Papers in Italian Archaeology VII

The Archaeology of Death
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edited by
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For the Accordia Research Institute
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Chemical characterization of EBA/MBA pottery from Ognina (Sicily).
A comparison of XRF and pXRF for analysis of ancient pottery

Davide Tanasi, Robert H. Tykot, Frederick Pirone, Erin McKendry

Summary
The archaeological exploration of the Ognina islet, near Syracuse, Sicily, carried out in 1964 provided the evidence of a long term prehistoric occupation between the Neolithic and the Middle Bronze Age. Maltese style ceramics were found in the Early and Middle Bronze Age layers. The small group of imports, belonging to the Thermi Ware, was found in connection with the local Castelluccian Ware (EBA), while the Borġ in-Nadur ware, also Maltese in style, was related with local Thapsos ceramics (MBA). In 2012, during a fieldwork, a large amount of ceramics were recovered, among which were some new examples of Thermi and Borġ in-Nadur wares, as well as large amounts of Castelluccian and Thapsos pottery. In order to ascertain whether the Maltese type pottery was imported from that small island, a program of archaeometric analyses was established. Diagnostic samples belonging to both Maltese-like and Sicilian pottery classes were analysed with destructive thin sectioning and X-ray fluorescence spectrometry (XRF) and subsequently with nondestructive pXRF together with a sample of Sicilian clay taken from the source closest to the islet. The analyses demonstrated that the two Thermi Ware samples were locally produced, while three out of four Borġ in-Nadur pieces were produced in Malta and one was produced in Sicily.

Keywords: Maltese pottery, Sicily, XRF, pXRF, imports, local imitations

1. pXRF or XRF that is the question?

The chemical characterization of archaeological materials has played an important role in the study of prehistoric exchange networks. In particular, the use of a portable or hand-held X-ray fluorescence spectrometer (pXRF) has become increasingly more popular in ceramic sourcing studies in recent years due to a number of advantages that include the ability to non-destructively analyze ceramic materials on location such as at excavations or at museums where ceramic collections are housed or on display and the overall affordability in analyzing a large number of artifacts within a relatively short period of time. While these advantages are attractive to researchers, it is important to note that non-destructively analyzing ceramic surfaces has a technical disadvantage compared to homogenized powder samples. However, a number of non-destructive ceramic studies have been performed taking into account the heterogeneous nature of clay types, surface treatment and decoration such as the application of slip or paint, and temper added (Speakman et al. 2011; McCormick 2013; Tykot et al. 2013; Ashkanani and Tykot 2013; Stremtan et al. 2014; Hunt and Speakman 2015). These studies provide examples in how to address the issue of heterogeneity in non-destructively analyzing only ceramic surfaces.

The portable XRF has brought sophisticated scientific equipment to museums and excavations with broad
enthusiasm and many expectations. It is truly the first laboratory machine to be widely adopted by the mainstream archaeological community, considered as the vehicle of a methodological revolution that only social theory had achieved previously, especially for its not being invasive or destructive. Yet, pXRF studies have been sparse, and adopting scientific methodologies has proven challenging in a discipline still divided between humanities and sciences. Furthermore, a common standard of practice is also lacking. For these reasons, the traditional XRF is generally preferred and conventionally considered apt to provide more reliable results. Both techniques have such pros and cons that it is definitely hard to make a choice among them. On one hand the pXRF enables rapid analysis of large numbers of sherds, because it is portable and non-destructive, which is important for identifying outliers and having statistically large groups to compare. But on the other it can’t be a total substitute for regular XRF (or Instrumental Neutron Activation Analysis or ICP) which may make even further subdivisions of pottery groups using trace elements below the pXRF’s detection limits (e.g. Lanthanum, Cesium, Barium and others).

2. A Mediterranean connection: Sicily and the Maltese archipelago in the Bronze Age

In the frame of the interconnections between Mediterranean prehistoric societies, the Sicily-Malta interaction has been a leitmotiv deriving from the geographic proximity of the two insular contexts and by a substantial cultural homogeneity over centuries. The evidence on which the academic debate is centered is often represented by Maltese ceramics found in Sicily and vice-versa, apparently pointing to a ‘mobility of goods’ rather than a ‘mobility of men’ as the main phenomenon of this interaction (Tanasi and Vella, 2014).

Pottery being the main indicator of exchanges, along with the inability of distinguishing between imports and local imitations due to the absence of archaeometric characterizations, has misled scholars, preventing us in having a correct perspective over this issue (Biehl and Rassamakin, 2008).

The archaeological implications of being not able to distinguish between those two classes of artifacts has left room in the studies of Sicilian and Italian prehistory to a proliferation of hypotheses about commercial and colonial routes especially with regard to a relationship with the Aegean (Tanasi, 2005). Those hypotheses supported and shared by the scholars for decades have been recently put aside after the spread of archaeometric analyses as a new starting point in the approach of the material culture.

In this perspective, the most important case study is represented by the evidence of Middle Bronze Age Sicily, where a large amount of Maltese pottery has been found mainly along the southeastern coast (Tanasi, 2008). The Middle Bronze Age in the Maltese archipelago is characterized by the Borġ in-Nadur culture, chronologically ranging between the second half of the 15th and the early 12th century BC and subdivided into three phases, Early, Middle and Late (Copat et al. 2013; Cazzella and Recchia, 2012; Tanasi, 2015a). This Maltese facies is partially coeval with the development of that of Thapsos, which represents all the cultural production of Middle Bronze Age Sicily.

The evidence of Borġ in-Nadur pottery in Thapsos contexts, often stressed as a critical phenomenon of cultural interweaving (Tanasi, 2010; 2011; 2014; 2015b), has pointed to a hypothetical strong commercial relation between the islands in this period. The discovery of Thapsos pottery in a few Maltese sites further supported this hypothesis (Tanasi, 2008).

However, the recent literature has pointed out how the simple macroscopic analysis of pottery and consequent fabric grouping can be systematically denied by archaeometric examination, emphasizing a need of abandoning the traditional archaeological approach of visual observation in the study of pottery (Maniatis, 2009). In this perspective, it appears obvious that in absence of proper archaeometric analyses carried out on the materials on which the Borġ in-Nadur – Thapsos frame is set, any further interpretations in this direction turn out to be totally pointless.

A rich assemblage of prehistoric pottery, collected during a recent fieldwork carried out on the islet of Ognina (Siracusa, Sicily) has been chosen as a case study for a comparative application of pXRF and XRF in order to assess the validity and reliability of the pXRF outcomes and to classify the fabrics and eventually to determine different geographic provenance. The scientific exercise has also shed some light on the complex question of Maltese presence in Sicily between the Early and Middle Bronze Ages, on the basis of the evidence coming from Ognina.

3. The islet of Ognina (Siracusa, Sicily)

The islet of Ognina 8 (Fig. 1) is located 9 km south of Siracusa and until the medieval period it was connected to the mainland by a narrow isthmus connected with the opposing canal harbor. The terrain of the islet has been drastically eroded over the centuries and nowadays the only preserved archaeological deposit is limited to its central part. In 1964, Luigi Bernabò Brea undertook the first archaeological explorations of the islet uncovering traces of occupation ranging from the Neolithic to the Middle Ages (Bernabò Brea, 1966). On the southern side of the islet, he documented 14 systems of aligned fish-farm tanks excavated in the rock and dated to the
The Archaeology of Death

EBA. The same chronology was assigned to an isolated rock-cut chamber tomb with a long entrance corridor and megalithic cover, found on the eastern side. In the central part of the islet, Bernabò Brea opened several test pits where he observed a stratigraphic sequence documenting a prehistoric settlement with main phases of occupation in the Neolithic, EBA, and MBA, with a gap in the Copper Age. The only structure identified was a complex building which developed through various phases into an early medieval Byzantine church. With regards to the prehistoric phases, the pottery collected belonged to five main classes: Stentinello, Castelluccio and Thapsos which are, respectively, Neolithic, EBA and MBA; and two further classes of uncommon ware which are traditionally interpreted as Maltese imports: Thermi Ware, found in connection with the local Castelluccian Ware (EBA), and Borg in-Nadur ware, related with local Thapsos ceramics (MBA) (Fig. 2).

The discovery of Maltese type pottery at Ognina is part of the well-known theme of the contact between the two islands during prehistory, an unavoidable phenomenon considering the proximity of Malta to Sicily and the outreach of their maritime transport (Tanasi, 2014).

The Thermi type ware, in first place considered a distinctive production of EBA Malta (Bernabò Brea, 1966), also attested in a few domestic contexts of southeastern Sicily (Palio 2008), has been recognized as the indicator of a mass migration from continental Greece and the Balkans which reached southern Italy, Sicily and Malta (Recchia and Fiorentino, 2015). Of the vast repertoire of shapes that this production shows in Malta, the Sicilian examples are limited just to one typology, the deep bowl with thickened rim decorated with incised and impressed geometric motifs.

The interpretation of the Maltese-type artifacts as imports resulted in the Ognina islet being considered a Maltese ‘colony’ in Sicily, a controversial hypothesis never dismissed (Bernabò Brea, 1966; Trump, 2004-2005).

This picture changed in the MBA with a substantial improvement in the presence of Maltese type pottery. Almost 100 examples of Borg in-Nadur type pottery summarizing the entire shape repertoire of this production on Malta have been identified in a number of domestic and funerary contexts of southeastern Sicily, testifying to the development of a tightening relation between the two islands in this period.
After almost fifty years since its exploration, in summer 2012, a team from Arcadia University, led by one of the authors (Davide Tanasi) undertook a survey of the islet and the adjacent coast in order to reappraise the evidence uncovered and only preliminary published by Bernabó Brea, and to create an archaeological map of this territory.

In terms of findings, the survey produced the same classes identified before and also a good number of Thermi and Borġ in-Nadur pottery.

But how can it be established whether the examples of Thermi ware and Borġ in-Nadur ware are imports from Malta and not products made in Sicily imitating Maltese ceramic prototypes?

4. Petrographic and chemical characterization through XRF

Moving on with that research question, a total of 63 samples of pottery, including Neolithic, Castelluccio, Thapsos, Thermi ware and Borġ in-Nadur pieces, out of a total of 95 diagnostic pieces collected on the islet of Ognina, were subjected to archaeometric analyses (Ranieri et al. 2015).

- 52 were examined with a portable X-ray fluorescence spectrometer (pXRF), and 11 with both a traditional X-ray fluorescence spectrometer (XRF) and by petrographic analysis of thin sections with optical microscopy (OM).
- 10 samples, including Sicilian and Maltese type pottery were analyzed with both pXRF and XRF techniques for comparing the results.
- The data-set included samples of Maltese and Sicilian clays and pottery from the Maltese site of Borġ in-Nadur.

The preliminary petrographic analyses pointed to a substantial difference between the group of Thermi ware type and Borġ in-Nadur type samples.

Sixty-nine samples, comprising 58 samples of ceramics and clays from the site of Borġ in-Nadur, and 11 pottery samples from Ognina and clays from various Sicilian districts, were analyzed with this Philips X-ray fluorescence spectrometer.
In order to highlight the compositional differences between materials coming from Sicily and Malta, chemical data have been treated through principal components analysis (PCA). The very promising results emphasized also a very good correspondence between the petrographic observation and the groups recognizable in the scatter plot. The variables having the highest variance were taken for setting up the discriminating triangular diagram Zr-Rb-La, in which more evident is both the discrimination between Maltese and Sicilian pottery and raw materials (Ranieri et al. 2015, p. 39, fig. 7).

5. Chemical characterization through pXRF

For ceramics in this study, careful attention was given to analyze ceramic surface with relatively flat areas and that showed no signs of slip or application of paint or decoration. Additionally, multiple spots on both the inside and outside surfaces of each sherd were analyzed and attention was given in order to avoid analyzing locations where there were visible inclusions.

A total of 48 ceramic samples from Borg in-Nadur, a prehistoric site located on the island of Malta, 49 ceramic samples discovered at Ognina, Sicily, 19 geological clay samples from two location on the island of Malta (Gnejna Bay and Selmun), and 4 geological clay samples taken from the northern outskirts of Siracusa, Sicily, were analyzed using a Bruker Tracer III-SD pXRF instrument. The Maltese samples represent three cultural phases, the Tarxien phase of Malta’s Temple Period, the Tarxien Cemetery phase (Early Bronze Age), and Borg in-Nadur phase (Middle Bronze Age). The Ognina samples represent two chronological periods, the early and middle Bronze Age. Analyses were conducted using the settings 40kV/10µA and filter (12 µm Al, 1 µm Ti, 6 µm Cu), providing greater precision and sensitivity for trace elements Th, Rb, Sr, Y, Zr, and Nb. The Bruker Tracer III-SD was positioned upright on a plastic stand and the samples carefully balanced on top (Fig. 3). Both the inner and outer surface and occasionally the edges for each of the ceramic samples

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<tr>
<td>Fabric</td>
<td>Medium coarse, with microfossil-rich groundmass grog and carbonation microfossil rich groundmass</td>
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<tr>
<td>Groundmass</td>
<td>Heterogeneous, with brownish-yellow color and medium micromass optical activity</td>
</tr>
<tr>
<td>Microstructure</td>
<td>Not preferentially oriented and single spaced channels, vughy and vescicles</td>
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<tr>
<td>Inclusions</td>
<td>Grog and fine quartz with a bimodal grain size distribution</td>
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<th>Borg in-Nadur</th>
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<tbody>
<tr>
<td>Fabric</td>
<td>Medium coarse; dominant grog and carbonatic microfossil-rich groundmass</td>
</tr>
<tr>
<td>Groundmass</td>
<td>Heterogeneous, with brownish-greyish color and high/medium high optical activity</td>
</tr>
<tr>
<td>Microstructure</td>
<td>Channels and planar voids with remains of carbonaceous material, vughy and preferentially oriented vescicles</td>
</tr>
<tr>
<td>Inclusions</td>
<td>Unimodal/bimodal grain size distribution, coarse grog with prevalently sub-angular shape and millimetric dimensions and common fine quartz</td>
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Figure 3. The Bruker Tracer III-SD portable X-ray fluorescence spectrometer (pXRF) analyzing a ceramic sherd at the National Museum of Archaeology, Valletta, Malta (photograph by Fred Pirone).
were analyzed for 120 seconds. Quantitative values in ppm for each trace element were obtained by calibrating the raw data using the 2008 MURR calibration software. These values were then analyzed statistically using principal components analysis with the IBM SPSS Statistics 23 software package.

Principal components analysis, using trace element data for Th, Rb, Sr, Y, Zr, and Nb obtained from the pXRF, was conducted on all the Maltese and Sicilian ceramic and geological samples. The results of this analysis show that the majority of the Maltese and Sicilian ceramics included in this study can be separated into groups based on whether the clay used to produce the ceramic wares was from a Sicilian or Maltese clay source (Fig. 4). The results further suggest that either raw clay materials or finished pottery moved with individuals traveling between Sicily and Malta during the Bronze Age and potentially prior in the Tarxien Phase of the Maltese Temple Period (Pirone, 2016). Variation within each of the groups consisting of ceramics made from either a Maltese or Sicilian clay source is best interpreted as the use of multiple clay outcrops in the pottery production that took place at either Borg in-Nadur or Ognina.

The above results were compared to the elemental compositional data for 69 samples, comprising ceramic and clays from Borg in-Nadur (58 samples) and pottery from Ognina (11 samples), analyzed with a Philips X-ray fluorescence spectrometer. The 11 Ognina samples included in this analysis were included also in the analysis used with the pXRF. The elemental compositions for Zr, Rb, and La showed the greatest variation and were therefore used to create a discriminating triangular diagram (Fig. 5). As with the results obtained using the pXRF, the analysis shows that the majority of the Maltese and Sicilian ceramics can be separated into groups based on the clay source used in the production of these wares. Both methods, either where the surfaces of the ceramics were analyzed non-destructively using the Bruker Tracer III-SD or when a powdered sample was taken for each of the ceramic sherds and analyzed by the Philips X-ray fluorescence spectrometer.
spectrometer, allow for distinctions to be observed between Maltese and Sicilian ceramics and clays.

6. Final remarks

Comparing the results, the distinction between the Maltese and the Sicilian groups appears strikingly clear with both techniques, with the only exception sample OG/151 which in the scatter plot of the data obtained with pXRF seems to be an outlier although it falls close enough to the group of the Maltese clays.

The analyses demonstrated that the two Thermi Ware samples were locally produced, while three out of four Borg in-Nadur pieces were produced in Malta and one was produced in Sicily. These results strongly suggest some mobility of artisans from Malta to Sicily, an interpretation that could reinforce the hypothesis of Ognina having been an emporium with different cultural groups present, if not a ‘colony’.

This study assessed an old interpretation adding targeted new data, which revealed the presence of both Maltese pottery imports and local imitations. Its methodology demonstrates also that pXRF analyses, despite some technical limitations, are effective in investigating certain research questions.

The chance to use a portable device with a non-destructive technique will allow us to move to the next step of this research which would be the analysis of all the Thermi ware type and Borg in-Nadur type pottery previously found at Ognina. This would verify the hypothesis of an exclusive Sicilian imitation of Maltese pottery in the EBA and of a greater likelihood to import Maltese pottery in Sicily in the MBA.
In this perspective, an extension of the analyses to all the Maltese type pottery from the other sites and mainly kept at the archaeological museum of Siracusa, will allow us to attempt a further distinction between imported products and local imitations on a larger scale. The more robust will be our data set of Maltese and Sicilian sampled ceramics and clays the more chance we will have to reconsider the problem of the relationship between Sicily and Malta between the EBA and MBA and to answer to the question whether it was characterized by cycles of mere exchanges of goods and mobility of people.

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