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# THE NEOLITHIC OBSIDIAN ARTIFACTS FROM ROCCAPALUMBA (PALERMO, ITALY): FIRST CHARACTERIZATION AND PROVENANCE DETERMINATION

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## ABSTRACT

This paper discusses the first geochemical characterization of obsidian fragments from the prehistoric site of Roccapalumba (Palermo, Italy). The Neolithic age of the prehistoric settlement was constrained by pottery and flint tools discovered in the same archaeological context. To define the provenance of the investigated obsidian artifacts major and trace element analyses have been carried out by using scanning electron microscopy (SEM-EDX) and inductively coupled plasma mass spectrometry (LA-ICP-MS). The comparison with literature data of the Central Mediterranean source areas, based on trace elements content and their ratios allow of constraining a provenance of the Roccapalumba obsidians from the Lipari Island. The obsidian lava flow from Gabelotto Valley is the most probable source of volcanic glass at Lipari and also the most exploited in the Mediterranean area for manufactured tools. The obtained results can contribute in reconstructing the trade/exchange and procurement relationships occurred between the prehistoric human groups inhabiting Sicily during Neolithic age.

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**KEYWORDS:** Roccapalumba, obsidian, Neolithic, SEM-EDX, LA-ICP-MS, trace elements, Lipari Island.

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## 1. INTRODUCTION

An assemblage of 85 lithic artifacts consisting of 34 obsidians and 51 flints has been brought to light in the prehistoric site of Roccapalumba (Fig. 1), a village located about 60 km away to the south east of Palermo, Italy, (Mannino, 1998, 2008; Gagliardo, 2005). The lithic artifacts were collected in 1980, by a student (author Francesco Italiano) of the Palermo University during a geological excursion at Castellaccio di Fiaccati nearby the village of Roccapalumba. This area was intended for quarry of limestones. Among the limestone blocks subjected to the extraction works, the obsidian and the flint artifacts were

found mixed with abundant ceramics fragments, which allowed archeologists to attribute the site to the Neolithic age, despite the settlement has never been found (Mannino, 1998). The discovered lithic and ceramic artifacts are presently kept at the “Antonino Salinas” Regional Museum of Palermo. Since those materials represent significant witnesses of cultural and social relationships by inhabitants of Sicily during prehistory, we carried out an archaeometric characterization of the obsidian artifacts together with typological analyses of the included flints, in order to establish their provenance and use.



*Figure 1. Castellaccio di Fiaccati, the hill near Roccapalumba (Palermo, Italy) where the Neolithic settlement was discovered in 1980 and where the obsidian tools of this study were collected (Images from Google Earth, modified by the authors).*

The Central Mediterranean is an interesting area for archaeological obsidian researches, because obsidian-bearing volcanoes are very few in this geographical area, being located far one from another. Such volcanoes are found in some Italian and Aegean islands. The latest studies related to obsidian artifacts suggested that in the Central Mediterranean, from Neolithic to the Middle Bronze Age (~6000-1300 BC), the main geological sources of raw materi-

al used to make obsidian cutting tools and weapons were located over some volcanic islands in Italy (Fig. 2), such as Monte Arci (Sardinia), Palmarola (Pontine Island), Lipari (Aeolian Islands) and Pantelleria (Pelagian Islands) and Greece (Islands of Melos and Giali; De Francesco *et al.*, 2008).

Each obsidian occurrence has a specific geochemical signature that can be constrained by different analytical methods, based on destructive, micro-

destructive or non-destructive analyses with the aim to determinate their major and trace elements contents. A variety of methods for multi-elemental characterization of obsidians including SEM-EDX, XRF, NAA, EMPA, LA-ICP-MS were used in the last decades (Cann and Renfrew, 1964; Hallam et al., 1976; Williams Thorpe et al., 1984; Francaviglia, 1984, 1988; Tykot, 1996; Barca et al., 2007; 2008; Crisci et al., 1994; Acquafredda et al., 1999; 2018; Bigazzi et al.,

2005; Le Bourdonnec et al., 2006; 2010). Therefore, tracing the geological source of any obsidian artifact allows reconstructing contacts and cultural exchanges between distant populations during prehistory. Sometimes it is also possible to discriminate among sub-sources of the same source-area using specific geochemical markers (Tykot, 2002, 2017, Freund et al., 2015).

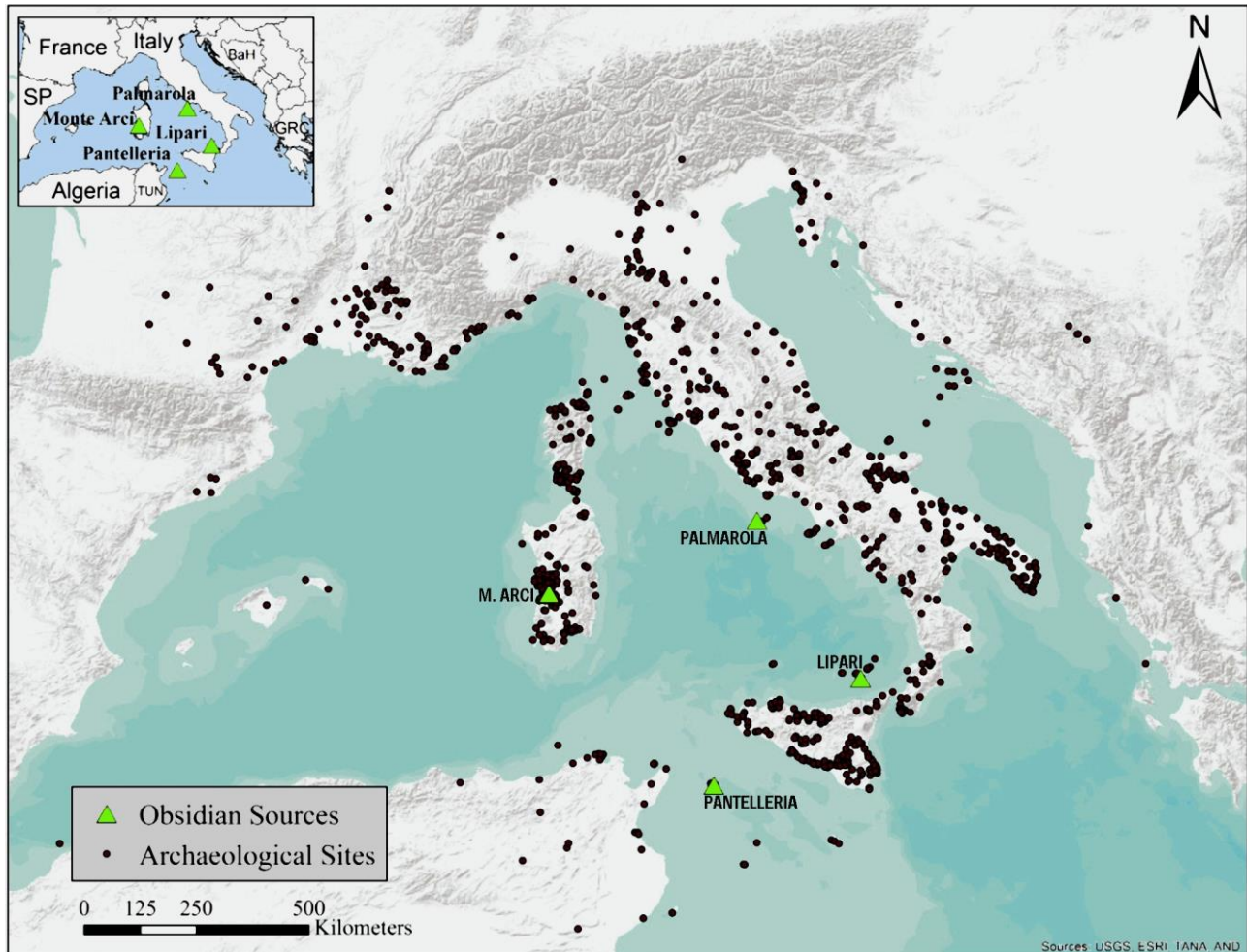


Figure 2. The four obsidian sources in Central Mediterranean and their distribution in archaeological sites from VI to II millennia BC (modified after Freund, 2017).

These studies highlighted the widespread diffusion of Central Mediterranean obsidians over about 1200 prehistoric villages, distributed in Italy, France, Slovenia, Croatia, Bosnia Herzegovina, Albania, Tunisia, Algeria, Malta and Spain. Interestingly, the Lipari obsidian was the most widespread in the Central Mediterranean area (Freund, 2017). Furthermore, in Sicily the diffusion of the Lipari's obsidian was predominant and in many cases exclusive. In some prehistoric sites of western Sicily relevant obsidian imports from Pantelleria Island, and occasionally from the most distant Palmarola Island were demonstrated (Francaviglia and Piperno, 1987; Francavi-

glia, 1988; Tykot et al., 2013; Foresta Martin et al., 2017).

Here we provide the first results of archaeometric-geochemical characterization of obsidians artifacts found at the Castellaccio di Fiaccati (Roccapalumba) site. To achieve this objective we carried out SEM-EDX and LA-ICP-MS analyses of obsidians with the aim to define the source area, and to provide new information to better constrain the commercial and cultural exchanges in Sicily during the Neolithic age. Moreover, our results may enhance the knowledge of the social complexity of Neolithic settlements in the widest context of the Central Mediterranean Basin.

## 2. THE SITE AND THE ARCHAEOLOGICAL EXCAVATION

Castellaccio di Fiaccati (lat 37°48'05.52"N, long 13°40'23.012"E), sometimes also called Le Rocche di Fiaccati (Mannino, 1998), is a hill rising 411 m asl, located to the East of the small village of Roccapalumba, about 20 km away from the northern coast of

Sicily and 60 km from Palermo city. The geographic site was ideal for a Neolithic settlement, being protected by Madonie mountain range and yet close to the Torto river valley, which offered a fast way to reach the Tyrrhenian Sea. Moreover, the Neolithic site was surrounded by wooded areas rich in hunting animals, timber and firewood (Fig. 3).



*Figure 3. View from the North of the hill of Castellaccio di Fiaccati (marked by the red arrow), site of the Neolithic settlement.*

From a geological point of view, the hill in which the Neolithic site stood is one of the many limestone blocks scattered in this area, which represents the remains of deep-water deposits formed in the open sea during Middle-Upper Jurassic and Cretaceous (Catalano *et al.*, 2011).

Local historians refer that the first written reports of prehistoric finds in Castellaccio di Fiaccati dates back to the end of XIX century. During that time, the landowner of the site, donated to the Archaeological Museum of Palermo some lithic instruments and ceramic artifacts found in his property (De Gregorio, 1917; Gagliardo, 2005). This donation is confirmed by an annotation written on the General Entrance Register of the Salinas Museum, dated April 10<sup>th</sup> 1901 (ASMARP, 1887). Nevertheless, from that time

onwards a systematic destruction of the site was carried out for both railway line construction and limestones quarry exploitation (Gagliardo, 2005). It was only soon after the report of a geologist in 1980 that the Soprintendenza ai Beni Archeologici of the western Sicily planned archaeological excavation surveys on the hill (Mannino, 1998, 2008).

The excavation was directed by Giovanni Mannino, assistant superior to the Archaeological Superintendency, with the collaboration of one of the authors of this article (F. Spatafora), and it was the first and nowadays, the only official intervention in that area. The archaeological excavation was carried out near the top of the hill of Castellaccio di Fiaccati, where the mining activity of limestones had highlighted a deep rift filled with thousands of ceramic

fragments. Inside the rift it was possible to collect a large quantity of pottery besides lithic and bone tools, all datable to the Neolithic age. However, within the discharge of these Neolithic materials it was not possible to highlight any stratigraphy or to understand its function. The presence of some vases with evident manufacturing defects and fragments that cannot be recomposed, suggests a dumping of waste and materials no longer functional for daily use. On the other hand, the numerous tools made of bone, flint and obsidian, and the presence of some

re-composable forms, also suggests a possible ritual use of that cavity (Mannino 2012).

Regardless of unpainted gray pottery, about one third of the recovered fragments is made of ceramic with engraved, embossed and graffiti decorations, attributable to the Stentinello facies (middle Neolithic, 6<sup>th</sup>-5<sup>th</sup> millennium BC). The figulina ceramic fragments are also significant, with a painted two-color or trichrome decoration, in some cases attributable to the so-called Capri style (Middle Neolithic, 5<sup>th</sup> millennium BC, Fig. 4).



Figure 4. Middle Neolithic vase with trichrome decoration collected at Castellaccio di Fiaccati-Roccapalumba during excavation in 1980. (Courtesy of Archaeological Museum Salinas, Palermo, Italy).

With regard to the shapes of the pottery, among the Stentinello ceramics various open vases are recognizable, including hemispherical cap bowls and truncated conical bowls, and a series of small closed forms such as collared olle and small continuous wall orbits. Articulated and complex are the decorations, consisting mainly of linear motifs, variously composed, on the surface of the vases. The painted ceramics, of excellent quality as regards the ceramic body that is hard, compact and sufficiently purified, is characterized by different shapes, including truncated conical cups, also on pedestals, collar olle, and slightly concave necks. As in the engraved and embossed ceramics, in this case too the decoration unfolds on the surface of the vases, mainly with linear motifs arranged in various ways, but also with zig-zag and fluted-on-the-border motifs typical of the so-called Capri style.

The excavations, unfortunately, did not bring to light any trace of the Neolithic village. However, considering the quality and quantity of the pottery

and of the tools recovered, it must have been populated by a community numerically consistent and technologically advanced, that had to possess wide and articulated cultural references, as can be inferred from the variety of the vascular repertoire.

Although it does not concern the Neolithic context, it should be noted that the archaeological excavation carried out on the top of the hill led to the discovery of a dwelling formed by quadrangular environments, and some nearby huts, likely belonging to the Norman farmhouse of Burgiseleth mentioned in a document dated 1170 (Mannino 1998).

## 2.1 Typological analyses of the lithic assemblage

The group of lithic artifacts includes 34 obsidians and 51 flints. The study was conducted on the basis of Laplace (1964) typological system. Fragments of cores and knapping wastes (platforms, flakes cores, debris) have been identified for both the raw materials and attesting the work in the settlement.

The obsidian group (Fig. 5, Table 1) include only 5 tools: 3 blade scrapers (L0); n. 1 backed blade (LD2); n. 1 flake scraper (E1). The other artifacts are n. 21 fragments of flakes not retouched, n. 1 fragment of core, n. 1 platform and n. 7 blades. Some of the blades are regular and thin bladelets (3 mm thick). The conservation is mostly fragmentary and only 8 pieces are integer.

The group of flint artifacts includes a few tools: n. 1 fragment of blade-sickle; n. 1 scarper with marginal retouch (R1); n. 1 simple burin. Most of the artifacts are characterized by n. 46 very little flakes (15-30 mm length), n. 1 core fragment, n. 1 thin blade (50 mm length).

*Table 1. Obsidian Typological analyses*

Number	Flake/Blade	Lithic typologie Laplace 1964	Conservation	Measures (mm)
1	Flake		Fragment	13x16x8
2	Flake		Whole	28x18x3
3	Flake		Middle distal fragment	15x18x3
5	Flake		Whole	22x20x5
6	Flake core		Whole	40x23x9
7	Flake		Whole	22x15x2
8	Bladelet		Middle fragment	20x12x3
9	Bladelet	Scraper (L0)	Middle fragment	28x12x3
10	Blade		Middle fragment	26x15x4
11	Flake		Middle fragment	20x25x5
12	Blade	Scraper (L0)	Middle fragment	24x16x3
13	Flake		Whole	25x17x3
14	Blade	Scraper (L0)	Middle fragment	28x16x3
15	Blade		Middle fragment	13x13x3
16	Core		Middle fragment	15x20x6
17	Flake		Whole	42x28x7
18	Flake		Middle fragment	28x18x6
19	Flake		Middle fragment	36x22x5
20	Platform		Whole	23x25x7
21	Flake		Middle fragment	19x31x9
22	Crested blade		Middle fragment	22x12x4
24	Bladelet		Middle proximal fragment	20x10x3
25	Blade		Middle fragment	12x12x3
26	Flake		Middle fragment	12x21x3
27	Flake		Middle fragment	22x11x4
28	Flake		Middle fragment	15x10x7
29	Flake		Middle fragment	12x15x4
30	Bladelet		Middle proximal fragment	25x10x2
31	Flake		Whole	19x7x2
32	Flake		Middle fragment	13x12x3
33	Flake		Middle fragment	7x14x4
34	Flake		Middle fragment	12x9x5
35	Blade	Backed blade (LD2)	Middle fragment	16x7x3
36	Flake	Scraper (E1)	Whole	7x11x5



*Figure 5. Some of the obsidian flakes and tools analysed in this study.*

On the surfaces of the flint artifacts deadlifts of circular shape are visible and probably caused by contact with fire. About raw material, were used flint nodules of different colors (yellow, red, grey, white, blond). Many of these artifacts show pseudo-retouch and abrasions due to reworking on the find site.

Features of the lithic industry suggest an intense knapping activity in the settlement. Some of the tools are probably wastes that could be in the fireplace. The type of tools, the presence of obsidian and sickle elements, even if in a small number, allow of attributing them to the Neolithic age (Martinelli, 1995; Martinelli and Quero, 2013).

### 3. ANALYTICAL METHODS

Thirty-four obsidian artifacts coming from the Roccapalumba archaeological site and presently kept at the "Salinas" Museum of Palermo, have been subjected to geochemical analyses. All obsidian specimens have been analysed to define major and trace element composition by means of Scanning electron microscopy (SEM-EDX) and Inductively Coupled Mass spectrometer with laser ablation (LA-ICP-MS).

#### 3.1. SEM-EDS

Scanning Electron Microscopy is an environmental ESEM-FEI Inspect-S equipped with a spectrometer Oxford INCA PentaFETx3 EDS, and a Si(Li) detector equipped with a ultra-thin window ATW2 (MIFT Department of the Messina University). Measurements were performed using a resolution of 137 eV at 5.9 keV. Data acquisition was made under environmental conditions, at a working distance of 10 mm with an acceleration voltage of 20 kV, counting times of 60s, approximately 3000 cps with dead time below 30%. The obtained semi-quantitative data were processed by INCA software Energy. This software uses the XPP matrix correction scheme developed by Pouchou and Pichoir (1984, 1985).

#### 3.2. LA ICP-MS

Trace elements composition of the obsidians was analysed at the INGV laboratory-Palermo using a GeoLasPro 193nm ArFExcimer laser ablation (LA) system, connected to an Agilent 7500ce quadrupole ICP-MS. The analyses were performed with a con-

stant laser repetition rate of 10 Hz, a fluency of 15 J/cm<sup>2</sup> and a He flux of 0.8 L/min in the ablation cell.

Despite the homogeneity of the obsidians, being composed of aphyric volcanic glass, the presence of some rare microlite is possible. Therefore, each sample was analysed with a 32 µm spot from 2 to 4 times in order to minimize possible errors due to the local heterogeneity. Total analysis time was 2 min per spot, including 1 min of background acquisition.

Glass reference material NIST612 was used as external standard and was measured at the beginning, in the middle and at the end of each analytical sequence. <sup>29</sup>Si, estimated by ESEM measures, was used as internal standard. The data were processed using the Glitter program (Van Achterbergh et al. 2001). The analytical accuracy (RSD%) was calculated by repeated analyses of the USGS basaltic reference glass BCR-2G, and resulted to be ≤5% for Sc, V, Co, Zn, Rb, Cs, Ba, La, Ce, Nd, U and Th, ≤10% for Li, Cu, Sr, Pr, Sm, Eu, Yb, Lu, Hf and Pb, ≤15% for Y, Cr, Zr, Gd, Tb and Ho, ≤20% for Ti, Tm and Fe.

### 4. DISCUSSION OF RESULTS

Thirty-four fragments of obsidian artifacts recovered from the prehistoric site of Roccapalumba have been analysed to trace back the source area through definition of physical/typological and geochemical properties of the materials. All the fragments are dated to the Neolithic age, on the base of typological features showed by flint artifacts also founded in the archaeological site. The obsidian fragments are black in color, variably transparent to opaque. They mainly include blade scrapers, backed blade and flake scraper; others artifacts are instead classified as not retouched flakes, fragment of core, platform and blades. The overall recognized types suggest that in the Neolithic settlement an intense knapping activity was carried out.

Taking into account that obsidian artifacts are well preserved, that sources are limited in number and that the compositional variability between different obsidian sources is high, it is possible to link artifacts with great confidence to specific sources using elemental analysis. SEM-EDX major element data of the Roccapalumba obsidians, expressed in wt % of oxides (SiO<sub>2</sub>, Al<sub>2</sub>O<sub>3</sub>, FeO, MgO, CaO, Na<sub>2</sub>O, K<sub>2</sub>O) are reported in Table 2.

Table 2. Major elements of obsidian set in wt% determined by SEM (PI=Peralcaline Index).

Sample/ Element	SiO <sub>2</sub>	Al <sub>2</sub> O <sub>3</sub>	FeO	MgO	CaO	Na <sub>2</sub> O	K <sub>2</sub> O	Na <sub>2</sub> O+K <sub>2</sub> O	PI
rocp1	76,43	12,5	1,32	0,18	0,71	3,4	5,47	8,87	0,71
rocp2	75,56	12,32	1,39	0,34	1,03	2,46	6,9	9,36	0,76
rocp3	76,41	12,41	1,41	0,1	0,79	3,32	5,56	8,88	0,72
rocp5	74,52	13	1,5	0,27	1,18	3,42	6,11	9,53	0,73
rocp6	76,35	12,45	1,32	0,03	0,73	3,69	5,43	9,12	0,73
rocp7	76,2	12,54	1,28	0,16	0,92	4,00	4,9	8,9	0,71
rocp8	76,62	12,28	1,35	0,06	0,75	3,4	5,53	8,93	0,73
rocp9	76,44	12,42	1,3	0,08	0,69	3,75	5,32	9,07	0,73
rocp10	76,38	12,55	1,23	0,23	0,73	3,84	5,04	8,88	0,71
rocp11	76,32	12,54	1,26	0,03	0,71	3,91	5,24	9,15	0,73
rocp12	75,78	11,96	1,77	0,11	1,79	3,25	5,34	8,59	0,72
rocp13	76,51	12,45	1,3	0,07	0,8	3,6	5,27	8,87	0,71
rocp14	76,38	12,28	1,32	0,1	0,79	3,76	5,37	9,13	0,74
rocp15	76,18	12,47	1,31	0,15	0,81	3,73	5,36	9,09	0,73
rocp16	76,01	12,6	1,35	0,11	0,78	3,93	5,23	9,16	0,73
rocp17	75,5	12,7	1,48	0,6	0,76	3,84	5,12	8,96	0,71
rocp18	76,59	12,45	1,28	0,1	0,73	3,76	5,08	8,84	0,71
rocp19	76,69	12,41	1,35	bdl	0,79	3,28	5,48	8,76	0,71
rocp20	76,69	12,5	1,37	bdl	0,8	3,47	5,24	8,71	0,7
rocp21	76,53	12,6	1,91	0,11	1,08	3,55	6,32	9,87	0,78
rocp22	76,47	12,5	1,42	bdl	0,79	3,13	5,71	8,84	0,71
rocp24	76,22	12,59	1,25	0,06	0,98	3,96	4,95	8,91	0,71
rocp25	75,96	12,33	1,3	0,11	1,09	3,81	5,38	9,19	0,75
rocp26	76,1	12,76	1,26	bdl	0,77	4,27	4,84	9,11	0,71
rocp27	76,18	12,73	1,31	0,02	0,74	3,86	5,17	9,03	0,71
rocp28	76,29	12,41	1,34	0,08	0,87	3,45	5,57	9,02	0,73
rocp29	75,72	12,31	1,25	0,01	1,82	3,92	4,97	8,89	0,72
rocp30	74,58	12,52	1,6	0,26	1,55	1,65	7,84	9,49	0,76
rocp31	76,44	12,41	1,34	0,03	0,83	3,76	5,19	8,95	0,72
rocp32	76,31	12,64	1,32	0,01	0,86	3,45	5,4	8,85	0,7
rocp33	75,99	12,64	1,39	0,15	1,01	3,48	5,34	8,82	0,7
rocp34	75,74	12,98	1,17	0,03	1,04	4,06	4,97	9,03	0,7
rocp35	75,99	12,79	1,23	0,06	0,74	3,96	5,22	9,18	0,72
rocp36	76,33	12,5	1,28	bdl	0,92	3,86	5,12	8,98	0,72

The studied obsidians show an average SiO<sub>2</sub> content of about 75 wt%, Al<sub>2</sub>O<sub>3</sub> ranging from 12.28 to 13.0%, and Na<sub>2</sub>O+K<sub>2</sub>O values from 8.59 to 9.87 wt%. The total alkali vs silica (TAS after Le Bas, 1986; inset of Fig. 6) classification indicates that all the analysed obsidians fall in the compositional field of rhyolites and are alkaline according to the Miyashiro classification (1978). In particular, all samples show potassic alkaline affinity according to La Maitre (2002) classification (Na<sub>2</sub>O-2 < K<sub>2</sub>O). Moreover, their peralkaline index P.I. = [(Na<sub>2</sub>O + K<sub>2</sub>O)/Al<sub>2</sub>O<sub>3</sub> molar ratio] is < 1 (from 0.70 to 0.78).

The provenance of the studied obsidians is investigated comparing their key major and trace ele-

ments with those of natural occurrences from the Tyrrhenian islands of Pantelleria, Palmarola, Lipari and Sardinia (M. Arci) and from the Aegean Islands of Melos and Gyalı (Crisci *et al.*, 1994; Acquafredda *et al.*, 1999; Tanguy *et al.*, 2003; Bigazzi *et al.*, 2005; Barca *et al.* 2007; 2008; De Francesco *et al.*, 2008; Tykot *et al.*, 2013; Tykot, 2017). The rhyolitic field of the TAS diagram is reported in Figure 6 with compositional fields of the Mediterranean occurrences. The studied samples fall near the Lipari field, but some samples plot in the area where the fields of Lipari, Palmarola and Monte Arci overlap.



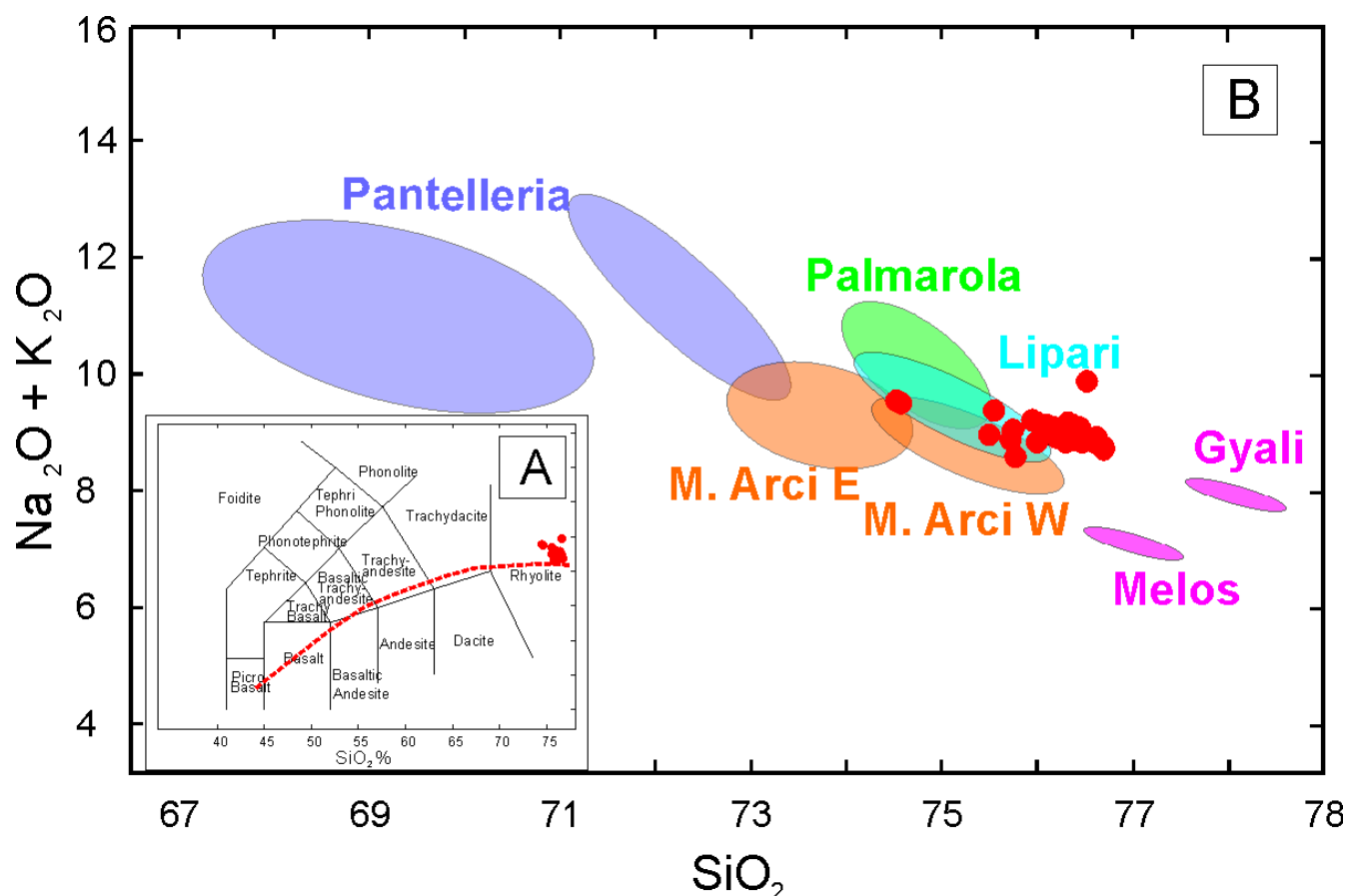


Figure 6 - A) Total alkali vs silica diagram (after Le Bas, 1986), with the discriminating boundary between alkaline and sub-alkaline magmas (dashed red line) after Miyashiro (1978) used to classify the studied Roccapalumba obsidians (red dot); B) Literature compositional fields of the main source areas from the Central Mediterranean area (data after Acquafredda et al., 1999; Le Bourdonnec et al., 2010 and De Francesco et al., 2008).

In order to better discriminate among these three possible source areas, values of the peralkaline index can be used. The low values of P.I. ( $< 1$ ) showed by the studied samples from Roccapalumba allow excluding the Palmarola obsidians, which are alkaline/peralkaline with  $P.I. \approx 1$  or slightly higher (Cadoux et al., 2005). Moreover, also Pantelleria obsidians can be excluded due to their strong peralkaline character and P.I. values  $> 1$  (MacDonald and Bailey, 1973). Despite the affinity with the Lipari obsidians showed by the majority of the studied samples, major elements appear not to be enough discriminative to constrain the source area. These preliminary results only allow to exclude Pantelleria and the Aegean islands as possible sources.

Several authors (e.g.: Crisci et al., 1994; Barca et al. 2007, 2008) demonstrated that some trace elements such as Nb, Sr, Zr, Rb, Yb and Y, are particularly indicative of petrogenetic processes that generated various silicic magmas. Since each magma batch had its own petrogenetic history, the abundances and ratios of these elements are different from one occurrence to the other therefore, they can help to distin-

guish, with high accuracy, the obsidian sources in the whole Mediterranean area.

To exclude M. Arci and other possible source areas, the use of trace elements abundances and their ratios as discriminative parameters represent the most reliable approach. The trace elements analyses of the obsidians artifacts from Roccapalumba performed by LA-ICP-MS, expressed in ppm, are reported in Table 3.

Cs vs Nb and Zr/Y vs La variation diagrams proposed by Barca et al. (2007) turned out particularly discriminative. On these diagrams, the obsidian samples from Roccapalumba ever fall in the field of the Lipari obsidians (Fig. 7A-B). Furthermore, on the Nb/Y vs Zr/Y and Nb/Sr vs Rb/Sr diagrams (Fig. 7C-D) by Acquafredda et al. (2018), the Roccapalumba obsidian artifacts fall again in the Lipari compositional field. Following this discrimination method, it can be confidently concluded that the studied obsidians come from Lipari Island.

In addition to these discrimination diagrams we propose the use of supplementary tools to better define or verify the source area.

*Table 3. Trace elements in ppm analysed by LA-ICP-MS (3 analyses per sample).*

Sample/Elements	rocp1	rocp2	rocp3	rocp5	rocp7	rocp8	rocp9	rocp10	rocp11	rocp12	rocp13	rocp14
Li	89.67	19.92	94.2	20.68	96.42	96.03	101.79	93.95	104.96	94.84	90.43	98.63
Be	7.22	6.04	5.48	5.76	7.21	7.21	8.46	6.58	7.71	7.4	6.11	7.47
B	205.43	215.47	212.37	214.47	225.01	241.51	231.53	214.4	233.23	214.54	207.2	216.34
Sc	11.66	10.75	8.75	8.02	7.95	6.44	7.49	16.8	5.69	6.47	7.12	6.05
Ti	438.16	428.87	438.45	450.11	430.69	465.92	483.93	330.22	380.68	318.56	479.36	389.67
V	0.49	0.66	0.53	1.34	0.59	0.58	0.55	0.55	0.62	0.63	0.58	0.49
Cr	1.6	bdl	1.76	1.52	1.89	bdl	2.22	bdl	1.71	1.3	bdl	1.29
Mn	449.62	448.68	462.31	441.45	471.47	454.12	477.58	451.02	493.14	460.8	449.86	450.33
Co	0.33	0.3	0.3	0.39	0.37	0.34	0.26	0.32	0.4	0.34	0.34	0.37
Ni	0.28	0.35	bdl	0.59	0.18	bdl	bdl	0.18	0.3	bdl	0.17	bdl
Cu	6.95	3.74	6.65	3.59	7.3	7.45	6.85	6.87	7.13	7.17	7.19	7.89
Zn	49.65	51.41	51.09	54.09	49.96	52.99	54.68	51.64	55.29	48.26	44.14	52.91
Rb	306.99	317.65	316.44	313.57	320.43	316.17	333.56	316.96	356.73	325.9	313.04	337.29
Sr	13.3	15.52	14.75	16.87	15.54	14.56	15.84	15.65	18.31	14.36	15.25	15.02
Y	33.26	36.52	36.29	39.28	39.68	36.93	40.92	40.62	46.92	37.17	37.41	39.29
Zr	138.41	150.41	148.53	156.91	161.07	150.42	162.49	164.9	186.95	146.89	148.65	162.04
Nb	32.19	32.33	33.59	32.54	32.68	32.09	33.55	33.81	38.84	33.08	32.15	34.02
Cs	16.06	16.53	16.83	16.81	17.34	16.75	17.56	17.34	20.01	18.63	16.58	18.3
Ba	13.43	16.79	13.68	16.79	14.06	14.06	14.82	15.15	17.38	14.87	12.76	14.45
La	47.42	50.97	52.32	55.05	54.14	50.07	53.75	56.21	66.23	51.43	51.7	55.36
Ce	100.28	102.36	106	104.97	106.43	102.79	109.2	109.85	129.45	112.32	102.62	112.73
Pr	10.06	10.88	10.97	11.49	11.26	10.8	11.32	11.73	13.41	11.44	10.5	11.75
Nd	34.81	37.67	39.23	39.93	39.91	36.59	39.49	41.22	48.69	38.22	37.59	41.05
Sm	7.27	7.9	7.77	8.27	8.13	7.28	8.07	8.34	9.74	7.65	7.82	7.78
Eu	0.11	0.13	0.14	0.12	0.12	0.12	0.13	0.13	0.14	0.1	0.12	0.13
Gd	6.1	6.81	6.82	6.73	7.21	6.41	6.86	7.68	8.18	6.43	6.29	6.57
Tb	0.9	0.95	1.01	1.11	1.02	0.97	1.04	1.09	1.24	0.96	0.97	1.07
Dy	5.72	6.24	6.5	6.72	6.67	6.26	6.56	6.9	7.92	6.62	6.03	6.53
Ho	1.16	1.18	1.31	1.39	1.4	1.29	1.36	1.51	1.72	1.28	1.29	1.34
Er	3.22	3.85	4.02	4.31	4.4	3.91	4.22	4.11	4.76	4.1	3.95	4.17
Tm	0.52	0.65	0.59	0.65	0.7	0.59	0.64	0.67	0.81	0.64	0.67	0.63
Yb	4.13	4.51	4.34	5.04	4.86	4.63	4.35	4.85	6	4.41	4.43	4.68
Lu	0.58	0.65	0.62	0.67	0.71	0.65	0.68	0.67	0.89	0.67	0.67	0.7
Hf	5.18	5.58	5.87	6	6.21	5.41	5.91	6.37	7.73	6.04	5.23	5.88
Ta	2.3	2.48	2.41	2.32	2.55	2.37	2.37	2.55	2.93	2.62	2.31	2.62
Pb	31.55	32.26	33.35	31.72	33.89	35.92	35.29	34.23	41.18	35.41	32.04	36.11
Th	41.28	46.04	46.75	49.34	49.5	45.09	48.65	51.31	64.14	47.93	46.01	50.28
U	14.46	15.05	14.85	14.93	15.42	14.83	15.66	16.01	19.59	16.12	14.25	16.12

Table 3. Continued

Sample/Elements	rocp15	rocp16	rocp17	rocp18	rocp19	rocp20	rocp21	rocp22	rocp24	rocp25	rocp26	rocp27
Li	109.02	91.51	98.43	108.27	101.55	104.03	98.24	94.87	92.89	93.89	91.84	98.69
Be	7.99	7.95	6.86	6.17	7.35	7.93	7.27	7.6	7.85	7.76	6.51	6.42
B	226.27	201.07	225.88	216.23	228.35	224.68	249.74	287.66	215.86	217.11	205.16	219.37
Sc	5.89	6.78	6.1	17.54	9.99	10.95	15.5	9.37	11.32	8.48	6.92	7.09
Ti	372.57	287.84	320.26	341.58	381.98	362.93	350.65	356.62	334.61	431.43	269.69	407.81
V	0.59	0.58	0.56	0.59	0.6	0.56	0.89	0.54	0.63	0.88	0.5	0.55
Cr	2.19	1.7	bdl	0.84	1.3	bdl	1.37	2.85	1.13	2.11	1.96	2.23
Mn	467.48	454.71	472.62	426.04	467.55	460.09	483.37	460.02	476.27	458.76	468.89	445.79
Co	0.31	0.33	0.33	0.33	0.29	0.31	0.36	0.33	0.36	0.39	0.36	0.27
Ni	0.29	bdl	bdl	0.19	bdl	bdl	0.28	bdl	0.18	0.23	0.42	0.17
Cu	6.79	6.74	6.62	6.13	6.91	7.18	7.18	6.24	3.93	6.35	6.59	7.04
Zn	53.46	44.09	49.9	53.96	54.02	52.47	54.02	51.94	53.87	52.17	43.42	43.83
Rb	339.84	321.42	333.81	333.63	336.77	324.75	351.94	324.82	331.02	298.91	313.45	328.59
Sr	15.54	15.29	15.84	14.52	15.37	15.03	16.99	15.91	17.9	14.41	15.46	14.39
Y	37.34	37.93	42.76	36.14	39.5	38.67	43.61	40.89	42.68	32.27	38.82	36.66
Zr	152.87	154.97	167.9	149.11	156.94	153.37	172.38	163.83	166.17	130.39	150.59	148.36
Nb	35.12	33.2	34.22	33.52	33.89	33.04	33.96	33.28	34.59	31.31	32.27	32.74
Cs	18.77	17.08	18.4	18.06	17.93	17.72	19.18	17.54	18.6	16.06	16.83	18.25
Ba	15.23	14.15	15.59	14.45	14.8	14.39	14.78	13.97	15.72	12.96	14.78	13.52
La	54.95	53.23	56.43	51.71	54.49	53.22	56.41	53.79	57.49	46.58	53.02	51.27
Ce	118.21	105.54	112.57	105.45	110.82	109.58	114.73	106.97	115.37	97.91	107.36	106.14
Pr	12.1	11.32	11.92	11.12	11.44	11.37	11.79	11.12	12.1	9.89	10.85	10.92
Nd	41.59	39.98	40.38	38.05	39.89	39.46	42.22	40.31	42.07	34.28	37.68	37.12
Sm	7.83	7.62	8.2	7.66	7.77	7.57	8.58	8.15	8.65	6.89	7.49	7.34
Eu	0.13	0.14	0.15	0.11	0.12	0.14	0.13	0.13	0.16	0.12	0.13	0.1
Gd	7.29	6.42	7.22	6.83	6.88	7.18	8.18	7.41	7.9	5.9	6.61	6.18
Tb	1.01	1.05	1.08	0.99	1.01	1.02	1.07	1.07	1.09	0.81	0.93	1
Dy	6.82	6.74	7.42	6.09	6.65	6.41	7.07	6.58	7.38	5.39	6.77	6.05
Ho	1.34	1.33	1.5	1.31	1.36	1.37	1.44	1.36	1.49	1.14	1.31	1.3
Er	4.21	3.96	4.55	3.79	4.12	4.09	4.18	4.15	4.52	3.56	4.05	3.85
Tm	0.64	0.68	0.71	0.61	0.67	0.64	0.67	0.66	0.68	0.52	0.63	0.59
Yb	4.89	4.31	5.14	4.49	4.8	4.54	5.09	5.13	5.06	3.76	4.45	4.43
Lu	0.7	0.65	0.8	0.64	0.67	0.69	0.7	0.68	0.75	0.54	0.69	0.68
Hf	5.82	5.98	6.76	5.67	5.96	5.92	6.49	6.11	6.54	4.83	5.82	5.56
Ta	2.67	2.46	2.68	2.49	2.48	2.54	2.56	2.49	2.6	2.14	2.34	2.36
Pb	38.07	34.59	37.01	36.51	35.74	35.27	38.17	39.3	37.54	31.38	33.49	32.65
Th	52.82	50.09	53.88	47.18	49.78	50.35	53.03	50.41	55.6	42.17	46.69	45.9
U	17.98	15.68	16.67	15.95	16.33	16.07	16.7	15.21	17.73	14.84	15.27	15.49

*Table 3. Continued*

Sample/Elements	rocp29	rocp30	rocp31	rocp32	rocp33	rocp34	rocp35	rocp36
Li	95.2	19.23	97.87	95.72	99.85	95.48	97.81	95.64
Be	6.17	6.56	9.72	7.41	8.5	7.72	6.56	6.32
B	206.84	210.96	222.71	214.83	229.08	251.74	206.15	213.93
Sc	6.98	11.61	9.93	14.37	6.29	11.52	7.04	7.52
Ti	350.76	414.46	437.64	350.9	320.65	395.97	447.6	434.99
V	0.63	0.61	0.61	0.58	0.64	2	0.59	0.55
Cr	bdl	1.26	1.64	2.08	1.21	4.32	bdl	bdl
Mn	453.05	453.02	466.37	488.65	466.43	561.66	462.07	457.96
Co	0.33	0.31	0.37	0.33	0.33	0.56	0.36	0.35
Ni	bdl	0.41	0.16	0.22	bdl	2.89	0.23	0.23
Cu	6.79	2.74	7.02	6.67	6.54	10.01	6.77	7.73
Zn	43.76	49.98	52.67	54.96	51.8	75.2	50.29	50.2
Rb	314.44	365.14	324.81	331.1	337.13	330.55	306.13	310.77
Sr	14.22	21.62	15.57	14.57	15.53	19.29	13.56	14.6
Y	34.64	34.26	41.57	40.16	40.09	42.14	31.78	33
Zr	138.06	139.09	161.53	158.44	158.83	162.05	129.76	135.71
Nb	31.65	31.57	33.09	34.11	33.94	34.18	31.81	31.91
Cs	17.24	15.77	17.56	17.82	18.64	18.57	15.97	16.84
Ba	13.18	45.98	14.22	13.5	15.33	20.18	12.92	13.54
La	48.52	50.07	54.77	55.71	55.56	56.2	45.58	49.13
Ce	101.83	103.27	109.5	111.59	113.38	115.96	96.6	102.56
Pr	10.19	10.85	11.99	11.75	11.91	11.55	9.9	10.69
Nd	34.47	37.87	40.93	40.7	40.87	41.85	33.98	35.97
Sm	6.8	7.5	8.07	7.84	7.96	8.41	6.31	6.98
Eu	0.11	0.12	0.14	0.11	0.13	0.17	0.1	0.15
Gd	6.17	6.24	7.37	7.74	7.07	7.72	5.67	5.93
Tb	0.91	0.98	1	1.13	1.06	1.1	0.87	0.88
Dy	5.93	5.93	6.71	6.54	7.14	6.69	5.32	5.66
Ho	1.18	1.21	1.37	1.41	1.4	1.46	1.11	1.16
Er	3.75	3.5	4.39	4.26	4.57	4.25	3.4	3.4
Tm	0.59	0.58	0.68	0.68	0.7	0.67	0.5	0.57
Yb	4.18	4.24	4.93	4.84	5.08	5.42	3.62	4.03
Lu	0.64	0.6	0.73	0.71	0.74	0.73	0.59	0.55
Hf	5.33	5.26	6.46	6.13	6.62	6.42	4.75	5.12
Ta	2.25	2.33	2.37	2.54	2.66	2.61	2.21	2.31
Pb	32.44	31.22	33.41	35.01	37.78	42.51	30.63	32.12
Th	42.43	44.49	51.36	50.98	54.64	38.37	40.81	43.08
U	14.44	14.81	16.16	16.55	17.76	38.67	14.23	14.9

A different way to characterize the compositions of volcanic rocks are normalised diagrams of incompatible elements and REE, in which each element abundance is divided by its concentration in chondrites of the primordial mantle.

Chondrite-normalised REE patterns are reported in Fig. 8A. The Roccapalumba obsidians closely match data of Lipari rhyolitic rocks, and are different from literature data of the other source areas plotted for comparison (Peccerillo, 2005; Forni *et al.* 2013). Both groups have similarly fractionated patterns en-

riched in LREE (Roccapalumba  $\text{La}/\text{Yb}_N=6.2$  to  $7.5$ ; Lipari rhyolites  $\text{La}/\text{Yb}_N=5.48$  to  $8.9$ ), relatively flat HREE (Roccapalumba  $\text{Tb}/\text{Yb}_N=0.8$  to  $1.03$ ; Lipari

rhyolites  $\text{Tb}/\text{Yb}_N=1.06$  to  $1.18$ ), and a strong negative Eu anomaly (Roccapalumba  $\text{Eu}/\text{Eu}^*=0.03$  to  $0.07$ ; Lipari rhyolites  $\text{Eu}/\text{Eu}^*=0.04$  to  $0.07$ ).

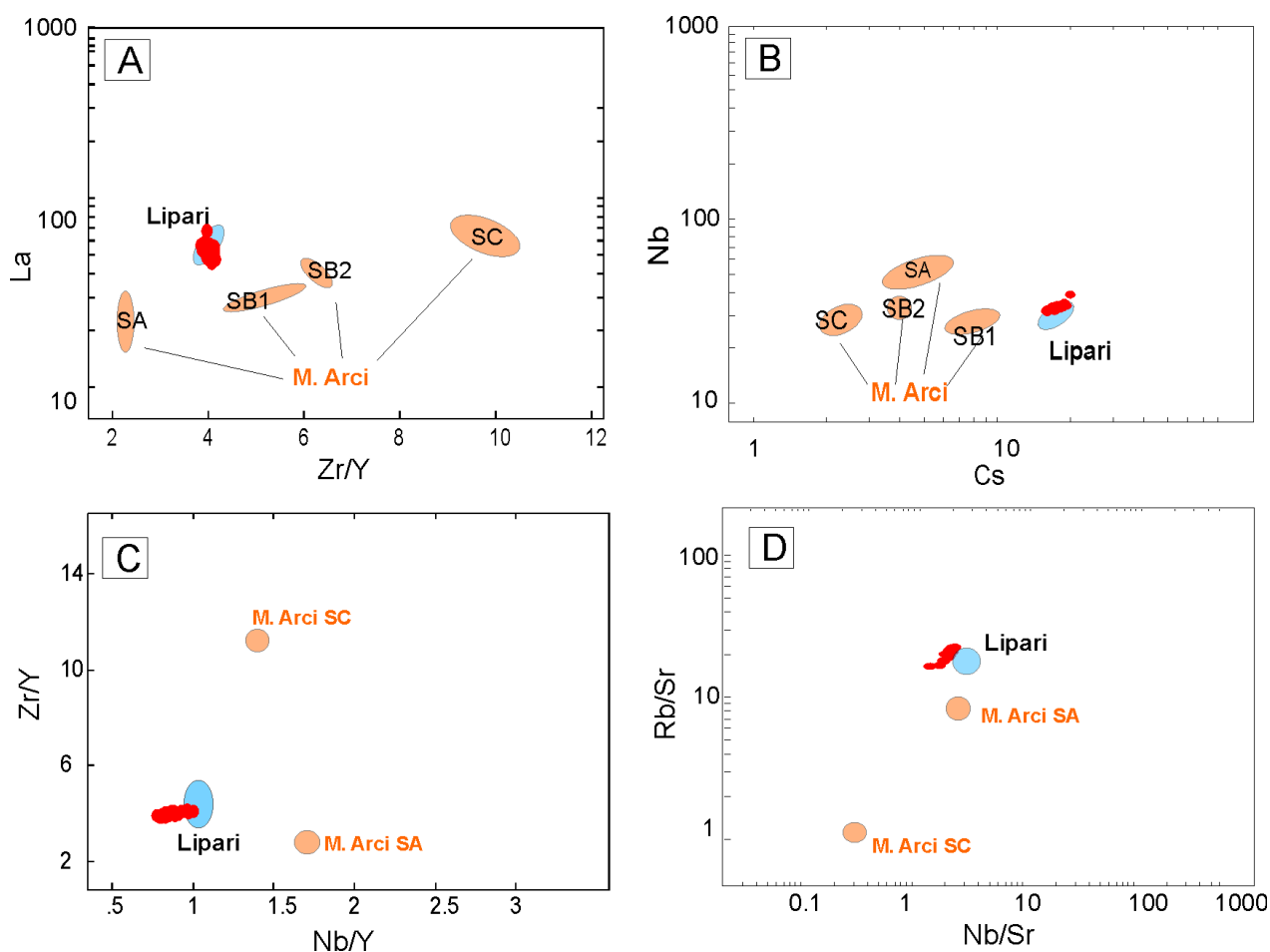


Figure 7 - A)  $\text{Zr}/\text{Y}$  vs  $\text{La}$  and B)  $\text{Cs}$  vs  $\text{Nb}$  discrimination diagrams by Barca et al., (2007); C)  $\text{Nb}/\text{Y}$  vs  $\text{Zr}/\text{Y}$  and D)  $\text{Nb}/\text{Sr}$  vs  $\text{Rb}/\text{Sr}$  discrimination diagram by Acquafredda et al. (2018). All the data are in ppm.

Patterns normalized against primordial mantle compositions of Roccapalumba obsidians are also very similar to the Lipari rhyolites and different from those of the other possible source regions. They show enrichment in some LILE (Cs, Rb and K), as well as a significant Sr (19-21 ppm) and Ba (12-45 ppm) depletion (Fig. 8B). They also show slight negative anomalies of Ta and Nb, and, to some extent, of Zr and Hf, along with a strong negative anomaly in Ti. As a matter of fact, the geochemical data of the archaeological Roccapalumba artifacts closely match those from the Lipari rhyolites, strongly supporting Lipari Island as the source of the investigated obsidian prehistoric artifacts.

Concerning the Lipari obsidians, recent volcanological studies (Forni et al., 2013) provided dating about all the obsidian outcrops of the island. The island of Lipari hosts many obsidians sources (Big-

azzi et al., 2005; Tanguy et al., 2003; Tykot et al., 2013; Tykot, 2017) but most of them, such as Forgia Vecchia, Rocche Rosse (1.6–1.4 ka), Lami (0.70 ka) and the historical lava flow from Rocche Rosse (1220±30 AD) are too young and could not exist in the Neolithic age. In this context, only the Pomiciazzo or Gabelotto obsidian flow dating back to 11.4 ka, 8.6 ka and 7.17 ka, could have been exploited as obsidian quarry in the Early Neolithic period. Indeed, it was in the Middle Neolithic (half of 6<sup>th</sup> millennium BC) identified with the Stentinello facies (Martinelli, 2016) that prehistoric populations began to settle in the Aeolian Islands and started the intense exploitation of obsidian as raw material to made tools. During Neolithic period the obsidian from Lipari is the main source diffused in the South of Italy (Acquafredda et al., 2018).

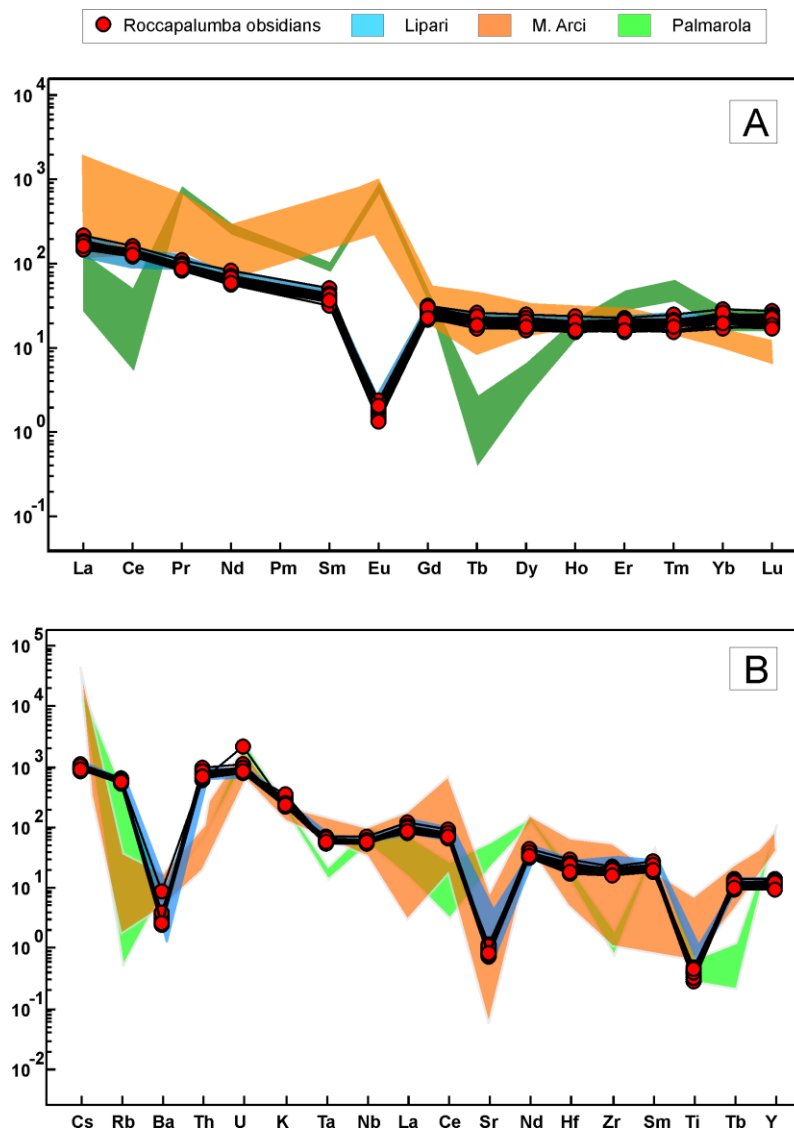


Figure 8 – A) Chondrite-normalised REE pattern (Boynnton, 1984) and B) Primitive mantle-normalized trace element spider diagram (Taylor and McLennan, 1985) of the Roccapalumba obsidians. For comparison patterns of the other possible source regions have been plotted (data from Peccerillo, 2005).

## 5. CONCLUSION

The studied Neolithic obsidian artifacts from Roccapalumba were attributed to obsidians traded from Lipari. They show geochemical fingerprint which is in agreement with that of other obsidian artifacts and obsidian rocks from Lipari Island reported in literature. Further confirmation about the Lipari provenance of obsidians is provided by the different REE elemental abundances and incompatible trace elements patterns of rhyolitic rocks from the Tyrrhenian source regions.

Recent researches on obsidian provenance in Sicily highlighted a clear geographic differentiation about the obsidian procurements. Eastern Sicily settlements made their imports exclusively from Lipari (Martinelli and Quero, 2013), while the settlements from Central-Western Sicily, supplied themselves either from Lipari and Pantelleria islands (e.g. Fran-

caviglia and Piperno, 1987; Francaviglia, 1988; Tykot *et al.*, 2013). Despite the geographical position of the investigated site (Central-Western Sicily), our results evidence that obsidian from Pantelleria Island was not exploited by prehistoric inhabitants of Roccapalumba settlement.

In general, the circulation of the obsidian raw material becomes consistent starting from the Stentinello facies when Lipari begins to be permanently inhabited (Cavalier, 1979, 1997; Bernabò Brea and Cavalier, 1995). Sicily and Calabria are directly involved through the exchange of other materials and the transport of obsidian blocks by sea. The Roccapalumba site is another witness of the obsidian diffusion in Italy (Tinè and Pessina, 2012). The analysis of the obsidian discovered in the Neolithic sites, far away from the source, allows tracing with better precision the area of its circulation.

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