Original Study

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Provenance Study of Prehistoric Ceramics from Sicily: A Comparative Study between pXRF and XRF

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Abstract: The 1964 archaeological exploration of the Ognina islet near Syracuse, Sicily, has provided evidence suggesting a long-term prehistoric occupation from the Neolithic to the Middle Bronze Age. Maltese style ceramics were found in Early and Middle Bronze Age layers. A small group of imports belonging to the Thermi Ware culture was found in connection with the local Castelluccian Ware (EBA), and Maltese style Borġ in-Nadur wares were recovered with local Thapsos ceramics (MBA). During fieldwork in 2012, large amounts of ceramics were recovered including 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 Malta, a program of archaeometric analyses was established. Diagnostic samples belonging to both Maltese-like and Sicilian pottery classes were analyzed with destructive thin sectioning and X-ray fluorescence spectrometry (XRF) and subsequently analyzed with non-destructive portable X-ray fluorescence spectrometry (pXRF) together with a sample of Sicilian clay taken from a clay source close to the islet. The analyses demonstrated that the two Thermi Ware samples were locally produced and three out of four Borġ in-Nadur pieces had a Maltese provenance while one of the four being 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 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 such as the

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application of slip or paint, decoration and temper added (Speakman et al. 2011; McCormick 2013; Tykot et al. 2013; Ashkanani & Tykot 2013; Stremtan et al. 2014; Hunt & Speakman 2015). These studies provide examples in how to address the issue of heterogeneity in non-destructively analyzing only ceramic surfaces.

pXRF has afforded the opportunity to bring sophisticated scientific equipment to museums and excavations and has created broad enthusiasm along with many expectations. Many researchers have considered pXRF a vehicle for a methodological revolution that only social theory had previously achieved. It is truly the first laboratory device to be widely adopted by the mainstream archaeological community especially with pXRF having the ability to non-destructively analyze artifacts. Yet, pXRF studies have been sparse and the adoption of new scientific methodologies has proven to be a challenge in a discipline still divided between the humanities and the sciences. Furthermore, a common standard of practice is also lacking. For these reasons, the traditional XRF is generally preferred and conventionally considered more apt to provide reliable results. Both techniques have pros and cons that make it hard to choose which device to employ in a research program. On one hand 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. On the other hand, it can't be a total substitute for regular XRF or other instrumental devices such as Neutron Activation Analysis or ICP, which may allow for 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

The Sicily-Malta interaction sphere has been a leitmotiv for the study of interconnections and mobility among Mediterranean prehistoric societies 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 individuals' as the main phenomenon of this interaction (Tanasi & Vella, 2014). Pottery has been seen as a main indicator of exchange among various communities; however, the inability of distinguishing between imports and local imitations due to the absence of archaeometric characterizations has misled scholars by preventing us from having a correct perspective onthis issue (Biehl & Rassamakin, 2008). The archaeological implications of not being able to distinguish between these 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). Many of those hypotheses that have been supported and shared by scholars for decades have been recently put aside due to the spread and application of archaeometric analyses.

It is within the context above that the Middle Bronze Age in Sicily serves as an important case study 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 & Recchia, 2012; Tanasi, 2015a). This Maltese facies is partially coeval with the development of Thapsos, which represents all the cultural production of Middle Bronze Age Sicily.

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

However, recent literature has pointed out how simple macroscopic analysis of pottery and consequent fabric grouping can be systematically denied by archaeometric examination, emphasizing a need to abandon 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 from which the Borġ in-Nadur-Thapsos frame is set, any further interpretations in this direction risks being totally pointless.

The rich assemblage of prehistoric pottery collected during recent fieldwork carried out at 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 the different geographic provenance for the recovered pottery. This scientific exercise has also shed some light on the complex question of the 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 (Fig. 1) is located 9 km south of Siracusa and until the medieval period it was connected to the mainland by a narrow isthmus 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 Bernabó Brea undertook the first archaeological explorations of the islet uncovering traces of occupation ranging from the Neolithic to the Middle Ages (Bernabò Brea, 1966; Cultraro, Crispino 2014). On the southern side of the islet, he documented 14 systems of aligned fish-farm tanks excavated in the rock, the chronology of which is still being debated. The same chronology was assigned to an isolated rock-cut chamber tomb found on the eastern side of Ognina having a long entrance corridor and megalithic cover. In the central part of the islet, Bernabó 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 has 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 Borġ 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 prehistoric theme of contact between the island of Sicily and the Maltese archipelago, which is an unavoidable phenomenon considering the proximity of Malta to Sicily and the outreach of their maritime transport (Tanasi, 2014).

The Thermi Ware, 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 an indicator of mass migration from continental Greece and the Balkans that reached southern Italy, Sicily and Malta (Recchia & 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 Maltese-type artifacts as imports has led some scholars early on to hypothesize that the Ognina islet was 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 Borġ 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 during this period. After almost fifty years since Ognina’s initial exploration, in the 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 further create an archaeological map of this territory. The 2012 survey produced the same classes identified before and also a good number of Thermi and Borġ in-Nadur pottery. However, questions with regard to whether the examples of Thermi Wares and Borġ in-Nadur wares are imports from Malta or products made in Sicily imitating Maltese ceramic prototypes arose.

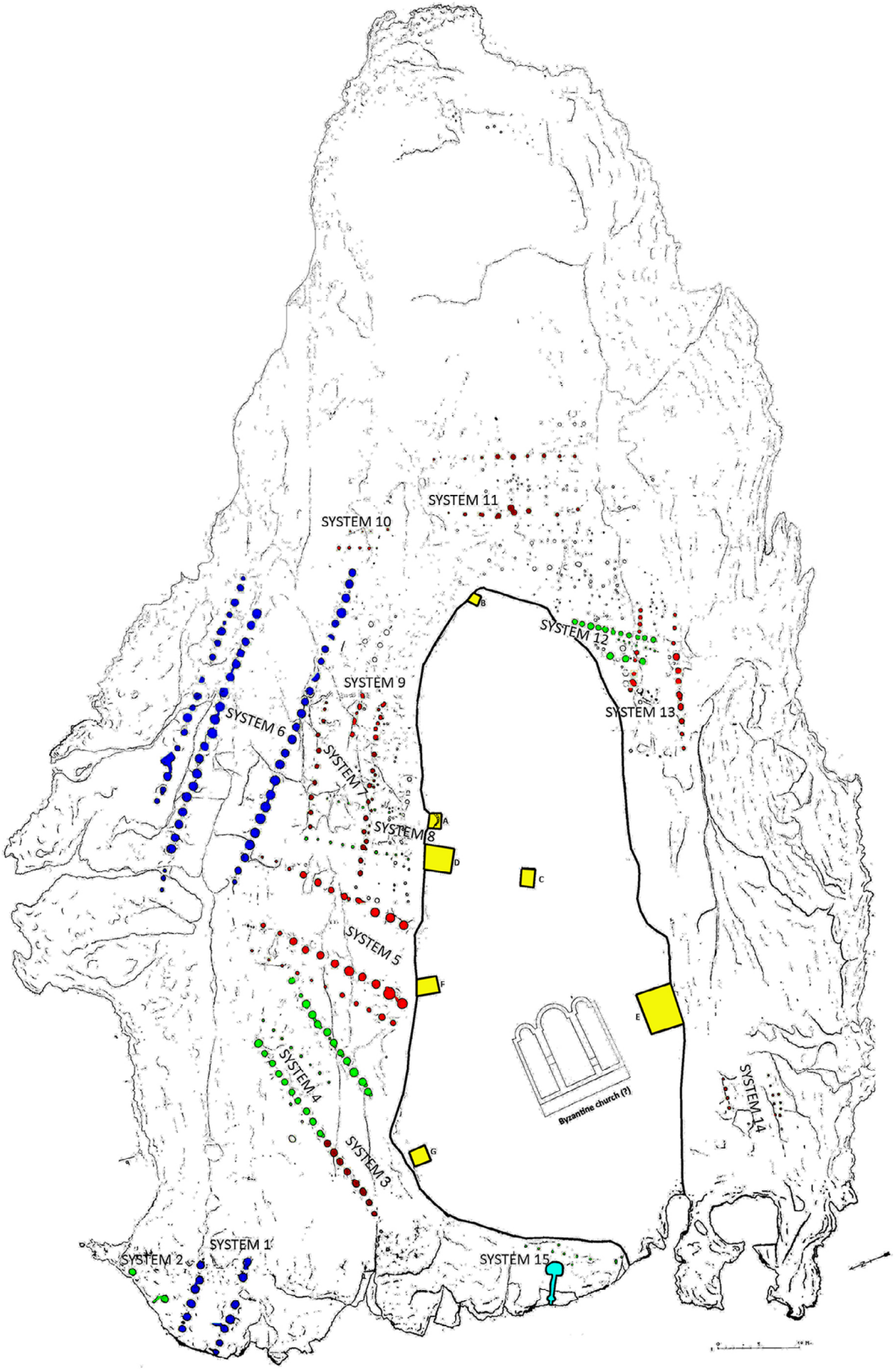

Figure 1. Plan of the islet of Ognina (after Bernabò Brea, 1966).

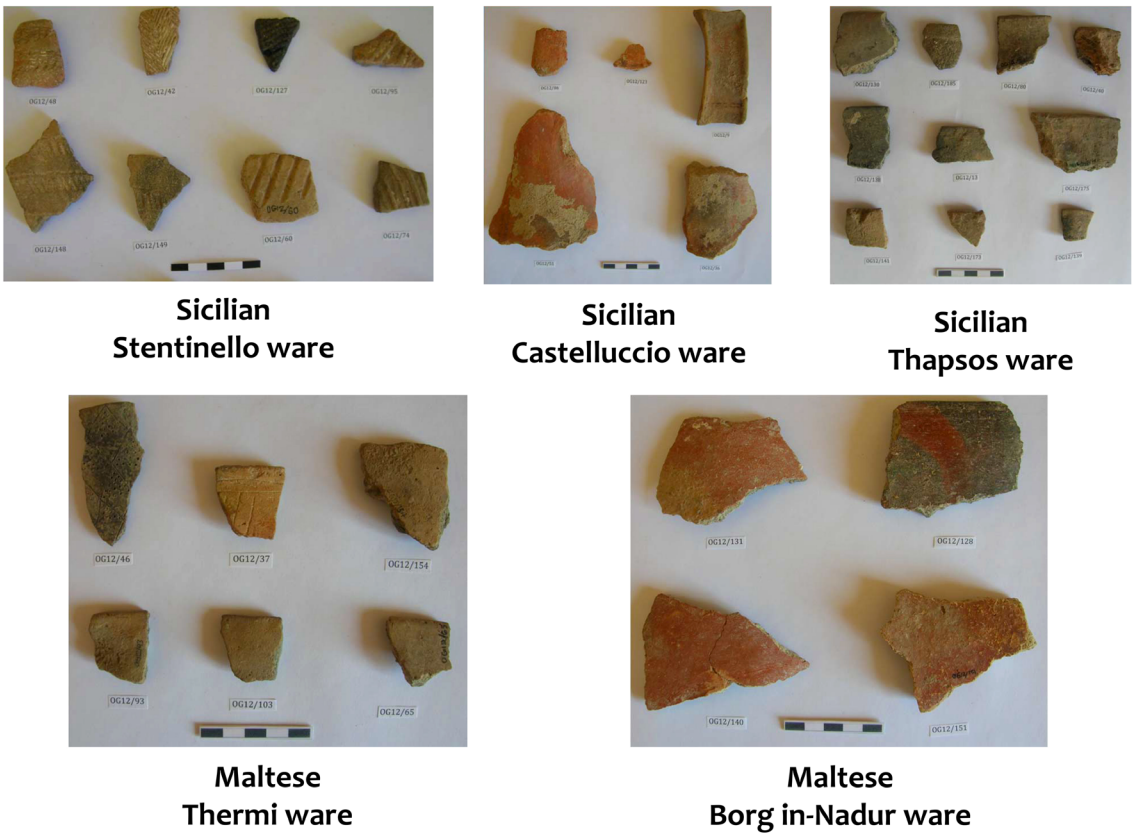

Figure 2. Prehistoric pottery classes from Ognina (fieldwork 2012): Stentinello, Castelluccio, Thapsos, Thermi Ware and Borġ in-Nadur ware (photo Davide Tanasi).

4 Petrographic and Chemical Characterization through XRF

In answering whether the examples of Thermi and Borġ in-Nadur pottery recovered at Ognina were either Maltese imports or local reproductions of Maltese ceramic wares, 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).

Fifty-two samples were examined with a portable X-ray fluorescence spectrometer (pXRF), 11 of which were tested by both a traditional X-ray fluorescence spectrometer (XRF) and by petrographic analysis of thin sections with optical microscopy (OM). Ten 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 Borg in-Nadur. The preliminary petrographic analyses pointed to a substantial difference between the group of Thermi Ware and Borġ in-Nadur type samples.

	Thermi Ware
Fabric	Medium-coarse, with microfossil-rich groundmass grog and carbonation micro-fossil rich groundmass
Groundmass	Heterogeneous, with brownish-yellow color and medium micromass optical activity
Microstructure	Not preferentially oriented and single spaced channels, vughy and vescicles
Inclusions	Grog and fine quartz with a bimodal grain size distribution

	Borġ in-Nadur
Fabric	Medium coarse; dominant grog and carbonatic microfossil-rich groundmass
Groundmass	Heterogeneous, with brownish-greyish color and high/medium high optical activity
Microstructure	Channels and planar voids with remains of carbonaceous material, vughy and preferentially oriented vesicles
Inclusions	Unimodal/bimodal grain size distribution, coarse grog with prevalently sub- angular shape and millimetric dimensions and common fine quartz

Sixty-nine samples, comprising 58 samples of ceramics and clays from the site of Borg in-Nadur, and 11 pottery samples from Ognina and clays from various Sicilian districts, were analyzed with this Philips X-ray fluorescence spectrometer (for tables with full chemical data see Ranieri et al., 2015). 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 results proved very promising and showed 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, which the discrimination between Maltese and Sicilian pottery and raw materials appears evident for both geographical locations (Ranieri et al., 2015, p. 39, fig. 7).

5 Chemical Characterization through pXRF

For ceramics in this part of the study, careful attention was given to analyze ceramic surfaces with relatively flat areas and that showed no signs of slip or application of paint or decoration. Following the example of Tykot et al. (2013:240), both the inside and outside surfaces and when possible the edge of each sherd were analyzed. Further, attention was given in order to avoid analyzing locations where there were visible inclusions. Geological clay samples for both Malta and Sicily were obtained and prepared for analysis using Molitor's preparation process (1988:154).

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 & Selmun), and 4 geological clay samples taken from the northern outskirts of Siracusa, Sicily, were analyzed using a Bruker Tracer III-SD pXRF instrument (Pirone 2017). 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 were analyzed for 120 seconds. Quantitative values for each trace element were obtained by calibrating the raw data using the 2008 MURR calibration software. The peak intensities for the K⟨ peaks of Rb, Sr, Y, Zr, Nb, and L⟨ peak of Th were calculated as ratios to the Compton peak of Rhodium and converted to parts per million (ppm). The calibrated values obtained for each of the trace elements were then averaged for each sample and are reported as the average of the measurements taken from the internal and external surfaces and when possible the edge of the sherd in Table 1. These values were then analyzed statistically using principal components analysis (PCA) with the IBM SPSS Statistics 23 software package.

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).

Table 1. Trace elemental compositions for all Maltese and 14 Sicilian (USF #'s 27212 to 27229) ceramic samples and geological clay samples (Pirone 2017). The remaining 35 Sicilian ceramic samples (USF #'s 18826 to 18857) were analyzed by Robert Tykot.

Site	Material	USF #	Sample ID	Phase	Th	Rb	Sr	Y	Zr	Nb
Borġ in-Nadur	ceramic	19079	BN 236a	Tarxien	10	90	494	24	137	17
Borġ in-Nadur	ceramic	19080	BN 236b	Tarxien	4	41	644	24	126	10
Borġ in-Nadur	ceramic	19081	BN 240a	Tarxien	7	62	529	22	87	10
Borġ in-Nadur	ceramic	19082	BN 241a	Tarxien	7	44	1258	13	90	2
Borġ in-Nadur	ceramic	19083	BN 246	Tarxien	9	89	645	19	134	13
Borġ in-Nadur	ceramic	19084	BN 255x	Tarxien	7	52	726	20	101	8
Borġ in-Nadur	ceramic	19085	BN 260	Tarxien	10	118	641	21	170	17
Borġ in-Nadur	ceramic	19086	BN 272a	Tarxien	11	74	354	22	105	12
Borġ in-Nadur	ceramic	19087	BN 272d	Tarxien	14	100	697	20	161	18
Borġ in-Nadur	ceramic	19088	BN 272f	Tarxien	12	74	708	24	186	17
Borġ in-Nadur	ceramic	19089	BN 273a	Tarxien	10	87	661	19	125	11
Borġ in-Nadur	ceramic	19090	BN 273b	Tarxien	9	65	432	19	105	10
Borġ in-Nadur	ceramic	19091	BN 277	Tarxien	11	94	849	21	167	13
Borġ in-Nadur	ceramic	19092	BN 281	Tarxien	13	79	556	27	122	11
Borġ in-Nadur	ceramic	19093	BN 282	Tarxien	10	71	638	20	104	9
Borġ in-Nadur	ceramic	19094	BN 284	Tarxien	8	96	692	22	132	11
Borġ in-Nadur	ceramic	19095	BN 294f	Tarxien	11	56	317	18	73	8

Site	Material	USF #	Sample ID	Phase	Th	Rb	Sr	Y	Zr	Nb
Borġ in-Nadur	ceramic	19096	BN 299a	Tarxien	7	63	465	17	84	7
Borġ in-Nadur	ceramic	19097	BN 300	Tarxien	11	66	401	22	131	13
Borġ in-Nadur	ceramic	19098	BN 301	Tarxien	7	64	628	24	127	12
Borġ in-Nadur	ceramic	19099	BN 302	Tarxien	7	69	492	18	97	9
Borġ in-Nadur	ceramic	19100	BN 303	Tarxien	9	85	578	21	128	10
Borġ in-Nadur	ceramic	19101	BN 18	Tarxien Cemetery	14	107	431	23	184	20
Borġ in-Nadur	ceramic	19102	BN 94	Tarxien Cemetery	13	94	664	22	163	17
Borġ in-Nadur	ceramic	19103	BN 143c	Tarxien Cemetery	14	82	657	24	154	16
Borġ in-Nadur	ceramic	19104	BN 186	Tarxien Cemetery	12	89	672	22	144	14
Borġ in-Nadur	ceramic	19105	BN 191	Tarxien Cemetery	12	119	630	22	176	15
Borġ in-Nadur	ceramic	19106	BN 304	Tarxien Cemetery	10	68	423	21	117	13
Borġ in-Nadur	ceramic	19107	BN 305	Tarxien Cemetery	10	116	753	22	170	18
Borġ in-Nadur	ceramic	19108	BN 306-2	Tarxien Cemetery	12	116	661	21	160	21
Borġ in-Nadur	ceramic	19109	BN 307	Tarxien Cemetery	9	98	554	23	150	17
Borġ in-Nadur	ceramic	19110	BN 308	Tarxien Cemetery	15	98	765	25	164	17
Borġ in-Nadur	ceramic	19111	BN 136a	Borg in-Nadur	19	122	884	24	168	19
Borġ in-Nadur	ceramic	19112	BN 136d	Borg in-Nadur	10	94	961	22	177	15
Borġ in-Nadur	ceramic	19113	BN 136e-f	Borg in-Nadur	14	90	273	27	195	25
Borġ in-Nadur	ceramic	19114	BN 141d	Borg in-Nadur	13	112	791	22	170	18
Borġ in-Nadur	ceramic	19115	BN 141e	Borg in-Nadur	14	118	678	25	203	20
Borġ in-Nadur	ceramic	19116	BN 148a	Borg in-Nadur	12	107	874	23	192	18
Borġ in-Nadur	ceramic	19117	BN 168	Borg in-Nadur	10	81	587	18	122	11
Borġ in-Nadur	ceramic	19118	BN 176	Borg in-Nadur	9	89	884	20	173	14
Borġ in-Nadur	ceramic	19119	BN 181a	Borg in-Nadur	11	99	782	24	163	17
Borġ in-Nadur	ceramic	19120	BN 181d	Borg in-Nadur	11	99	889	22	165	19
Borġ in-Nadur	ceramic	19121	BN 190	Borg in-Nadur	13	93	450	21	150	18
Borġ in-Nadur	ceramic	19122	BN 199	Borg in-Nadur	11	90	673	22	161	16
Borġ in-Nadur	ceramic	19123	BN 208	Borg in-Nadur	11	99	627	22	137	14
Borġ in-Nadur	ceramic	19124	BN 209	Borg in-Nadur	23	81	534	24	147	15
Borġ in-Nadur	ceramic	19125	BN 225-223	Borg in-Nadur	11	92	626	20	153	17
Borġ in-Nadur	ceramic	19126	BN/P7	Mycenaean (BN)	10	138	514	21	125	7
Ġnejna Bay	clay	19440	Geological Clay		10	88	739	19	111	9
Ġnejna Bay	clay	19441	Geological Clay		9	100	637	16	122	15
Ġnejna Bay	clay	19442	Geological Clay		9	99	711	16	117	10
Ġnejna Bay	clay	19443	Geological Clay		8	96	638	18	114	13
Ġnejna Bay	clay	19444	Geological Clay		9	92	806	14	104	10
Ġnejna Bay	clay	19445	Geological Clay		7	97	669	15	97	11
Ġnejna Bay	clay	19446	Geological Clay		13	129	442	21	126	14
Ġnejna Bay	clay	19447	Geological Clay		9	123	454	21	149	13
Ġnejna Bay	clay	19448	Geological Clay		12	137	457	22	134	16
Ġnejna Bay	clay	19449	Geological Clay		11	127	493	21	129	15
Selmun	clay	19450	Geological Clay		6	84	820	15	117	10
Selmun	clay	19451	Geological Clay		7	83	880	17	135	12
Selmun	clay	19452	Geological Clay		13	83	915	14	134	7
Selmun	clay	19453	Geological Clay		5	68	937	19	122	7
Selmun	clay	19454	Geological Clay		8	76	1163	15	129	7
Selmun	clay	19455	Geological Clay		9	106	439	20	120	14
Selmun	clay	19456	Geological Clay		10	97	518	16	108	10
Selmun	clay	19457	Geological Clay		9	82	559	18	108	11
Selmun	clay	19458	Geological Clay		10	93	410	18	105	15
Ognina	ceramic	18826	12_105-29	Middle Bronze Age	10	84	422	21	183	17
Ognina	ceramic	18827	12_117-16	Middle Bronze Age	9	69	301	20	149	11
Ognina	ceramic	18828	12_130-1	Middle Bronze Age	9	90	710	20	156	14
Ognina	ceramic	18829	12_13-6	Middle Bronze Age	12	85	420	24	233	16
Ognina	ceramic	18830	12_137-19	Middle Bronze Age	8	98	399	22	204	16
Ognina	ceramic	18831	12_138-5	Middle Bronze Age	10	87	460	21	177	16
Ognina	ceramic	18832	12_139-10	Middle Bronze Age	9	88	267	21	202	15
Ognina	ceramic	18833	12_144-13	Middle Bronze Age	10	95	566	20	180	14

Site	Material	USF #	Sample ID	Phase	Th	Rb	Sr	Y	Zr	Nb
Ognina	ceramic	18834	12_147-8	Middle Bronze Age	11	90	424	23	260	12
Ognina	ceramic	18835	12_162-21	Middle Bronze Age	9	94	336	23	216	14
Ognina	ceramic	18836	12_16-27	Middle Bronze Age	13	93	537	24	247	16
Ognina	ceramic	18837	12_163-14	Middle Bronze Age	11	83	444	24	212	19
Ognina	ceramic	18837	12_163-14	Middle Bronze Age	11	83	444	24	212	19
Ognina	ceramic	18838	12_169-15	Middle Bronze Age	10	86	304	23	251	16
Ognina	ceramic	18839	12_170-17	Middle Bronze Age	9	74	421	20	195	18
Ognina	ceramic	18840	12_173-9	Middle Bronze Age	9	83	388	21	241	16
Ognina	ceramic	18841	12_175-7	Middle Bronze Age	9	92	468	23	186	19
Ognina	ceramic	18842	12_185-2	Middle Bronze Age	6	56	493	19	123	9
Ognina	ceramic	18843	12_3-24	Middle Bronze Age	12	95	431	23	190	18
Ognina	ceramic	18844	12_39-30	Middle Bronze Age	10	93	425	20	209	18
Ognina	ceramic	18845	12_40-4	Middle Bronze Age	9	87	395	22	188	15
Ognina	ceramic	18846	12_49-11	Middle Bronze Age	10	76	560	23	216	14
Ognina	ceramic	18847	12_55-31	Middle Bronze Age	7	83	1770	13	139	7
Ognina	ceramic	18848	12_58-26	Middle Bronze Age	9	91	473	21	209	16
Ognina	ceramic	18849	12_61-12	Middle Bronze Age	8	96	438	23	194	15
Ognina	ceramic	18850	12_6-20	Middle Bronze Age	8	96	538	20	182	12
Ognina	ceramic	18850	12_6-20	Middle Bronze Age	8	96	538	20	182	12
Ognina	ceramic	18851	12_62-25	Middle Bronze Age	8	73	321	22	180	13
Ognina	ceramic	18852	12_63-23	Middle Bronze Age	10	99	463	22	203	16
Ognina	ceramic	18853	12_67-32	Middle Bronze Age	12	83	502	21	211	16
Ognina	ceramic	18854	12_7-28	Middle Bronze Age	11	72	358	26	191	16
Ognina	ceramic	18855	12_79-22	Middle Bronze Age	12	101	563	22	205	21
Ognina	ceramic	18856	12_80-3	Middle Bronze Age	10	86	419	21	199	14
Ognina	ceramic	18857	12_94-18	Middle Bronze Age	9	80	340	23	211	14
Ognina	ceramic	18857	12_94-18	Middle Bronze Age	9	80	340	23	211	14
Ognina	ceramic	27212	OG 12/140	Borg in-Nadur	13	98	492	21	134	13
Ognina	ceramic	27213	OG 12/148	Thapsos	6	40	252	18	140	8
Ognina	ceramic	27214	OG 12/60	Thapsos	6	56	494	17	98	6
Ognina	ceramic	27216	OG 12/94	Thapsos	9	72	293	23	182	15
Ognina	ceramic	27217	OG 12/154	Thermi	8	80	284	22	184	16
Ognina	ceramic	27218	OG 12/160	Thapsos	10	77	213	24	223	14
Ognina	ceramic	27219	OG 12/128	Thapsos	11	92	594	22	148	18
Ognina	ceramic	27221	OG 12/46	Thermi	9	74	333	19	142	13
Ognina	ceramic	27223	OG 12/163	Thapsos	10	63	406	20	163	14
Ognina	ceramic	27225	OG 12/151	Borg in-Nadur	7	67	1562	12	103	2
Ognina	ceramic	27226	OG 12/131	Borg in-Nadur	12	100	456	20	133	13
Ognina	ceramic	27227	OG 12/36	Castelluccio (EBA)	13	79	406	19	147	12
Ognina	ceramic	27228	OG 12/51	Castelluccio (EBA)	11	81	351	21	157	16
Ognina	ceramic	27229	OG 12/47	Thapsos	11	58	313	19	122	11
North Outskirts of Siracusa	clay	27243	Geological Clay		8	33	484	16	68	1
North Outskirts of Siracusa	clay	27244	Geological Clay		6	31	493	14	69	5
North Outskirts of Siracusa	clay	27245	Geological Clay		8	33	724	16	52	6
North Outskirts of Siracusa	clay	27247	Geological Clay		11	33	485	14	53	3

Prior to running the PCA, the values for each of the trace elements were transformed using base log.10. A PCA was then run and the number of components extracted was based on an Eigenvalue greater than one. The results of this PCA indicated that the trace elements loaded on one of two components with Sr primarily loading on the second component (Table 2). Further, the PCA shows 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, 2017). 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.

Table 2. PCA Component matrix showing how the trace elements load on each component. Principal component scores were determined from the log transformation of the trace elemental compositional data for each sample listed in Table 1.

Component Matrix		
	Component	
	1	**2**
Log(10) Th	.645	.261
Log(10) Rb	.728	.528
Log (10) Sr	-.335	.867
Log(10) Y	.834	-.285
Log(10) Zr	.831	-.048
Log(10) Nb	.915	.017

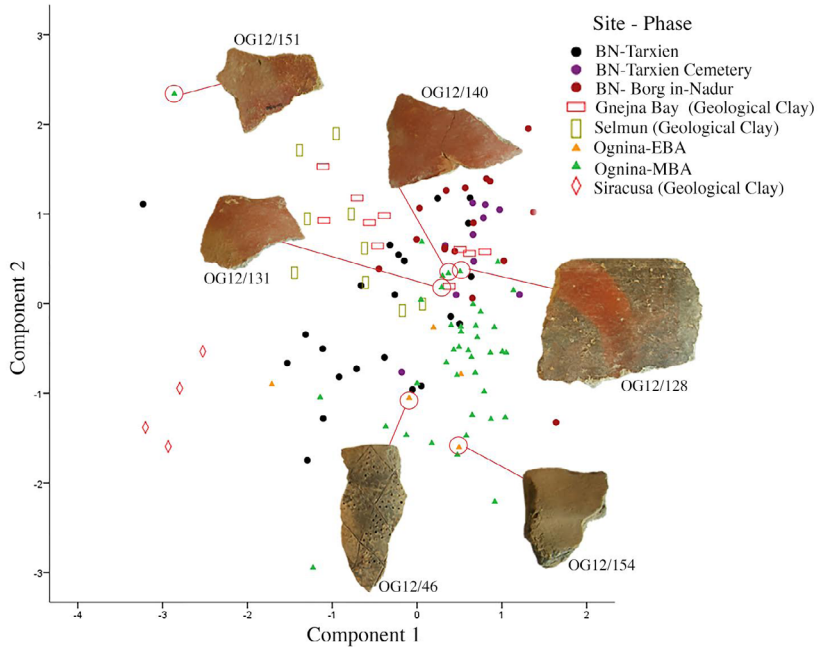

Figure 4. Bivariate scatterplots of principal components scores for all Maltese and Sicilian ceramic and clay samples.

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 conducted with 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 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, allow for distinctions to be observed between Maltese and Sicilian ceramics and clays.

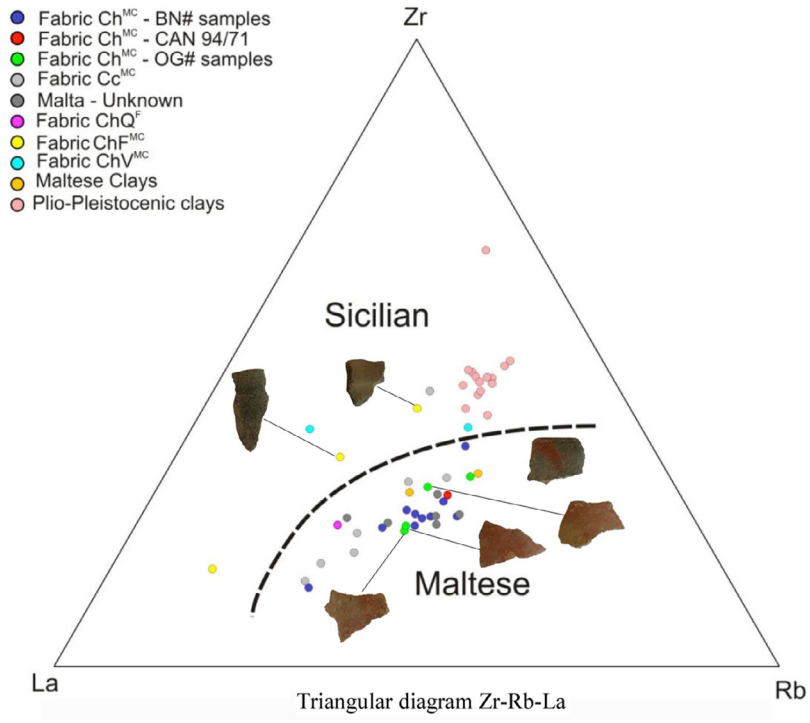

Figure 5. Discriminating triangular diagram of Zr, Rb, and La for all Maltese ceramic and clay samples from Borg in-Nadur and ceramic samples from Ognina.

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 being sample OG/151, which in the scatter plot of the data obtained with the 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 Borġ in-Nadur pieces were produced in Malta and one was produced in Sicily. These results seem to suggest there was human mobility, perhaps the ceramic artisans, who may have either carried with them finished pottery or clay materials from Malta to Sicily. This study further assessed the hypothesis based on macroscopic observation that Ognina served as an emporium with different cultural group present. The elemental compositional data from both the XRF and pXRF revealed the presence of both Maltese pottery imports and local imitations of Maltese ceramics.

Despite some technical limitations, this study has demonstrated the potential usefulness of pXRF analyses as an effective device in investigating certain research questions such as those relating to trade, interaction and mobility of people and artifacts among various locations. pXRF, therefore, appears useful as a non-destructive technique allowing us to critically investigate evidence previously obtained by visual inspection and traditional archaeological methods. In the present study, pXRF offers an effective method to study the other Thermi Ware artifacts and Borġ in-Nadur pottery previously found at Ognina. This would further verify the hypothesis of an exclusive Sicilian imitation of Maltese pottery in the EBA and of the greater likelihood for Maltese pottery being imported into Sicily during the MBA.

Additional research using pXRF in analyzing all the Maltese type pottery from the other sites, which for the most part these ceramic artifacts are mainly kept at the archaeological museum in Siracusa, will allow us to further distinguish between imported products and local imitations on a larger scale. In this regard, pXRF affords an opportunity to have a more robust data set of Maltese and Sicilian sampled ceramics and

clays by having the ability to analyze a greater number of ceramic artifacts due to the portability and cost point of using a pXRF on location non-destructively. This will allow us to further reconsider the nature of the relationship between Sicily and Malta between the EBA and MBA and to further answer the question whether this relationship was characterized by cycles of mere exchanges of goods or the mobility of people.

References

Ashkanani, H. & Tykot, R.H. (2013). Interregional interaction and Dilmun Power in the Bronze Age: A provenance study of ceramics from Bronze Age sites in Kuwait, and Bahrain using non-destructive pXRF analysis. In R.A. Armitage & J.H. Burton (Eds.), *Archaeological Chemistry VIII. ACS Symposium Series* 1147: 245-267.

Bernabò Brea, L. (1966). Abitato neolitico ed insediamento maltese dell'età del Bronzo all'isola di Ognina (Siracusa) e i rapporti fra Sicilia e Malta dal XVI al XIII sec. a. C. *Kokalos* 12: 41-69.

Biehl, P.F., Rassamakin, Y.Y. (2008). *Import and Imitation in Archaeology*. Langenweißbach: Beier & Beran.

Cazzella, A. & Recchia, G. (2012). Tas-Silg: the Late Neolithic megalithic sanctuary and its re-use during the Bronze Age and the Early Iron Age. *Scienze dell'Antichità*, 18: 15-38.

Copat, V., Danesi, M., Ruggini, C. (2013). Late Neolithic and Bronze Age pottery from Tas-Silg sanctuary: new research perspectives for the Maltese prehistoric sequence. *Rivista di Scienze Preistoriche 18*: 39-63.

Cultraro, M. & Crispino, A. (2014). Ognina (Siracusa, Prov. di Siracusa), in Notiziario di Preistoria e Protostoria 1.IV, pp. 112-114.

Hunt, A. & Speakman, R. (2015). Portable XRF Analysis of Archaeological Sediments and Ceramics. *Journal of Archaeological Science* 53: 1-13.

Maniatis, Y. (2009). The Emergence of Ceramic Technology and its Evolution as Revealed with the use of Scientific Techniques. In Shortland A.J., Freestone, I.C., Rehren, T. (Eds.), *From Mine to Microscope: Advances in the Study of Ancient Technology*. Oxford: Oxbow Books, 1-18.

McCormick, D.R. (2013). *Exploring Potential Applications of Portable X-ray Fluorescence on Earthen Materials from Southeast Mesoamerica*. MA thesis, Department of Anthropology, University of South Florida, Tampa, Florida. http://scholarcommons.usf.edu/etd/4540.

Molitor, M. (1988). *Pots and Potters of Prehistoric Malta*. Unpublished Ph.D. Dissertation. University of California, Los Angeles.

Palio, O. (2008). Ognina, Malta e l'Egeo, in Bonanno, A., Militello, P. (Eds.), *Malta negli Iblei. Gli Iblei a Malta*. Palermo: Officina di Studi Medievali: 7-19.

Pirone, F. (2017). *Trade, Interaction and Change: Trace Elemental Characterization of Maltese Neolithic to Middle Bronze Age Ceramics Using a Portable X-ray Fluorescence Spectrometer*. PhD dissertation, Department of Anthropology, University of South Florida, Tampa, Florida.

Raneri, S., Barone, G., Mazzoleni, P., Tanasi, D., Costa, E. (2015). Mobility of men versus mobility of goods: archaeometric characterization of Middle Bronze Age pottery in Malta and Sicily (15th-13th century BC), *Periodico di Mineralogia* 84.1: 23-44.

Recchia G. & Fiorentino, G. (2015). Archipelagos adjacent to Sicily around 2200 BC: attractive environments or suitable geo-economic locations? *Tagungen des Landesmuseums für Vorgeschichte Halle* Bd. 12/I+II: 1-15.

Speakman, R., Little, N.C., Creel, D., Miller, M.R., & Iñañez, J.G. (2011). Sourcing ceramics with portable XRF spectrometers? A comparison with INAA using Mimbres pottery from the American Southwest. *Journal of Archaeological Science* 38: 3483-3496.

Stremtan, C., Ashkanani, H., Tykot, R.H. & Puscas, M. (2014). Constructing a Database for pXRF, XRD, ICP-MS and Petrographic Analyses of Bronze Age Ceramics and Raw Materials from Failaka Island (Kuwait). In R.B. Scott, D. Braekmans, M. Carremans & P. Degryse (Eds.), *Proceedings of the 39th International Symposium for Archaeometry.* Leuven, Belgium: Centre for Archaeological Sciences, KU Leuven, pp. 274-279.

Tanasi, D. (2005). Mycenaean Pottery Imports and Local Imitations: Sicily VS Southern Italy. In Laffineur, R., Greco, E. (Eds.), *Emporia. Aegeans in Central and Eastern Mediterranean. Aegaeum* 25: 561-569.

Tanasi, D. (2008). *La Sicilia e l'arcipelago maltese nell'età del Bronzo Medio*. Palermo: Officina Studi Medievali.

Tanasi, D. (2010). Bridging the gap. New data on the relationship between Sicily, the Maltese Archipelago and the Aegean in the Middle Bronze Age. *Mare Internum* 2: 111-119.

Tanasi, D. (2011). The Prehistoric Pottery. In Tanasi, D., Vella, N.C. (Eds), *Site, Artefacts, Landscape: Prehistoric Borġ in-Nadur, Malta*. Monza: Polimetrica.

Tanasi, D. (2014). Uniti e divisi dal mare. Mobilità di uomini e merci tra la Sicilia e l'arcipelago maltese nel II millennio a.C. In Congiu, M., Micciché, C., Modeo, S. (Eds.), *Viaggio in Sicilia. Racconti, segni e città ritrovate*. Caltanissetta: Sciascia, 99-116.

Tanasi, D. (2015a). The pottery from the excavation campaign of David H. Trump (1959) at the settlement of Borġ in-Nadur. In Tanasi, D., Vella, N.C. (Eds.), *The late prehistory of Malta: essays on Borġ in-Nadur and other sites*. Oxford: Archaeopress, 35-98.

Tanasi, D. (2015b). Borġ in-Nadur pottery abroad: a report from the Sicilian necropoleis of Thapsos and Matrensa. In Tanasi, D., Vella, N.C. (Eds.), *The late prehistory of Malta: essays on Borġ in-Nadur and other sites*. Oxford: Archaeopress, 173-184.

Tanasi, D., Vella, N.C. (2014). Islands and mobility: exploring Bronze Age connectivity in the south-central Mediterranean. In Van Dommelen, P., Knapp B. (Eds.), *The Cambridge Prehistory of the Bronze and Iron Age Mediterranean*. Cambridge: Cambridge University Press, 184-201.

Trump, D.H. (2004-2005). Ognina: a puzzling prehistoric site in Sicily. *Malta Archaeologica Review* 7: 55-60.

Tykot, R.H., White, N.M., Du Vernay, J.P., Freeman, J.S., Hays, C.T., Koppe, M., Hunt, C.N., Weinstein, R.A., & Woodward, D.S. (2013). Advantages and Disadvantages of pXRF for Archaeological Ceramic Analysis: Prehistoric Pottery Distribution and Trade in NW Florida. In Armitage, R.A. & Burton, J.H. (Eds.), Archaeological Chemistry VIII. (ACS Symposium Series). Washington, D.C.: *American Chemical Society*, 233-244.