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The cupellation of argentiferous lead in Mesogeia, East Attica, during the Final Neolithic/ Early Bronze Age periods – The cupellation workshop at Lambrika

ABSTRACT: Excavations in southeast Attica have uncovered a wealth of metallurgical finds relating primarily to cupellation and to a lesser extent copper working. Workshop installations were only identified at Lambrika, but significant quantities of litharge are also known from the sites of Merenda, Gyalou and Zapani. Litharge is characteristically scarce at the extensive, nodal settlement of Koropi. The finds from Merenda and possibly Gyalou place the beginnings of cupellation in this region at least to the mid-4th millennium BC, extending continuously among the different sites to the EHII phase. The bowl-shaped litharge is the most common type of litharge identified here with the characteristically arranged ten depressions on the top surface appearing to be a chronologically later (mature EH I/EH II) feature, possibly also spatially restricted to the Laurion/Lambrika zone. The identification of litharge does not intrinsically testify to in situ practice of cupellation, as the material is known to have had secondary usages in antiquity, not least as a raw material for lead production. The quantity of finds and other considerations, however, suggest that cupellation workshops existed on almost all the sites considered here, even if actual remains of installations have only been identified at Lambrika. The finds from southeast Attica testify to the significance of the Laurion deposits already for the earliest Aegean silver production and allow for direct study of this technology.

KEYWORDS: PREHISTORIC, SILVER, BOWL-SHAPED LITHARGE, LAMBRIKA, LAURION, TECHNOLOGICAL CHANGES

Introduction

The earliest evidence for metals and metallurgy in the Aegean dates at least back to the 5th millennium BC and involves the production of a few copper-based artefacts, primarily known from Northern Greece, for example from Promachonas, Servia, Sitagroi, Dikili Tash and Makrygialos (Zachos, 2010, p.81; Tsirtsoni, 2016, p.21; Malamidou, 2016, p.311, fig. 26). Apart from copper-based artefacts, excavations have also uncovered remains of extractive metallurgy such as slags and metallurgical ceramics. On the contrary, finds from Southern Greece are fewer for these periods, examples including a copper needle from the Kitsos Cave dating to the middle of the 5th millennium BC (Bourhis, et al., 1981, p.425 no. 4, fig. 288) as well as the copper axe from Spata (Phelps, et al., 1979, p.176, no. 120, pl. 22.1; Ζάχος, 2010, figs. 6-6β, cat. no. 70) which is a chance find or the result of looting.

Regarding silver, the earliest known artefacts are also dated to the 5th millennium BC and are items of jewellery finds found primarily in Southern Greece (Ζάχος, 2010, p.88); for example in Salamina (Dimakopoulou, 1998, p.64, no. 62), Alepotrypa, Diros (Papathanassopoulos, 1996, p.227, nos. 41–43); Παπαδημητρίου and Τσιρτσώνη, 2010, cat. No.51), Amnissos, Heraklion (Μαρινάτος, 1930, p.97, fig. 9). Compared to copper, the earliest extractive metallurgy of silver in the Aegean is presently attested a little later towards the end of the Neolithic period in the mid–4th millennium BC. It is at this period that the earliest known mining galleries are dated in the Lavreotiki (Lohmann, 1993, p.75, pl. 72.3 [Mokriza], p.88, pl. 76.4 [Rimpari], p.486, pls. 129.2,3 [Kastela TH 49]; Lohmann, et al., 2002; Lohmann, 2005, p.128) and at Ayios Sostis, Siphnos (Gropengiesser, 1986; 1987).

Prehistoric remains of silver production from the immediate vicinity of the extensive Laurion metalliferous zone are understandably very rare. Later, intense activities in antiquity and primarily in the modern period at Laurion will have largely destroyed this evidence (Kovo ϕ á γ o ς , 1980; K α k α β o γ i α vv η ς , 2005). As such, the finds discussed



Fig. 1: Map of Attica presenting sites discussed in this paper (source: Archive of the Ephorate of Antiquities of East Attica, Copyright © Hellenic Ministry of Culture and Sports).

in this paper, discovered mainly in the nearby Mesogeia region, constitute an important witness to the technology of silver production in this period as well as the spatial and overall organization of these activities.

The finds presented here relate to the last step of silver production, the cupellation, where the silver is separated from the argentiferous lead metal, with lead being oxidized to litharge, the main by-product of this procedure (Κονοφάγος, 1980, p.304). Litharge is by far the main type of metallurgical find we come across in prehistoric contexts from southeast Attica. Lead slags or other relevant remains of smelting are essentially absent. We discuss primarily the finds from four sites from the broader Mesogeia area, (Fig. 1), Lambrika, Merenda, Gyalou and Koropi while we consider also comparatively the finds from the site of Zapani (Ανδρίκου, 2010; Andrikou, et al., this vol. pp.57-72). We present first the archaeological evidence for these sites and subsequently discuss the characteristics of the litharge fragments and other relevant finds in each case.

Lambrika

The discovery of the Lambrika workshop during rescue excavations in 2002¹ (Douni and Kakavogianni, 2002, p.195), opened the way to the direct study of prehistoric silver production in the Mesogeia plain and more broadly in the region of southeast Attica. Lambrika constitutes to this date the only clear cupellation workshop in Attica (Kakavogianni, et al., 2006; 2008; K α k α β α γ i α ν γ , et al., 2009) up to the Classical period. Together with the plethora of other finds coming from the other neighbouring sites, they allowed us to recognize and study new types of litharge, particularly the bowl shaped with or without depressions, as well as to trace a chronological evolution or change in the technology of cupellation in the region.

Lambrika is situated in the southeast foothills of Hymettus (Fig. 1), in the site of the ancient demos Lamptrai $K\alpha\theta \dot{u}\pi\epsilon\rho\theta\epsilon v$. Lamptrai means shiny. Could the toponym have its roots in a very old memory of the shining silver which was produced here? The site is located at a



Fig. 2: General plan of the EH settlement at Lambrika (source: Archive of the Ephorate of Antiquities of East Attica, Copyright © Hellenic Ministry of Culture and Sports). distance of 1.8 km from the other EH settlement of Lamptrai $Y\pi \epsilon v \epsilon \rho \theta \epsilon v$ at Kontra Gliate (Kiafa Thiti) (Lauter, 1996) and 5 km from the extensive nodal EH settlement of Koropi (Κακαβογιάννη, 1993; Κακαβογιάννη and Ντούνη, 2009; Avδρίκου, 2013). The settlement extends on a low hill (Fig. 2), the south part of which has been destroyed during the construction of the Vari-Koropiou highway, many years ago. It presents two chronological phases. During the EH I period, the settlement encompasses the metallurgical workshop (Fig. 3), which is found in the southeast part of the hill, whilst to the south of the workshop a small house was uncovered as well as ditches with refuse deposits from both the settlement and the workshop, as attested by the large number of litharge fragments. During the EH II period the settlement extends to the west of the workshop and to the north of the Varis Koropiou highway. Excavations revealed part of a road, as well as remains of houses. In some cases, small cavities carved into the bedrock were identified as possible hearths and along with the litharge fragments they point to the practice of cupellation during this phase but to a smaller extent than appears to have been taking place during the EH I.

With regards to the EH I period finds, the excavated cupellation workshop extends to an area of approximately 70 m² (Fig. 4), whilst it seems that part of it had been destroyed by the construction of the adjacent highway. It consists of a cluster of three pits of circular shape, of which the largest one is located in the middle and has a diameter of 1.5 m and depth of 0.5 m. On its circumference and between this largest pit and the two smaller circular shaped ones, are two clusters of cavities, three towards the north and two towards the south. These have a diameter of roughly 17 cm and a depth of 10 cm. These cavities bear a whitish coating (Fig. 5).

The workshop is defined from the north by an enclosure made of wooden stakes, while north of the largest pit there was part of a building that continued to the north, outside the excavated area. Northeast of the cluster of pits and east of the building there was a large ellipsoidal pit, 5 m in diameter, while to the northeast a natural submersion was exposed, part of which had been cut during the initial opening of the highway. At the surface of the broader workshop area and more specifically within the large ellipsoidal pit, at its northeast part, hundreds of fragments of bowl-shaped litharge were collected (Fig. 6), obviously waste coming from the workshop activities, together with pottery of the EH I period (Fig. 7) and fragments of grinding stones.



Fig. 3: Plan of the workshop at Lambrika (source: Archive of the Ephorate of Antiquities of East Attica, Copyright © Hellenic Ministry of Culture and Sports).



Fig. 4: Photo of the workshop at Lambrika (source: Archive of the Ephorate of Antiquities of East Attica, Copyright © Hellenic Ministry of Culture and Sports).



Fig. 5: Cupellation hearth from Lambrika (source: Archive of the Ephorate of Antiquities of East Attica, Copyright © Hellenic Ministry of Culture and Sports).



Fig. 6: Litharge fragments exposed during excavation of the workshop (marked with yellow labels) at Lambrika (source: Archive of the Ephorate of Antiquities of East Attica, Copyright © Hellenic Ministry of Culture and Sports).



Fig. 7a: EHI pottery from the workshop at Lambrika (source: Archive of the Ephorate of Antiquities of East Attica, Copyright © Hellenic Ministry of Culture and Sports).

The small house found 15 m to the south of the cupellation workshop (Fig. 8) also dates to the same period. The building had well-built walls, a paved floor and special care had been taken to protect the foundations of the building externally from the rainwater. A large quantity of pottery of various shapes and types (Fig. 9) was collected from the house, as well as many other finds (5 spindle whorls, obsidian tools and grinding tools, animal bones) that relate to the habitation of the building (Κακαβογιάννη, et al., 2009, fig. 5). In the southeast part of the EH I settlement and at a small distance from the above remains, at least two large ditches were exposed, 15 m in length, 4 m in width and 3 m in depth. Their original use possibly relates to collecting water from the small streams of the area. Large quantities of EH I pottery sherds (Fig. 10), stone tools, one intact bowl-shaped litharge (Fig. 11) and many litharge fragments were found in their fills.



Fig. 7b: EHI pottery from the workshop at Lambrika (source: Archive of the Ephorate of Antiquities of East Attica, Copyright © Hellenic Ministry of Culture and Sports).



Fig. 8: Photo of the EHI house at Lambrika (source: Archive of the Ephorate of Antiquities of East Attica, Copyright © Hellenic Ministry of Culture and Sports).



Fig. 10: EHI pottery from ditches at Lambrika (source: Archive of the Ephorate of Antiquities of East Attica, Copyright © Hellenic Ministry of Culture and Sports).

Therefore, it is worth highlighting, that litharge fragments were concentrated in special parts of the settlement and were not found scattered throughout. Apart from the area of the metallurgical workshop, litharge was also found in the ditches. It seems that inside these ditches, inhabitants have discarded the material from the workshop (litharge, stone tools, and sherds of pottery). On the other hand, it is noteworthy that, despite the proximity of the EH I house to the metallurgical workshop, no litharge fragments were found within or in the immediate vicinity of the former.

The litharge fragments

There are about 1,500 litharge fragments (or 160 kg) from Lambrika and by far the most common type is the so



Fig. 9: Large part of pithos from the EHI house at Lambrika (source: Archive of the Ephorate of Antiquities of East Attica, Copyright © Hellenic Ministry of Culture and Sports).

called bowl-shaped with the characteristic ten depressions on the upper surface. Similar finds were recovered from Zapani (Andrikou, et al., this vol.). The bowl-shaped type, with or without depressions, was known before from a few examples (Kakavogianni, et al., 2008, fig. 15) but the contexts were always either unknown or unclear and the finds had been thought of as belonging to the classical period (Kovoφάγος 1980, pp.367–369, fig. 16–4, 16–5). The finds from Lambrika and subsequently from Zapani place this type of litharge with certainty to the EH I and EH II periods in southeast Attica.

The preservation state of the Lambrika litharge finds varies. The average size of the fragments is 6 by 4 cm, while the largest fragment measures 11 by 8 cm. The intact bowl (Fig. 11), as well as several others large enough, allow us to reconstruct with confidence the original shape of these remains. Their diameter varies between 6 to 12 cm and the height of their walls between 1 and 2 cm. The weight of the intact litharge is 500 g. Very characteristic are depressions found on their upper surface. It is obvious that each bowl had always 10 depressions arranged in three parallel rows of three- four- three depressions and the dimensions of these were 0.6-1.6 cm diameter and 0.01-0.08 cm depth.

The dimensions of the bowl-shaped litharge finds correspond very well to the dimensions of the small coated cavities cut into the bedrock surrounding the large pit in the Lambrika workshop area. Furthermore, the analytical data suggest a close relationship between the chemical composition of the lining of these cavities with the bowlshaped litharge fragments (Georgakopoulou, et al., in





Fig. 11: Bowl-shaped litharge from Lambrika (source: Archive of the Ephorate of Antiquities of East Attica, Copyright © Hellenic Ministry of Culture and Sports).

Fig. 12: Bowl-shaped litharge with flat rectangular shape from Lambrika (source: Archive of the Ephorate of Antiquities of East Attica, Copyright © Hellenic Ministry of Culture and Sports).

prep). The litharge bowls should thus more correctly be identified as litharge-impregnated hearth lining, as proposed by E. Pernicka and his colleagues on the basis of similar finds from Habuba Kabira in Syria (Pernicka, et al., 1998). The process they described was relatively simple and certainly different to what we encounter in the case of Classical Laurion. A well-formed calcium- and silicarich lining, leaving, at the end of the process, the less reactive silver metal to collect on the top, was applied, or was placed internally to the rock-cut cavities. The ten depressions must have been pre-formed on this lining or the cupel prior to firing. The solid silver-rich lead metal was subsequently placed within these cavities along with the fuel and heated under an oxidizing atmosphere. The forming litharge was continuously absorbed by the lining leaving, at the end of the process, the less reactive silver metal to collect on the top, most likely concentrating within the ten depressions. There are no ceramic or other finds with evidence for a lead-rich coating to suggest that any other vessels were involved in the process.

It is noteworthy, that among the bowl-shaped fragments, there is additionally a unique specimen (Fig. 12) that has a flat and rectangular shape. It has eight shallow depressions in two series of four, and another less clear one below. In at least five of these depressions there are smaller rounded depressions marked by a litharge rim at its inner side This is a peculiar and so far unique find. A possible interpretation is, that these depressions which must have been formed naturally are the imprints left by the separated silver globules, which were too small to fill the entire space of the pre-formed depressions on the lining(see also below for the single depressions noted on the Merenda and Gyalou finds). It is evident that at Lambrika there was an outdoor cupellation workshop. The large amount of metallurgical remains (roughly 160kg in weight) suggests that the primary aim of the workshop was the production of silver by cupellation. On the basis of the workshop layout (Fig. 4), we propose that the metallurgists were standing in the large pit in the centre, to facilitate their work within the small cavities. Preliminary calculations have shown that despite the extent of the Lambrika workshop and the large amount of litharge recovered, so far unparalleled anywhere else in the Old World, the final product of silver from several centuries of production may be impressively small, even if we accept a large error margin (Georgakopoulou, et al., 2020). This observation certainly highlights the significance and value of silver in prehistory.

Merenda

Between 2000 to 2004, extensive archaeological research at the nearby site of Merenda, Markopoulo (Fig. 1), brought to light another interesting metallurgical assemblage, that includes the earliest securely dated litharge fragments known to date from southeast Attica. The prehistoric settlement of Merenda (Kακαβογιάννη, 2009, pp.55–57, fig. 2 (no. 2); Kακαβογιάννη, et al., 2009) is dated from the FN to the EH II period (Kakavogianni, et al., 2016, pp.442–443). In the first two occupational phases assigned to the FN/EH I and the EH I period, the settlement is characterised by underground chambers cut into the soft bedrock, organised in six clusters (A – Σ T), which were used as residences (Κακαβογιάννη, et al., 2009, figs. 1–3, 7, 8, 12, 14; Kakavogianni, et al., 2016, figs. 6, 7). During the third phase, in the EH II period, habitation was limited to few underground chambers and to two small free-standing buildings, built above ground (Κακαβογιάννη, et al., 2009, pp.169–171, figs. 1, 15).

Eighty litharge fragments (in total 9 kg) were recovered from the entire Merenda settlement. However, the vast majority of the material is dated to the FN/EH I and EH I phases. A large number (34) were found in the fills of chambers A, B, Γ , Δ and E. Following their use, these chambers had been filled with waste from the settlement and among these materials also the litharge fragments were found, which were mainly bowl-shaped.

Amongst the material discovered in the chambers, particularly important is the identification of litharge fragments in a closed context from cluster B of the subterranean chambers (Kakavogianni, et al., 2016, pp.445–447), which, according to the latest evidence



Fig. 13: Fragments of bowl-shaped litharge from Merenda (source: Archive of the Ephorate of Antiquities of East Attica, Copyright © Hellenic Ministry of Culture and Sports).



Fig. 14: Fragment of bowl-shaped litharge with a single depression on upper surface from Merenda (source: Archive of the Ephorate of Antiquities of East Attica, Copyright © Hellenic Ministry of Culture and Sports).

from radiocarbon dating, date to the middle of the 4th millennium BC (Maniatis, et al., 2016, p.58; Tsirtsoni, 2016, p.454). These are the earliest securely dated litharge fragments known to date and constitute the earliest direct evidence for silver production through cupellation in the Aegean. Furthermore, a small fragment of a flat litharge was found in a small, isolated building in close vicinity to the settlement, which was characterized as a small 'workshop' due to the nature of the finds that included a large number of stone tools.

In the third phase of the settlement (EH II period), in close vicinity (northeast) to a small free-standing house, an important obsidian and flint tool production workshop (Spiliotakopoulou, 2020) was brought to light. In the same context, several metallurgical remains were uncovered relating mainly to copper (clay moulds and copper slags) as well as a lead clamp, a small amorphous lead fragment and few fragments of litharge. The latter are all very small and fragmentary and their original shape cannot be reconstructed.

At the Merenda settlement we recovered primarily bowl-shaped litharge fragments (76 out of 80) and a small number of flat fragments. Intact bowl-shaped litharge examples were not found at Merenda; the largest preserved fragment measures 9 by 6 cm and provides substantial information concerning its original shape - its diameter would have been approximately 10 cm and its height about 2 cm. The average size of the litharge fragments is relatively small, roughly 4 by 3 cm. The vast majority of the fragments has a flat upper surface (Fig. 13), with no evidence for the characteristic depressions, which, as was outlined before, is a distinctive element of the finds from the nearby workshop of Lambrika. In three cases, however, the litharge fragments display a single large depression on their upper surface (Fig. 14). Contrary to the multiple depressions seen in the litharge fragments of Lambrika, these single depressions on the Merenda specimens could be a natural result of the cupellation process, marking the spot, where the silver was concentrated at the end of the process (see for example Bayley and Eckstein, 1997; Renzi 2013, p.54, fig. 4.1) or performed to collect the silver during the process of cupellation.

In the flat litharge fragments, a different type, or a different procedure cannot be distinguished, as their number and their state of preservation is poor. It is noteworthy that their thickness, which is less than 1 cm, is smaller than that of the bowl-shaped litharge of Lambrika and of some small pieces of so-called plate-shaped litharge known from other sites (Kakavogianni, et al., 2008).

The archaeological context of the litharge fragments in the excavated area at Merenda do not allow for identifying a particular workshop area, as diagnostic installations were not recognized and there were no particular concentrations of these waste materials that stood out, as is the case with Gyalou, discussed below. Litharge fragments at Merenda are found dispersed throughout the excavated area and across all chronological phases. Their poor preservation in comparison to the other sites discussed here is also noteworthy. Nevertheless, the number of specimens is substantial, particularly if we compare it to the small number of litharge from the large settlement of Koropi (see below).

We therefore suggest, even in the absence of clear installations, that towards the middle of the 4th millennium BC, there was another cupellation workshop in the broader area of Merenda, whose litharge finds differ from those of the workshop of Lambrika. Although the litharge at Merenda is also bowl-shaped, the characteristic depressions are absent. The significantly smaller amount of litharge found here, may reflect the fact that the actual workshop area has not been identified, or alternatively, might indicate a smaller scale of production, possibly only serving the needs of the settlement itself.

Gyalou

The settlement of Gyalou at Spata (Fig. 1) is a small, flat settlement of approximately 3 acres, dating to the FNL/EHI period (Γ κινάλας, Στάθη and Ζγουλέτα, 2015; Georgakopoulou, et al., 2020). In the western part of the settlement, excavation revealed mainly groups of pits which were used as storage areas, refuse pits etc.. Aside from stone tools and pottery, several litharge fragments were recovered from some of these pits.

Fifty-one litharge fragments (8 kg) were recovered in the Gyalou settlement and all were found in the western nuclei and particularly its northern part, in Sector 14. Excavations in this sector revealed a complex of six pits of different sizes. The largest is Pit 6 of ellipsoid shape and large dimensions (2 by 2.3 m), but relatively shallow depth (0.66 m). In its interior there was a grey-brown fill, whilst on its sides there was a groove, perpendicular to them. The largest number of litharge fragments was collected from the interior of this pit together with masses of clay, pottery, stone tools (obsidian and grinding tools) and animal bones. Pit 1, of approximately circular shape and smaller dimensions (diameter of 1 m), had at its bottom a floor made up of semi-worked stone. Initially it probably served a storage use, but subsequently became a refuse pit filled with pottery, stone tools and a few litharge fragments.

All litharge fragments from Gyalou are of the bowlshaped type (Fig. 15). Their preservation is quite good. Although there are no completely preserved pieces, the average size preserved is quite large around 7 by 5 cm. The largest preserved fragment measures 12.3 by 9.8 cm. Their estimated diameters range between 11–22 cm, whilst their height is approximately 2.5 cm. They have a flat upper surface (Γκινάλας, Στάθη and Ζγουλέτα, 2015,



Fig. 15: Fragments of bowl-shaped litharge from Gyalou (source: Archive of the Ephorate of Antiquities of East Attica, Copyright © Hellenic Ministry of Culture and Sports).



Fig. 16: Fragments of clay moulds from Koropi (source: Archive of the Ephorate of Antiquities of East Attica, Copyright © Hellenic Ministry of Culture and Sports).



Fig. 17: Fragment of bowl-shaped litharge from Koropi (source: Archive of the Ephorate of Antiquities of East Attica, Copyright © Hellenic Ministry of Culture and Sports).

p.344, fig. 23), whilst their bottom surface is rough with evidence of burning and attached soil. Approximately one third of these (15 examples) bear on their upper surface a single central depression, similar to that seen in some cases at the Merenda material. The central depression has a diameter that ranges between 1.4-3.8 cm and a depth of 0.2-0.3 cm. As in Merenda, this could be the negativeprint of the silver which agglomerates by cupellation, rather than pre-formed depressions on the cupellation hearth lining, as proposed for the case of Lambrika. The excavations at Gyalou did not bring to light a single example with multiple depressions, as known from Lambrika and Zapani.

Although, similarly to Merenda, clear workshop features were not identified in the excavated settlement, the evidence for *in situ* cupellation is substantial. It includes a large number of litharge fragments and their concentration in a single part of the settlement, specifically its northern side, as well as their spatial connection with a group of pits, which might be connected to the metallurgical activities based on the associated finds and evidence for burning.

Koropi

The extensive and nodal settlement of Koropi, situated at a distance of 5 km from Lambrika (Fig. 1), shows a different picture in terms of the presence and distribution of metallurgical remains. Such finds were retrieved from the earliest strata dating to the EH I period (western part of the settlement) (Kakavogianni, et al., 2008; Kακαβογιάννη and Ντούνη, 2009, p.392). They include a fragment of a bowl-shaped litharge without depressions, a few copper slags and 23 clay moulds (Fig. 16). They were mainly found in secondary deposits in the filling of a room together with a great quantity of pottery, animal bones, shells and stone tools.

Metallurgical finds dating to the subsequent EH II period were recovered from the western, eastern and northern nuclei of the settlement. In the filling of the subterranean chambers (northern nucleus), three small fragments of plate-shaped litharge were collected, as well as clay crucibles, clay moulds and two large copper slags, one of which is rounded and appears to have been used as a pestle (see for comparison Georgakopoulou, 2013, p.672 and references within). A copper slag and a clay mould were collected from the eastern part of the excavated street.

Several clay moulds and two litharge fragments were found scattered in the eastern and western part of the settlement, where complexes of streets with adjacent houses were discovered as well as a large communal building and areas for the processing and production of stone tools.

In total six litharge fragments were found in the excavated area, a number surprisingly small compared to the previous sites, especially if the large extent of the excavated area is taken into account (Georgakopoulou, et al., 2020). Only one of them is clearly of the bowl-shaped type (Fig. 17), most probably without the characteristic depressions. In contrast to the surprisingly small number of litharge fragments, evidence for copper working activities is particularly striking at Koropi, mainly at the early phase of the settlement (EH I period).

Discussion

From simple bowls to bowls with depressions – an Attic/Lavreotic innovation

Similar to the scanty early lead-silver mining evidence in the Lavreotiki region, the earliest litharge fragments known from southeast Attica date to the Final Neolithic period, in the middle of the 4th millennium BC and were found at Merenda (Tsirtsoni, 2016, p.450, n. 113). Perhaps the material from Gyalou dates to the same period. However, the only clear cupellation workshop known presently, at Lambrika, is dated to the following period, the EH I, at the end of 4th/beginning of the 3rd millennium BC. Its discovery in 2002 opened a new chapter in the study of archaeometallurgy in Attica. The earliest litharge finds from the extensive Koropi settlement also date to the EH I, as does the single example from the cemetery of Tsepi, Marathon (Παντελίδου-Γκόφα, 2005, pp.68, 323, 345-349, pl. 8) as well as all the litharge fragments recovered from Asteria, Glyfada (Καζά-Παπαγεωγίου, 2014, fig. 6). The dating of a second potential cupellation workshop found at Zapani (Andrikou, et al., this vol.) also seems to extend back to this period. So far, it constitutes the earliest preserved workshop from within the Lavreotiki.

The EH I thus emerges as a period of intense silver production activity in southeast Attica. This picture somewhat changes in EH II. Aside from the possible workshop at Zapani, which continues to operate, evidence from the settlement of Lambrika, suggests that metallurgical activities also continue there, but on a much smaller scale. Remains associated with lead-silver metallurgy of this period have also been discovered in different areas in Attica: litharge fragments in Koropi, Ayia Marina at Koropi, Provatsa on Makronissos (Spitaels, 1982, p.158), Kalyvia Melissourgou (Τσαραβόπουλος, et al., 2001, p.186), two so-called lead ingots from Rouf (Πετριτάκη, 1980, p.174, pl. 48a) and the leaf-shaped lead object from Askitario at Raphina (Θεοχάρης, 1953-1954, p.75). The earliest exploitation of Mine No. 3 at Thorikos has been dated to this period, although the ceramic evidence suggests possibly earlier works (Spitaels, 1984, 166; Νάζου, 2013).

During the following Middle Helladic period, litharge fragments have been found in settlements such as Velatouri at Keratea (Kakavogianni, et al., 2008, p.50, fig. 10; Κακαβογιάννη and Ντούνη, 2009, pp.393–395) and Velatouri at Thorikos (Servais, 1967, p.22, fig. 16). Perhaps the examples mentioned above found in private collections or in classical contexts also date to prehistoric periods.

With reference to the morphological aspects of litharge, the bowl-shaped type is by far the most abundant and characteristic type of the prehistoric period. It is related to a specific technological process that becomes evident in the Lambrika workshop. All the other types, like plate-shaped litharge, flat litharge or lumps are so few that we cannot draw any conclusions concerning the technology involved (Kakavogianni, et al., 2008). As for the morphology of the bowl-shaped litharge, in the light of the present finds, we can distinguish two categories: bowl-shaped without depressions (Merenda type) and with multiple shallow depressions on their upper surface (Lambrika type). In the first category, some specimens bear on their upper surface a single central depression. The archaeological evidence proves that there is a chronological difference between the two categories, at least between the material discussed in this paper; the bowlshaped specimens without depressions being older than the Lambrika type. It seems that the older material (FN) in Merenda and Gyalou is always without depressions, while the depressions appear from the more mature EH I phase, attested at Lambrika. However, the appearance of depressions may also have a restricted spatial distribution within Attica, concentrating primarily in the Lavreotiki and extending to Lambrika. This would be supported by their absence not only from adjacent sites such as Merenda and Koropi but also from other sites at a longer distance such as Tsepi or Asteria.

Bowl-shaped litharge cakes are also known from Aegean sites outside Attica such as at Final Neolithic Limenaria on Thasos (Papadopoulos, 2008; Neranztis and Papadopoulos, 2013; Νεραντζής, 2017; Bassiakos, et al., 2019) and EBII Akrotiraki on Siphnos (Papadopoulou, 2011; Μπασιάκος, et al., 2013; Παπαδοπούλου, 2013). None of these show any evidence for the multiple depressions on the upper surface, proving that these are so far at least a regional, Attic/Lavreotic innovation. The reason behind the clearly deliberate presence of these depressions leaves many open questions. Was the aim to recover silver in small globules of specific sizes? This is a tempting suggestion with interesting implications for early systematic recovery and distribution of precious metals in specific weights and sizes. Presently we have no evidence for the existence of such small silver globules in the prehistoric Aegean, how ever, this cannot be considered negative evidence, as these could have been remelted to make artefacts. Alternatively, or additionally, would these depressions have improved silver recovery by exposing slightly more lining surface and promoting the soaking of the lead oxide (Rehren and Klappauf, 1995)? Or finally, is this Attic technological particularity not driven at all by functionality, but rather reflects an attempt at a distinctive cultural differentiation by the Attic metalsmiths? Planned future systematic experiments will attempt to address these questions.

Presence of litharge in the sites mentioned above – identification of cupellation workshops

Litharge may be present in an archaeological context either as waste, or as raw material for secondary use, with several instances known from antiquity (see discussion and references in Georgakopoulou, 2007, pp.394-395). Specifically in the Aegean, there is clear evidence that litharge was used as a pigment at the site of Akrotiri on Thera, based on remains of the material identified on a significant number of stone tools dating from the Early to the Late Bronze Age (Sotiropoulou, et al., 2010). Similar evidence has not been identified in Attica, but a systematic study remains to be done. Earlier analyses of litharge and lead and silver artefacts from the Bronze Age Aegean have not favoured the idea that litharge was recycled back to lead metal in the prehistoric Aegean (Gale and Stos-Gale, 1984; Pernicka, et al., 1983). The relatively high silver content of the analysed lead artefacts (above 0.01%) compared with the analyses of contemporaneous litharge finds suggested that lead was produced by smelting separate batches of silver-poor lead ores, rather than by reduction of the litharge produced as a by-product of silver production from silver-rich lead ores. It should, however, be noted that published analyses of lead, silver and litharge finds from the Attica region are so far scarce and the pattern remains to be tested here.

Thus, the identification of litharge fragments on a site does not necessarily suggest in situ cupellation, but the possibility that these have been transferred there for secondary usage needs to be considered. Useful deductions are to be drawn when considering the archaeological contexts of these finds. The quantity of litharge and the workshop installations at Lambrika leave no doubt that a cupellation workshop existed on the site during the EH I and possibly during the EH II. Similarly, Zapani also stands out as a cupellation workshop, based on the high relative number of litharge compared to the excavated area (Georgakopoulou, et al., 2020), despite the lack of identified installations so far. In any case, the unsophisticated nature of these installations means that their preservation in most cases is rather unlikely. Although significantly less, both in absolute terms and relative to excavated areas, the amounts of litharge from Merenda and Gyalou are substantial and were most likely disposed of from a nearby cupellation workshop, as yet unidentified. The smaller quantities suggest activities on a much smaller scale or perhaps carried out at a longer distance from these settlements or over a shorter period than those of Lambrika or Zapani.

The situation at Koropi is, however, somewhat different and the strikingly small number of litharge compared to the size of the excavated settlement could either suggest that cupellation was practiced rarely and in a very small scale here, or alternatively that the litharge was present as raw material for other uses. What these possible uses may have been remains to be addressed through the thorough study of the range of finds from the settlement. Regarding the possibility that the litharge fragments served there as raw material for recycling and re-melting back to lead metal, another parameter should be noted. The analysis of litharge fragments from Attica, undertaken so far (Georgakopoulou, et al., in prep) showed that these are not composed of pure lead oxide but bear a significant content of other gangue oxides. In principle pure re-melting of these materials under a reducing atmosphere would not produce pure metallic lead unless they were properly smelted removing the gangue oxides by slagging. The lack of such slags from the excavated sites in southeast Attica may constitute an additional argument against the proposal that litharge was re-melted back to lead metal during this period.

The wealth of finds from southeast Attica testify to the significance of the rich metalliferous deposits of Laurion already from the earliest stages of silver production in the Aegean. The inhabitants of this region not only knew of these resources, but also systematically exploited them, as is clearly evident from the organized Lambrika workshop. It seems that Lavreotic silver and lead were distributed across the Aegean. The finds discussed here illustrate a unique Attic/Lavreotic technological trajectory which is currently being defined more specifically both temporally and spatially.

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We mourn the sudden loss of our colleague Myrto Georgakopoulou. She was an eminent scientist and her work has had a transformative effect on our understanding of prehistoric metallurgy and Laurion's important role in prehistoric and later periods.

Notes

 The archaeologists K. Douni, P. Michailidi and F. Nezeri participated in the excavations at Lambrika. The archaeologists M. Nazou and Efth. Kakavogiannis catalogued the litharge and other finds.

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