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War and commercial exchanges in the late medieval Aegean: investigating the metal provenance of the Chalcis hoard

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Abstract

At the end of the Middle Ages the Aegean was an active trade area, connected to numerous exchange networks. Arms and armor were strategic products, exchanged over extra-regional distance through the Mediterranean trade. To shed light both on military goods manufacturing and Aegean trade networks, this paper focuses on the archaeometric examination of thirteen medieval pieces of armor found in Chalcis (Euboea Island, Greece), then a Venetian hub. Our aim was to perform a provenance study of the metal, based on the LA-ICP-MS analysis of trace rare earth elements in the slag inclusions of the metal artefacts, combined with statistical approaches to compare the chemical signatures with published reference groups. Until now most of these objects were considered imported from northern Italy, whereas the chemical approach highlighted several subsets. Seven samples have a chemical signature compatible with a reference group of Italian style defenses, while others present different chemical tracers such as high Titanium content, that could indicate the use of regional metal resources (Macedonia). These results bring new insight into the Aegean trade network opening the door to more extensive studies in the area.

Keywords : Archaeometallurgy, iron artefacts, arms and armor, provenance study, Middle Ages

1. Introduction

The late medieval Aegean was marked by multiple conflicts for the control of land and sea territories and saw encounters between various communities (Byzantine, Latin, Turkish...). In this context, arms and armor were commodities of crucial importance, exchanged through the Mediterranean trade but also through the looting and reuse of weapons collected by the soldiers on the battlefield. While written sources provide crucial information on the trade routes (Sprandel, 1970; Valérian, 2017), they remain insufficient to assess the importance of these exchanges on a regional scale. Hence artefacts are precious witnesses, whose study is essential to detail the morphology of exchange networks. Although archaeometric investigations have been performed on European armor for several decades (Bérard, 2019; Bérard et al., 2020; Böhne, 1961; Brewer, 1981; Imiołczyk et al., 2020; Kraner et al., 2019; Leever, 2005; Williams, 2011, 2003; Williams et al., 2012; Williams and De Reuck, 1995), few were made on medieval ones. Hence, the hundreds of armor discovered at Chalcis (Ffoulkes and Traquair, 1911; Kontogiannis, 2012) constitute valuable evidence to document the eastern Mediterranean network exchanges and the history of armor making.

This set of artefacts includes over 400 objects, the majority of which are typo-chronological dated to the 14th and early 15th centuries. The set was discovered in 1840 in the ancient fortress of Chalcis (Euboea Island) when, according to the historian Jean-Alexandre Buchon, a wall (whose present location is unknown) collapsed. At the end of the Middle Ages, Chalcis was a Venetian hub named Negroponte. The terminus ante quem of the hoard is dated to 1470, when the city fell to the Turks (Ffoulkes and Traquair, 1911; Kontogiannis, 2012). Soon after their discovery they were first stored in Athens in the Ethnological Museum where in 1919-1920, Bashford Dean, curator at the Metropolitan Museum of Art (New York), acquired about two hundred pieces (Dean, 1925). Following his death in 1928 they were kept in the MET. Today the hoard is dispersed among several collections, most of the pieces being kept in the MET and in the National Historical Museum (Athens).

The majority of the pieces were originally identified by scholars as of western European manufacture (Dean, 1925). Of important interest are the armorers marks still preserved on some pieces, attributed to Italian workshops (see an example Figure 1, “A” showing a mark attributed to the Milanese Missaglia workshop), leading to assume that almost all of the hoard was Italian made and imported by the Venetians. Nevertheless, this Italian-centric perspective needs to be reexamined. Some of the marks applied has not been linked to any known workshop or armorer. Furthermore, the mobility of skilled Italian craftsmen is attested in many parts of Europe (La Rocca, 2017; Painsonneau, 2004, p. 25; Reppel, 2008) but also in the Aegean, notably in Genoese and Venetian territories (Balard, 1978; Racine, 1995;

Thiriet, 1975). In addition, it is possible that Italian-style objects not bearing marks were made by local craftsmen reproducing the Italian style. Part of the artefacts shows particular stylistic features only known from Chalcis and other Aegean sites, that do not fit an Italian-style, (such as the two pieces sallet shown on Figure 1, "C"). Their distinctive morphology and multi-piece assemblage have led to the assumption of a regional production, especially in Dalmatia (Blair, 1982), while some objects of Turkish style (kept at the National Historical Museum in Athens) would rather suggest a Middle East origin (Kontogiannis, 2012). The complexity of the hoard also underlines its historical importance, as it reflects war practices (such as looting), the mobility of weapons and soldiers and the variety of communities involved in the medieval Aegean.

Investigations of the ferrous alloys can provide answers to these open questions. Although a first metallographic examinations has been already published on a few Chalcis samples (Williams, 2003), crucial questions regarding the nature and origin of the metal used remains unexplored. Several methodologies have been proposed to shed light on the metal provenance based on the comparison of isotopic signature (Brauns et al., 2020, 2013; Degryse et al., 2007; Dillmann et al., 2017; Milot et al., 2021, 2016; Schwab et al., 2006), major (Blakelock et al., 2009; Buchwald and Wivel, 1998; Charlton et al., 2012) and lithophile trace elements ratios (Coustures et al., 2006, 2003; Desaulty et al., 2009; Disser et al., 2016; Leroy et al., 2012, 2011; L'Héritier et al., 2020, 2016; Pagès et al., 2022) in slag inclusions (SI). The latter approach is based on the follow-up of the chemical signature from the initial ore to the SI that remains entrapped in the metal artefact. During the bloomery process used to transform ore into metal, some of the chemical elements ("non reduced compound" NRC) are not reduced and remain in the same proportion in the slag left as a waste at the ironmaking site. This chemical signature is also preserved in the small parts of the slag entrapped in the metallic matrix, under the form of inclusions. Thus, by determining the chemical composition of SI both in terms of major and lithophile trace elements and by comparing it with available reference data, it is possible to evaluate the compatibility of the ferrous alloys. Several analytical techniques have been used, tailored to those SI (< 30µm, see Figure 2) found in armor, such as µ-XRF (Leroy et al., 2011) and LA-ICP-MS (Bérard et al., 2020). The latter methodology has recently allowed to quantify about fifteen trace elements in armor samples, highlighting ancient exchange networks and trade, such as in Lombardy, a reputed area of steel and arms and armor manufacture (Bérard et al., 2020), and from which some of the Chalcis pieces may have been produced.



Figure 1 – Examples of armor from Chalcis examined, “A” upper arm defense with an Italian mark detail (Milanese Missaglia workshop) shown on the reverse side (MMA 29.150.57), “B” brigandine plate (MMA 29.150.90ii) with a mark M surmounted by a star, “C” Helmet defense (MMA 29.158.42) with specific stylistic features different from Italian-style armor of the same period and a mark S surmounted by a crown

In this paper, we present a first provenance study of thirteen armor from Chalcis in the collection of The Metropolitan Museum of Art (New York), based on the analysis of lithophile trace elements in SI, combined with statistical approaches. Our aim is to estimate the variability of the supply sources through the investigation of the metal and compare their chemical signature with reference sets of European armor previously defined (Bérard et al., 2022, 2020).

2. Set of artefacts

Thirteen artifacts were selected for analysis (see Table 1), according to the condition of their metal surfaces. Three of the marks applied on the pieces studied (MMA 29.150.57, MMA 29.150.18, .115, and MMA 50.160) have been identified in the literature devoted to Italian armor (Boccia, 1982). The others remain unidentified but the brigandine breastplates marks (MMA 29.150.90ii, MMA 29.150.90oo, MMA 29.150.90t) and mark on piece MMA 29.158.42 are assume to be of Lombard origin (Eaves, 2014 ; Boccia et al., 1980). Piece MMA 29.150.18, .115 is a composite piece of the lower vambrace (forearm defense) 29.150.18 and the couter (elbow defense) 29.150.115. The two elements belonging to the Chalcis hoard were united in the MET.

As ancient ferrous alloys can be heterogeneous and likely to present several chemical families of SI, sampling size and location were chosen to ensure a minimum alteration for the artefact while guaranteeing the representativeness of the results. Thus, delamination areas on the plates have been avoided and sample size was fixed to about 6mm long and 2mm wide, the thickness of the sample corresponds to that of the armor plate. Samples were collected with a small rotary cutting tool and a diamond-coated blade.

Table 1 – Pieces selected for study

Identification Number	Category	Sub-category	Date	Type of mark	Sample(s) number	Additional information
MMA 2005.466.4	Torso defence	Small plate from brigandine	ca. 1450	-	2005-466-4	
MMA 29.150.105	Torso defence	Coat of Plates, or Brigandine	ca. 1400	-	29-150-105	
MMA 29.150.18, .115	Arm defence	Forearm defense (Vambrace)	15th century	Split cross with double S	29-150-18.22	
	Arm defence	Elbow defense (Couter)	15th century		29-150-18.1	
MMA 29.150.57	Arm defence	Upper arm defense (Rerebrace)	ca. 1430–40	M with cross and split leg (attributed to Missaglia)	29-150-57	
MMA 29.150.90ii	Torso defence	Large plate from brigandine	1400–1450	M surmounted by a star	29-150-90ii	
MMA 29.150.90oo	Torso defence	Large plate from brigandine	1400–1450	M surmounted by a star	29-150-90oo	
MMA 29.150.90t	Torso defence	Large plate from brigandine	1400–1450	Letter B mark	29-150-90t	
MMA 29.158.42	Head defence	Great sallet	possibly 1420–50	S surmounted by a crown	29-158-42	stylistic features only known from Chalcis and the Aegean
MMA 49.120.7	Head defence	Lower plate of a bascinet	early 15th century	-	49-120-7	stylistic features only known from Chalcis and the Aegean
MMA 50.160	Arm defence	Left vambrace	ca. 1400	YO with cross and split leg	50-160g	
MMA 49.120.5	Head defence	Lower plate of a bascinet	early 15th century	-	49-120-5	stylistic features only known from Chalcis and the Aegean
MMA 49.120.8	Head defence	Lower plate of a bascinet	early 15th century	-	49-120-8	stylistic features only known from Chalcis and the Aegean

MMA unacc#1	Torso defence	Small plate from brigandine	ca. 1450	-	unacc1	
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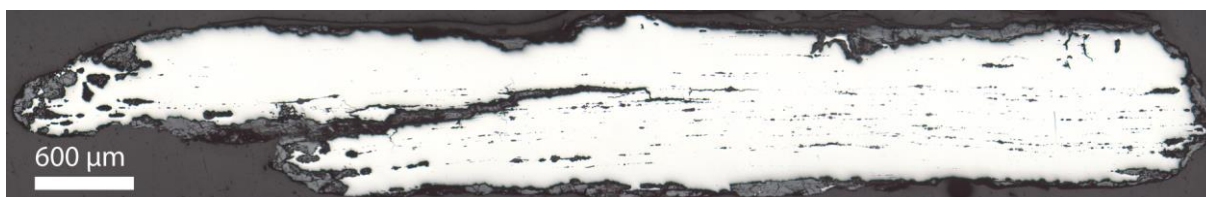


Figure 2 – Optical micrograph of a sample cross section (29.150.90t) showing slag inclusions entrapped in the metallic matrix.

3. Materials and methods

Samples were embedded in epoxy resin and studied in cross section. They were grinded using abrasive papers (MICRO-MESH™ REGULAR, grade 1800-4000). Final polishing was performed using diamond polishing media of 9μm, 3μm and 1μm. The major compositions of the SI were determined using Energy Dispersive Spectrometry (EDX with silicon drift detector) coupled with a Scanning Electron Microscope (SEM-FEG JEOL 7001-F) at the LAPA-IRAMAT-NIMBE laboratory (France). A probe current of about 8 nA and a 15 kV accelerating voltage were used. To pursue a provenance study using lithophile elements in SI, it is necessary to determine the major elements compositions of a statistically representative number of SI (a minimum of 40 SI is recommended (Dillmann and L'Héritier, 2007)). In our case, this condition was met by analyzing the chemical composition of the SI on the entire cross section of the samples. The protocol described in Bérard et al., (Bérard et al., 2020) was followed to both detect the SI by image analysis using the Aztec particle analysis software (Oxford company), and estimate the compositions of each SI on the whole surface of the sample. Concentrations were determined after normalization of the total signal (considered to be 100%), of the set of elements known as major components of iron slags (O, Na, Mg, Al, Si, P, S, Cl, K, Ca, Ti, V, Cr, Fe, Mn).

Phosphorus is a strong chemical marker frequently found in ancient iron alloys (L'Héritier et al., 2019; J. W. Stewart et al., 2000, 2001). Its presence in significant amount (> 0.1 wt.%) in the metal matrices was checked on samples showing significantly elevated levels in some SI (> 2wt.%), using a microprobe with a wavelength dispersive X-rays spectrometer (SX 100 CAMECA, accelerating voltage 15kV, current probe about 200nA, ICMPE, Thiais, France).

In Europe, during the late Middle Ages, two main ironmaking processes were used: the direct (single step: bloomery furnace) and indirect (two steps : blast furnace and finery) processes. However, no provenance study based on the analysis of lithophile trace elements in SI can be pursue for metal

associated with the indirect process, as the NRC-based signature of the ore (smelting system) is lost during the separation between the metallic phase and the blast furnace slag during smelting. (Dillmann and L'Héritier, 2007; Disser et al., 2014). In order to determine if the SI families related to the smelting slag have a composition compatible with the bloomery process, the procedure proposed by Dillmann and L'Héritier (Dillmann and L'Héritier, 2007) and completed by Disser et al., (Disser et al., 2014), was followed. Contrary to other inclusion families generated by the use of fluxes during forging or by fragmentation of the SI, SI related to the smelting slag are characterized by constant NRC ratios (Dillmann and L'Héritier, 2007) (in particular %Al₂O₃/%SiO₂, %MgO/%Al₂O₃, %K₂O/%CaO).

Recycling practices, by welding several pieces of metal, also need to be addressed. Recent work has shown that, in cases where the metal is derived from the direct reduction process, recycling does not irreparably alter the chemical signature of the objects, thus allowing provenance studies to be carried out (M. L'Héritier and P. Dillmann, 2009). Such practices can be identified by the presence of spatially distinct SI families in the sample. Hence, the following procedure has been applied for each SI family identified in the samples, representative of one or more smelting slags in case of welding.

After selecting SI coming from the smelting stage, an average weighted major element content was calculated for each sample (Desaulty et al., 2009; Dillmann and L'Héritier, 2007; Leroy et al., 2012). This transformation evaluates a “surface weighted average composition” of all the inclusions analysed on a cross section to facilitate comparisons between the samples.

$${}^{\circ}\mathbf{E}^{*} = \sum_{i=1}^n ({}^{\circ}\mathbf{E}i \times \frac{Si}{St})$$

Where ${}^{\circ}\mathbf{E}i$ is the composition of a given inclusion, Si the surface of the inclusion analysed, St the sum of all the inclusion surfaces.

Discrimination between the two smelting processes (bloomery or indirect process) was made using a reference database (see Supplementary materials) (Dillmann and L'Héritier, 2007; Disser et al., 2014).

For each sample compatible with the bloomery process, a dozen of the biggest SI was chosen for analysis by LA-ICP-MS. Analyses were made at the Centre Ernest- Babelon-IRAMAT (Orleans) with a sector field ICPMS Element XR (Thermo Scientific) coupled to an excimer laser (ArF*, 193 nm, RESolution S155-E, Resonetics). The methodology published in Bérard et al., (Bérard et al., 2020), was followed to calculate relative concentrations of trace elements in small slag inclusions (<30µm) found in armor samples, especially Y, Nb, Ba, La, Ce, Nd, Sm, Eu, Yb, Hf, Th, U. SI containing at least one of these elements below the quantification limit (10 × SD) were excluded for provenance analysis comparison by statistical approaches (Bérard et al., 2020).

Iron oxides are differentially reduced during smelting, according to the thermodynamic conditions in the furnace. Therefore, their absolute concentrations in the slag and in the SI being variable, they cannot be directly compared, and a transformation of the data is required. We have chosen to apply a log-ratio data transformation already used for the purpose of ancient ferrous alloys provenance study (Dillmann et al., 2017; Disser et al., 2016; Leroy et al., 2012).

$$X_E = \text{Log}([E]) - \frac{1}{N} \sum_{k=1}^N \text{Log}([E_k])$$

Where:

X_E is the transformed value for each element

$[E]$ is the measured concentration for each element

Finally, chemical signatures were compared by combining unsupervised multivariate approach (Principal Component Analysis, PCA, and PCA combined with Hierarchical Clustering Analysis, HCA) and a supervised approach (Linear Discriminant Analyses, LDA), using R software (MASS FactoMineR and gg dendro packages). The potential advantages and drawbacks of each method for provenance analysis in archaeometallurgy have been widely discussed previously (Disser et al., 2016; Leroy et al., 2012; Tomczyk et al., 2023). PCA analysis aims at representing the global dispersion of multidimensional clusters and provide information regarding the influence and relations of the variables on the cloud projection. As the last principal components are likely to contain noise, PCA can be combined with HCA to consider only the principal components that contain relevant information (approximately 90% of the total variance) (Disser et al., 2016). In this study we choose to conduct HCA based on Euclidian distance using the Ward method of agglomeration, as advised by previous studies focusing on iron provenance (Dillmann et al., 2017; Disser et al., 2016). Nevertheless, PCA results may be dominated by variables or dataset with highest variance, leading to an important overlapping. On the contrary, supervised analysis such as LDA may improve discrimination between the clusters, providing that reference data of chemical signatures have been thoroughly established (Disser et al., 2016; Leroy et al., 2012). L'Héritier et al., 2020 (L'Héritier et al., 2020) showed the interest to cross several approaches (supervised and unsupervised) to discriminate between several hypotheses in the case of ferrous alloys provenance study. Building on this work we choose to compare the results given by both LDA, PCA and PCA-HCA approaches to establish three levels of provenance compatibility between two clusters:

- high compatibility: clusters are not separable by both PCA and LDA approaches;
- compatibility probable (clusters presenting chemical similarities) : clusters separable by a LDA approach but not with PCA approach;

- incompatibility: clusters separable by both LDA, PCA and PCA-HCA approaches.

4. Results and discussion

First, we will consider the major elements chemical tracers such as phosphorus and manganese. As shown in Table 2, phosphoric iron was evidenced for two samples (unacc1 and 49-120-5). The majority of the samples are characterized by low phosphorus content ($\%wt.P^* < 0.5$) and high manganese content ($\%wt.Mn^* > 1$) in the SI. Two samples (unacc1, 2005-466-4) are also distinguished by a high titanium content ($\%w.Ti^* > 1$)

Trace element ratios were compared for samples compatible with the bloomery process and exempt from values below the quantification limit ($10 \times SD$) for each measured trace element (dataset are available in Supplementary materials). Thus samples 49-120-5, unacc1 and 49-120-8 that did not meet these conditions are excluded in the following.

A Principal Component Analysis made on the transformed value of the trace elements concentration confirmed that one sample (2005-466-4), present a different chemical signature from the other samples, in particular in terms of Th, U and Hf contents (see Figure 3). The other principal components are shown in Supplementary materials.

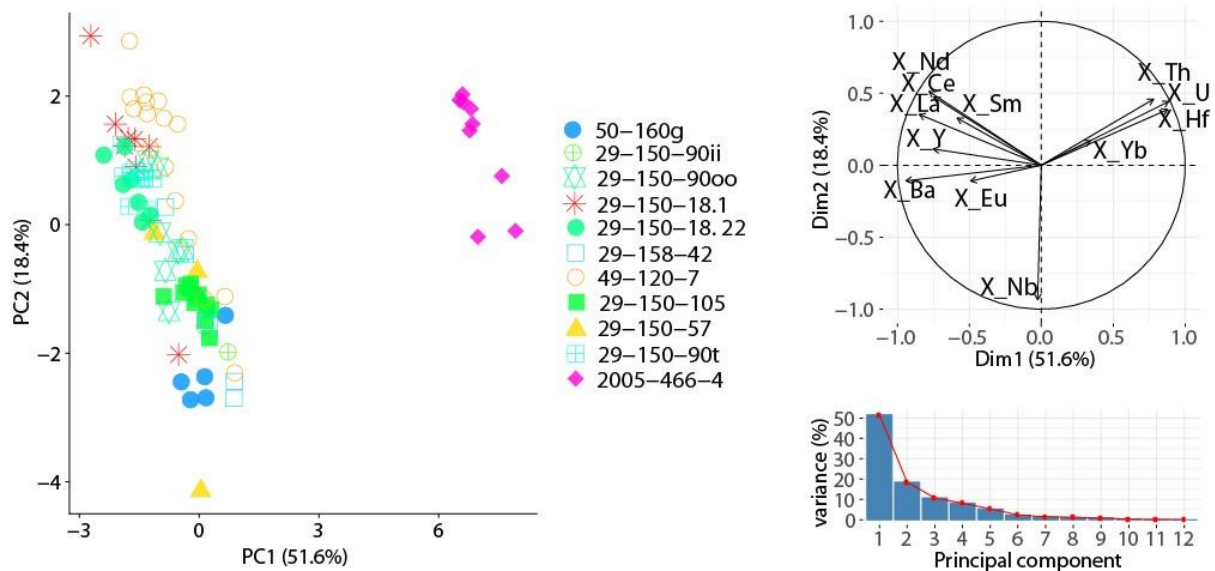


Figure 3 - PCA carried out on the transformed data of SI originating from Chalcis armor projected onto PC1 and PC2 (other components are available in Supplementary materials)

As the majority of the Chalcis armor was assumed to be of Italian manufacture, their chemical signature has been compared with a reference dataset of Italian-style armor analyzed by Bérard et al., (Bérard et al., 2020). The chemical signature of this group has been fully characterized and constitutes a first

reference set associated with an Italian armor style. Except for two samples, PCA results show that the Chalcis armor and the reference dataset of Italian-style armor overlap into the same cluster, suggesting a common provenance of the metal. Sample 2005-466-4 is clearly isolated and characterized by the presence of X_Th, X_U and X_Hf, opposed to X_Ba variable(see Figure 3). After excluding sample 2005-466-4, sample 29-150-90t is slightly detached from the reference dataset of Italian-style armor, particularly by the X_Hf variable (see Figure 5). To confirm this observation, a HCA was made on the principal components of the PCA that cumulate about 90% of the variance, using the Ward's method. Sample 29-150-90t is isolated on one main branch (see Figure 6).

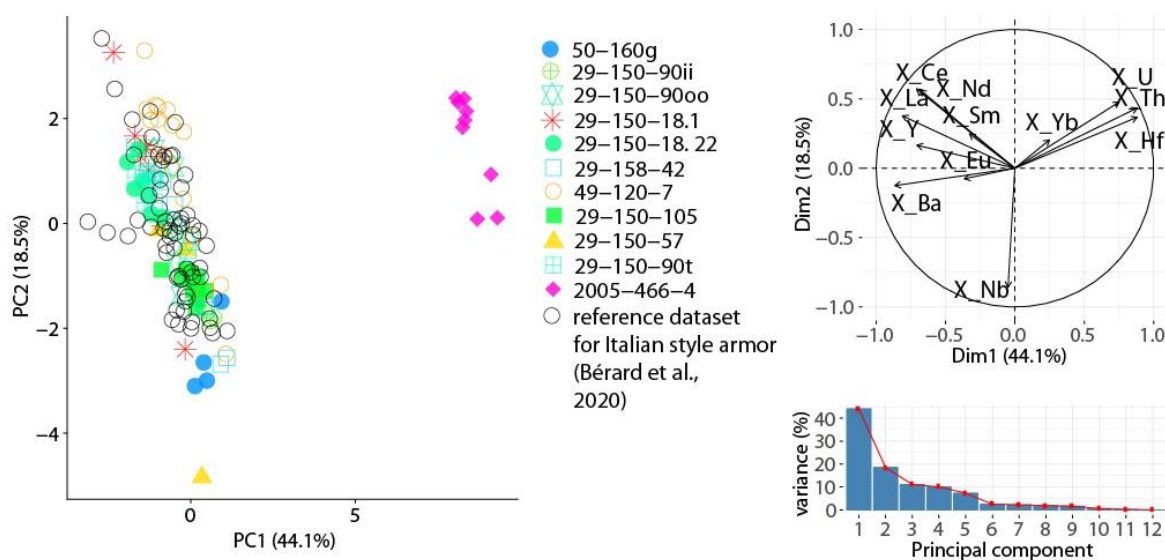


Figure 4 - PCA carried out on the transformed data of SI originating from Chalcis armor and the reference dataset for Italian style armor (Bérard et al., 2020) projected onto PC1 and PC2

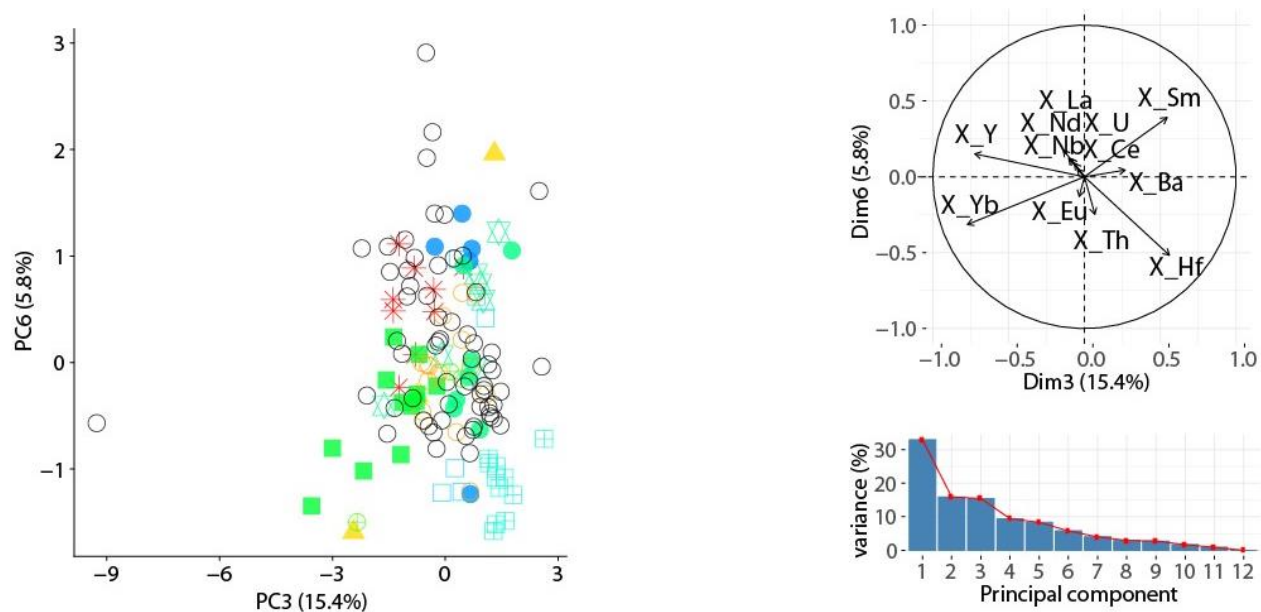


Figure 5 - PCA carried out on the transformed data of SI originating from Chalcis armor (sample 2005-466-4 excluded) and the reference dataset for Italian style armor (Bérard et al., 2020) projected onto PC3 and PC6

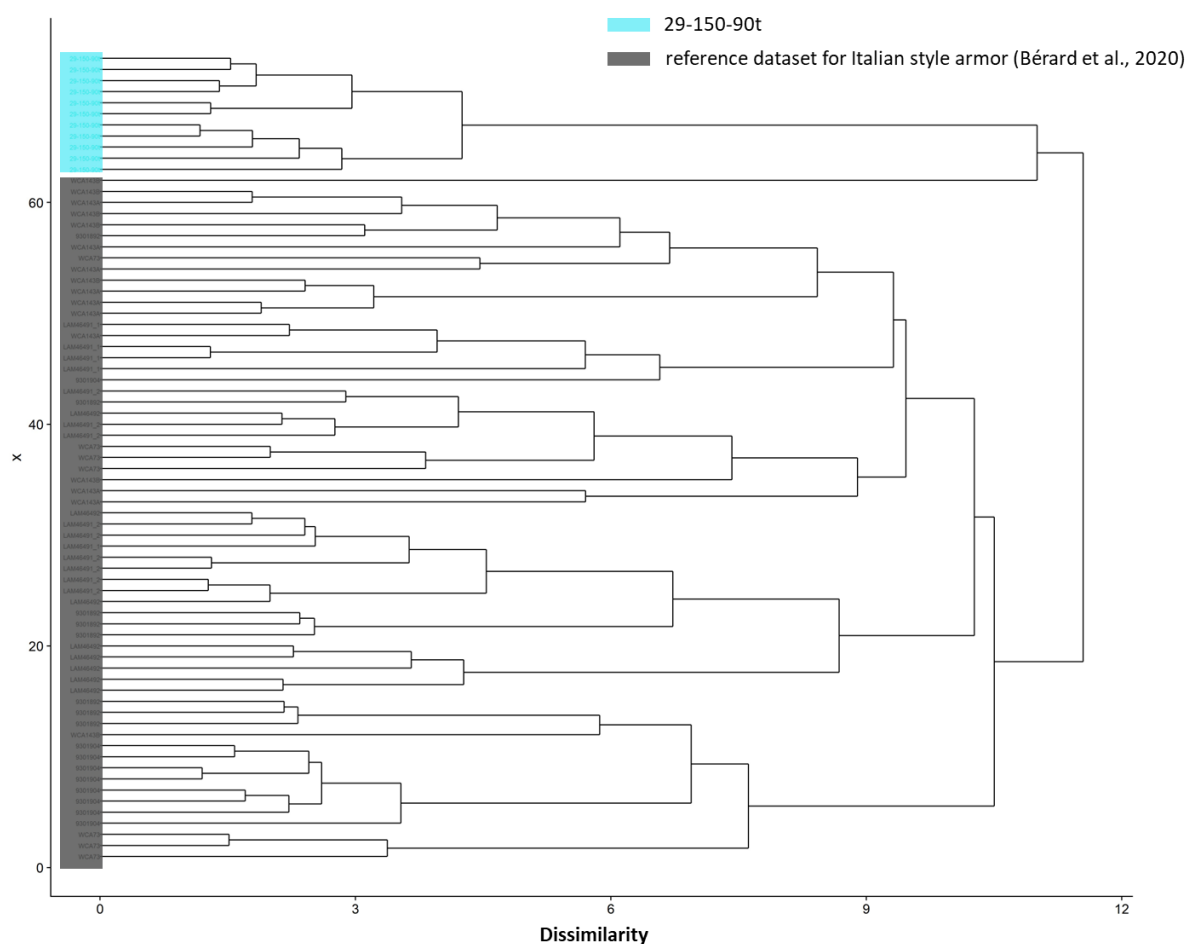


Figure 6 - dendrogram showing the clustering performed on the results of the PCA on sample 29-150-90t and the reference dataset for Italian style armor, by taking into account the first seven PC which express 90% of the variance.

256 LDA confirms these observations. Inclusion of each sample was individually compared with the
257 reference dataset for Italian style armor (Figure 7). Sample 2005-466-4 is the most distant from the
258 reference dataset, followed by sample 29-150-90t. Two other samples are clearly separated from the
259 reference set by LDA: samples 29-150-18.1 and 29-150-57. On the contrary other samples cannot be
260 separated from the reference dataset for Italian style armor. A summary of the provenance
261 compatibility considering both LDA and PCA-HCA results is presented in Table 2.

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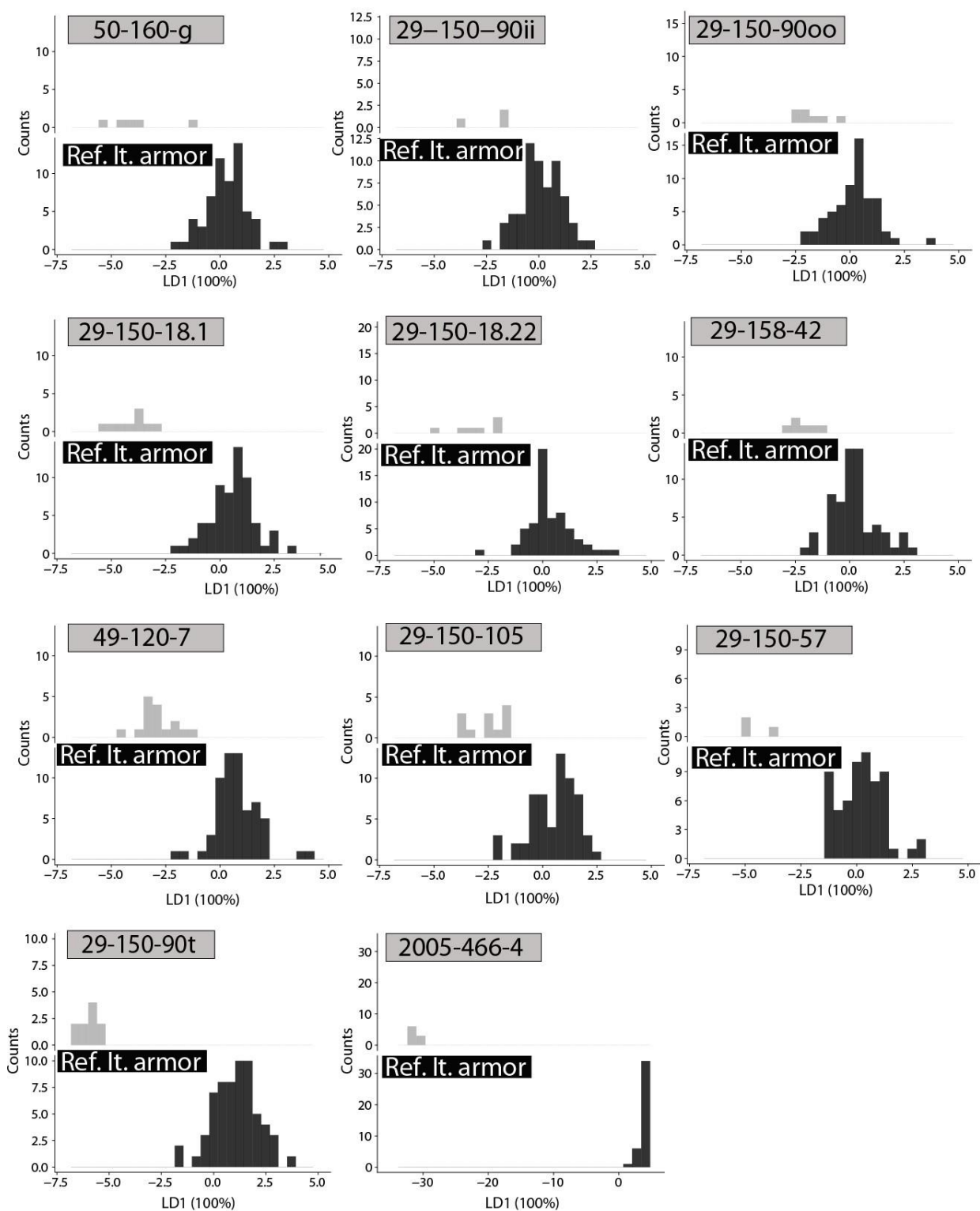


Figure 7 - LDA carried out on the transformed data of SI originating from Chalcis armor plates and from the reference dataset of Italian style armor published in Bérard et al. 2020.

Table 2 – Summary of the provenance study: LDA and PCA approach comparison, Comparison of average weighted Mn, P and Ti content (E* see Methodology) of the SI originating from the Chalcis armor, phosphorus presence in significant amount (>0.1 wt.%) in the metallic matrix.

Column 6 : High compatibility with reference dataset of Italian style armor (not separable by a LDA or PCA approach)
Column 7 : Chemical similarities with reference dataset of Italian style armor : compatibility probable (Separable by a LDA approach but not with PCA approach)
Column 8 : Not compatible with reference dataset of Italian style armor (Separable by a LDA and PCA approaches or composed of phosphoric iron)

Sample number	%wt. Mn*	%wt. P*	%wt. Ti*	Phosphoric iron (%wt.P > 0.1 in metallic matrix)	Column 6	Column 7	Column 8
50-160g	5.4	0.6	0.1		X		
29-150-90ii	6.3	0.1	<LD		X		
29-150-90oo	5.5	<LD	0.2		X		
29-150-18.1	10.6	0.2	0.3			X	
29-150-18.22	1.5	<LD	0.2		X		
29-158-42	6.7	0.6	0.2		X		
49-120-7	4.6	<LD	0.2		X		
29-150-105	3.7	0.2	0.1		X		
29-150-57	5.9	0.2	0.2			X	
29-150-90t	2.0	<LD	0.2				X
2005-466-4	0.4	0.2	2.8				X
unacc1	0.6	2.5	1.4	X			X
49-120-5	7	0.2	0.1	X			X
49-120-8	0.2	0.8	0.2				

The provenance study conducted here strengthens the hypothesis of an Italian manufacture for five marked armor elements (MMA 50.160.g, MMA 29.150.90ii, MMA 29.150.90oo, MMA 29.150.18, .115 forearm defense, MMA 29.158.42), as they share a common chemical signature, compatible with a reference dataset for Italian style armor according both to PCA and LDA analysis. Two other pieces, unmarked, have also a chemical signature compatible with this group (MMA 49.120.7, MMA 29.150.105), while two pieces (sample 29-150-57 and sample 29-150-18.1), show chemical similarities with this group (PCA compatibility), consequently their compatibility was deemed probable. This slight difference in chemical signature for piece MMA. 29.150.57, bearing a Missaglia mark (Milanese workshop) could illustrate the chemical variability of the Lombard iron-making production area. The variation of chemical signature for samples 29-150-18.22 and 29-150-18.1 is in good accordance with the history of the whole piece (MMA 29.150.18, .115). As specified in the “Set of Artefact” section,

piece MMA 29.150.18, .115 is an assemblage of two Chalcis pieces: a forearm bearing an Italian mark (sample 29-150-18.22), whose chemical signature have a high compatibility with the reference dataset of Italian style armor and an unmarked couter (sample 29-150-18.1).

Further research is needed to identify precisely the provenance of the metal, although several hypotheses can be made. Chalcis being a strategic location for Venice in the Aegean, the city received war material from the Republic particularly during the expansion phase of the Ottoman Empire (Braunstein, 1966; Mazou, 2018). The manufacture and commercial exchanges of weaponry was a twofold challenge for Venice. Their sale was highly lucrative, encouraging trade that was not, however, in line with the imperatives of defense. Venice imported iron from various regions, in particular for the Arsenal's imposing needs. Iron from regional resources (in particular Belluno, Zoldano and Agordino) and more distant iron-producing areas such as Styria and Carinthia manufactured in Frioul, are mentioned in historical sources (Braunstein, 1965). During the 15th century, Venice also extended its influence over the valuable Lombardy iron resources of Bergamo and Brescia, which produced highly-quality steel (Braunstein, 2001). The Republic encouraged the development of metallurgical and arms and armor manufacturing activities in the area to secure regular supplies of iron and military equipment (Gaier, 1995). Northern Italy constituted an important arms and armor manufacturing area, Milan being the most renowned, but in competition with Brescia, especially under Venetian influence. Milanese and Brescia arms manufacturers had a privileged relationship with Lombardy's iron producers (Braunstein, 2001; Menant, 1988; Pyhrr et al., 1998; Verna, 2001). One of the most important outlets for Lombardy steel was the arms industry, nevertheless, supplies coming from elsewhere may have been used by Italian armorers.

Styria, Carinthia and Italian Alps are iron-making areas where the practice of direct reduction is attested, exploited manganese-rich deposits (Dillmann et al., 2011; Sprandel, 1968). In addition, Lombardy is characterized by high levels of manganese (average: 5 wt%), barium (average: 4690 $\mu\text{g.g}^{-1}$) but also low phosphorus content (average <0.3 wt%) ores (Bérard et al., 2020; Leroy, 2010). Those chemical tracers were also highlighted by the provenance study of the Chalcis pieces compatible with the reference dataset of Italian style armor (see and Table 2). Therefore, Lombardy is a prime candidate for iron supply of those Chalcis pieces, testifying to Venice's influence in northern Italy and the shipment of military equipment from this region to the Venetian Aegean territories.

The statistical approaches highlighted two isolated chemical signatures (MMA 2005.466.4 and MMA 29.150.90t), whereas armor MMA 29.150.90t shows some chemical similarities with the reference dataset of Italian style armor. If the attribution of the pieces' mark to an Italian workshop is correct, the provenance study could illustrate the use of metal supplies outside Lombardy by Italian craftsmen, derived from manganese rich ores. The other piece (MMA 2005.466.4) is characterized by specific chemical tracers very different from Italy, such as high amount of Ti in SI (>2%wt) as well as by X_{Hf},

X_U and X_{Th} variables (see Figure 3). Important quantities of Ti were also found in another small plate brigandine (MMA unacc#1). The use of titanium-rich magnetite sands as iron ore source for bloomery furnaces were already reported. Most of the references point to Africa and Asia (Ige and Rehren, 2003; Iles and Martínó N-Torres, 2009; Killick and Miller, 2014; Park and Rehren, 2011) the Black Sea (Tylecote, 1981) and to northern Greece (Nerantzis, 2009; Nerantzis et al., 2022; Photos, 1987), suggesting Ti as a chemical tracer in the SI. Studies of northern Greece black sands mineralogy have highlighted the enrichment of certain elements (Filippidis et al., 1997; Peristeridou et al., 2022), such as Hf, Th, U and Ti (several hundreds of $\mu\text{g}\cdot\text{g}^{-1}$ for Th, U and $5076 \mu\text{g}\cdot\text{g}^{-1}$ in zircon mineral fraction (Filippidis et al., 1997)). These chemical tracers, also observed in sample 2005.466.4, could constitute a first indication of provenance. In the late Byzantine period, magnetite sands were important iron sources (Photos, 1987). Numerous slag heaps in the Vrontou attest to the continuation of iron mining and smelting activities in Macedonia during the Ottoman period (Nerantzis, 2009; Photos, 1987). Therefore, Macedonia could be potential candidate for iron supply for those pieces. Further analyses, involving the comparison of trace elements, currently lacking for the Macedonian iron slags will be required to validate this hypothesis and attest the use of Balkans ores resources in the production of these pieces. If confirmed it would give new insights on arms manufacturing activities in the Balkans and the Aegean, far less documented than those in Western Europe during the late Middle Ages. Iron smelting activities continued in Macedonia during Ottoman times therefore an oriental origin should not be excluded, especially since Turkish style pieces (kept at the National Historical Museum in Athens) are reported among the Chalcis Hoard (Ffoulkes and Traquair, 1911; Kontogiannis, 2012). If confirmed, it could testify not only to the mobility of armor and soldiers, but also to the reuse of military equipment as war-booty by late Middle Ages armies.

Among the Chalcis pieces studied, four helmets (MMA 49.120.5, MMA 49.120.7, MMA 49.120.8, MMA 29.158.42) share common morphological features, known only from Chalcis and other Aegean sites, leading to suspicion of a common provenance. Claude Blair suggested a Dalmatian origin for those pieces supported by the comparison of a high relief in Sibenik showing helmets with similar morphological features. He pointed out the importance of Dubrovnik manufacturing center, renowned for its firearms production, to support his hypothesis (Blair, 1982; Petrovic, 1975). Based on the composition of the metal and the provenance analysis, one single piece (MMA 49.120.5) is made with phosphoric iron derived from the use of a phosphoric ore. Two of them (MMA 49.120.7 and MMA 29.158.42) shows a chemical signature with the reference dataset of Italian style armor. (Bérard et al., 2020). The last piece of this group, MMA 49.120.8, did not met the conditions to pursue a provenance study based on the comparison of trace elements ratios but may also have a different provenance characterized by low manganese content (<1wt.%). Despite having common morphological features, the provenance study demonstrates that these four helmets were not made with the same raw

material. It may suggest that they were made in a workshop using various metal supplies or in different workshops. Further research would be needed to decipher whether a Dalmatian provenance for those pieces, as suggested by Claude Blair, could be confirmed. The presence of phosphorus, as chemical tracer for piece MMA 49.120.5, could be a first indication in the future for dataset comparison. Furthermore, the compatibility with reference dataset of Italian style armor for pieces MMA 49.120.7 and MMA 29.158.42 could point to Italian workshops or could also evidence a use of Italian metal supply in the Aegean or Balkan areas. Italian merchants, in particular Venetians, had indeed a very active role in the eastern Mediterranean iron trade routes (Sprandel, 1970). The provenance study brings thus new insight on the medieval network's exchanges in this region.

5. Conclusions

The provenance study of the Chalcis armor reinforces the hypothesis of the existence of different subsets within the large medieval Chalcis defense hoard and suggest that several manufacturing workshops or areas were involved. Seven samples have a common chemical signature compatible with a reference dataset for Italian style armor. This result strengthens the hypothesis of an Italian manufacture for these pieces, whose two of which are unmarked. Lombardy appears to be a prime candidate for the metal's origin underlining the influence of Venice in this area. Another sample taken from small plate brigandines show a different chemical signature characterized by high Ti content. Comparison with literature indicate that Ti-rich iron sources occur in northern Greece (Macedonia) suggesting the possible use of local ores in the production of these pieces. If confirmed, it would constitute a first step towards future research aiming at understanding workshop practices, arms manufacture and trade in the Aegean region during the Middle Ages. To that end, these first results will serve as a basis for a more extensive study on the Chalcis Hoard which will gather a thorough investigation on the nature, heat treatments and origin of the metal combined with other type of historical sources.

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