

## Compositional Analysis of Sixth–Fifth Century BC Silver Coins from the Larnaca Hoard (*IGCH* 1272) (Cyprus) using pXRF Spectrometry

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The elemental composition of 436 silver coins from a hoard of the fifth century BC, minted by the Iron Age Cypriot city-kingdoms of Kition, Idalion, Lapethos, Paphos and Salamis, as well as a small number of coins from unidentified mints, was determined by pXRF spectrometry in order primarily to study the silver alloys used for their manufacture. The specific technique was applied because it allows for a non-destructive analysis, taking into consideration its inherent limitations as a surface analysis technique and the possibility of the existence of surface enrichment elements that do not reflect the materials' actual bulk composition. This is the first time such a large number of ancient Cypriot silver coins has been analyzed providing numismatists and archaeologists with new insights into this important component of ancient material culture. The results of the compositional analysis show that the various groups of coins were made of a similar Ag-Cu alloy with

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silver concentration in the range of 96.5–98.5%. The proportion of copper, larger than the approximately 0.5% that would be expected from ordinary ancient methods of refining silver, is likely justified as an attempt to increase the hardness of the alloy and to improve its casting and minting processing, rather than as an attempt of debasement or a form of adulteration. Furthermore, the analysis has shown that most of the analyzed coins have a lead concentration below 0.5%, indicating a very efficient silver refining process. Gold is in most instances present in a concentration range between 0.1 and 0.5%. The presence of detectable bismuth in most of the coins, at lower concentrations than gold, provides information about the type of ores that were used for the production of silver metal. The interpretation of the chemical elements content reveals similarities and differences between the mints of the Cypriot city-kingdoms and the several coin issues.

## 1. INTRODUCTION

Coinage in Cyprus was an early phenomenon. The fact that in the Persepolis foundation deposits, which were buried in the late sixth century BC, three out of the five Greek coins were from Cypriot mints (Kraay 1976; Kagan 1994) comes as an interesting observation of how early the Cypriot monetary phenomenon was.

During the Archaic and Classical periods (eighth–fourth centuries BC) the island was divided into a number of kingdoms (Gjerstad 1948; Stylianou 1989; Iacovou 2002; Iacovou 2008; Iacovou 2014; Satraki 2012) and to most of them have been attributed coinages with coin legends in the local script (Fig. 1). The study of the coinages of the city-kingdoms that were included in hoards is of great significance, as it provides researchers with important information regarding coin circulation and the dating of the various coin issues (Pilides and Destrooper-Georgiades 2008; Markou 2011c). Following the successful analytical study and publication of gold coins issued by the local kings of the Iron Age Cypriot city-kingdoms via a handheld pXRF (Markou et al. 2014), a new research project was launched. The new project focuses on the silver coin issues of the local Cypriot kings of the Archaic and early Classical period that were included in well-dated hoards. In this paper we present and discuss the results of the analysis of the silver coins from the Larnaca Hoard (IGCH 1272), one of the most important hoards of the early fifth century BC on Cyprus.

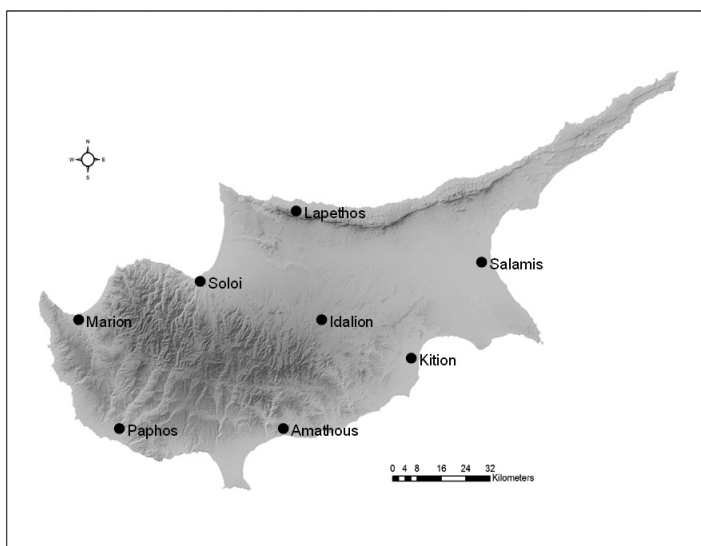


Figure 1. Map of located mints on Cyprus.

### The Larnaca Hoard (IGCH 1272)

The Larnaca Hoard was discovered accidentally in July 1933 during digging for the foundations of a new wing of the municipal hospital of Larnaca (Dikaios 1935; Robinson 1935; Destrooper-Georgiades 1984). It originally contained at least 700 coins (Kraay 1976), but just before the local antiquities authorities reached the discovery site, a significant number of the silver coins was kept or sold by the workers who discovered them. Most of the scattered coins were recovered in the following months, but 99 coins found their way to the London antiquities market and two coins came into the possession of a Cypriot resident in South Africa (Dikaios 1935).

The coins of the specific hoard were all Cypriot *sigloi* (weight ca. 11 g), for the most part well preserved, and have been dated to the latter part of the sixth century and the first part of the fifth century BC. The combined coin die study of silver issues of the royal coinages of Cyprus is currently in progress, but based on some preliminary observations, it appears that the silver coins of the Archaic and early Classical period were following a common “local” weight standard based on a *siglos* of *circa* 11 grams that was divided into thirds, sixths, twelfths, and so on (Markou 2011b). The *terminus ante quem* of the Larnaca Hoard is around 480 BC or perhaps a little later. The coins (a total of 564) were initially attributed

by Porphyrios Dikaios to several city-kingdoms of Cyprus as follows: Kition, 3; Idalion, 36; Lapethos, 143; Paphos, 292; Salamis, 51; uncertain series, 29; and unidentified due to their poor state of preservation, 10 (Dikaios 1935). Interestingly, despite the fact that the hoard was discovered in the modern city of Larnaca, which was built on the location of the ancient city of Kition, the majority of these coins has been attributed to cities other than Kition. The re-examination of 468 coins from the hoard (Destrooper-Georgiades 1984) has shown that more than three coins can be attributed to the city of Kition. In fact, in the Cyprus Museum collections there are eight coins from the hoard, which can be attributed to Kition, and they are all included in the present study (LH1–8).

The coins of the Larnaca Hoard cover a very important historical period and reveal the complexity of Cypriot coinage of the late Archaic–Early Classical period. A number of historical events took place in the Eastern Mediterranean at this time and it is reasonable to expect that they would have also affected the monetary practice of the Cypriot city-kingdoms. Cyprus was encompassed in the Persian Empire in 525 BC and formed part of the fifth satrapy of the Persian Empire, along with Phoenicia, Syria and Palestine. Some years later, in 498 BC, most of the Cypriot cities joined in the Ionian Revolt against the Persian Empire, and together attacked the city of Amathous (Hdt. 5.104.3). Following the collapse of the revolt, Persian authority continued to rule the island of Cyprus, down to the middle of the fourth century BC (Stylianou 1989; Kagan 1999; Iacovou 2008).

### **The procurement of silver for the production of coins**

As has already been mentioned, the Larnaca Hoard consists exclusively of silver coins. But what was the origin of the silver used to produce these coins? Was silver available to the island and easily accessible to the Cypriot kings or did silver have to be imported? Cyprus is well known for its mineral wealth and its massive copper sulfide deposits, which were extensively exploited from antiquity up to the present (Constantinou 1992). Silver, on the other hand, does not appear to have been exploited on the island in ancient times. The reason behind this is that silver is found in Cypriot ores in a colloidal form and in very low concentrations that are not visible to the naked eye and, thus, cannot be collected by panning (Bear 1963). Instead, silver must be extracted through a series of fairly complex metallurgical procedures, none of which are recorded on the island (Kassianidou 2012).

Therefore, we must seek the sources of the silver used to produce the Larnaca Hoard coins overseas. In the Eastern Mediterranean region, important silver ore deposits are located in Laurion (Attica) and in the Aegean area, specifically on

the islands of Siphnos (Gale and Stos-Gale 1981; Pernicka et al. 1985) and Thasos (Grabolle 1988; Papadopoulos 2008). Silver production at Laurion, at the site of Thoricos, had already begun in the second half of the fourth millennium (Kakavogianni et al. 2008), reaching its peak in the fifth and fourth centuries BC, before it was finally abandoned in the first century BC (Conophagos 1980). Anatolia also possesses numerous important argentiferous ore deposits that were extensively exploited in antiquity (Bayburtoğlu and Yildirim 2008; Moorey 1994).

No argentiferous deposits are known from the Levant or Mesopotamia, although from the end of the eighth century BC Phoenicia was supplying both of these regions with raw material, including silver (Aubert 2001). The need for silver, made even more imperative due to the pressing demands of the Assyrian Empire, is believed to have led the Phoenicians to the Iberian Peninsula and the extremely rich silver deposits (Aubert 1993; Eshel et al. 2019) of the so-called “Iberian Pyrite Belt,” which extends from Seville to the south of Lisbon (Salkield 1987). It is possible that Iberian silver could have reached Cyprus through the extended trade networks of the Phoenicians, in exchange for Cypriot copper or locally produced luxury goods, such as metal vessels (Kassianidou 1992). The Phoenicians established a foothold on Cyprus in the ninth century BC (Gjerstad 1948; Teixidor 1975; Aubert 1993; Stylianou 1989; Iacovou 2006) and the presence of a Phoenician royal authority in Kition is well attested from the fifth to the fourth century BC (Gjerstad 1948; Iacovou 2014).

Moreover, a possible source of silver could have been the island of Sardinia, where significant argentiferous lead-zinc ore deposits can be found (Valera et al. 2005). Indeed, in antiquity Sardinia was well known as a source of lead and silver, and was given the name of ἀργυρόφλεψ νήσος or “the island of silver veins” (Schol. Plat. *In Tim.* 25b) by the ancient Greeks. Sardinian silver could have reached Cyprus through trade and in exchange for Cypriot copper (Kassianidou 2006).

### Aim of the research study

The aim of the present study is the non-destructive pXRF chemical analysis of the silver coins of Larnaca Hoard and the determination of their composition, in order to identify the silver alloys chosen for their manufacture. Determining the silver alloys utilized will allow us to answer important archaeological questions and provide essential new information regarding the specific coin issues of the Cypriot mints included in the hoard. The non-destructive pXRF technique was chosen due to the fact that it was not permitted to (a) take the coins out of the museum for conventional (and usually destructive) laboratory analysis, and

(b) sample them, which would have made possible the application of additional analytical techniques. We are aware of the limitations of pXRF technique regarding surface versus bulk composition, but the high concentration of silver in the assemblage under study seems to minimize the enrichment effect. It has been argued that silver-enriched surface layers are not found in alloys with 96–98 wt% Ag (Ager et al. 2013).

It is important to note that no analytical study of silver coins from the Iron Age Cypriot city-kingdoms has been undertaken on this scale before. Only two studies of the gold coins of the Iron Age Cypriot city-kingdoms have been conducted in the last 20 years. The first one was conducted on 53 coins from the collection of the Cabinet des Médailles de la Bibliothèque Nationale de France in Paris, with the application of two different analytical techniques, Proton Activation Analysis (PAA) (Gondonneau and Amandry 2002) and Laser Ablation Inductively Coupled Plasma Mass Spectrometry (LA-ICP-MS) (Gratuze et al. 2004). The results from both analytical techniques were included together with supplementary examples analyzed by the same methods in 2011 (Markou 2011a). The second study was performed on 48 coins from the coin collections of the Department of Antiquities of Cyprus (27 coins) and the Bank of Cyprus Cultural Foundation (21 coins) using pXRF spectrometry (Markou et al. 2014).

A number of studies on silver coins either from hoards or large coin collections, dated from the fourth century BC up to the seventeenth century AD, from several areas and with the application of various techniques, among them XRF spectrometry, have been published the last years (Constantinescu et al. 2003; Civici et al. 2007; Pitarch and Queralt 2010; Rodrigues et al. 2011). Some of the most important issues that the specific studies address are: (a) the concentration of copper and other elements in the silver coins, (b) the provenance and methods of production of the silver metal, and (c) the advantages and limitations of the analytical techniques applied on the surface and bulk analysis of the coins.

The current analytical approach focuses on the study and interpretation of the compositional results of the main types of coins, which are present in numbers higher than 5 (the smallest group is the Kition group with eight coins) rather than individual pieces or groups of two to three coins. We opted for this approach as it allows for more statistically meaningful results, not affected by small sample size. The current study does not address the question of provenance, as it is well known that a simple chemical composition analysis is not adequate to determine the origin of the raw material (Pollard and Bray 2014), namely the silver metal used to produce the coins. Lead Isotope Analysis (LIA) is currently the best technique for studying metal provenance, although it is not without limitations (Pollard and Bray 2014). Lead Isotope Analysis was, however, not

possible in the case of the assemblage under study, as we were not allowed to take any samples.

## 2. MATERIAL AND METHODS

### 2.1. Silver coins

The assemblage analyzed for this study includes 436 Cypriot silver *sigloi* attributed to several city-kingdoms. The coins are distributed as follows: Kition, 8; Idalion, 29; Lapethos, 107; Paphos, 224; Salamis, 46; unidentified series, 22. The eight coins of Kition, issued by early kings, are dated to the end of the 6th – beginning of the fifth century BC and they illustrate a recumbent lion turning his head on the obverse and a smooth reverse (no type imprinted on it). The iconography of the lion is quite common in Cyprus and is attested at several unknown mints of the fifth century BC, as well as in Amathous, where it becomes the exclusive numismatic type (Amandry 1984; Amandry 1997; Markou 2015). In the case of Idalion there are two groups of coins: the first group (Idalion 1 henceforth) contains 21 coins, issued by an unknown king at the beginning of the fifth century BC and depicting a sphinx with curved wings seated right on the obverse and an incuse square on the reverse. The second group (Idalion 2 henceforth), composed of 8 coins issued by an unknown king Ki(-) after 480 BC, depicts a sphinx with curved wings seated right on the obverse and a lotus flower on two spiral tendrils on the reverse side.

The coins from the kingdom of Lapethos form the second largest group in the Larnaca Hoard. There are four different types: the first and probably earliest issue is represented only by two specimens, issued by an unknown king beginning of fifth century B), and has a kneeling-wounded (?) giant (the type is poorly preserved) on the obverse and a running-kneeling Herakles with bow and club on the reverse (Robinson 1935; Kagan 1994). One early example of this type was included in the Persepolis foundation deposit (ca. 511 BC) (Kraay 1976). Similarly, the second issue is represented only by two specimens, issued by an unknown king (ca. 525 BC) with an unrecognizable pattern on the obverse side and the bearded head of Herakles on the reverse (Robinson 1935; Zapiti and Michaelidou 2010; Markou 2015). The remaining 102 coins belong to two large groups, both issued by an unknown king(s), dating to the beginning of the fifth century BC (after 490 BC). They depict the head of Aphrodite facing to the right on the obverse side and the head of Athena in crested Corinthian helmet, facing either to the right (this group of 56 *sigloi* will be named Lapethos 1 henceforth) or to the left (this group of 46 *sigloi* will be named Lapethos 2 henceforth) on the reverse side.

The 224 coins of Paphos comprise the largest assemblage of the hoard. Of these, 223 were issued by King Pu(-) or Pny(-) (early fifth century BC) and depict a bull standing to the left on the obverse and a head of an eagle facing to the left on the reverse. One early example of this type was included in the Persepolis foundation deposit (ca. 511 BC) (Kraay 1976). The only example issued by King A(-) (early fifth century BC) depicts a bull walking to the left on the obverse and the same iconography as the other coins of Paphos on the reverse. There are three different types of coins from Salamis: the first group of 31 coins (Salamis 1 henceforth) was issued by King Evelthon (end of sixth century BC) and depicts a ram lying to the left on the obverse side and a smooth reverse side (no type imprinted on it), while the second group of 14 coins (Salamis 2 henceforth) was issued by Evelthon's Successors (early fifth century BC) and depicts a ram lying to the left on the obverse and an ankh in an incuse square on the reverse side. There is only one coin issued by King Nicodamos or King Evanthes (second quarter of the fifth century BC or earlier) that depicts a ram lying to the left on the obverse side (same iconography as the other coins of Salamis) and the head of a ram facing left on the reverse.

The Larnaca Hoard also includes some coins, which cannot be attributed with certainty to a kingdom, but form three different groups, while there are two coins, which are unique. The first group consists of two coins, minted by an unknown king, probably dated to the first quarter of the fifth century (Kagan 1994). These depict a lion forepart on the obverse and a Gorgon head on the reverse. The second and larger group (Unidentified 1 henceforth) consists of 12 coins, issued by an unknown king or kings (there are five different versions of the same iconography on both the obverse and the reverse side of the coins), dated to the first quarter of the fifth century (Kagan 1994; Zapiti and Michaelidou, 2010), and they depict a lion head with open jaws on the obverse and a bull head on the reverse side. The third group (Unidentified 2 henceforth) consists of 6 coins, minted by an unknown king, probably dated to the first quarter of the fifth century (Kagan 1994). These depict a lion head (similar to that of the second group) on the obverse and an octopus on the reverse side. The octopus depiction, most probably copied from Eretria coins, is suggestive of a connection with the Ionian Revolt and precisely to 499/8 BC, when Cyprus participated for a short time in the movement (Robinson 1935; Kraay 1976). It has been suggested that the city-kingdom of Kourion could have minted the last two groups of coins (Kagan 1999).

The main groups of the analyzed coins (obverse and reverse sides) are shown in Figure 2.



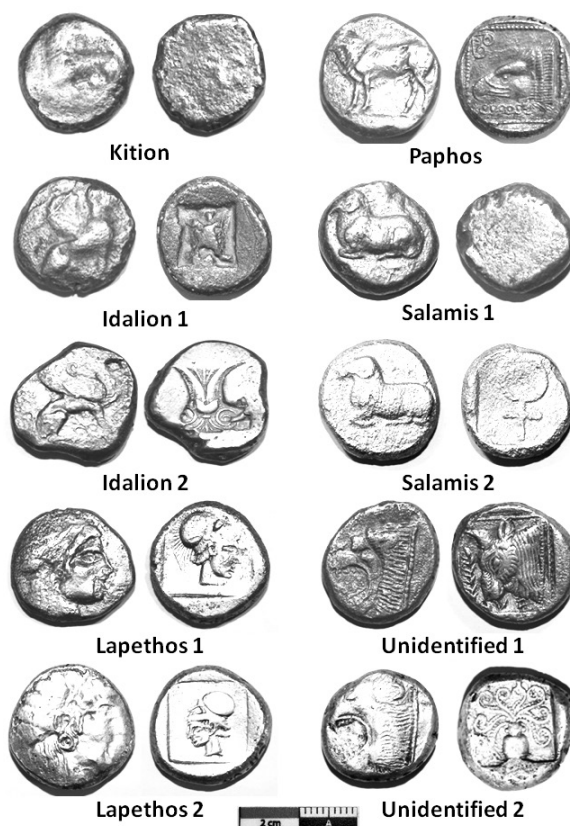


Figure 2. The main groups of the analyzed coins.

## 2.2. Chemical analysis and methodology

The numismatic study of the silver coins included the determination of the weight, diameter and axis. Some general characteristics are presented in Table 1 (below). Moreover, photos of both sides of the coins, obverse and reverse were taken, in order to mark the selected areas of analysis and to avoid areas with surface corrosion. Generally, all the studied coins were in a very good condition and free from surface depositions and corrosion, as the hoard had previously been treated by a museum conservator. Areas on the surface of the coins where some corrosion was still visible were avoided for analysis, although measurements were conducted on a small number of corroded areas as a means of recording differences between the corroded and uncorroded areas on the same coins. Due

to the very good preservation condition of the coins and the very thin layer of the corrosion (usually defined by black coloring of the surface of the coins), the differences in the composition between the various analyzed areas of the same coin in most of the cases were very small and did not affect the general composition of the coins (only the concentration of iron and lead was slightly higher on the darker areas).

Table 1. Overview of the characteristics of the Larnaca Hoard coin groups

Coin Group	Number of Coins	Weight (g)	Diameter (mm)	Period
Kition	8	9.99–10.79	18–20	End of 6th–beginning of 5th century BC
Idalion 1	21	9.43–10.91	20–24	Beginning of 5th century BC
Idalion 2	8	9.83–11.30	21–24	After 480 BC
Lapethos 1	56	9.14–11.26	20–25	Beginning of 5th century BC, after 490 BC
Lapethos 2	46	7.23–11.21	20–25	Beginning of 5th century BC, after 490 BC
Paphos	224	9.30–11.19	18–25	Early 5th century BC
Salamis 1	31	8.75–11.17	18–22	End of 6th century BC
Salamis 2	14	9.39–11.09	20–23	Early 5th century BC
Unidentified 1	12	9.79–10.86	19–23	First quarter of 5th century BC
Unidentified 2	6	9.43–10.74	21–22	First quarter of 5th century BC

The analytical measurements were made using a portable, handheld Innov-X Delta Energy-Dispersive XRF analyzer (pXRF). Energy-Dispersive X-ray Fluorescence (EDXRF) spectrometry is a well-known, non-destructive, fast and multi-element analytical method. The portable XRF was selected for the *in situ* analysis and the determination of the chemical composition of the coins as the only non-destructive and non-invasive analytical technique available. Thus, the application of other more reliable laboratory analytical techniques, such as Laser Ablation Inductively Coupled Plasma Mass Spectrometry (LA-ICP-MS) or Proton Activation Analysis (PAA), which have lower detection limits than the EDXRF technique, especially for trace elements (Guerra 2008), were not an option in our case. The portability and non-destructive nature of portable handheld XRF has resulted in the widespread use of the specific technique for the chemical analysis of ancient precious artefacts, despite the fact that it only provides a chemical profile for the surface, which may not be representative of the whole (Ager et al. 2013). However, one should bear in mind that particularly with re-

guards to silver coins, there are certain conditions that may result in increased values of the items' silver content as far as surface composition is concerned (Schmitt-Korte and Cowell 1989; Butcher and Ponting 1995; Butcher and Ponting, 2012; Ager et al. 2013). These conditions pertain particularly to the alloy composition, means of manufacture, burial conditions and the extent of cleaning and polishing of the artefact (Tate 1986; Araújo et al. 1993; Craddock et al. 1998; Guerra 1998; Cowell and Hyne 2000; Karydas et al. 2004; Ager et al. 2013).

The instrument used in our analyses is equipped with a 4W, 50kV tantalum anode X-Ray tube and a high performance Silicon Drift Detector (SDD) with a resolution of 155 eV (Mo-K $\alpha$ ), covered by a 20-mm detector window. The X-rays are emitted by a miniaturized X-ray tube, located in the internal structure of the instrument, behind a polypropylene film window. The diameter of the X-Ray beam, adjusted by the use of a collimator, is 3 mm.

Six measurements, three on each side, covering a large part of the surface, were performed on each of the coins and the elements concentrations represent the average values of all measurements. This way of measurement takes into account surface irregularities and heterogeneity effects of the coins (Al-Kofahi and Al-Tarawneh 2000; Constantinescu et al. 2003). Measurement time for each spot analysis was 70 seconds.

The analytical mode chosen is Alloy Plus. For this mode Beam 1 (40 kV) analyses the elements Ti, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, As, Hf, Ta, W, Re, Au, Pb, Bi, Zr, Nb, Mo, Pd, Ag, Sn and Sb, whereas Beam 2 (10 kV) is used for the determination of Mg, Al, Si, P and S. The calibration of the instrument was done by the Fundamental Parameters (FP) method designed by the manufacturer (Innov-X). This method calculates chemistry from the spectral data, without the requirement of stored fingerprints. More specifically, Fundamental Parameters involves iterative corrections to raw X-ray counts based on the measured chemical composition, accounting for expected differences in various X-ray phenomena like X-ray emission, diffraction and secondary fluorescence (Frahm 2013).

A silver certified reference material (CRM) 133X AGQ1 (MBH ANALYTICAL LTD, England) was used to test the analytical procedure and the accuracy of the applied analytical mode. The results of the analysis of the silver certified reference material along with the uncertainty and the detection limits of the instrument are provided in Table 2 (below). There was a good agreement between the recommended and measured concentrations for Ag, but higher concentrations for Cu, Au and Pb. The differences were taken into consideration and small corrections were made on the final values of Cu, Au and Pb concentrations of the analyzed coins.

Table 2. The results of the analysis of the silver certified reference material

Element	Silver certified reference material 133X AGQ1		Uncertainty (%)		Detection Limits (ppm)
	Certified value (%) $\pm$ std	Measured value (%) $\pm$ std	Absolute	Relative	
Ag	96.972 (bal)	96.44 $\pm$ 0.3	0.532	0.05	200
Cu	2.532 $\pm$ 0.016	2.64 $\pm$ 0.05	0.108	4.2	200
Au	0.251 $\pm$ 0.003	0.26 $\pm$ 0.01	0.09	3.6	200
Pb	0.245 $\pm$ 0.002	0.26 $\pm$ 0.02	0.015	6.1	250
Fe		0.05 $\pm$ 0.005			250

### 3. RESULTS AND DISCUSSION

Before presenting the results of the chemical analysis of the silver coins, it will be very helpful to provide some information about the cupellation process, the main method of silver extraction and purification during antiquity, and how this process could have affected the composition of the coins and the presence or absence of certain elements.

This process was used in antiquity, as indeed until today, in order to extract the silver from argentiferous lead ores such as galena, the lead sulphide, or cerussite, the lead carbonate (Nriagu 1985). Lead was also used to collect silver from polymetallic ores (Craddock, 1995; Anguilano et al. 2010). Cupellation was and is still used as an assay method for the determination of silver content in ores and metals (Tylecote 1987; Craddock 1995; L'Héritier et al. 2015). The result is metallic silver of high purity. More specifically, the lead cupellation process involves the oxidation of the silver-containing lead bullion in air at a temperature of about 900–950° C, producing silver metal and litharge (lead oxide, PbO). The litharge is absorbed by the reaction vessel, usually made of clay and/or bone ash, called a cupel, while the silver is left within the vessel (Tylecote 1987; Martín-Torres et al. 2009). Trace elements that were initially contained in the lead bullion are separated from the silver because of their affinity to oxygen. Litharge dissolves and absorbs the oxides of most of elements including silicon (Si), calcium (Ca) and iron (Fe). On the other hand, noble metals like platinum (Pt), palladium (Pd) and gold (Au), and nonreactive metals like bismuth (Bi) tend to remain in the silver bullion (Craddock 1995; Karydas et al. 2004).

Besides silver (Ag), copper (Cu), lead (Pb), gold (Au) and bismuth (Bi), which are related to the silver ores and metallurgy (Butcher and Ponting 2012), iron (Fe) and in some coins small amounts of zinc (Zn) were also detected. Iron

will not be included in the discussion as it is most often the result of surface contamination or found as inclusions in the coins (Constantinescu et al. 2003; Civici et al. 2007; Pitarch and Queralto 2010). The concentration ranges of the elements Ag, Cu, Pb, Au and Bi, which are found in most of the coins, are presented in Table 3, with the number of coins analyzed from each group in parentheses.

The results of the chemical analysis show that all coin groups from the various mints under examination are made of a similar silver-copper (Ag-Cu) alloy and in each group most of the coins have a silver concentration in the range 96.5–98.5%. The two groups of coins from Salamis differ, as most examples from both groups have a silver concentration, which is higher than 98% (Table 3, below).

Copper is the main alloying element of the silver metal used for the manufacture of the analyzed coins. Some scholars have argued that copper concentrations in silver (extracted from most types of ores) can reach 1.5%. A copper concentration higher than 1.5% is, therefore, believed to indicate a deliberate addition (Ogden 1992). Others, however, state that the usual natural concentration levels of copper in extracted silver do not exceed 0.5% (Tylecote 1987; Craddock 1995; Karydas et al. 2004). On the other hand, the results of the analysis here show a relatively low copper concentration, which does not support the possibility of an attempt at debasement or some form of adulteration. Therefore, the fact that the concentration of copper is at a higher level than expected after an efficient purification method (around 0.5% Cu) can be justified as a deliberate addition in an attempt to increase the hardness of the alloy and to improve its mechanical properties for the manufacture of coins (Karydas et al. 2004). Moreover, the strong negative correlation between Ag and Cu shown in the plots of silver and copper content of the coins (Fig. 3, below) provides further support to the notion of a deliberate addition of copper in the alloy.

The average copper concentration of the analyzed coins is in the range 1.15–1.9%, with the exception of the two groups from Salamis where the copper concentration is 0.65 and 0.7% respectively (Table 3, below). Interestingly, the issues of the same city-kingdoms have similar or very close average concentrations of copper (e.g., Idalion 1 and Idalion 2 groups have average concentrations 1.7 and 1.55%, respectively). Let us assume that the highest concentration of copper in the silver that can be considered not to have been a deliberate addition is 1.5% (Ogden 1992). In this case, we can divide the coins for each series into those that do and those that do not exceed this limit. We see that Cu concentrations above 1.5% are found in 2 out of 8 coins of Kition (25% of the total), 8 out of 21 coins of Idalion 1 (38% of the total), 4 out of 8 coins of Idalion 2 (50% of the total), 15

Table 3. Elemental concentration range and average value of the analyzed coins from different groups measured by pXRF

Coin Group	Ag (%)			Cu (%)			Pb (%)			Au (%)			Bi (%)		
	Min	Max	Average	Min	Max	Average	Min	Max	Average	Min	Max	Average	Min	Max	Average
Kition (8)	96.8	99	97.65	0.45	2	1.4	0.2	0.8	0.4	0.1	0.8	0.3	0.05	0.09	0.07
Idalion 1 (21)	95.5	98.7	97.25	0.8	3.5	1.7	0.2	1.1	0.5	0.1	0.8	0.35	0.05	0.2	0.1
Idalion 2 (8)	95.3	98.1	96.75	0.95	2.2	1.55	0.4	0.9	0.55	0.1	1.9	0.9	0.05	0.2	0.1
Lapethos 1 (56)	96.5	98.8	97.6	0.4	3.6	1.3	0.15	0.8	0.4	0.2	0.7	0.4	0.05	0.4	0.1
Lapethos 2 (46)	96	99	97.65	0.3	2.3	1.15	0.15	0.9	0.45	0.1	0.8	0.45	0.05	0.2	0.07
Paphos (223)	94	99.1	97.1	0.4	3.8	1.9	0.15	1.4	0.5	0.1	1.2	0.25	0.05	0.5	0.1
Salamis 1 (31)	93.5	99.6	98.3	0.05	1.5	0.65	0.07	1.6	0.4	0.1	1.5	0.4	0.05	0.3	0.1
Salamis 2 (14)	97.5	99.3	98.7	0.1	1	0.7	0.1	1	0.3	0.05	0.6	0.3	0.05	0.2	0.1
Unidentified 1 (12)	95.8	99.1	97.1	0.3	2.5	1.4	0.2	1.4	0.75	0.1	0.9	0.35	0.05	0.3	0.15
Unidentified 2 (6)	96.6	99	98	0.06	2.8	1.2	0.1	0.5	0.3	0.05	0.4	0.25	0.07	0.2	0.15

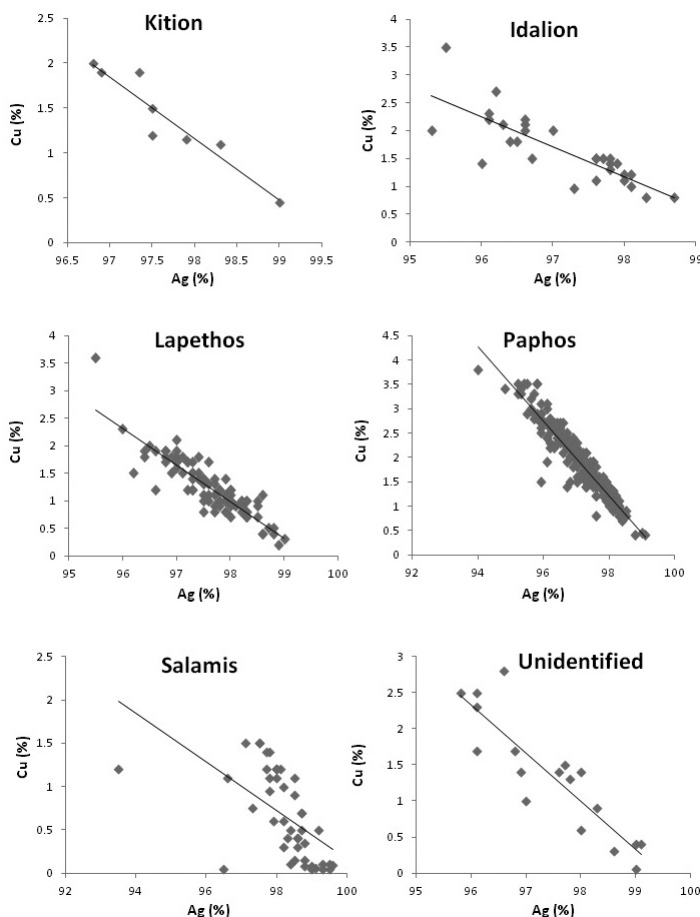


Figure 3. Silver and copper content of the coins from different groups.

out of 56 coins of Lapethos 1 (27% of the total), 9 out of 46 coins of Lapethos 2 (20% of the total), 139 out of 224 coins of Paphos (62% of the total), none of the coins of Salamis, 5 out of 12 of Unidentified Mint 1 (42% of the total) and 1 out of 6 of Unidentified Mint 2 (17% of the total) (Table 4, below). Based on this, the assemblage assigned to Salamis is particularly interesting: all coins, from both issues, have a copper content lower than 1.5%. Equally interesting is the assemblage from Paphos as more than half of the coins (62% of the total) have a copper content above 1.5%, possibly indicating a rather different manufacturing technique for the alloy, compared to the rest of the studied mints.

Lead (Pb) may have been introduced to the silver alloy either as a result of the smelting procedure of the silver ores and the associated cupellation method or from the deliberate addition of copper. In the first instance, the lead concentration in the silver alloy depends on the type of mineral that was used for the production of silver. If argentiferous lead ores, and particularly galena (PbS), cerussite ( $\text{PbCO}_3$ ) and anglesite ( $\text{PbSO}_4$ ), were smelted (the case of Laurion, Greece), the lead contents in the silver alloy could range between 0.05% and 2.5% (Gale and Stos-Gale 1981) or between 0.001% and 3% (Moorey 1985; Karydas et al. 2004). Tylecote (1987) has mentioned a range of Pb concentration between 0.1 and 1%; the concentration of copper in the case of silver coming from argentiferous galena is usually no more than 0.5% (Gale and Stos-Gale 1981; Tylecote 1987). Moreover, in the case of argentiferous jarosite ores (hydrated sulphates) (the case of Rio Tinto, Spain), the use of lead was obligatory in order to collect the silver (Craddock 1995). Generally, it is well accepted that a low lead concentration (below 0.5%) indicates a good refining process (Civici et al. 2007). It is worth mentioning that the addition of lead for the accumulation of silver from some types of minerals (a process called liquation) could be very misleading in provenance studies on silver artifacts using lead isotope analysis, in cases where they take for granted that lead originates with the silver (Craddock 1995).

In this study, most of the analyzed coins have a Pb concentration below 0.5%, indicating a very efficient silver refining process. More specifically (Table 4), a Pb concentration  $\leq 0.5\%$  was detected in 6 out of 8 coins from Kition, 20 out of 29 coins from Idalion (15 out of 21 coins for Idalion 1 and 5 out of 8 coins for Idalion 2), 78 out of 104 coins from Lapethos (42 out of 56 coins for Lapethos 1 and 35 out of 46 coins for Lapethos 2), 145 out of 224 coins from Paphos, 36 out of 46 coins from Salamis (24 out of 31 coins for Salamis 1 and 12 out of 14 coins for Salamis 2), 4 out of 12 coins from Unidentified Mint 1 and 6 out of 6 coins from Unidentified Mint 2. So, the more efficient refining process was observed in the coins of the two groups from Salamis and the second group of unidentified coins (Unidentified Mint 2), while the least efficient process was noted in the coins of Idalion 2, Paphos and Unidentified Mint 1.

Gold (Au) was detected in all coins, ranging from 0.05 up to 1.9% (Table 3, above), although in most coins, its concentration varies between 0.1 and 0.5%. The case of the Idalion coins is very interesting in this regard. In the Idalion 2 group, 6 out of the 8 coins have a gold concentration above 0.6% (Au: 0.65–1.9%, the remaining two coins have 0.1% Au). On the contrary, 18 out of the 21 coins of Idalion 1 group have a gold concentration in the range of 0.1–0.4% (the remaining three coins have 0.7–0.8% Au). These differences in the gold concentra-



Table 4. The comparison of Cu, Pb, Au concentration thresholds and Bi, Zn presence for each group

Coin Group	Cu (%)		Pb (%)		Au (%)		Bi		Zn	
	< 1.5%	> 1.5%	≤ 0.5%	> 0.5%	≤ 0.5%	> 0.5%	Detected	Undetected	Detected	Undetected
Kition (8)	6	2	6	2	7	1	6	2	0	8
Idalion 1 (21)	13	8	15	6	18	3	21	0	3	18
Idalion 2 (8)	4	4	5	3	2	6	6	2	1	7
Lapethos 1 (56)	41	15	42	14	46	10	50	6	7	49
Lapethos 2 (46)	37	9	35	11	35	11	39	7	3	43
Paphos (224)	85	139	145	79	216	8	215	9	45	179
Salamis 1 (31)	31	0	24	7	22	9	30	1	1	30
Salamis 2 (14)	14	0	12	2	9	5	9	5	1	13
Unidentified 1 (12)	7	5	4	8	10	2	12	0	1	11
Unidentified 2 (6)	5	1	6	0	6	0	6	0	0	6

tion between the two groups most likely indicate two different silver sources for the coin issues of Idalion. In the other groups, the gold concentration shows the same trend where the majority of the coins have a gold content below 0.5% and only a small number of coins exceed that concentration. Specifically, as shown in Table 4, in Kition, 7 out of the 8 coins have a gold content in the range 0.1–0.4% (the only exception has 0.8% Au), in Lapethos 1, 46 out of the 56 coins have a gold content in the range 0.2–0.5% (the rest 10 coins have 0.55–0.7% Au), in Lapethos 2, 35 out of the 46 coins have a gold content in the range of 0.1–0.5% (the remaining 11 coins have 0.6–0.8% Au), in the large group of Paphos, only eight coins have a gold content above 0.5%, in Salamis 1, 22 out of the 31 coins have a gold content in the range of 0.1–0.5%, in Salamis 2, 12 out of the 14 coins have a gold content in the range of 0.05–0.45% (the two exceptions have 0.6% Au), in Unidentified Mint 1, 10 out of the 12 coins have a gold content in the range of 0.1–0.45% (the two exceptions have 0.6% and 0.9% Au) and in Unidentified Mint 2 the six coins have a gold content in the range 0.05–0.4%. So it is obvious, in the cases where we have two mints from the same city, that only in the case of Idalion we can suggest the differentiation of the two mints. In the cases of Lapethos, Salamis and the unidentified mints, the two mints have a similar gold concentration range.

But how can the presence of gold in silver coins be explained? Even though ancient silver commonly contains substantial concentrations of gold (Au) and its presence was almost certainly known in antiquity, along with the technology of separating the two precious metals, there seems to have been little effort to recover the gold (Craddock 1995). Specifically, the gold contained in the original argentiferous ores survives the melting process and the manufacture of the coins and is present in the silver metal in its original amount. The gold content of silver depends on the type of ore that was smelted for the production of silver. If argentiferous galena was employed, silver would contain zero up to about 1% Au (Gale and Stos-Gale 1981; Karydas et al. 2004), while the use of oxidized lead ores, like cerussite and anglesite, would have resulted on a gold content of 0.1 up to about 0.5% (Meyers 1983; Craddock 1995; Karydas et al. 2004). Another possible silver source could have been electrum, the natural gold-silver alloy (Craddock 1995; Ramage and Craddock 2000; Butcher and Ponting 2012). In electrum, the concentration of silver varies enormously but typically lies between 5% and 40% by weight (Tylecote 1987; Wallace 1987; Ramage and Craddock 2000). The specific alloy was applied in its natural form for the manufacture of the first coins (electrum issues) in the area of Asia Minor (Lydia) in the seventh century BC or it was processed in order to provide pure gold and pure silver for coins in the sixth century BC (Wallace 1987; Ramage and Craddock 2000).

Similarly to lead, bismuth (Bi) can be found in the silver metal either from the primary source of silver or as an accidentally added impurity. In the first case, if dry ores (ores which contain very small or no amount of elements like copper, lead or zinc) (Hayes and Waldemar Lindgren 1908) or native silver was used for the production of silver metal the bismuth concentration would be less than 0.05% (Craddock 1995; Karydas et al. 2004). If argentiferous galena was used, then the concentration of bismuth in the silver would be between 0.1 and 1% (Gale and Stos-Gale 1981; Karydas et al. 2004). In the second case, bismuth would have entered the silver metal through the deliberate addition of copper, if it was present in the original copper ores. Based on the results of the chemical analysis and the relatively low copper content in the silver coins in most of the analyzed cases, along with the suggested use of pure copper, it is highly possible that bismuth was entering the silver metal through the cupellation process. Bismuth can, thus, be considered as an ore source tracer for silver material (L'Héritier et al. 2015). The analysis of the assemblage under study, showed that bismuth is not present in all the coins. It was found in 6 coins from Kition (0.05–0.09%), in all coins from Idalion 1 (only 6 out of 21 coins have a Bi content higher than 0.1%), in 6 coins from Idalion 2 (only 1 coin exceeds 0.1%), in 50 coins from Lapethos 1 (only in 7 coins Bi content exceeds 0.1%), in 39 coins from Lapethos 2 (only in 4 coins Bi content exceeds 0.1%), in 215 coins from Paphos (49 coins have Bi content 0.15–0.3%, only one coin has 0.5% Bi), in 30 coins from Salamis 1 (only in 9 coins Bi content exceeds 0.1%), in 9 coins from Salamis 2 (only in 2 coins Bi content exceeds 0.1%), and in all coins from Unidentified 1 (in 5 coins Bi content exceeds 0.1%) and Unidentified 2 (in 4 out of the 6 coins Bi content exceeds 0.1%) (Table 4, above). If we convert the presence of bismuth into percentages of the analyzed coins (Kition, 75%; Idalion 1, 100%; Idalion 2, 75%; Lapethos 1, 89%; Lapethos 2, 85%; Paphos, 96%; Salamis 1, 97%; Salamis 2, 64%; Unidentified 1, 100%; Unidentified 2, 100%), we see a clear differentiation between the two Idalion issues (similar to the case of gold) and the two issues of Salamis, a relatively small differentiation in the case of Lapethos and no differentiation in the case of the two unidentified issues. Moreover, if we observe the number of coins that have a bismuth concentration above 0.1% we can see that the two unidentified issues have the higher percentage, 41% and 66%, respectively, followed by Salamis 1 (30%) and Idalion 1 (28.6%).

And what about zinc (Zn)? The intensely oxidizing cupellation process would totally eliminate any zinc (Craddock 1995), thus the presence of even small traces of this metal in silver, would indicate that the zinc entered the silver metal most probably through the addition of copper. In fact, the small amount of zinc found in some of the coins (62 coins, 0.05–0.2%), and the absence of tin possibly

indicate that copper was added as pure metal and not as copper alloy, a practice known from Roman times onwards (McKerrell and Stevenson 1972; Civici et al. 2007). In our case, the addition of copper as pure metal, and not as a copper alloy, seems to have started much earlier (fifth century BC). Also, the possibility that part of zinc could be attributed to external pollution, like in the case of iron, cannot be rejected on current evidence. In the present study, zinc was detected in 4 coins from Idalion (3 of Idalion 1 and 1 of Idalion 2 group, 0.05–0.07% Zn), 10 coins from Lapethos (7 of Lapethos 1 and 3 of Lapethos 2 group, 0.05–0.2% Zn), 45 coins from Paphos (0.05–0.2% Zn), 2 coins from Salamis (one from each group, 0.05% Zn) and in one coin from Unidentified Mint 1 (0.05% Zn), while the specific metal was not detected in the cases of Kition and Unidentified Mint 2 groups (Table 4, above).

In closing, it is worth mentioning the results of the analysis of the two small groups of coins that are probably attributed to Lapethos (kneeling-wounded (?) giant/running-kneeling Herakles and unrecognizable pattern/bearded head of Herakles) and the small unidentified group (lion forepart/gorgon head) that were not included in the general discussion and presentation of the results. Starting with the first small Lapethos group, the results of our analyses showed similar composition for all coins and element values very close to the average values of the two bigger groups from Lapethos (Ag: 97.7% and 97.4%; Cu: 1.2% and 1.4%; Pb: 0.15% and 0.5%; Fe: 0.25% and 0.15%; Au: 0.55% and 0.4%; Bi: 0.15% and 0.08%; no Zn). Almost identical is the composition of the two coins of the second small group that was attributed to Lapethos, even though it shows slightly higher silver and lower gold and bismuth concentrations (Ag: 98.5% and 98.2%; Cu: 1% and 1.1%; Pb: 0.2% and 0.15%; Fe: 0.08 and 0.15%; Au: 0.15% and 0.3%; Bi: 0.05%; no Zn). As for the unidentified group, the analysis showed a similar composition for the two coins, with relatively high silver content and low concentrations for the remaining elements (Ag: 98.3% and 98.5%; Cu: 1% and 0.9%; Pb: 0.3% and 0.2%; Fe: 0.15%; Au: 0.15% and 0.1%; Bi: 0.1%; no Zn).

#### 4. CONCLUSIONS

The analytical study of the silver coins in the Larnaca Hoard, by means of a non-destructive portable XRF methodology, has generated useful new information with regards to the composition and manufacture of Cypriot silver coins dating to the sixth–fifth century BC. This is the first time ever that such a high number of Cypriot coins of this period, from a secure archaeological context, has been analyzed in any way.

The pXRF analysis of the 434 silver coins has shown that all the groups of coins are made of a similar silver-copper (Ag-Cu) alloy despite originating from different mints. Furthermore, the majority of coins from every analyzed group is characterized by silver concentration in the range 96.5–98.5%. Only in the two Salamis groups the concentration of silver is higher than 98% (in the majority of coins). With regards to copper—the main alloying component—of much interest are the cases of Salamis, where all coins, from both issues, have a copper content lower than 1.5%, and the case of Paphos where more than half of the coins (62% of the total) have a copper content above 1.5%, indicating a much different manufacture technology for the alloy, compared to the rest of the studied mints. The copper content indicates that this element was deliberately added to silver, most probably for the improvement of silver processing and the increase of its hardness rather than for debasement or adulteration reasons.

The concentration of lead provides useful information with regards to the efficiency of the silver production method. Most of the coins analyzed in this study have a lead concentration below 0.5%, suggesting a very efficient silver refining process. The most efficient silver refining process was observed in the coins of the two groups from Salamis and the Unidentified 2 group, while the coins of Idalion 2, Paphos and Unidentified 1 groups are the result of the least efficient refining process.

Gold (Au) and bismuth (Bi) are two elements that originated in the primary source of the silver minerals and can provide some information about the possible origin of the silver. The two elements can also reveal similarities and differences between the several coin issues coming from the same city. Starting with gold, the majority of the analyzed coins have a gold concentration in the range of 0.1 to 0.5%. The most interesting conclusion we can reach based on the gold content of the silver coins is that the two coin issues of Idalion (different gold concentration range, possibly due to different silver sources) are differentiated from one another, while in the cases of Lapethos, Salamis and the Unidentified mints, the two different issues have a similar gold concentration range. As for

bismuth, the results of the analysis show that there is a differentiation between the two issues of Idalion (as in the case of the interpretation of the gold content) and between the two issues of Salamis, a relatively small differentiation in the case of Lapethos coin issues and no differentiation between the two Unidentified issues.

The sources of silver that were used for the manufacture of the coins is a critical issue and one of the main still unanswered archaeological questions regarding Cypriot archaeology. In our study, the results of the analysis and mainly the presence and concentration levels of lead, bismuth, and, principally, gold, suggest that the silver metal of the majority of the coins was most probably processed by argentiferous galena. The closest source of argentiferous galena to Cyprus is the Lavrion mine in Attica area (Athens, Greece), a mine that was highly active during the period that the mints of Larnaca Hoard are covering (sixth–fifth centuries BC) (Conophagos 1980; Kary et al. 2004) and, thus, a possible source for at least part of the raw metal used for the specific coins. At this point we should underline that Athenian coins—as well as Aeginetan coins—have been found in Cyprus mainly as overstrikes, which means that those foreign coins circulated on the island and were used as flans for the minting of local coins (Milne 1945; Destrooper-Georgiades 1996; Markou 2011c). The Athenian coins were made of silver from Laurion and could have been melted and used—as metal—for the issuing of the local Cypriot coinages at different times. However, it must be kept in mind that during this period we also note the documented presence of extensive trade networks organized by the Phoenicians between Cyprus and the rest of the Mediterranean. The establishment and maintenance of such networks in the area indicates that other silver production sources must not be excluded without prior investigation.

Finally, it is worth repeating that the studied coins albeit from different mints were buried together at the same time and they were cleaned, restored and stored in the same way and under the same conditions. Because of that and despite the fact that the applied surface analytical technique is not considered to be the most appropriate one for the study of coins, the pXRF study of the specific Cypriot silver coins has been a very fruitful exercise. By undertaking this study, we have been able to provide numismatists and archaeologists with new and significant information with regards to the minting technology of a series of silver coins produced during a very interesting period in the history of the Cypriot city-kingsdoms.

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## CONTENTS

Editorial Committee	v
A. CHARALAMBOUS, E. MARKOU AND V. KASSIANIDOU. Compositional Analysis of Sixth–Fifth Century BC Silver Coins from the Larnaca Hoard (IGCH 1272) (Cyprus) using pXRF Spectrometry	1
LLOYD W. H. TAYLOR. On the Reattribution of some Byblos Alexanders to Arados II	31
ANEURIN ELLIS-EVANS. The Late Hellenistic Tetradrachms of Parion and Lampsakos	93
D. ALEX WALTHALL AND ANNE E. TRUETZEL. New Evidence for the Introduction of the Roman <i>Denarius</i> System: An Unpublished Hoard of Republican Bronzes from Morgantina (Sicily)	127
SETH BERNARD. The Imperial <i>Victoriatus</i> in New Inscriptions from Pompeii and London	157
RYAN H. WILKINSON. Identifying Ancient Coins Deposited with Modern Ships’ Ballast: A Problem for Distribution Studies?	169
JACK NURPETLIAN. Emesan Tetradrachms of Caracalla: An Investigation of their Symbols	179
GEORGE WATSON. Kraft in the 21st Century: A New Listing of Shared Dies in the Roman Provincial Coinage	219
CRISTIAN MONDELLO. Re-reading the So-called “ <i>Asina</i> Tokens”: Religious Diversity in Late Antiquity	273
IRENE SOTO MARÍN. Coin Molds and a Decentralized Monetary Policy in Tetrarchic Egypt	313
ANTONINO CRISÀ. Construction Sites, State Collections, Sixteenth-Century Spanish and Maltese Coins: The Via Maqueda Hoard in Context (Palermo, 1872)	339
GUY CHAMBERLAND. François Declos, Barber? A Reconsideration of the F D Countermarks from Trinidad	389
HEINZ TSCHACHLER. When the Magic Has Gone: Coin Collecting and the Senses	415



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