. All sample sets were built with ceramics of presumed Aeginetan provenance.

Ceramic sherds were prepared for hornblende analysis in a fashion similar to that employed for the Aegina dacitic lava samples. From each sherd a thin (approximately 0.125 to 0.25" thick) slab was prepared using a slow-speed water cooled high precision diamond saw. This slab was then sent off for commercial hi-polished thin section preparation. The analytical procedures, conditions and strategy for analyzing hornblende crystals in the sherds were identical to those described above. Within a given sherd, a distinction was made between hornblende phenocrysts in dacitic rock fragments and individual hornblende mineral fragments. Identical procedures for quality control and data normalization were also employed.

6.0 - Hornblende Na_2O-K_2O Variations in the Aegean Arc

The South Aegean Volcanic Arc consists of several volcanic islands that are a result of northward subduction of the African Plate beneath the Aegean Plate (Figure 1). Variations in the Na_2O and K_2O (and therefore Na_2O/K_2O) of erupted magmas in many subduction zones are known to vary considerably (e.g. Dickinson and Hatherton, 1967; McDonald et al., 2000). The Aegean arc is no exception with significant along-arc and across-arc variations in K_2O (and therefore Na_2O/K_2O) (e.g. Keller, 1982). Given that different volcanic islands have different Na_2O/K_2O ratios, this feature should be reflected in the compositions of hornblende crystals in erupted lavas and/or ash deposits on different islands. It turns out that hornblende is an important mineral only in the western portion of the arc including the islands/centers of Santorini, Melos, Methana, and Aegina.

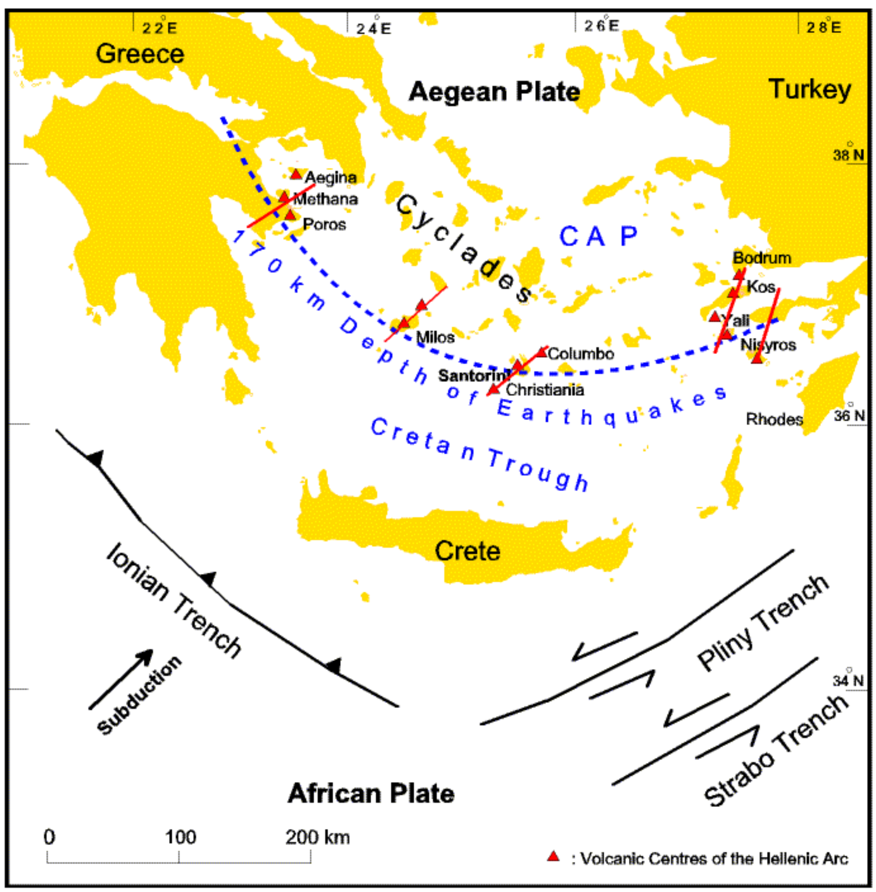


Figure 1. Tectonic Map of the Aegean Volcanic Arc.

Figure 2 shows a plot of Na_2O and K_2O abundances of individual hornblende crystals in volcanic rocks from each of four islands/centers: Aegina, Methana, Melos, and Santorini. From this diagram it is seen, first, that the hornblende crystals from each of the islands display a significant amount of chemical variation. Aegina and Santorini define distinct fields that are different from one another as well as Melos and Methana. On the one hand, hornblendes from Methana and Melos cannot be distinguished from one another on the basis of Na_2O-K_2O. However, it is also apparent that the hornblende crystals from Aegina define a field that is characterized by higher K_2O abundances than the other islands.

The numerical data in the searchable database (see on SAVA database website) or the excel data files (see under supplementary materials) can be used to constrain the provenance of a given ceramic artifact of volcanic origin by the Na_2O-K_2O relations of their hornblende mineral fragments. For example, when specific sherds (e.g. K132 or HP 294) are compared with the numerical data for each island, one can see that K132 could not have been derived from Melos, Methana or Santorini, but could have been derived from Aegina. HP294 could not have come from Aegina or Santorini, but could have come from Melos or Methana. Though not completely satisfactory, the Na_2O-K_2O relations of hornblende crystals in erupted volcanic rocks can be used to "fingerprint" the specific island that they come from.

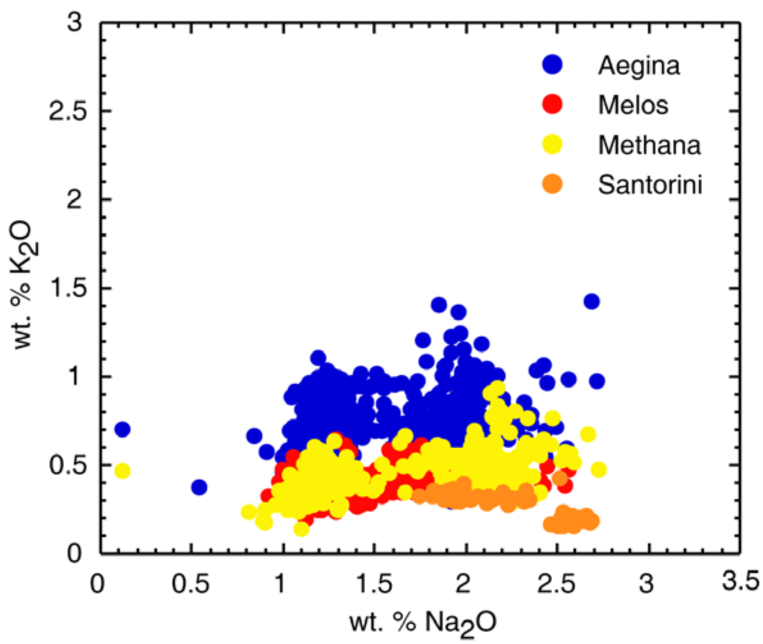
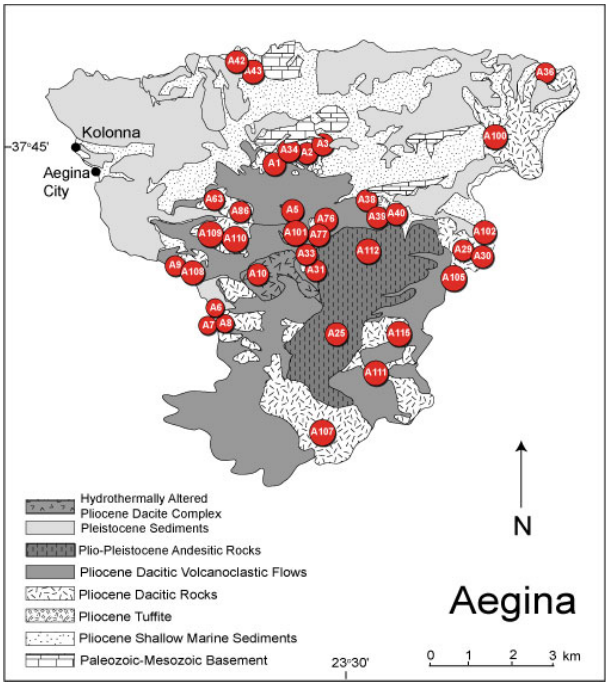


Figure 2. Na2O-K2O of individual hornblende phenocrysts in volcanic rocks from the islands or centers of Aegina, Melos, Methana and Santorini.

7.0 - Hornblende Na_2O-K_2O Variations for Aegina Island

Much of our work has concentrated on provenancing ceramic artifacts, identified as Aeginetan Ware, to the island of Aegina. Consequently, our mineralogical studies from Aegina are summarized in more detail. The Aegina Island reference data set consists of several thousand analyses of hornblende phenocrysts and/or mineral fragments from 37 samples of dacite lava and 17 samples of volcanic ash collected from across the island (see Figures 3 and 4 for sample locations). For dacite lavas, the geologic base map of Dietrich et al. (1991) was used as a sampling guide. The strategy was to collect at least one and preferably more than one sample from each of the 8 dacite units identified and mapped.



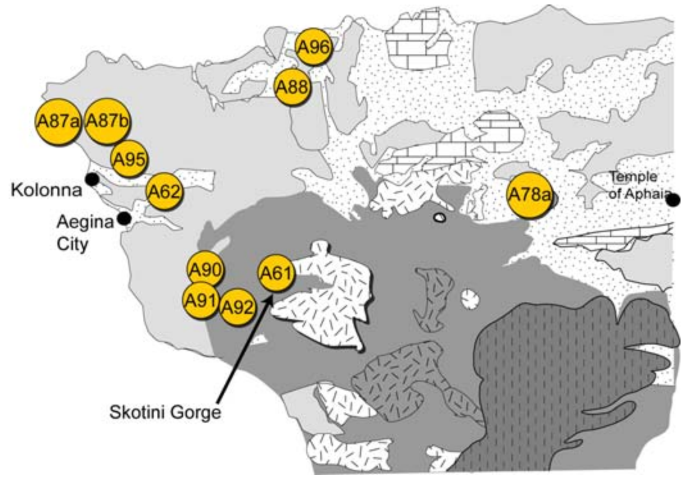


Figure 3 (left) – Map of Aegina with rock sample locations.

Figure 4 (right) – Map of Aegina with clay-rich sediment sample locations.

Our own field work led to the identification of a widespread volcanic ash horizon (now altered to clay) in the northwestern part of the island. The horizon is exposed continuously along the shoreline and at isolated inland localities. Samples (see Figure 4) were collected from 4 shoreline and 3 inland localities. At any given locality multiple samples were collected along a vertical section that covered the entire thickness of the horizon.

8.0 - Provenancing Results

Figure 5 shows a plot of Na_2O-K_2O on which are shown the compositions of all analyzed hornblende crystals from the Aegina lavas (rocks) and clay deposit (mineral and rock fragments). When taken all together, the hornblende results define a weak bi-modal distribution with the first mode centered around 1.25% Na_2O and 0.7% K_2O and the second mode centered around 1.9% Na_2O and 1.0% K_2O. The results indicate a fairly tight clustering of data points with only a few spurious “outliers”. We assume that this tight cluster represents the range of hornblende compositions on the island of Aegina. The compositions of hornblende crystals in any ceramic with an Aeginetan provenance should fall within the ranged defined by the first mode and, preferably, within that defined by the second mode as well. Any ceramic with hornblende compositions that fall outside this cluster are assumed to have been produced somewhere other than Aegina.

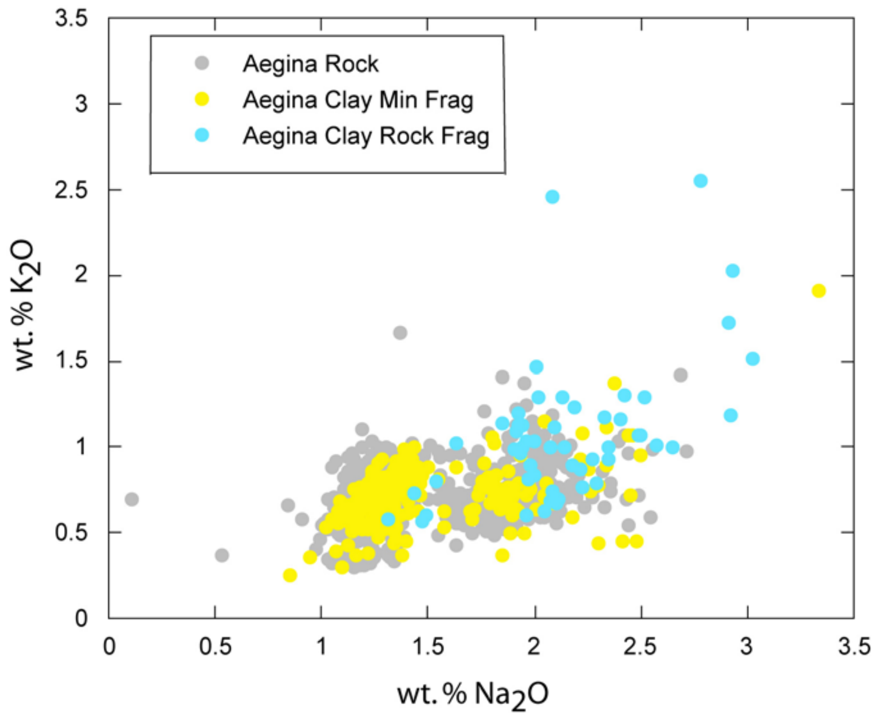


Figure 5 – Composite Na_2O-K_2O diagram showing the compositions of individual hornblende phenocrysts and mineral fragments from multiple dacitic lava flows and a clay-rich altered volcanic ash deposit on Aegina Island.

8.1 *Nature of the Data*

We have opted not to use a statistical "confidence" bubble to delimit Aeginetan provenance. Instead, Brophy has used only the raw amphibole data from Aegina rocks, clay-rich sediments and rock fragments within sediments in his interpretation (see figure 5). This may appear to be a conservative approach to provenancing, but grew out of archaeologists' need to document each ceramic on a one-to-one basis (see discussion 9.1).

There are other ways in which the data files can be organized. In some of the Aegina clay-rich sediment samples amphibole composition is modal. There are some sherd samples that also contain only one mode of amphiboles (see plots under supplementary materials). At this time, these ceramics have been provenanced to Aegina. In addition, the Aegina unit from which sample A112 was taken is considered by Dietrich (1991) a hi-alumina basalt. It can be removed from the database as the geochemical compositions only reflect dacites.

8.2 *Further Provenance of Ceramic Artifacts to Aegina*

The underlying premise of our work is that any ceramic produced on Aegina must: (1) be derived from volcanic clay sources, and (2) have hornblende crystals that fall within the range of hornblende compositions displayed by lavas and/or volcanic ash deposits on Aegina. This work has led to a working definition for Aeginetan ceramics that is based, in part, on the compositions of hornblende crystals present in a given artifact (Brophy et al., in prep.) Two parts of the working definition are pertinent to this report:

1. The fabric must be of volcanic origin such as that shown in Figure 6. A typical volcanic fabric consists of fine grained altered volcanic ash with or without

mineral fragments of feldspar, quartz and/or hornblende and with or without rock fragments consisting of the same minerals.

2. The Na_2O-K_2O characteristics of any constituent hornblende crystals must fall within the range of potential source rocks on Aegina Island shown in Figure 5.

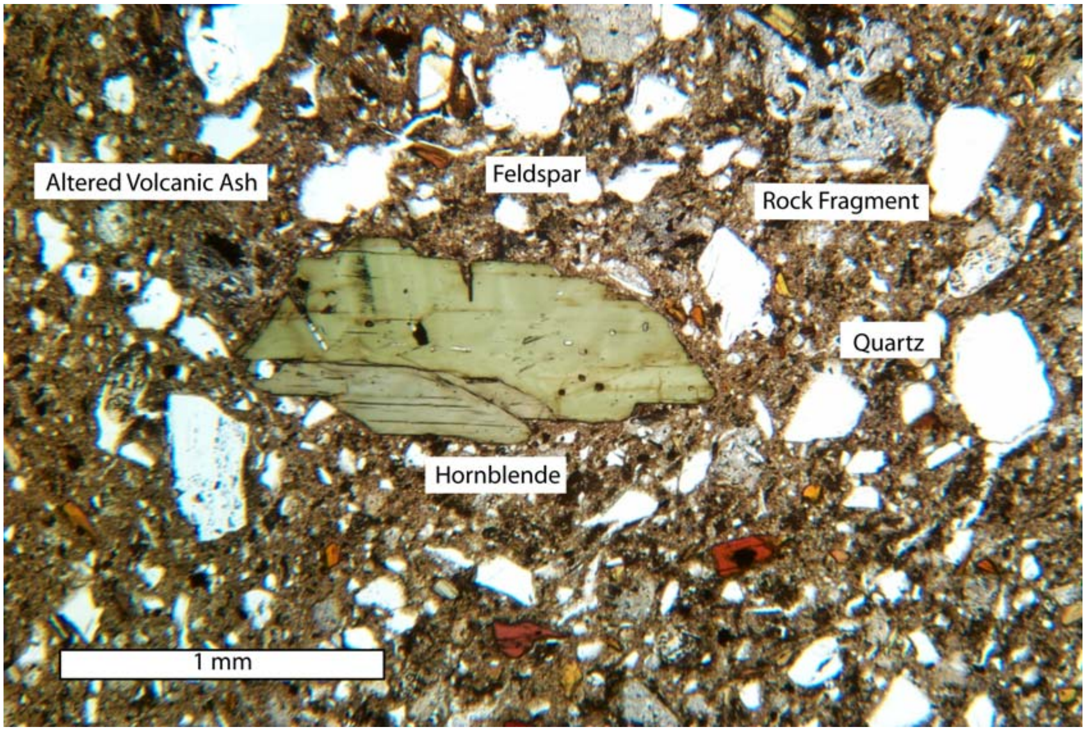


Figure 6. Photomicrograph that shows those features which constitute a "volcanic fabric".

The identification of any given sherd as being "from Aegina" or "non-local to Aegina" is based on criteria 1 and 2 above. For each sample a 5-step procedure was followed:

Step 1 – Thin sections were prepared from all sherds

Step 2 – All sherds were examined with a petrographic microscope

Step 3 – If a given sherd had a volcanic fabric, it went to the next step. If it did not have a volcanic fabric (typically a metamorphic fabric), it was immediately labeled as "non-local to Aegina".

Step 4 – The remaining sherds were separated into those that contained hornblende and those that did not. Sherds that did not contain hornblende were tentatively labeled as "from Aegina".

Step 5 – The remaining hornblende-bearing sherds were analyzed with an electron microprobe (EMP) to determine the compositions of multiple hornblende crystals within each sherd. The Na_2O and K_2O abundances were then plotted on a Na_2O-K_2O diagram (see plots under supplementary materials). If the hornblende compositions in a given sherd fell within the range for Aegina (see Figure 5), it was labeled as "Aegina". If the compositions fell distinctly outside of the range for Aegina, it was labeled "non-local to Aegina".

9.0 - Discussion

9.1 Significance of Individual Artifacts

As a provenance study, this work demonstrates the power of an approach that characterizes multiple individual mineral grains in ceramic fabrics and compares them to geological samples. This is an innovative approach that makes the mineral grains within the sherd the target population in contrast to bulk characterization studies, in which the target population consists of the sherds themselves. The power of the approach is that it opens up the possibility of addressing source assignment even for single sherds (see plots under supplementary materials).

For example, if we had shown all amphibole data from ceramics of a particular class or type (e.g. Aeginetan Kitchenware, Aeginetan Plain, and Aeginetan Matte-Painted) with a single symbol, it would make it impossible to assess whether there might be differences between individual ceramics from the same class or type. There could be subtle differences in fabrics which could not be discerned because of the use of the same symbol for samples from the same type. Essentially, what this practice does is make the amphiboles in a given ceramic "type" the target population rather than the amphiboles in an individual sherd. So, we are asked to agree that types are as real as the discrete sherds themselves.

9.2 The Study of Emergent Complexity: An Enlarged Definition

The original archaeological problem for this project was the quantitative verification of an Aeginetan provenance for the presumed archaeological distribution of Aeginetan Ware (AW) in the Greek Bronze Age. One of the more recent goals of our study has been to develop a quantitative definition for Aeginetan Ware that will permit specific archaeological problems to be solved that warrant a comparative mechanism for interpretation. A study of emerging social complexity is one of those interpretive problems.

Distribution and craft specialization are two important observational criteria for the emergence of a complex society. As an example, the distribution of AW in the Aegean is an archaeological representation of the spatial and temporal extent of not only the ceramic's production and exchange, but evidence for the emergence of complex cultural organization. Presumably, the distribution of excavated or surveyed artifacts can define the spatial extent, the temporal duration, and the intensity of social interaction for a given society. The underlying assumption is that the greater the spatial distribution over time, the greater the organizational complexity of the society. As only observational data, there is no mechanism for site to site comparison of these criteria. We are merely describing how a specific observed assemblage is distributed in space and time. It is an idealized form of distribution. In order to explain a process of related observations from site to site, we need an extended definition for AW – one that can incorporate quantitative data. Not a definition that would replace existing definitions, but would enlarge the interpretive ability of the definition as a whole. Quantitative data can help us to explain the actual distribution of this ware and the specialization that the ware implies.

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