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ORIGINAL PAPER

Provenance and technology of fourth–second century BC glass from three sites in ancient Thesprotia, Greece

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Abstract

Thesprotia, one of the most remote regions in Greece, was inhabited from as early as the Palaeolithic period. The particular geomorphological terrain, with the mountainous and fragmented landscape, has been determinant in the formation of economic and social institutions throughout antiquity. Thesprotia was gradually developed into an important node of communication and transport of goods to the West and the mountainous hinterland of Epirus. During the second half of fourth century BC, socioeconomic changes occurred in the region and small villages were joined to form the first organised settlements. Elea, Gitana and Dymokastro were founded within a few years from one another, during the fourth century BC. Built at geographically crucial locations that ensured the control of the valleys or the riverside crossings and sea routes, they evolved gradually into political, economic and administrative centres for the surrounding areas. In the present study, 56 samples of glass, excavated from these three sites in Thesprotia, are investigated using analytical techniques (SEM-EDX and LA-ICP-MS). The chemical compositions of the samples show significant differences in raw materials used and provide evidence for provenance for the artefacts. This is the first study to examine Hellenistic glass from within a region of northern Greece. The results are compared with other published compositional data for Hellenistic glass. The analytical results for the majority of glass samples from the three sites in Thesprotia show with high probability a Levantine origin and therefore also possibly for the artefacts themselves. This confirms the archaeological record of trade in other materials/objects, while a small group of glasses from Gitana in Thesprotia were made in Egypt.

Keywords Natron glass · Hellenistic period · Thesprotia · Greece · Trace elements · Chemical composition · SEM-EDX · LA-ICP-MS

Introduction

During the last decade, analytical studies of ancient glass dating to between the seventh and first century BC in the Greek region have been the focus of the scientific community. The researchers have paid attention mainly to the Archaic and Classical period (Brill 1999; Triantafyllidis 2000; Oikonomou et al. 2008; Zacharias et al. 2008a,b; Sokaras et al. 2009; Beltsios et al. 2012; Oikonomou et al. 2012; Triantafyllidis et al. 2012; Cheilakou et al. 2012; Oikonomou et al. 2014; Palamara et al. 2015; Blomme et al. 2016 2017; Oikonomou and Triantafyllidis 2018) and little is known about Hellenistic glass raw materials, technology and provenance (Rehren et al. 2005; Brill and Stapleton 2012; Connolly et al. 2012; Oikonomou 2018; Smirniou et al. 2018; Oikonomou 2019). The focus of this article is an analytical study of Hellenistic glass excavated from the three sites of Thesprotia region, in Greece, in order to redress the balance of our knowledge of glass technology and provenance from this time period. The main objectives of this work are to shed light to the glass technology used in this period and to try to suggest a provenance for the glasses. Furthermore, potential trade routes and connections between these major sites with Egypt and Levant will be investigated using an archaeometric approach.

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Glassmaking evolved as a trial and error procedure, and after long experimentation, ancient glassmakers were able to mix the correct proportions of raw materials to produce the base glass. Base glass was produced by mixing two (or three) main components (Brill 1988). Using more than three components would add complexity to the procedure and finding the correct proportions to produce glass would be rather difficult. The two main components used in ancient glassmaking were sand or quartz pebbles that acted as the glass former and the alkalis, mineral or plant ash that acted as a flux to lower the melting point of the network former.

Sand or/and quartz pebbles provide silica (SiO_2) which is the main component of glass. Sand, a less pure raw material than quartz pebbles, introduces other elements in the main glass composition as impurities such as iron (Fe_2O_3) and especially alumina (Al_2O_3) associated with feldspars. Most of the glass of the first millennium BC was manufactured with the fusion of sand.

The sand sources suitable for glassmaking are very few and include places across the Mediterranean lands from the West to the East, as well as in Egypt, the Levant and inland locations (Brill 1988, 1999; Degryse 2014, Henderson et al. 2020). Apart from Fe_2O_3 and Al_2O_3, sand can also contain trace elements such as Ti, Cr, Zr, La and Nd. The different proportions of the aforementioned elements will form part of the final glass compositions since their concentrations don't change significantly through glass fusion, especially since they are non-volatile elements. Therefore, by investigating the relationships between these elements, we can get information about the use of different raw materials and attempt to determine their provenance.

Flux was also a necessary ingredient in ancient glassmaking. It could either be a mineral or a plant ash (Henderson 2013, 22-55). The mineral used extensively in ancient glassmaking referred to as 'natron' was formed when certain lakes evaporated. The most likely source of evaporitic minerals during antiquity was the region of Wadi Natrun, in northern Egypt, located approximately 100 km from Cairo. Except for this major source, other possible locations could be found in Tarabiya (Nile Delta), al-Kab in upper Egypt or Bi'r Natrun on the route to Sudan (Shortland et al. 2006), el-Barnugi (Jackson et al. 2018) and possibly, but less likely, Pikrolimni Northern Greece (Devulder et al. 2014). Recent data has also revealed natron sources in Anatolia having very specific chemical characteristics (Dardeniz 2015). The use of such minerals as a flux in glassmaking in the Mediterranean during antiquity happened especially from the middle of first millennium BC till the end of first millennium AD) (Henderson 2013, 92-97). These mineral deposits contain various minerals in different proportions such as natron ($Na_2CO_3.10H_2O$), trona (Na_2CO_3.-$NaHCO_3.2H_2O$), burkeite ($Na_6CO_3.2SO_4$) and/or halite (NaCl) (Devulder et al. 2014).

The current study aims to investigate the technology of fourth–second century BC glasses found in the three locations in Thesprotia and by examining the major, minor and trace levels of element oxides in the glasses and suggest the raw materials used to make them. Moreover, for the first time, we investigate compositional variations in glasses found in a region of northern Greece, Thesprotia. The compositional variations provide a provenance for the glasses and therefore potentially the objects too. This in turn will provide evidence of trade between the three cities and the outside world.

Archaeological background

The glass finds investigated in this article come from three very important sites for the archaeology of Western Greece Elea (DMS: 39°26'24.1"N 20°33'00.8"E), Gitana (DMS: 39°34'15.2"N 20°15'39.4"E) and Dymokastro (DMS: 39°19'48.9"N 20°17'37.2"E) which are located in the region of Thesprotia in northwestern Greece (Fig. 1). Thesprotia, a territory with the geopolitical importance in the western part of Epirus, was inhabited from as early as the Palaeolithic period. According to ancient sources, Thesprotians, one of the three most important tribes in Epirus along with Molossians and Chaones, till the end of the fifth century BC–beginning of the fourth century BC, were organised in smaller tribes and they lived in small unprotected settlements (Hammond 1967; Dakaris 1972). Their economy was mainly agricultural. In the second half of the fourth century BC, they organised themselves politically into the Thesprotian League and they participated at the Epirotic Alliance. Later on, they formed part of the Koinon of the Epirotes (233 BC), with which their history was associated with thereafter. In the second half of the fourth century BC, they founded big cities with walls, such as Elea and Gitana; this was accompanied by a population rise. Thesprotians had trade connections mainly with Corfu island and Corinth. Both of these settlements were colonists of the west coast of Epirus from as late as the Archaic period.

The time of most prosperity for the whole Epirotic region, including Thesprotia, was the period from the fourth–third century BC until the Roman occupancy in 167 BC. Some historical geopolitical conditions such as the Peloponnesian war, the decline of Polis-Kratos and the rise of political coalitions allowed Thesprotians to prosper through to the Hellenistic period.

One of the most important settlements that Thesprotians founded, a little before the fourth century BC, was the settlement of Elea. It is located on a naturally fortified plain, at an average height of 500 m above sea level and controlled the trading connections across a wide plain. The settlement flourished during the third to second century BC in the Hellenistic period and was destroyed in 167 BC by the

Fig. 1 Map of Epirus region indicating the three major cities of ancient Thesprotia during Hellenistic period

Romans. Elea seems to have been the seat of the Thesprotian League for several decades till 330 BC when the seat moved to the other big city in the region, Gitana (Riginos and Lazari 2007).

Gitana extends across the southwestern slope of a mountain, while the Kalamas river (the ancient Thyamis river) served as natural protection and borders the city at its western and southern sides. Gitana served as the political centre and seat of the Thesprotian League from the time of its establishment in the second half of the fourth century BC until its occupation by the Romans in 167 BC (Kanta-Kitsou 2008).

The third important settlement during the Hellenistic period in Thesprotia was Dymokastro, a coastal fortified settlement founded during the second half of the fourth century BC. The settlement flourished during the Hellenistic period until its devastation by the Romans in 167 BC. The importance of the settlement is testified by the continuation of habitation till the first century AD and its key coastal position secured control over maritime routes along the Ionian Sea (Lazari et al. 2008).

The three cities were connected through a vast number of inland routes including low-cost routes according to GIS studies (Liakos and Vasiliadis 2008). In addition, the three cities, having access to the Ionian Sea, would have been connected with various sea routes. Gitana had access to the sea through the Kalamas river which was in its control during the late Classical and Hellenistic period and Elea through the Acheron river and its port at the delta of the river. Dymokastro, being a coastal settlement, had its own port and was the only Thesprotian city which developed intense naval activity. Gitana and Elea, excepting the sea route connection, were also possibly connected with an inland route (through the Vasilaki hills) where diachronic archaeological remains were found. Furthermore, a possible inland route to connect Elea and Dymokastro has been suggested by Liakos and Vasiliadis (2008). It is worth noting that Thesprotia as part of the greater Epirus region during the Hellenistic period had close relations with the two major artistic centres of this era, Alexandria in Egypt and Taranto in Italy, which are historically and archaeologically well documented (Lévêque 1957; Franke 1961; Hammond 1967; Cabanes 1976).

All the above prove that the three cities had an important role in the trade of goods, across Epirus and the Greek mainland, and this study aims to investigate how the three centres in Thesprotia impacted on the commercial networks of this period through the extensive archaeometrical study of glass and its provenance.

Materials and methods

In the present study, 56 fragments of glass vessels are investigated using SEM-EDX and LA-ICP-MS (the analytical data obtained can be found in the Supplementary Material Table I).

All samples were excavated from the three major sites of ancient Thesprotia discussed above, Elea, Gitana and Dymokastro. The dating of some samples is rather ambiguous. The archaeological context of the excavations (mainly dated by pottery) and the fact that the three cities prospered during the Hellenistic period (fourth–second century BC) indicate that the samples probably date to this period. From 168 to 167 BC onwards, Thesprotia was destroyed by the Roman invasion and the cities were mostly abandoned. However, there is evidence that Dymokastro was occupied in later periods, and this is reflected in the chemical composition of a group of five samples (DY12a, 14, 15, 23, 28) which comes from poorly stratified contexts and is probably of a later (perhaps early Roman) date. The samples tested consist of various types of glass vessels including cast, grooved and ribbed bowls, while there is a substantial number of fragments of less diagnostic vessels, which are most likely part of cast vessels, typical of the Hellenistic era (Table 1). The coloration of the samples varies: they can be divided in 4 main colours: olive green, dark blue, brown and colourless transparent. The vessel type, colour, sample number and origin of the samples are listed in Table 1. In Supplementary Material (Table II), there is a detailed catalogue of the sampled fragments including photographs.

Table 1 General description of the glass fragments

Region	Sample no.	Typology	Colour
Elea	EL.4	Ribbed bowl	Violet
($n = 6$)	EL.5	Grooved bowl	Violet
	EL.6	Non-diagnostic	Violet
	EL.7-9	Non-diagnostic	Colourless
Gitana	GT.10, 24-27, 41	Non-diagnostic	Deep blue
($n = 17$)	GT.11	Non-diagnostic	Blue opaque
	GT.28-30	Non-diagnostic	Turquoise
	GT.31	Grooved bowl	Violet
	GT.32, 33a	Non-diagnostic	Amber
	GT.33b, 34	Non-diagnostic	Olive green
	GT.35	Non-diagnostic	Aqua blue
	GT.36	Grooved bowl	Colourless
Dymokastro	DY.1-2	Mosaic glass	Multicolour
($n = 33$)	DY.5	Ribbed bowl	Colourless
	DY.4, 6, 47-48	Cast or sagged bowl	Colourless
	DY.7, 25	Ribbed bowls	Amber
	DY.13	Ribbed bowl	Olive green
	DY.9, 49-50	Non-diagnostic	Aqua blue
	DY.10	Non-diagnostic	Transparent
	DY.11,12b	Non-diagnostic	Transparent
	DY.8, 12a, 22-23	Non-diagnostic	Amber
	DY.14-15, 27-28, 53	Non-diagnostic	Olive green
	DY.21,46	Non-diagnostic	Deep blue
	DY.24	Grooved bowl	Amber
	DY.26,52	Non-diagnostic	Green
	DY.31-35	Non-diagnostic	Colourless

Photographs of the samples can be found in Supplementary Material (Table II)

A JEOL (JSM-6510LV) scanning electron microscope was used for the detection of major and minor elements in the 56 glass samples. The scanning electron microscope is equipped with an energy dispersive X-ray spectrometer made by Oxford Instruments. All samples were analysed under high vacuum, with an operating voltage at 20 kV and working distance for each sample of 15 mm. The calibration of the system was performed with geological standards and the accuracy/precision was established by analysing standard reference materials (NIST SRM620, SRM1831 and SRM612). The analyses of the standards are in close agreement with the expected values and are presented in Table 2. The relative error between the expected and measured values is approximately 5% for most of the oxides. Due to submicrometer beam size, 5 analyses of 300 s were performed on each sample and the mean value was calculated for each element.

Trace element characterization was performed by laser ablation inductively coupled plasma mass spectrometry (LA-ICP-MS). The ablation was conducted with a NewWave UP193FX excimer (193 nm) laser system, with built-in microscope imaging, which was coupled to an Agilent 7500 series ICP-MS. Laser ablation craters were set at 70 μm, the laser being fired for 45 s at 10 Hz and a typical fluence of 2.8 Jcm^{-2}. Data was collected in a time-resolved analysis mode, with a gas blank being measured before a series of ablations on glass samples; calibration standards and quality control standards were carried out. Calibration standards bracketed the samples and QC over a period of 1 h or less. Calibration of the system was performed using NIST SRM610 trace element glass standard. The measured and expected values are

Table 2 Measured and expected values of major and minor oxides for standard reference materials

Sample	Na_2O	MgO	Al_2O_3	SiO_2	SO_3	K_2O	CaO
SRM620	14.19	3.66	1.72	73.59	0.21	0.39	6.27
Expected	14.39	3.69	1.8	72.08	0.28	0.41	7.11
r.e.	*1.4*	*0.8*	*4.4*	*2.1*	*25.0*	*4.9*	*11.8*
SRM1831	13.67	3.67	1.20	74.03	0.27	0.29	6.9
Expected	13.32	3.51	1.21	73.08	0.25	0.33	8.2
r.e.	*2.6*	*4.6*	*0.8*	*1.3*	*8.0*	*12.1*	*15.9*
SRM612	13.94	-	2.06	72.93	-	-	11.08
Expected	13.7	-	2.03	72.1	-	-	11.9
r.e.	*1.8*	-	*1.5*	*1.2*	-	-	*6.9*

The expected values for SRM612 were provided by GeoRem (Jochum et al. 2011). *r.e.* relative error presented in Table 3. NIST SRM612 was used for quality control purposes (Table 3).

Both techniques were applied on small fragments, cut in cross sections, which were embedded in resin blocks resulting in a fresh flat glass surface. The resin blocks were further treated with silicon carbide papers and polished using diamond pastes down to 1 μm removing any corrosion layers.

Results and discussion

Major oxides

The majority of samples from Thesprotia are soda-lime-silica glasses. They contain SiO_2 of between 51.1% and 73.6 % wt. and Al_2O_3 of between 1.7 and 2.6 % wt. (Fig. 2). In the same plot, there are six samples (GT11, 27-30 and DY23) which have lower SiO_2 levels (below 66% wt.), while four samples were excluded from the calculations of minimum and maximum values of Al_2O_3, having very unusual concentrations; two of the samples have low levels of Al_2O_3 (0.4–0.6 % wt.) and the rest have high levels of Al_2O_3 (~ 4% and ~ 15% wt.). The low Al_2O_3 (DY 26 [0.61%], 52 [0.59%]) suggests that quartz pebbles were the silica source; these samples also show typical low amounts of trace elements, such as La and Nd (La: 2.2–2.3 and 1.6–1.5 mg/kg, respectively) which supports this interpretation; provenance of quartz sources though is more difficult than sand because different quartz sources may have similar impurity patterns (Henderson 2013, 309). The third exception (DY27) contains relatively high Al_2O_3 (4.0 % wt.), low SiO_2 (51.1% wt.) and a significant amount of PbO (26.1% wt.), making it a sample with a rather unusual (dating possibly to the seventeenth–eighteenth century) chemical composition. Finally, the fourth exception (DY53) has a strangely very high Al_2O_3 (15.6% wt.), possibly indicating the interaction of glass with the crucible. In addition, it exhibits very low CaO (3.5% wt.) and almost no Fe_2O_3 (0.06% wt.).

Furthermore, all samples from Thesprotia have a mean CaO value of 5.9 % wt. which can be considered quite low compared to some natron glass compositions. However, the majority of samples have values between 5 and 8% wt. (Fig. 3). On the same plot, there are 12 samples (GT10, 11, 27-30, 41, DY50, 53, EL4-6) which have lower CaO content (≤ 5% wt.). These samples (from now on referred to as 'low Ca') have an average CaO content of 3.95% wt. which is close to a group of samples Nenna et al. (2000) identified as having an Egyptian origin (av. 3.7% wt.) and more specifically coming from the area of Wadi Natrun and Alexandria in Egypt. Moreover, the majority of low CaO glasses contain low SiO_2 levels as discussed above. It is of interest to investigate further the relationship between SiO_2, Na_2O and CaO (which are normally accounted for circa 90% or more of glass

Table 3 Measured and expected values (mg/kg) of trace elements for standard reference glass NIST612 by LA-ICP-MS

Sample	Li	B	Ti	V	Cr	Mn	Co	Ni	Cu	Zn	As	Rb	Sr	Y	Zr	Sn	Sb	Ba	La	Ce	Nd	Pb	Th	U
NIST 612	39.5	34.9	40.7	37.8	35.0	37.6	34.2	38.8	37.2	43.4	33.3	31.4	77.8	38.2	38.1	38.4	33.1	39.3	35.6	37.7	34.4	38.0	36.6	36.3
Expected	40.2	34.3	44.0	38.8	36.4	38.7	35.5	38.8	37.8	39.1	35.7	31.4	78.4	38.3	37.9	38.6	34.7	39.3	36.0	38.4	35.5	38.6	37.8	37.4
r.e.	*1.7*	*1.7*	*7.5*	*2.6*	*3.8*	*2.8*	*3.7*	*0.0*	*1.6*	*11.0*	*6.7*	*0.0*	*0.8*	*0.3*	*0.5*	*0.5*	*4.6*	*0.0*	*1.1*	*1.8*	*3.1*	*1.6*	*3.2*	*2.9*

The expected values for SRM612 were provided by GeoRem (Jochum et al. 2011). *r.e.* relative error

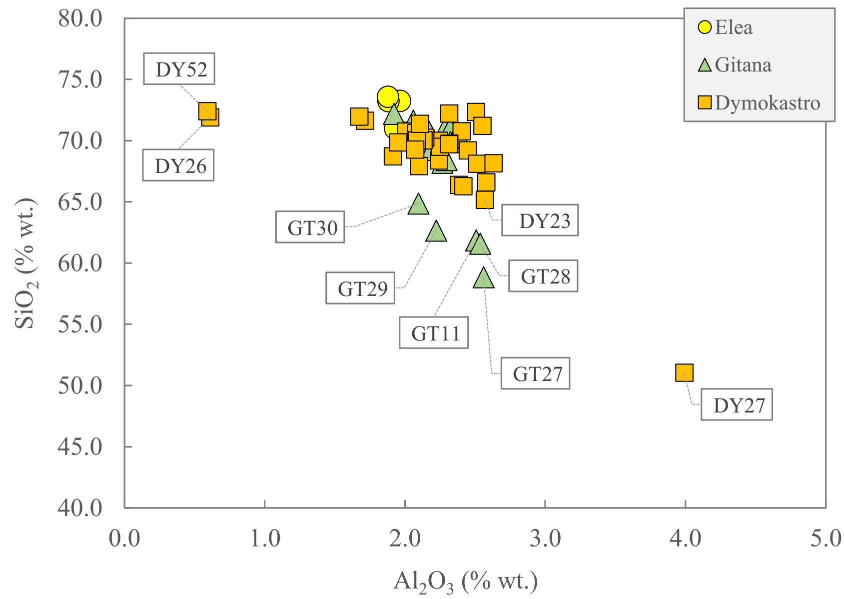


Fig. 2 Al_2O_3 vs SiO_2 for the samples of the three Hellenistic cities

compositions) in Thesprotia samples and to compare them with published data for other eastern Mediterranean Hellenistic glasses. More specifically, in the ternary graph of Fig. 4, the glass from Thesprotia is compared to glass from the island of Rhodes (Brill 1999; Triantafyllidis 2000; Rehren et al. 2005; Brill and Stapleton 2012), from the site of Pherai, in Thessaly, central Greece (Connolly et al. 2012), from the House of Orpheus at Paphos in Cyprus (Cosyns et al., 2018), from the Hellenistic site of Jebel Khalid in Syria (Reade and Privat 2016) and from Beirut (Thirion-Merle 2005) (see Table 4). The data have been normalised to 100% and for clarity the mean values are plotted for each region. As it can be seen in Fig. 4, there is a clear distinction between the two groups of samples based on CaO levels: the 'low Ca' samples from Thesprotia showing similar chemical characteristics to Egyptian samples and the other Hellenistic glasses with higher CaO levels.

Flux

Natron is a purer source of sodium than plant ashes and contains relatively low levels of both MgO and K_2O (usually below 1.5% wt.). As it can be seen in Fig. 5, all samples are plotted in the low MgO low K_2O area (MgO < 1.4% and K_2O < 1.3%), indicating the use of evaporitic minerals as a flux.

It is interesting to see in Fig. 5 that there are two distinguishable groups. The majority of samples show a positive but weak correlation. There are also 8 samples, DY12a, 14, 15, 23, 27, 28,

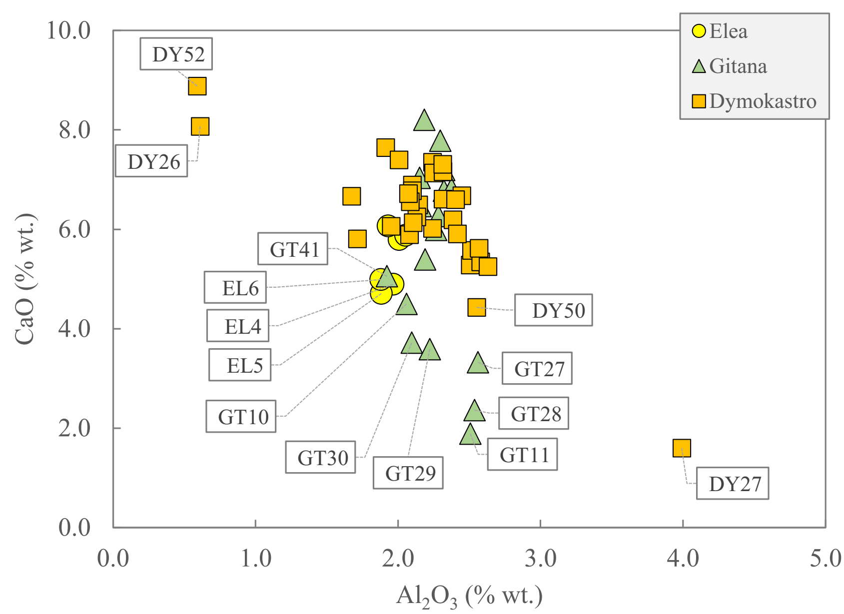


Fig. 3 Al_2O_3 vs CaO for the samples of the three Hellenistic cities

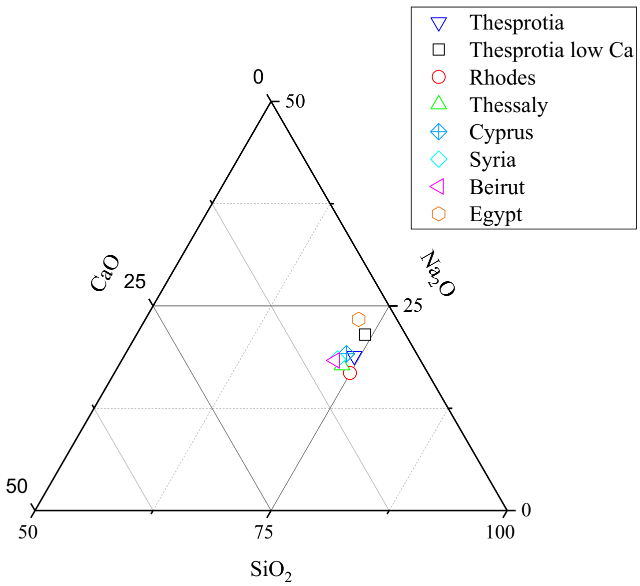


Fig. 4 Ternary plot of SiO_2, Na_2O and CaO. All data are normalised to 100%

GT11 and 28 (in the ellipse in Fig. 5), that form a separate non-correlated group, having relatively low K_2O but elevated MgO contents. These glasses have mean values of 0.36 % wt. K_2O and 1.04 % wt. MgO, compared to 0.88 % wt. and 0.57 % wt. for the majority of samples. This second group may have been manufactured with natron with different chemical characteristics than the natron used for the majority of glass. It is interesting to note that glass from Wadi Natrun has similar values for K_2O and MgO (av. 0.34 % wt. and 0.9 % wt., respectively; see Table 4) (Nenna 2000), and in addition, Jackson et al. (2018) suggest that natron coming from el-Barnugi (a natron source close to Naucratis in Egypt) can add an excess of MgO of around 0.3% wt. which could explain the somewhat elevated values of MgO for this group of samples. This is a clear distinction though it can be also attributed to different processes in the purification of evaporate minerals as Shortland et al. (2011) have suggested. Other possibilities include seasonal harvesting of evaporitic minerals which could provide differences in the chemical composition (Shortland 2004) and potentially the elevated MgO is caused by the introduction of an impurity during a glass working process (e.g. fuel ashes) (Paynter 2008). Moreover, the samples with non-correlated MgO/K_2O concentrations are not the same as those with low CaO levels in Fig. 3 and therefore need to be investigated further with trace element analysis. Five of these samples in the uncorrelated group in Fig. 5 are probably of a later production date than the majority of (Hellenistic) glasses investigated here (see below).

Trace element analysis

A very useful tool in the provenancing of ancient glass is trace element analysis. This can provide a fingerprint for the raw

Table 4 Analytical data coming for comparison

Oxide	Thesprotia	Low Ca	Rhodes	Thessaly	Cyprus	Syria	Beirut	Egypt
	(*n* = 46)	(*n* = 12)	(*n* = 48)	(*n* = 18)	(*n* = 54)	(*n* = 59)	(*n* = 47)	(*n* = 7)
Na_2O	18 ± 3	19 ± 3	16 ± 2	16 ± 2	18 ± 1	17 ± 1	17 ± 1	21 ± 2
MgO	0.6 ± 0.3	0.7 ± 0.2	0.6 ± 0.3	0.8 ± 0.7	0.6 ± 0.1	0.6 ± 0.2	0.6 ± 0.3	0.9 ± 0.3
Al_2O_3	2 ± 2	2.2 ± 0.3	2.1 ± 0.4	2.3 ± 0.4	2.2 ± 0.1	2.2 ± 0.3	2.4 ± 0.2	2.7 ± 0.7
SiO_2	70 ± 4	68 ± 6	70 ± 3	66 ± 7	69 ± 2	67 ± 2	69 ± 1	67 ± 3
K_2O	1 ± 1	0.6 ± 0.2	0.9 ± 0.3	0.8 ± 0.6	0.6 ± 0.1	0.9 ± 0.2	0.7 ± 0.3	0.34±
CaO	6 ± 1	4 ± 1	8 ± 2	8 ± 1	7 ± 1	9 ± 1	9 ± 1	4 ± 2
Fe_2O_3	0.5 ± 0.4	0.2 ± 0.1	0.7 ± 0.5	0.5 ± 0.4	0.3 ± 0.1	0.6 ± 0.2	0.4 ± 0.3	1.1 ± 0.6
Reference	This study	This study	Brill 1999, Triantafyllidis 2000, Rehren et al. 2005, Brill and Stapleton 2012	Connolly et al. 2012	Cosyns et al. 2018	Reade and Privat 2016	Thirion-Merle 2005	Nenna et al. 2000

SD standard deviation

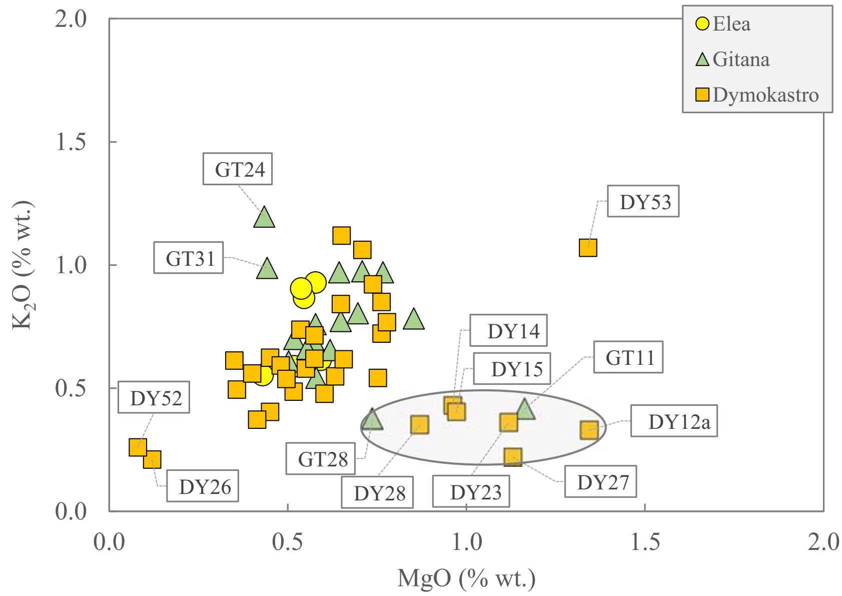


Fig. 5 MgO vs K_2O. The majority of Thesprotian samples have a positive but weak correlation while the second group has a distinct position on the plot

materials used to make glass and potentially the provenance for it since they reflect the local geochemistry of the raw materials used. Trace elements such as Nd, Ti, La, Cr and Zr can have similar values both in coloured and colourless (Late Bronze Age) glass samples (Shortland et al. 2007) and therefore similar raw materials were used to make both. Since natron is rather a pure raw material, many trace elements in natron glass are associated with the mineral impurities in sands such as feldspars, zircons, monazites, chromite and titanite. These are related to the formation age of the sand (Henderson 2013, 57-62).

Distinctions between samples from Thesprotia can be observed in Fig. 6 according to different Th/Zr and La/Ti ratios.

According to the graph, there are three subgroups with two outliers (DY27, DY53). The majority of the samples are plotted in a group on the right side of the graph with La/Ti ratio values greater than 140. In this group, there might be two subgroups consisting of samples with La/Ti ratio around 150 (a tight cluster with green triangles, mainly samples from Gitana) and the second one with samples having La/Ti values greater than 160 (consisting mainly of samples from Dymokastro and Elea). The second group, having values of La/Ti lower than 140, consists of four samples. Two (DY26 and DY52) of these also have low Nd concentrations and La levels of ~ 2 mg/kg; both have very low Al_2O_3 (~ 0.6% wt.) and Fe_2O_3 contents (< 0.1%) as discussed above. In addition, these two samples show extremely high values of Cr ~ 1300

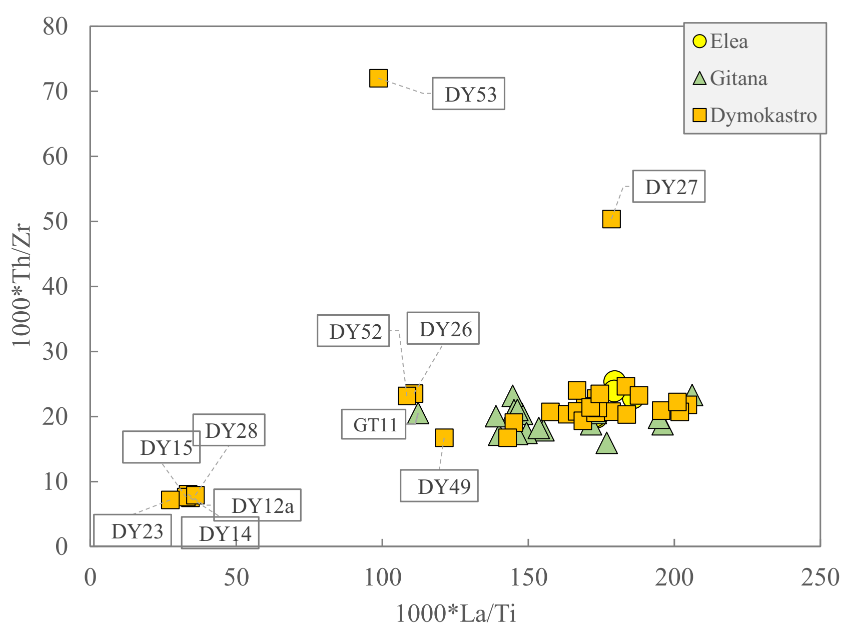


Fig. 6 A bi-plot of the ratios La/Ti versus Th/Zr in the samples analysed

mg/kg which could suggest the use of a different sand source. They also have low MgO and K_2O (~ 0.1 and ~ 0.2 % wt., respectively) indicating a different alkali source. The third group consists of five samples having the lowest values for both ratios. Furthermore, these samples show an elevated amount of Fe_2O_3 (> 1.5 % wt.) which can be linked to either the sand as an impurity or to the colourant used (these samples are amber brown and olive green). They seem to have characteristics similar to HIMT glasses as indicated by various scholars (Foy et al. 2003; Nenna 2014; Ceglia et al. 2015; Freestone et al. 2018, Bertini et al. 2020); i.e. high iron (average at 1.4% wt.), high manganese (average at 1.4% wt.) and high titanium (average at 0.3% wt.) but may be earlier than the suggested production date for HIMT. They are not similar to early Roman emerald green glasses since these belong to the plant ash glass technology (Jackson and Cottam 2015). These samples were excavated in Dymokastro from poorly stratified contexts and therefore their dating is rather insecure. It is worth noting that Thesprotia was destroyed by the Romans in 168 BC including the major cities of Elea, Gitana and Dymokastro. Therefore, the presence of these samples very likely relates to late Roman activities and possibly habitation of the area. Among the three cities, only in Dymokastro there are indications of occupation after the 168 BC. This is because Dymokastro holds a very strategic geographical position in the area (it was the main port of Thesprotia) controlling the sea routes of the Ionian sea (Lazari et al. 2008, Oikonomou 2019). Therefore, we may assume that these samples belong to a later period (after 168 BC) from the majority of the samples. Whatever the scenario, this is another indication of possible Egyptian origin for a few samples, since HIMT glass is believed to have originated from Egypt (Gratuze and Barrandon 1990; Freestone et al. 2000; Foy et al. 2003). Regarding the two outliers (DY27, DY53), both have notable concentration of minor oxides, e.g. Al_2O_3, CaO and MgO, as noted above.

The distinction between major production zones (e.g. Egypt and Mesopotamia) can be provided by investigating the relationship between ratios of specific trace elements, such as Cr/La and Zr/Ti, as it was demonstrated by Shortland et al. (2007) for LBA plant ash glass. The question is: does this distinction hold for natron glasses? In Fig. 7, the majority of Thesprotian samples are divided in two distinct groups. The first group shows low Cr/La ratios (~ 1.5%) and variable Zr/Ti ratios, while the second group shows variable Cr/La ratios, whereas the Zr/Ti ratios vary slightly (Zr/Ti ~ 80–120). This differentiation into two groups indicates that two different sand sources were used to make the glasses. The glass from Thesprotia is compared with already published data for natron glasses from eighth–fourth century BC Macedonia (Blomme et al. 2017), Hellenistic Thessaly (Smirniou et al. 2018) and early Byzantine Israel (Phelps et al. 2016).

The elemental ratios plotted in Fig. 7 are characteristics of the silica sources used (either crushed quartz or sand) and were used originally to distinguish between Egyptian and Mesopotamian geological variations used to make Late Bronze Age plant ash glasses. Even though Thesprotian glasses are natron glasses, we examine the possible relationships between the silica sources used in the Late Bronze Age groups and ours, at least for glasses made in Egypt. Therefore, its utility was tested by plotting the data along with appropriate published data for natron glasses.

It can be seen that some natron glasses from the primary glass making sites of Apollonia and Bet Eli'ezer fall close to the 'Egyptian' field for Late Bronze Age glasses, with the same Zr/Ti and slightly higher Cr/La ratios: most LBA glasses contain below 2.0 Cr/La. There are also Apollonia and Bet Eli'ezer glasses with much higher Cr/La ratios. The data for Egypt II glasses also fall outside the Late Bronze Age 'Egyptian' field, having higher Cr/La ratios. The other Thesprotian glasses have much higher 1000*Zr/Ti ratios than the LBA glasses that constitute the original 'Mesopotamian' field, the maximum Mesopotamian Zr/Ti value being 60. It is therefore necessary to use the plot of these elemental ratios in a somewhat different way from its use for provenancing plant ash glasses.

According to Fig. 7, the first group of Thesprotia glass with low Cr/La values correlates well with the coeval glass from Thessaly and some Macedonian glasses. These samples slightly overlap with the raw furnace glass from Apollonia which has somewhat lower values of the Zr/Ti ratio.

This compositional distinction between higher and lower Cr/La ratios could be interpreted in two possible ways: either a different sand source was used to make glass from Thesprotia, Thessaly and Macedonia (with higher Zr/Ti values) and Apollonia and Bet Eli'ezer (with lower Zr/Ti values) or we have a similar Syro-Palestinian sand source but with a contribution of Zr/Ti which is not associated with the raw materials. However, although this last suggestion has been made in relation to small-scale plant ash glass production within crucible (Rehren and Pusch 2007), natron glass was made on a far larger scale in massive tank furnaces where any contribution from the furnace walls would be minimal or non-existent. Since sand was used to make natron glass, no impurities would be derived from tools used for crushing quartz pebbles. Therefore, the first interpretation is the more plausible.

The second group of the samples from Thesprotia with elevated Cr/La values overlaps with samples labelled Egypt II, actually HIMT glass found in Israel but thought to have been made in Egypt. The samples in this group can probably be further subdivided into two groups, one with Cr/La values of between 3 and 8 and the other one with values between 9 and 15, though this may be a false distinction and only more analytical results for similar glasses will be able to provide evidence as to whether this is a true distinction.

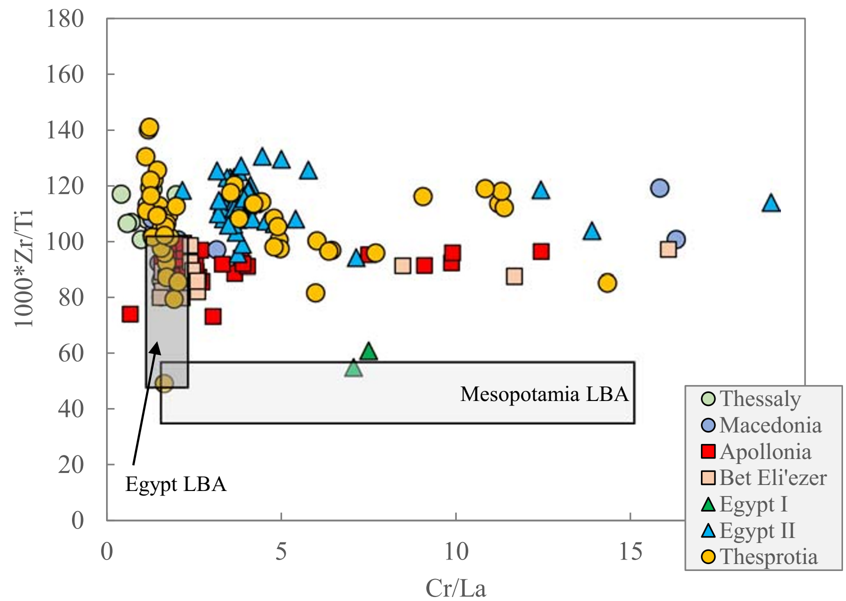


Fig. 7 Bi-plot of Zr/Ti and Cr/La ratios for natron glasses from different locations compared with Late Bronze Age plant ash glasses published by Shortland et al. (2007)

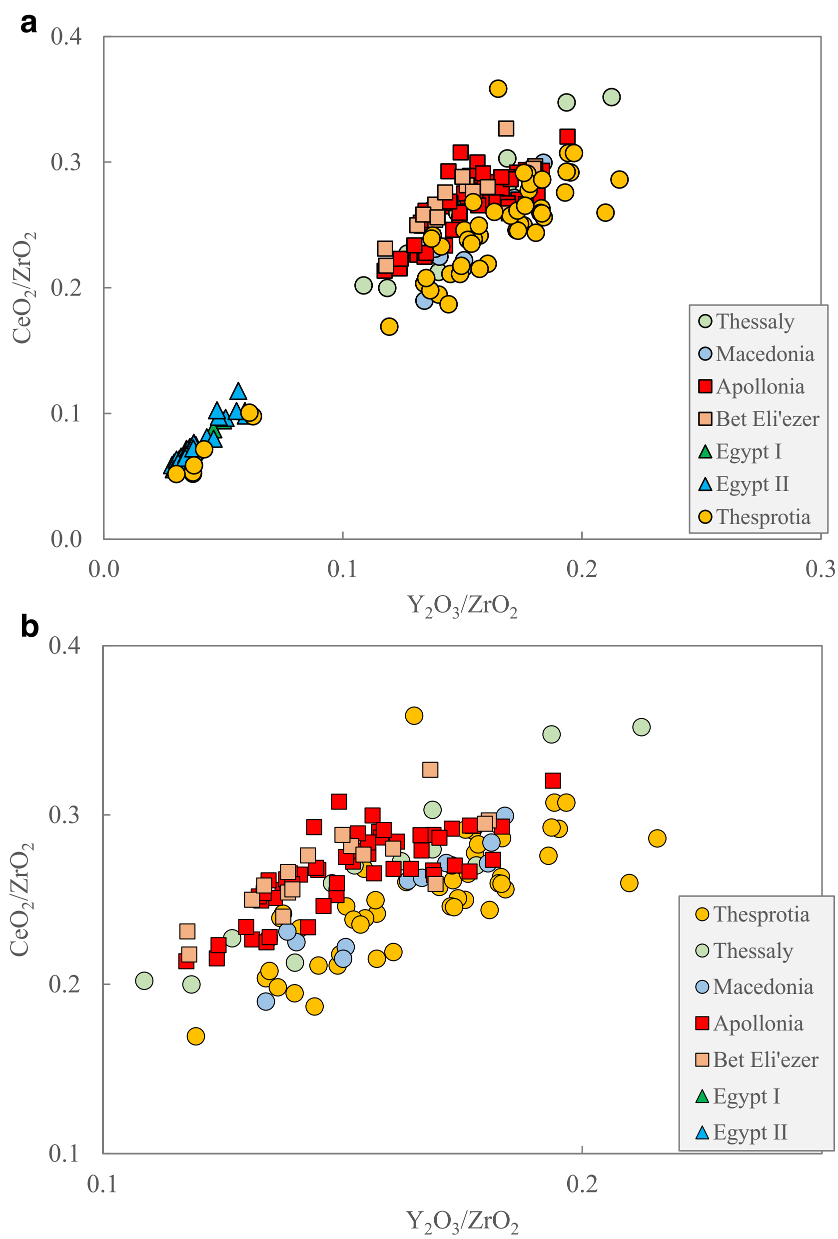
In Fig. 8, Y_2O_3/ZrO_2 and CeO_2/ZrO_2 for the Thesprotia samples are plotted. Here the glass divides into two groups, one with similar values to Egypt II glass (area below 0.1 for both ratios) and the other in a similar position to glasses from Apollonia, Bet Eli'ezer and Thessaly but with slightly different ratios, with Macedonian glasses being quite similar to Thesprotian ones. In Fig. 8a (close up of Fig. 8), glasses from Apollonia and Bet Eli'ezer are clearly seen to be slightly different from Thesprotia glass and the latter has the closest compositional links to Macedonian glass and a few Thessalian glass. Therefore, it is likely that Thesprotian glasses were manufactured with sand from two different sources, Egypt and the Syro-Palestinian coast being the most likely. This may be interpreted geochemically as follows: Ce and Y are both REE elements, with Y mimicking the middle (Dy-Ho) REE elements due to its atomic radius (Chapman et al. 2016). As such, a positive correlation is to be expected; by normalising against ZrO_2, if the Ce and Y are mainly derived from zircons, we are removing the effect of the abundance of zircons within the sand source and this different ratio suggests that zircons are derived from a different rock source in the two groups. Therefore, firstly the Egypt I/II and associated Thesprotia samples are derived from sand where the zircons have a lower content of total REE + Y than those from Apollonia, Bet Eli'ezer, Thessaly, Macedonia and the remaining Thesprotia glasses. Secondly, this group with higher total REE + Y can be further split between (a) Apollonia, Bet Eli'ezer and Thessaly and (b) Macedonia and Thesprotia, where *a* has a greater proportion of CeO_2 than Y_2O_3. This arises from a difference in light REE versus middle and heavy REE in the original crystallisation of zircon or the possibility of a Ce anomaly associated with Ce being able to take the 4 + valence state as well as the 3 + state which all REE (expecting Ce and Eu) may take depending on redox conditions at the time of crystallisation. This simplification may be further perturbed by the possibility that another REE rich phase may be in the sand source, which would inherently provide different proportions of CeO_2 to Y_2O_3. However, this is complicated by the likelihood that a significant proportion of Ce was derived from other heavy mineral phases to Y, such as monazite, a light REE/Th phosphate and garnet providing a significant amount of middle REE including Y (Ayres and Harris 1997).

Statistical analysis

Taking into consideration the major, minor and trace elements associated with sand with an assumed shell component, such as Al_2O_3, CaO, Fe_2O_3, Ti, Zr and Sr, we have applied multivariate statistics to further investigate the data. Thesprotia data is compared with the same natron glasses as plotted in Figs. 2, 3, 5, 7 and 8 dating to earlier and later periods (Phelps et al. 2016; Blomme et al. 2017; Smirniou et al. 2018). The elemental data obtained from the SEM-EDX and LA-ICP-MS were expressed in mg/kg, transformed into base-10 logarithmic values and submitted to a variance-covariance matrix PCA employing algorithms in the STATISTICA 8 software.

In Fig. 9, we have plotted PC1 against PC3 since it clearly separates the samples into groups; PC1 against PC2 did not provide useful information. According to the plot, the majority of Thesprotian samples correlate well with the glass from Apollonia, Bet Eli'Ezer, Thessaly and Macedonia. However, a number of Thesprotian samples are outliers. In particular, there are 7 samples (GT.11, 27–30, DY27, 50, 53) which plot in the upper right corner of the graph.

Fig. 8 The correlation between the Y_2O_3/ZrO_2 and CeO_2/ZrO_2 ratios. Thesprotia glass fall into two groups; the majority of glass samples have relatively high ratios (above 0.1) while a small group of seven samples has low ratios (below 0.1). (a) The area of Fig. 8 with Y_2O_3/ZrO_2 ratios above 0.1 in which the majority of Thesprotia glass is plotted



They are positioned there because of the higher alumina content and lower silica content (Fig. 10) and we have identified all of them earlier as outliers. The two samples (DY26 and DY52) are the ones with very low alumina content and were attributed in a quartz raw material rather than sand. Finally, and more interestingly, the small group of samples (DY. 12a, 14, 15, 23 and 28) which plot together with Egypt I glass are clearly very distinctive. It is significant that the Egypt I samples, with the highest Ti and Zr levels, are earlier than Egypt II samples and have the closest match to Thesprotia glasses. This therefore shows that they can be attributed with certainty to an Egyptian origin, largely due to differences in Fe, Ti and Zr concentrations (Fig. 10), typical of HIMT-type material.

Conclusions

The movement of glass, both raw and worked, could potentially occur in a complex way due to increasingly easy communication and interaction throughout the Mediterranean area and the adjacent countries during the Hellenistic period. In this period, glass was both produced and worked, in few specialised glassmaking centres. Archaeological evidence for secondary glass workshops in Greece is limited to few places such as Rhodes (Weinberg 1969; Rehren et al. 2005; Triantafyllidis et al. in press), Delos (Nenna 1998), Olympia (Schiering 1991) and Macedonia (Nenna 1998). A possible example of a primary glass workshop is that on Rhodes although the evidence is equivocal (Henderson 2013, 246-7)

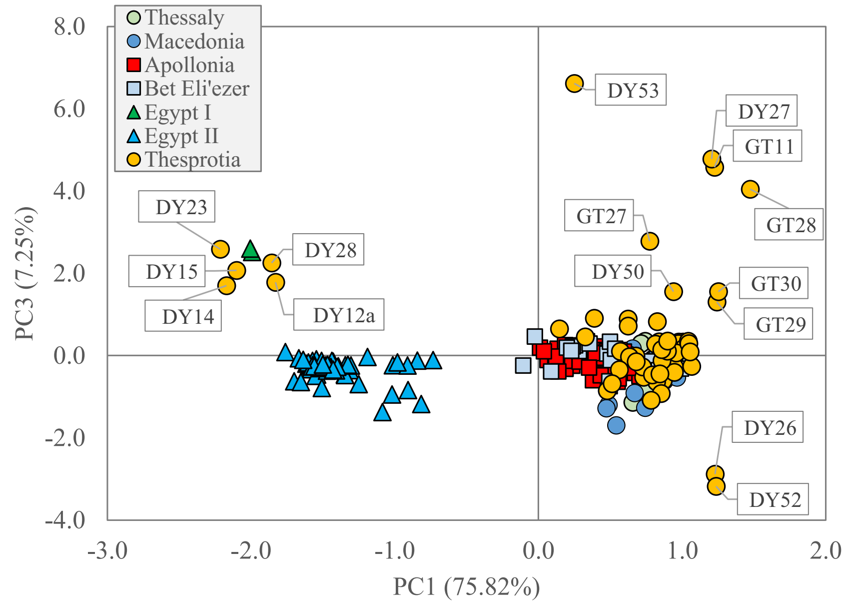


Fig. 9 Plot of the first (PC1) and the third (PC3) principal components for Thesprotia samples and published data

and according to a recent study the sands on Rhodes are not suitable for glassmaking (Blomme et al. 2016). It is therefore likely that the glass worked there was imported.

In addition, various scholars have proposed Hellenistic glass workshops in diverse places of the eastern Mediterranean and the adjacent areas. More specifically in Alexandria, the capital of Ptolemaic Egypt, founded by Alexander the Great in 332–331 BC (Harden 1980), in the wider area of Magna Graecia in southern Italy (Grose 1989), in Amathus, Cyprus (Cosyns and Nys 2010), in Beirut, Lebanon (Foy 2005), and on the Syro-Palestinian coast

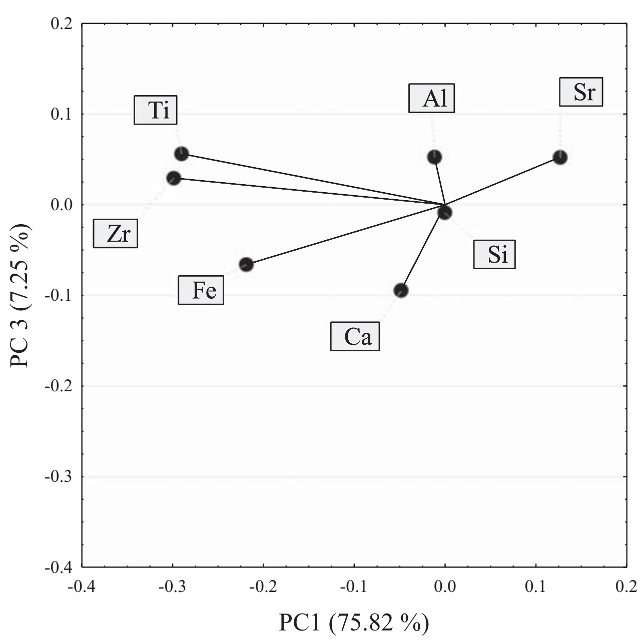


Fig. 10 PCA loadings' plot showing the effect of elemental variables on PC1 and PC3 axes

(Nenna 1998). Indeed, there is clear archaeological evidence for late Hellenistic primary glass production on a massive scale in tank furnaces excavated in Beirut (Kowatli et al. 2008, Henderson 2013, 213-222). According to Pliny (Natural History 5.76), the production of high-quality clear glass was taking place in Sidon, a Phoenician city on the Syro-Palestinian coast which became a permanent member of the Hellenistic Kingdoms. Most of the clear glass formed into objects in the Hellenistic period and later was probably imported from Sidon (Stern and Schlik-Nolte 1994, 108-109). The Syro-Palestinian coast was ruled by the kingdoms of Ptolemy in Egypt and the Seleucids in Syria, providing easy access to the glass from Syro-Palestine to the glassworkers around East Mediterranean.

The analytical data provided in this article show that Thesprotian glass is a typical natron glass using sand as the source of silica and natron as the alkali. The major, minor and trace element characterisation of glass from the three sites in Thesprotia has provided us with valuable information about the source of silica. The data suggest that there are at least three sources (of the raw materials) for Thesprotian glass. Some glass from Thesprotia seems to have a Syro-Palestinian origin from an area which may have been near the later sites of Apollonia and Bet Eli'ezier. Along with Thessalian and Macedonian glass, there are clear similarities. However, we have shown that trace element signatures and PCA provide evidence for distinctive compositional variations away from the main compositional group of glass from Apollonia and Bet Eli'ezer. These variations may be attributable to the changes in the geochemistry of the Syro-Palestinian sand deposits over time between the second century BC and the seventh–ninth centuries AD.

Yet other Thesprotian glasses are quite distinct from the other glass compositions considered here. The provenance of this second compositional type is currently rather obscure and should probably be sought in Hellenistic glassmaking centres in Syro-Palestine or Egypt, or possibly totally different locations such as Italy, Macedonia or Asia Minor. A detailed isotopic research could be useful in determining where these glasses were made. The third glass source is clearly in Egypt. These glasses from Thesprotia have a particular natron glass fingerprint characteristic of El Barungi and exhibit low lime contents, a characteristic of Egyptian samples from the Wadi Natrun region.

As discussed above, Thesprotia glass comes from at least three primary glass production sites. The location of the secondary glass workshop(s), where exactly the objects were made, is unknown and more difficult to determine. However, the fact that glass from three different regions (one of them still unknown) reached the lands of Thesprotia shows that the region was open to diverse commercial activities and probably was part of a wide-ranging trade network, highlighting its socioeconomic importance during this period.

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