Bronze Age pottery from the Aeolian Islands: definition of Temper Compositional Reference Units by an integrated mineralogical and microchemical approach

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Abstract An integrated microchemical–petrographic approach is here proposed to discriminate the provenance of archaeological pottery artefacts from distinct production centres. Our study focuses on a statistically significant sampling (n = 186) of volcanic temper-bearing potteries representative of the manufacturing and dispersion among the islands of the Aeolian Archipelago during the Bronze Age. The widespread establishment of new settlements and the abundant recovery of Aeolian-made ceramic in southern Italy attest for the increased vitality of the Archipelago during the Bronze Age. The increasing importance of the Archipelago is documented by the importation of Mycenaean pottery (Late Helladic I–II) during the late stages of the Capo Graziano culture.

1 Archaeological framework

The Bronze Age marks the renaissance of the Aeolian Islands. The Archipelago, is a key archaeological context since Early Neolithic because of the availability of obsidian in Lipari, the only island continuously inhabited as a central place in the Archipelago. The islands knew a long decline during the Copper Age because of the progressive abandon of lithic technologies.

The Archipelago is located in a strategic position in the southern Tyrrenian Sea (Fig. 1) allowing the control of maritime circulation between Aegean, eastern and western Mediterranean, Sicily and peninsular Italy [1–3] i.e. on the main routes pointing to the Sardinian mineral resources. Consequently, new settlements appeared on the islands at the onset of the Bronze Age, during the Capo Graziano culture (Early to Middle Bronze Age 2; 2300–1430 BC).

The increasing importance of the Archipelago is documented by the importation of Mycenaean pottery (Late Helladic I–II) during the late stages of the Capo Graziano cul-
ture at Lipari, Filicudi and Stromboli (Capo Graziano II; 1700–1430 BC; [4–8]).

At the same time Aeolian pottery is documented in maritime exchanges as for instance the finding of the shipwreck of Pignataro di Fuori [9, 10] near the Lipari coasts, unique evidence for the Italian Bronze Age, and several occurrences of pottery with the peculiar Capo Graziano style in Sicily [11–18] and Italian mainland [19–23]. The typical Aeolian pottery classified as impasto ware, resulting by mixing clay and a relatively coarse temper, is shaped by mould or coil, burnished and decorated with incised or impressed geometrical patterns. The social organisation of this pottery production can be classified at the domestic workshop level [24, 25]. Capo Graziano settlements, typically composed of oval stone huts, are initially situated (Capo Graziano I; 2300–1700 BC) on coastal plains such as the Contrada Diana in Lipari [26] and Filo Braccio in Filicudi [27, 28]. Starting from the late Capo Graziano culture, a general movement of the villages is observed, possibly induced by defence needs (Capo Graziano II; 1700–1430 BC). Naturally defended places, such as the Acropolis of Lipari [1], the Montagnola of Filicudi [27] and the plateau of San Vincenzo in Stromboli [8, 29] are settled at this time. During Capo Graziano II, pottery is characterised by a typical decorative style with incised zig-zag (a stylised representation of the sea), lines and alignments of points.

2 Rationale and sampling

Since late 1960s John Williams analysed the whole pottery sequence (from Early Neolithic to Late Bronze Age) efficiently discriminating Aeolian productions from the imported extra-Aeolian ones based on petrographic observations [30, 31]. The distinction is made possible by the different petrographic character of tempers being of volcanic origin those of the Aeolian Archipelago while containing sedimentary and metamorphic clasts those from the Calabrian and Sicilian grounds. Volcanic tempers used for Aeolian pottery production are in fact sands deriving by the natural dismantling of the rock units outcropping nearby a given workshop. Aeolian Islands consist of seven stratovolcanoes belonging to an active, subduction-related magmatic arc (200 km long) growing in the southern Tyrrhenian Sea (Fig. 1). Subaerial volcanism of the Aeolian Islands having started at ∼0.22 Ma BP is still quiescent at Lipari and Vulcano and persistently active at Stromboli [32]. Basaltic to rhylotic extrusive series are present on all islands. Lava and pyroclastic products belong to calcalkaline (CA), high potassium calcalkaline (HKCA), shoshonitic (SHO) and, more rarely, potassic (KS) affinities [33, 34].

According to Williams studies, local productions prevailed during Early and Middle Bronze Age, with evidence of self-sufficient productions in Lipari and Filicudi, later replaced by imported and/or local products using imported clay from northern Sicily [30, 31, 35, 36]. The high probability of common clay sourcing makes difficult to distinguish the intra-archipelago workshops based on bulk compositional analyses because tempering may not significantly affect group distinctiveness until large amount of temper are added [37, 38]. On the other hand, if tempers are locally added they would reflect the local geochemical fingerprint. Provenance studies should thus focus on this component in order to track the location of the manufacturing centres. A study of potteries from the same region has, however, demonstrated that petrography alone is not sufficient for a correct provenance attribution even when tempers of volcanic origin are used [39]. This is mainly due to the similar compositional evolution of the volcanic activity in the region that makes challenging a safe attribution to a compositional group based on the pure petrographic characters even for a well trained observer.

Moving from this background we set up a microchemical approach to assess the robustness of petrographic classification with the aim of defining Temper-based Compositional Reference Units to allow easy attribution of a given sample to a production area and/or craft tradition. This study re-considers samples from Williams collection along with additional samples from Lipari Museum and two new excavations (San Vincenzo, Stromboli [8] and Filo Braccio, Filicudi [28]) and few occurrences from the shipwreck of Pignataro di Fuori recovered off the coasts of Lipari. Pottery from Stromboli, the northernmost island of the Archipelago, has never been analysed before. The investigated collection includes pots of different shapes and functions covering
the entire Capo Graziano chronological sequence in Lipari 
\((n = 46)\), Filicudi \((n = 56)\) and Stromboli \((n = 80)\), plus few 
from the shipwreck of Pignataro di Fuori \((n = 4)\).

Samples with non-volcanic-temper, abundantly found at 
Stromboli and more rarely on the other islands, attesting a 
Calabrian and/or Sicilian provenance, are not investigated 
in the present paper.

3 Methods

Mineralogy and petrography were determined on thin petro-
graphic sections by a polarising microscope. Point counting 
(at least 500 points for each thin section) was performed on 
139 thin sections, whose area is sufficiently large to ensure 
a modal distribution representative of sample variability.

Modal distribution data have been treated by multivariate 
statistics in order to get a classification of pottery reflecting 
the compositional variability and the archaeological site of 
provenance. Principal Component (PCA) and Discriminant 
Analysis (DA) have been performed using SPSS 17.0 statis-
tical package (Table II, electronic supplementary material).
This approach has been helpful for the definition of the com-
positional groups and the identification of possible imports.

Major element composition of minerals and glasses was 
assessed by electron microprobe. Analyses were carried out 
with the Superprobe Jeol JXA 8200 at the Eugen F. Stumpfl 
Laboratory of the Leoben University (Austria). Analytical 
conditions are the following: acceleration voltage 15 kV, 
beam current 10 nA, focused beam (≈1 µm diameter), peak 
counting time 20 s and background 10 s. Analytical data 
were corrected through the ZAF method. A set of natural and 
synthetic standards have been used for internal calibration.

In-situ trace elements have been measured at Centro In-
terdipartimentale Grandi Strumenti (CIGS) at the Universi-
ità di Modena e Reggio Emilia using a Nd:YAG deep UV 
(213 nm) New Wave Research UP-213 laser ablation sys-
tem (LA) coupled to a Thermo Fisher Scientific X-SeriesII 
Induced Coupled Plasma Mass Spectrometer (ICP-MS). In-
strumental drift correction was computed by linear correct-
ion of measured analyte intensities among repeated mea-
urements of the NIST 612 glass. \(^{44}\)Ca has been routinely 
employed as internal standard based on electron microprobe 
CaO contents. LA–ICP–MS spectra were obtained with unit 
mass resolution. They require few if any interference correct-
dues, to very low molecular and doubly charged species 
production. In this study ThO\(^+\)/Th\(^+\) was maintained below 
0.5 % and Ba\(^{2+}\)/Ba\(^+\) below 0.1 %. Analytical routine com-
prises 100 µm pre-ablation scan (dwelltime: 2 s, 5 Hz 
laser fire, laser fluency 18 \(\div\) 20 J/cm\(^2\)) followed by 80 µm 
ablation scan (dwelltime: 30 s, 20 Hz laser fire, laser flu-
cy 18 \(\div\) 20 J/cm\(^2\)). Data reduction performed with Plasma 
Lab\(^\circ\) software, by Thermo Scientific.

4 Results

4.1 Mineralogy and texture

In his pioneering works, Williams [30, 31] proposed a clas-
sification based on petrographic criteria, identifying three 
main groups named A, B and C. Group A (with nine sub-
groups AI to AIX) includes pottery of Aeolian production 
bearing volcanic-derived tempers, ranging from basaltic-
andesitic to rhyolitic in composition [33, 34]. Group B con-
sists of pots imported from North Sicily and Calabria, con-
taining clasts of metamorphic and plutonic rocks. Potteries 
of group C are interpreted as Aeolian production but char-
acterised by mixed temper of both metamorphic or plutonic 
and igneous origin. Here we focus on samples pertaining 
to petrographic group A to check the robustness of the pet-
rographic classification and infer possible intra-archipelago 
relationships. We build over Williams’s work keeping when 
possible his structure and definitions. His classification can 
be tested and improved by a statistical approach considering 
quantitative parameters defined by petrography and point 
counting such as: abundance of mineral and glass phases, 
compositions and abundance of lava clasts (e.g. basalt and 
basaltic andesite vs. andesite and rhyolite), modal ratios of 
plagioclase/pyroxene, mineral/lavas and glass/lavas. There 
result a total of 13 different components to operate for statis-
tical analyses (Table I, electronic supplementary material).
Statistical distribution have been computed on 139 samples 
(see methods), analyses parameters and output are resumed 
in Table II (electronic supplementary material). When sam-
iples are divided by their site of sampling, multivariate anal-
yses show a good separation for the three islands and some 
“intruders” representing possible exchanges among the three 
sites (i.e. Lipari, Filicudi and Stromboli). Plots of PCA and 
DA (Figs. 2A–2B) show a clear distinction of Lipari popu-
lation (abundance of glass) from Filicudi and Stromboli 
(abundance of lava clasts). Notwithstanding the overlapping 
of Filicudi and Stromboli shown in the PCA plot, some com-
positional differences (clinopyroxene at Filicudi and pumice 
at Stromboli) are emphasised by DA where the two Islands 
result more separated (80 % of cross validated grouped cases 
correctly classified).

PCA and DA based observations along with modal min-
eralogy and texture allow clustering the investigated sam-
ples in four groups. Three of them: AI, AIV and AVIII 
strictly correspond to Williams description [35], while a new 
group AX is characteristic of Stromboli. Simple pie dia-
ograms of modal distribution of the main characters reveal 
the basic differences among the groups (Fig. 3). Compo-
sitional groups clearly show up when PCA is run by dis-
regarding the site of sampling (Figs. 2C–2D). Under these 
conditions PCA shows a significant separation of groups AI 
and AX while group AVIII overlaps with AIV (in particu-
lar the AIV from Filicudi). The difference among the four
Fig. 2  (A) Principal Component Analyses (PCA) based on 13 measured petrographic components (mineral, rock and glass clast abundance) grouped based on their site of sampling.  
(B) Discriminant Analyses (DA) of the same population showing a good separation between Lipari population and the other two islands (Filicudi and Stromboli). (C) PCA and (D) DA of the whole population without the limit of the sampling site. The four different compositional groups (AI, AIV, AVIII and AX) are clearly separated. Compositional groups are defined in the text.

Groups arise from DA (Fig. 2D). Here the group AIV is in an intermediate position between AX and AVIII allowing the use of these statistical set to help classifying a given sample.

Iteratively crossing PCA and DA analyses and petrographic observation we refined the compositional characters of each group estimating 98% of cross validated cases are correctly classified in the petrographic groups. Results are resumed in Table 1. The lower cross validation when PCA and DA account for the site of sampling attests for dispersion of elements pertaining to a given compositional group in other sites. Accordingly statistic disregarding the site of provenance clearly separates the compositional groups. It appears that AI mainly occurs on Lipari and AIV is restricted to Filicudi and Stromboli with the majority on the former island. AVIII is characteristic of Filicudi while AX is typically found at Stromboli.

The resulting petrographic characters of these groups can be confidently described as follows.

Group AI: Temper is characteristically rich of colourless fresh volcanic glass fragments and pumices (Figs. 3, 4A and 4B) and less plagioclase and pyroxene phryic lava clasts (basaltic andesite and andesite) with cryptocrystalline groundmass. Y-shaped glass shards (Fig. 4B) or perlite fragments (Fig. 4C) can be also present. Plagioclase, the most abundant mineral phase (Fig. 3) is strongly zoned and sieve-textured, enriched in glassy inclusions (Fig. 4D). Pyroxene (both monoclinic and orthorhombic), locally shows resorption embayments and poikilitic inclusions. Few dacitic to rhyolitic clasts containing quartz and K-feldspar are also present (Fig. 4E). Olivine (generally altered to iddingsite), sanidine, xenoliths of quartzite and scoriae rarely occur. Hydrous phases are almost lacking, except of rare horn-

| Group | Lipari | Filicudi | Stromboli |
|-------|--------|----------|-----------|
| Group AI | 27 | 3 | 2 |
| Group AIV | 38 | 14 |
| Group AVIII | 1 | 11 |
| Group AX | 2 | 41 |
blende. Temper grains are subangular, generally finer than medium sand. This group is mainly present in Lipari, subordinately in Filicudi and Stromboli (Table 1).

Group AIV: Temper mostly consists of mineral phases and lava clasts with very low to negligible glass content (Fig. 3). Minerals are plagioclase, clinopyroxene and scarce orthopyroxene. Hydrous minerals are represented by euhedral brown hornblende and subordinate green hornblende and biotite. They usually show opaque rims (Fig. 4F). Andesitic lava clasts with micro- to cryptocrystalline groundmass and phenocrysts of plagioclase, pyroxene and hydrous minerals are also present. Olivine (iddingsite), quartzite xenoliths and scoriae may locally occur. When present, volcanic glass shows sub-rounded morphologies, with orange and brown colours and palagonitic devitrification. Temper grain size do not exceed medium sand with unimodal or bimodal distribution. This group is well attested both in Filicudi and Stromboli (Table 1).

Group AVIII: Large abundant grains (up to 2 mm) of plagioclase, clinopyroxene and orthopyroxene, along with some pyroxene-plagioclase glomerocrysts represent a distinct feature for this group (Fig. 3). Lava clasts (basaltic to basaltic andesite lavas; Fig. 4G) with oxidised or crypto- to microcrystalline groundmass are
mainly characterised by pyroxene as single phenocrysts or glomerophyric aggregates, together with iron oxides and plagioclase. Glassy clasts (scoriae and palagonite) are very rare. Hydrous phases are absent or limited to few green hornblende grains. Pyroxenes, both rhombic and monoclinic, may represent up to 34% (average 27.3%, Fig. 3) of the temper, largely prevailing over plagioclase (average 17.6%, Fig. 3). The temper does not exceed medium sand grain size, it is poorly sorted or with bimodal distribution. This group is typical of Filicudi pottery, mainly from the earlier village of Filo Braccio. One sample is from Lipari (Table 1).

Group AX: Temper contains abundant plagioclase, less clinopyroxene, orthopyroxene and hydrous phases. Green hornblende without opaque rims (Fig. 4H) is more abundant than any other group (Fig. 3). Lava clasts (basaltic andesite to andesite) are characterised by subrounded morphologies and large clasts (up to 7 mm), with pyroxene phenocrysts and cryptocrystalline groundmass. Pumices, quartzite xenoliths and spherulitic glass are frequent. Iddingsitised olivine is rare. The temper is poorly sorted and coarse with clasts often larger than 2 mm and subangular morphologies. Samples belonging to this group are found in the archaeological site of Stromboli with the exception of two from Filicudi (Table 1).

4.2 Mineral chemistry

Group separation can be independently verified based on major and trace element variability of the observed temper phases. If petrographic groups reflect local productions then the chemical composition of the tempers must reflect the composition of local lava suites i.e. the geochemical flavour of that given volcanic centre. Hence we performed micro-chemical in-situ analyses by Electron Microprobe (major elements) and Laser Ablation ICP-MS (trace elements) on clinopyroxene, plagioclase and hornblende.

Lipari samples (AI) show a bimodal distribution for clinopyroxene in the Na$_2$O vs. FeO$_{tot}$ diagram (Fig. 5): one region includes samples from the shipwreck of Punta Pignataro along with AI samples from Stromboli and Filicudi. The compositional field of samples pertaining to group AIV from Filicudi and Stromboli completely overlap with group AVIII from Filicudi and the AX group from Stromboli. In general major elements for both ortho- and clinopyroxenes do not allow separate groups AIV, AVIII and AX.

Trace elements in clinopyroxene give indication for a different distribution of group AX. Group AIV from Filicudi and Stromboli are not resolvable from AVIII. However, in all trace element systematic they present an unimodal distribution while group AX samples always trend bimodally with a cluster plotting toward markedly enriched compositions. Rare Earth Elements content of clinopyroxene is a useful tool to distinguish the characters of parental lavas. Figure 6 shows the relationship between Eu$^*$ and Sm$_N$, where Eu$^*$ is a measure of the depth of the Eu anomaly calculated as Eu$_N$/((Sm$_N$ - Gd$_N$)$^{1/2}$) and the “N” subscript indicates that values are normalised to chondrite composition [40]. When clinopyroxene crystallises along with or after plagioclase, Eu in the residual melt is preferentially extracted from the melt with respect to the other REEs because of the compatible behaviour of Eu$^{2+}$ in plagioclase. Samples from Li-
Fig. 7 Compositional variability of K\textsubscript{2}O vs. Ca\# in plagioclase of the four volcanic temper-based Aeolian pottery groups sampled at the different archaeological sites. Ca\# = CaO/(CaO + Na\textsubscript{2}O + K\textsubscript{2}O) here used as a proxy of anorthite content.

Fig. 8 Compositional variability in hornblende defined as FeO\textsubscript{tot}/TiO\textsubscript{2} vs. MgO/K\textsubscript{2}O. Large symbols represent the average and 1\sigma error bar relative to petrographic groups AIV, AVIII and AX. Subgroups AIV from Stromboli and Filicudi are plotted separately although they show no statistically significant separation.

5 Discussion and conclusions

Based on modal mineralogy and texture of the Aeolian Bronze Age pottery we recognised four statistically separated groups (AI, AIV, AVIII and AX) among the volcanic temper-bearing pots collected at Lipari, Filicudi and Stromboli archaeological sites. These groups we confidently assume as Temper Compositional Reference Units for the specified context [41]. They match those proposed by Williams [31, 35] with the addition of a new unit (AX) [42] recently found in the Stromboli archaeological site of S. Vincenzo [8]. The inferred petrographic subdivision is independently tested by mineral chemistry (major and trace elements) of representative phases such as clinopyroxene, plagioclase and hornblende, giving a robust statistical framework for sample identification.

Unit distribution among the islands mainly match with the provenance sites (Fig. 2, Table 1). The geographical distribution of AI, AX and AVIII is almost exclusively restricted to the localities where they have been collected, respectively Lipari, Stromboli and Filicudi (Fig. 2, Table 1). A minor amount of samples pertaining to a given unit is found in a different island attesting for an active intra-archipelago exchange network. Their limited number is, however, not statistically relevant to infer reliable exchange fluxes. Overall, collected data support the hypothesis of independent, domestic-workshop, production centres in each insular community. This is coherent with the observation that Italian Bronze-age communities locally produced their own pottery because of a poorly hierarchical society where work division was not yet completely settled.
The existence of a flourishing production in Filicudi is also suggested by the local production of a unique masterpiece: the cup with an incised decoration representing a complex pattern with sea waves, human figures and boats [28].

A controversial case is that of unit AIV. Stromboli and Filicudi AIV subsets completely overlap in all mineral compositional fields (both major and trace elements) and petrography, strongly supporting their provenance from a single production centre, independently from their occurrence in different Aeolian Islands. Samples of this unit have been found at Filicudi (73 % of the total) and Stromboli (27 %). This observation may attest for a strong exchange network between these two islands. To date archaeological evidences suggest the occupation of Filicudi to predate the settlement of Stromboli [8, 28]. This may alternatively suggest that the abundance of Filicudi-made pottery at Stromboli could be evidence for import from Filicudi to Stromboli before the onset of a local manufacturing resulting in production of AX pottery.

Other considerations arise by comparing AIV and AVIII distribution in Filicudi. Their difference only lies in mineral modal distribution and petrographic characters (Fig. 3) while completely overlap in the compositional systematic (Figs. 5–8). This aspect may suggest the adoption of different formulae in a given workshop oriented to obtain different technical properties or esthetical effects linked to the transition from the lower village of the phase CGI (where AIV and AVIII are alternatively employed) to the upper village of the phase CGII (where AIV predominates).

Overall, there is no correspondence between composition and shape/function of the vessels attesting for a general adoption of the same pasteware for the whole local production set (mainly bowls, cups, and jars).

Crossing petrographic data (modal mineralogy and texture) and mineral chemistry is a reliable approach to unravel the provenance of an artefact in a network of different production centres and commercial exchanges. Provenance of a pottery, even a small shard, can be robustly inferred by this approach. On the other hand some potentialities ask for further extend elemental analyses to the whole population of mineral, glass and lava clasts used as temper and compare them with the compositional variability of the volcanic rocks of the Archipelago.

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