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The emergence of metal use in Greek Eastern Macedonia during the Neolithic period (late 6th–5th millennia BC)

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ABSTRACT – Copper, gold, and silver artefacts, together with evidence of metallurgical activities, have been retrieved from Late Neolithic strata in several settlements in Greek Eastern Macedonia. Recent excavations at Dikili Tash revealed that gold was further used in paints for the decoration of pottery. It appears that the area's inhabitants had a great familiarity with different metals and the distinct stages of the production-elaboration processes, including those interfering with other chaînes opératoires. Considering also the results from geological research, we propose a reflection on the socio-economic role of metal production and consumption for these societies, in their broader Balkan context.

KEY WORDS – metalworking; Eastern Macedonia; Greece; Late Neolithic

Začetek uporabe kovin v grški vzhodni Makedoniji v obdobju neolitika (pozno 6.–5. tisočletje pr. n. št.)

IZVLEČEK – V več naseljih v grški vzhodni Makedoniji so bili v pozno neolitskih plasteh skupaj z dokazi o metalurških dejavnostih odkriti bakreni, zlati in srebrni artefakti. Nedavna izkopavanja v Dikili Tashu so pokazala, da se je zlato uporabljalo v pripravi barv za okraševanje keramike. Zdi se, da so prebivalci tega območja dobro poznali različne kovine in faze procesov priprave in izdelave, tudi tiste v drugih operacijskih sekvencah. Ob upoštevanju rezultatov geoloških raziskav, predlagamo razmislek o družbeno-ekonomski vlogi proizvodnje in uporabi kovin v širšem balkanskem kontekstu.

KLJUČNE BESEDE – obdelava kovin; vzhodna Makedonija; Grčija; pozni neolitik

Introduction

Northern Greece has been diachronically associated with the exploitation of its mineral wealth. Greek Eastern Macedonia in particular, *i.e.* the area between the Strymon and Nestos river valleys including the nearby Thasos island (Fig. 1), is known for its numerous polymetallic resources, which have been intensively exploited from Antiquity until modern times, with gold, silver, copper, lead and iron being the main metals extracted. Although we still

lack direct evidence about their exploitation in prehistoric and protohistoric times (with the remarkable exception of the Palaeolithic ochre mine at Tzines, Thasos, around 20 000 years ago; *Koukouli-Chryssanthaki, Weisgerber 1999*), we have good reasons to believe that many of the gold, silver and copper artefacts retrieved in recent decades from a number of Neolithic, Chalcolithic and Bronze Age sites in the area were produced from local resour-

ces. Moreover, recent analytical studies in artefacts and metallurgical remains support the use of local ores on Thasos island (Bassiakos et al. 2019).

The geological setting of the area, its metallic resources

From a geotectonic point of view, Greek Eastern Macedonia belongs to the Rhodope Massif, which possesses several small- to large-scale magmatic-hydrothermal ore mineralizations (Melfos et al. 2002; Melfos, Voudouris 2017). Most of these occurrences present traces of ancient mining (Wagner, Weisgerber 1988; Koukouli-Chrysanthaki 1990; Vavelidis et al. 1995; 1996a; 1996b; Chiotis et al. 1996; Vaxevanopoulos 2017a; Vaxevanopoulos et al. 2022). Precious metal mineralizations also appear in the Serbo-Macedonian Massif at its borders with Rhodope Massif in the Strymon valley. The Strymon river is well known for its placer gold from ancient writers. Pseudo-Aristotle describes the gold nuggets found in the riversides after heavy rains in the Paeonian territory (*De Mirabilibus Auscultationibus* 45).

The Pangaeon mountain is often mentioned by ancient writers, and ores rich in gold, silver and copper were extracted in many mining areas during Antiquity (Vaxevanopoulos 2017a; 2017b). Copper rich mineralizations and metallurgical areas with copper being one of the main extracted metals, especially in Roman times, have also been recorded (Vaxevanopoulos et al. 2018). Placer gold has been identified at several spots in the surroundings of the mountain. Alluvial gold is further reported in the streams crossing the region between Pangaeon and Symvolon (Baker et al. 1992), and limonitic veins with gold occurrences have been located in Alistrati, at the feet of the Menoikion mountain (Vavelidis et al. 1995). Further north, on the Angistrion and Orvilos mountains, several gold and silver deposits have been recorded and gold-bearing veins in marble have also been exploited (Chiotis et al. 1996).

Several mineralizations rich in gold and silver have also been identified in the region of Palea Kavala, on the Lekani Mountains, which are considered as the ancient Skapti Yli described by Thucydides (Photos et al. 1989; Koukouli-Chrysanthaki 1990).

More than 150 occurrences rich in Fe, Cu, Ag and Au have been recorded, whereas underground galleries are widespread in this area and can be dated from the 6th century BC until the Ottoman period (Vavelidis et al. 1996a; 1996b).

Finally, Thasos island, which as previously mentioned offers the earliest evidence for exploitation of metallic minerals in the Aegean, also has a number of gold, silver-lead and copper deposits. Gold deposits are located mainly on the eastern coast of the island where ancient galleries have been studied (Wagner et al. 1979; 1981; Vavelidis et al. 1988a; 1988b). Ancient exploitation of silver has been studied in the Marlou-Kourlou, Koumaria and Vouves mining areas, in the opposite southwestern part of the island, with numerous underground galleries, shafts and surface extraction (Hauptmann et al. 1988; Pernicka, Wagner 1988; Pernicka et al. 1992; Wagner, Weisgerber 1988; Sanidas et al. 2018).

Brief history of archaeological research in the area

The first Neolithic settlements were established in the area (Fig. 2) in the second half of the 7th millennium BC (Lespez et al. 2013; Maniatis 2014, 2017). The plains of Serres and Drama were ideal for human occupation as they offered abundant arable land and extended pastures, with sizable rivers or perennial sources of water present (Fotiadis 1985; Andreou et al. 1996). Relative proximity to the wooded slopes of the surrounding metalliferous mountains as well as to the Northern Aegean coastline must have added considerably to the subsistence/economic potential of the area.

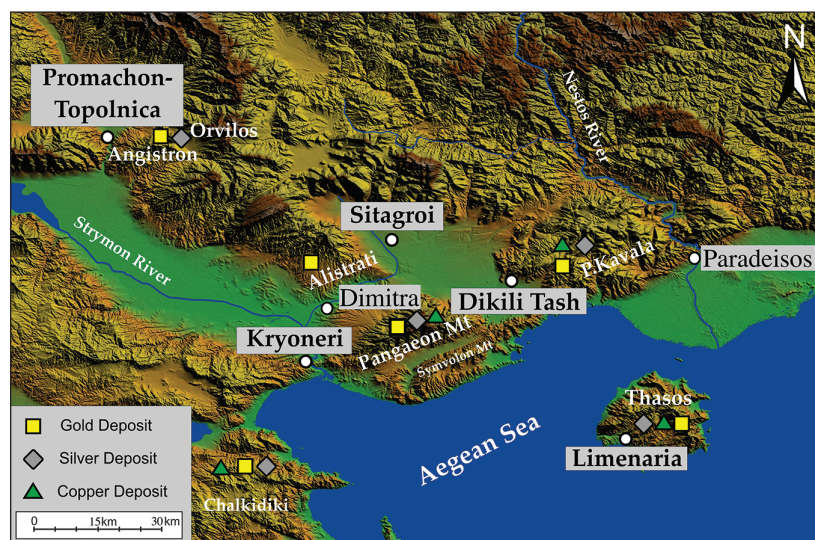


Fig. 1. Map of Greek Eastern Macedonia showing the main locations of gold, silver and copper deposits.

Although both plains comprise many important Neolithic (late 7th–mid 4th millennium BC) sites (*Grammenos, Fotiadis 1980; Grammenos 1991.120–126; Koukouli-Chryssanthaki et al. 2008*), only a few have been properly investigated. Promachon-Topolnitsa, on the west bank of Strymon on the Greek-Bulgarian border, is the only one excavated systematically to a large extent in the Serres basin (*Koukouli-Chryssanthaki et al. 2007*). Smaller scale excavations have been conducted in the cave Orpheas-Alistrati (*Kontaxi et al. 2004*) and the open-air sites at Agio Pnevma (*Tolia-Christakou, Siopi 2008*), Amphipolis-Hill 133 (*Lazaridis 1964; 1965*), Dimitra (*Grammenos 1991; 1997*) and Kryoneri (*Malamidou 2007; 2016*). In the plain of Drama, important systematic excavations have been conducted at the tell sites of Sitagroi (*Renfrew et al. 1986; Elster, Renfrew 2003*) and Dikili Tash (*Treuil 1992; Koukouli, Romiopoulou 1992; Darcque et al. 2020* with references; and www.dikili-tash.fr), whereas smaller investigations

took place at Polystylo (*Mylonas, Bakalakis 1938. 109–111*), Arkadikos-Drama (*Touloumis, Peristeri 1991; Peristeri 2002; 2004*) and in the cave of Maaras at the Angitis sources (*Trantalidou et al. 2005*). In the small valley of Pieria, between the Pangaeon and the Symvolon mountains, only the site of Akropotamos has undergone some investigation (*Mylonas 1941*). A small-scale excavation was further conducted at the site of Paradeisos, in the Nestos river valley (*Hellström 1987*). The sites of Kastri-Theologos (*Koukouli-Chryssanthaki 1974; Koukouli-Chryssanthaki, Papadopoulos 2016.342–349*) and Limenaria (*Papadopoulos, Malamidou 2012*) are the only two Neolithic sites excavated so far on Thasos island. Limited remains of the final stages of the period have also been found in the neighbouring site of Agios Antonios (*Maniatis et al. 2015.810–811*).

Most of these sites have provided evidence in connection with metals or metallurgy, in the form of fi-

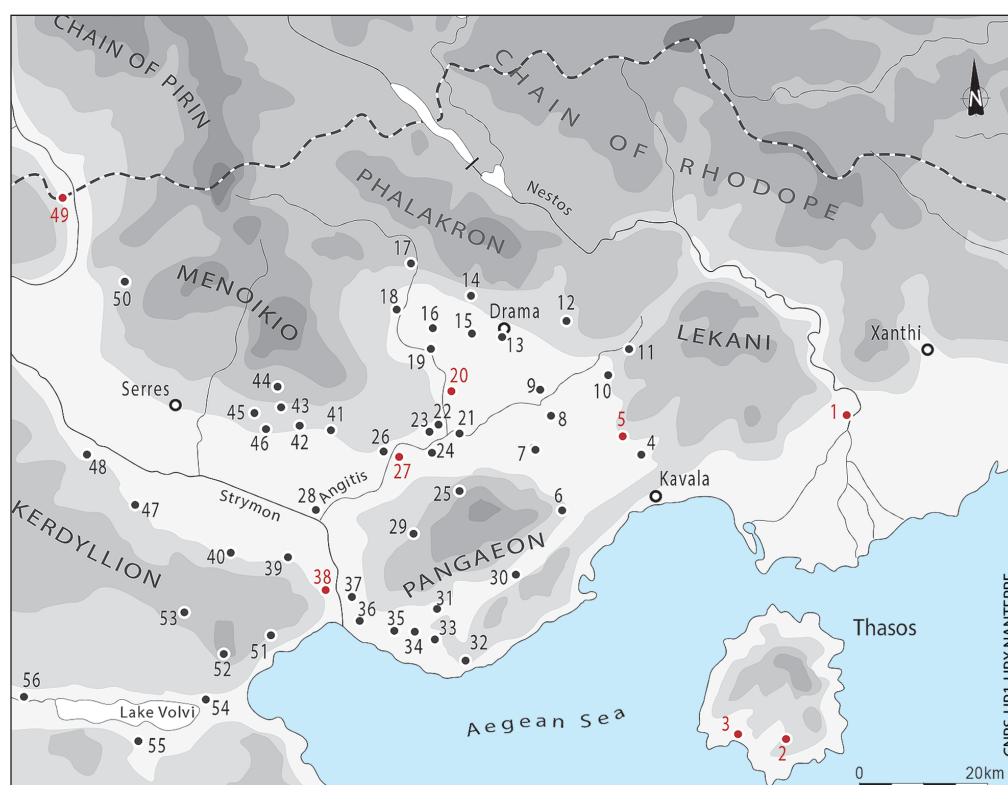


Fig. 2. Map of the area with the main Neolithic and Copper Age sites: 1 Paradeisos, 2 Kastri, 3 Limenaria, 4 Polystylo, 5 Dikili Tash, 6 Eleftheroupoli, 7 Kalamonas, 8 Kalambaki, 9 Doxato, 10 Kefalari, 11 Adriani, 12 Kallifytos, 13 Arkadikos-Drama, 14 Xeropotamos, 15 Mylopotamos, 16 Petroussa, 17 Maaras-Angitis Sources, 18 Kali Vrysi, 19 Megalokambos, 20 Sitagroi, 21 Mavrolefki, 22 Symvoli, 23 Orpheas-Alistrati Cave, 24 Angista R.S.-Paliokostra, 25 Nea Bafra, 26 Airi Bairi, 27 Dimitra, 28 Fidokoryfi, 29 Mikro Souli, 30 Moustheni, 31 Podochori, 32 Loutra Eleftheron, 33 Akropotamos, 34 Kokkinochori, 35 Galepsos, 36 Ofrynio, 37 Amphipolis-Hill 133, 38 Kryoneri, 39 Kastanochori, 40 Zervochori, 41 Tholos, 42 Toumba, 43 Pentapoli, 44 Agio Pnevma, 45 Fakistra Chryssou, 46 Chryssos R.S., 47 Vergi, 48 Strymoniko, 49 Promachon-Topolnitsa, 50 Katarraktes-Sidirokastro, 51 Asprovalta-Agia Lydia, 52 Asprovalta-Platoma, 53 Arethoussa, 54 Mikri Volvi, 55 Nea Apollonia, 56 Profitis. Sites no. 51 to 56 belong administratively to the region of central Macedonia. Red dots indicate sites with evidence of metal objects and/or metallurgy.

nished objects, metal-processing installations, instruments or by-products. Part of the evidence relates with later occupation layers (Early or Late Bronze Age), frequently present at the same sites, but much of it comes from secure stratified Late Neolithic deposits and even, exceptionally, from in situ contexts (Aslanis, Tzahili 1990). Archaeometric examination of the related finds has produced valuable information on the technology of early copper, gold and silver exploitation in the area (Mirtsou et al. 1997; Koukouli-Chrysanthaki, Bassiakos 2002; Renfrew, Slater 2003; Koukouli-Chrysanthaki, Papadopoulos 2009.2; Papadopoulos 2008.67; Bassiakos 2012; Nerantzis, Papadopoulos 2013; Bassiakos et al. 2019).

Aim of the paper

Our aim is to go through this available disparate information in order to produce a comprehensive synthesis of our present knowledge on metal production and use in Greek Eastern Macedonia during the late 6th and 5th millennia BC, which is an important formative period not only here but across the entire Balkan peninsula (Pernicka 1993; Mazanova 2004; Jovanović 2009; Šljivar 2006; Hansen 2013; Kunze, Pernicka 2020a; Radivojević, Roberts 2021) and the Aegean (Zachos 1996a; Papadatos et al. 2004; Bassiakos, Catapotis 2006; Kakavogianni et al. 2008; Zachos 2010), as well as in Anatolia and the Near East (Efe 2002; Maddin et al. 1999; Özdoğan, Parzinger 2000; Rothenberg, Merkel 1998; Thornton 2001; Yalçın 2000; Yener 2000). More recent and still unpublished data are also included, mainly from the recent works at the tell settlement of Dikili Tash. We focus on technological information about the *chaîne opératoire* of the different metals, from the acquisition of ores and minerals from local sources and extractive metallurgy through smelting, to the production of finished objects. We combine material and earth sciences with well-established archaeological facts from individual sites, thus hoping to identify patterns of human behaviour in connection with other activities (e.g., other *chaînes opératoires*), in order to better understand the functioning of Neolithic metallurgy and its social background (cf. Storberg 2002.469; Ottaway, Roberts 2008; Bartelheim et al. 2015). By privileging well-dated and secure excavation contexts we try to gather pertinent evidence about metal resource exploitation and consumer choices, taking into consideration the overall practices of local populations, as well as possible networks of contacts and exchange at various geographical scales during the aforementioned period.

Data presentation

Copper

Promachon-Topolnitsa

Copper artefacts and the remains of copper-processing were found in levels of phase Promachon III in the Greek sector, corresponding roughly to the first half of the 5th millennium BC. Excavations yielded several malachite beads (Koukouli-Chrysanthaki et al. 2007.51, Fig. 8), one copper pendant, copper fragments and slags. One heavily burned clay crucible containing traces of copper smelting was found at the bottom of a small pit (*ibid.* 48–51, Fig. 7.1, 2, 4). The surrounding area also revealed traces of copper on the floor. In trench B, a series of hollows was discovered in the floor with successive layers of burnt clay in the interior (Fig. 3), which offered clear evidence for copper extraction. These features resemble similar constructions at Dikili Tash (see below). According to preliminary analyses (optical microscope, XRF and SEM), a special feature of copper production at Promachon-Topolnitsa is the use of a secondary copper ore (carbonate) of high pureness, consisting solely of malachite with >95% CuO. Such a pure copper ore, with no alumina-calcium-silica and iron admixtures, allows the production of copper at temperatures lower than 1000°C without slag being produced (Koukouli-Chrysanthaki, Bassiakos 2002; Bassiakos et al. 2019). Although relevant analysis of the aforementioned pure copper ores is still ongoing, it is clear that the metallurgical activity in Promachon-Topolnitsa is closely related to technological traditions of copper production in the Balkan hinterland (Pernicka et al. 1997; Gale et al. 2003; Borić 2009; Radivojević et al. 2010; Radivojević, Rehren 2016; Rehren et al. 2020; Radivojević, Roberts 2021).

Dimitra

Metal finds from Dimitra are proportionally numerous (Grammenos 1997.Pl. II) as all the soil from the two trenches investigated was sieved. Neolithic strata yielded 38 copper objects – 21 beads, one fish-hook (*ibid.* 51, Pl. 36.2), five parts of pins or wires, and 11 undefined fragments. The majority come from 5th millennium BC layers, but some certainly date to the second half of the 6th millennium (*i.e.* five beads from ‘Middle Neolithic’ layers in trench II). Fourteen copper artefacts have been analysed with several methods, including emission spectroscopy for the estimation of their qualitative composition, proton induced x-ray emission (PIXE) to determine their quantitative elemental composition, and

x-ray radiography in order to confirm the internal metallic structure of the objects (Mirtsou et al. 1997). One copper artefact was sectioned for metallographic examination to obtain information about the manufacturing procedure. All samples had different compositions, with arsenic, zinc and bismuth present in various proportions. Copper beads were produced by cold hammering and annealing at about 600°C.

Apart from the metal objects, small amounts of poly-metallic ore (rich in copper, iron, lead) have been collected from both trenches, but they are not yet analysed. No melting or smelting installations have been identified in the excavated areas. Consequently, there is still no direct evidence for on-site metallurgical activity, and we are not aware which metal source was exploited.

Kryoneri

Three copper pins/awls, two fragments of copper rings and some ore fragments were found in Late Neolithic deposits (mid-5th millennium BC), from both domestic context and waste pits (Malamidou 1997.518, Fig. 11; 2007.302, Fig. 8; 2016.312, Fig. 26). Preliminary examination of two pins under an electron-scanning microscope (SEM) by Yannis Maniatis at the Laboratory of Archaeometry of the NCSR Demokritos showed that they are made of pure copper. No further analysis has yet been conducted.

Sitagroi

The finds of phases Sitagroi II and III (late 6th and 5th millennia BC) include a copper pin, an awl, five beads and a few copper fragments or lumps (Renfrew, Slater 2003.305, 319–320, Fig. 8.1.a-d,f-g). In addition, a marked concentration of sherds with copper incrustation was detected in square MM layer 20 and adjacent levels (phase Sitagroi III). Four sherds with copper remains from square MM, layers 61 and 60, probably belong to an earlier context. Given the nature of the ceramics (coarse, with rough surfaces and curved walls) and the distribution of the remains (mostly inside and on the broken edges), they are interpreted as parts of crucibles or as sherds used to remove or to hold back dross during metal pouring (Fig. 4) (Renfrew, Slater 2003.303, 312, Fig. 8.4). One of the best-preserved is a fragment of an oval-shaped crucible (dimensions c. 9x6cm, wall thickness up to 1.7cm) (*ibid.* 306, Fig. 8.4.I, Pl. 8.10). No moulds were recognized.

Sixty-eight samples from Neolithic phases, either metallic or non-metallic but presumably related to metal technology, were sectioned and analysed on a



Fig. 3. Promachon-Topolnitsa, series of hollows in trench B with evidence of use for copper extraction (© Ephorate of Antiquities of Serres).

Microscan V Microprobe Analyser (*ibid.* 301–302). It should be kept in mind that these analyses were conducted in the late 1960s using a first-generation microprobe analyser that had limited levels of precision and sensitivity. The five non-metallic samples from the Sitagroi I levels provided no evidence of metalworking. Four out of the 12 analysed samples from the phase II levels were made of copper with no intentional alloying, while the other eight were non-metallic, showing no evidence of on-site smelting. By contrast, phase III finds (11 copper objects and 40 sherds with copper deposits) show clear evidence of smelting and casting. The very low concentrations of impurities in some samples may indicate that smaller pieces of native copper were melted together in order to form an object, whereas objects with higher impurity levels may well be the product of smelting from ores. Other specimens show evidence of cold working and annealing. Three objects contained tin, but not in percentages as high as those normally associated with deliberate alloying, and two contained lead in small amounts.

The question arises of where the activities of melting or smelting, casting, cold working and annealing took place. Five separate contexts yielded sherds with adherent copper deposit. The richest one is, as already mentioned, square MM level 20, which is a rich fill including pottery, figurine fragments, bone, clay and stone objects, as well as two finished copper objects (the roll-headed pin and the awl or fish-hook). All fragments with copper deposit were concentrated around an installation, consisting of a ba-

ked clay surface surrounded on three sides by a packed clay wall and open to the east (Renfrew et al. 1986.212, Fig. 8.19, Pl. XXXVII. 2). This may be interpreted as a pyrotechnological installation. Perhaps crucible melting was carried out in such places, although the smelting process was probably undertaken at or near the mining areas from which the copper ore was obtained.

Dikili Tash

A relatively small number of copper artefacts were found during the first excavation program, conducted by Jean Deshayes and Dimitrios Theocharis between 1961 and 1975. A small copper bead (Séféridès 1992.115, Pl. 146.a) was the only artefact retrieved from a phase I context (end of 6th millennium BC), roughly contemporary with Sitagroi II. The levels of Dikili Tash phase II (contemporary with Sitagroi III) in the sectors excavated by the French team yielded nine copper awls or pins (*ibid.* 115–116, Pl. 146.b, 199.e,f), but no ornaments. Two unstratified copper roll-headed pins from the same sectors might also come from levels of this period (*ibid.* 118–119, Pl. 147.b,c; 200.b).

Five pins from phase II have been analysed by atomic absorption spectroscopy (AAS). They all contain minor percentages of lead, iron, silver, and nickel, and in two cases tin (*ibid.* 114, Tab. 12). All these elements can be present in small percentages in copper oxides such as malachite.

A few occurrences of copper oxides and copper fragments are reported in the archives of Jean Deshayes in contexts of both phase I (square X29-niv. 13) and II (squares W29-niv. 9 and W30-sol 6). Interestingly, the last of these contexts shows heavy traces of combustion around a clay-plastered pit, which has been interpreted as a possible hearth (Treuil 1992. 23). A heavily burnt sherd with copper traces is also recorded in phase II (square X30-niv. 7).

Sectors I and II, excavated by Theocharis in 1961 and 1967, respectively, have yielded some 10 copper artefacts and a sherd with copper incrustation, most probably a crucible fragment, and these are all unpublished. At least two of them (one pin and a flat sheet of copper) found in sector II/1967 could date from the Late Neolithic.

The second and third excavation programs (1986–2016) added further copper objects to the site's inventory. Parts of four pins or awls were retrieved from late-5th millennium contexts in sector 6 (which

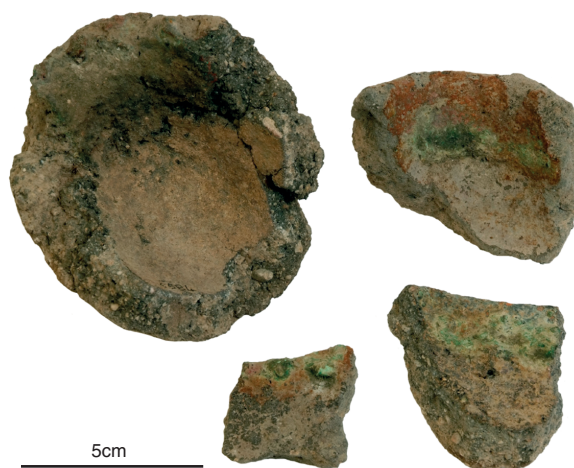


Fig. 4. Sitagroi, fragments of crucibles (© Ephorate of Antiquities of Drama).

is an extension of Theocharis' sector II), although none in an entirely secure position. A fifth one was stuck on the outer surface of a sherd found in a slightly earlier level in sector 5 (level V/East/1, c. 4700–4500 BC). A copper bead (Fig. 5) was found near a hearth or oven in House 2, again in sector 6.

The best-contextualized copper artefacts are however two other beads and an awl (Fig. 6) found in the neighbouring House 1, whose destruction has been dated to the years 4340–4260 cal BC. They come from two distinct groups of ornaments and raw materials found on the house floor, less than 2m apart from each other. The first group, from which the beads come, contained some 270 items in total, mainly clay, shell and stone beads and a few pieces of unworked shell, whereas the second group containing the awl comprised no less than 1000 beads of stone, a dozen of spondylus bracelets, hundreds of perforated plaques and pieces of boar tusks, graphite cones, and also a few gold ornaments (Darcque et al. 2012–2013. 753–758; 2014.605–608; 2015; 2020.256–265, Fig. 6–39 to 6–48, 274–275, Fig. 6–64). One of the two copper beads (actually a sheet of copper folded in the shape of a cylinder) was caught in the hole of a stone bead. Analysis with a portable XRF conducted in 2013 by Sarel Shalev (Haifa University) confirmed that both beads were made from pure copper (the awl was not analysed).

A few more copper artefacts were retrieved from sector 2 at the southern periphery of the tell, but their chronology is not safe as they all come from colluvia with later components (Darcque et al. 2020. 86–87, Fig. 3–52.b–d). The only exception is a small copper bead, which comes from a colluvium securely dated to the 5th millennium BC.

From the same layer of sector 2 also comes a fragmented clay mould (Fig. 7), the first discovered at Dikili Tash (*ibid.* 75–76, Fig. 3–39.e). It is made from well-refined clay and has fine incisions on all sides. The shape of the cavity on its upper surface suggests that it was probably used for the casting of chisels – a type of object common in assemblages of this period in both the Aegean and Balkans (*Chernykh 1978.Pls. 10, 12; Zachos 2010.86, Fig. 6–6*). XRF-analysis detected copper traces in the cavity's walls.

No crucible fragments have been found so far. One vitrified sherd with a copper deposit on its internal surface was found in the area to the southwest of House 3 in sector 6, but very near the surface.

The existence of melting or smelting installations in Dikili Tash is debated. A series of small cavities whose inner surfaces were covered with successive layers of burned clay plaster, alternating with ash layers and bearing traces of intense fire, have been interpreted as possible metallurgical structures. All of them were found in layers of phase Dikili Tash I (three in level XIV of square W30, one in level XIII of square W30 and two in level 11 of square X30) (*Séfériadès 1983.647; Treuil 1992.21–23, Pls. 13.B, 15, 32.A,B; Séfériadès 1992.115*). However, unlike Promachon-Topolnitsa (see above), no trace of slags or other elements related to metallurgical activity have been detected within or nearby these structures.

Paradeisos

The Late Neolithic levels at Paradeisos yielded two copper needles, slightly bent, 7 and 7.2cm in length respectively (Fig. 8) (*Hellström 1987.85, Fig. 48.18, 19*). Some other shapeless copper fragments were collected. No other indications of metallurgical activity were recorded.

Kastri Theologos-Thasos

Two copper pins were retrieved from Late Neolithic levels at Kastri (mid-5th millennium BC). Analysis



Fig. 5. *Dikili Tash, sector 6, copper bead found next to a hearth or oven in House 2* (© Dikili Tash Project-EFA).

has demonstrated the existence of arsenical copper, probably of Thasian origin, although this could not be safely attributed to any of the known extraction sites on the island (*Koukouli 1992.677*).

Limenaria-Thasos

Excavations of Neolithic strata at Limenaria brought to light a significant number of finds associated with copper related metallurgical activities. The recovered evidence includes copper artefacts, slags, and numerous fragments of hematite/limonite lumps, some containing secondary cupriferous minerals, mainly malachite and azurite. A malachite bead was recovered from an upper layer in the Konstantinidis plot, dating from the end of the 6th or the beginning of the 5th millennium BC (Fig. 9) (*Papadopoulos 2008.64, Fig. 2, p. 67; Papadopoulos, Malamidou 2012.41*). It is a pierced piece of malachite, made simply by percussion and polishing.

Twenty samples from slags and/or ore lumps, coming from various layers of the excavated areas, were studied with optical microscopy (OM), scanning electron microscopy (SEM-EDX), x-ray diffraction spectroscopy (XRD), and neutron activation analysis (NAA) (*Bassiakos 2012; Bassiakos et al. 2019*). Analyses showed that copper slag samples derive from reduction smelting to obtain copper. A notable heterogeneity in their texture, either micro-morphological or chemical, seems to have resulted from inefficient reducing conditions, suggesting a certain insuff-



Fig. 6. *Dikili Tash, sector 6, copper beads (a-b) and copper awl (c) from House 1* (© Dikili Tash Project-EFA).



Fig. 7. Dikili Tash, sector 2, fragmented clay mould with incised decoration on all sides (© Dikili Tash Project-EFA).

efficient control of furnace operation with some loss in the gaining of metallic copper. Despite that, these slags offer solid proof of a successful metallurgical process for efficient production of metallic copper. Until recently a similar heterogeneity in smelting slags from copper extraction was only noted at Aegean sites dated to the late 4th and 3rd millennia BC, such as Kephala-Petras, Crete (Catapotis et al. 2011; Papadatos et al. 2007) and Kythnos, Cyclades (Bassiakos, Philaniotou 2007), but this is now also reported from the late 5th millennium BC site of Akladi Cheiri in South-East Bulgaria (Rehren et al. 2020).

One sample represents a case of slagged lining deriving from the furnace's internal surface. Slag fragments like this are common within or very close to the smelting furnace from numerous other sites of early copper pyrometallurgy in the Aegean (Georgakopoulou 2005). Therefore, locating finds of this kind at Limenaria would suggest that the main smelting site, from which the slags under study derive, was not far from the excavated sectors. The numerous slagged ceramic sherds found in a pit at Akladi Cheiri have also been attributed to the lining of a nearby smelting hearth (Rehren et al. 2020; Radivojević, Roberts 2021.221).

As for the numerous lumps or fragments of iron ores from the same excavated layers, they most probably derive from local sources and could be explained as waste products of a final enrichment stage of a cupriferous iron ore (cf. Bassiakos, Catapotis 2006),



Fig. 8. Paradeisos, copper needles (© Ephorate of Antiquities of Kavala).

although they could also have been waste products of a different enrichment process for the extraction of a red or yellow pigment.

Taken altogether, the analytical results strongly suggest that the metalworkers exclusively utilized the local mineral resources for copper production. They demonstrated significant skill and succeeded in extracting metallic copper from the locally available polymetallic (and poor in cupriferous secondary ores) mineral resources.

Silver – lead

Limenaria

An unexpectedly early metal find, a small fragment of a silver pin, was discovered in a late Middle/early Late Neolithic horizon (second half of the 6th millennium BC) at the Lioudas plot (Fig. 10) (Papadopoulos, Malamidou 1997.836–837; Papadopoulos 2008.65, Fig. 3). Compositional data and microstructural examination have shown that the pin was shaped by hammering of a lump of silver. Its internal micro-morphology reveals that the silver is heterogeneous and consists of pure metal in which semi-translucent, minute pieces of slag are embedded. It is therefore presumed that the silver used to make the pin is not native, but the product of an extraction from argentiferous lead ore (Bassiakos 2012.210–211).

No silver finds are recorded from the 5th millennium, which is altogether less well documented on the site. However, there is firm evidence for silver extraction practice from a context of the early 4th millennium BC ('Final Neolithic'). Three litharge fragments coming from Markoulis plot belong to a piece of the 'shallow bowl' type, which derives from lead-silver extraction process (Fig. 11) (Papadopoulos 2008.66, Fig. 5.a-b, 68–69; Papadopoulos, Malamidou 2008.431). Compositional analysis and microstructural examination of these fragments suggest extraction and treatment of local Pb/Zn/Ag ores



Fig. 9. Limenaria, malachite bead (© Ephorate of Antiquities of Kavala).

for the production of silver artefacts (*Bassiakos 2012.208–210; Bassiakos et al. 2019*). The circular, discoid morphology and the chemical composition of this type of litharge fragments do not relate to the platy or tubular litharge deriving from the well-known cupellation process of historic times (*Bassiakos et al. 2013*). The process of early silver production that leaves behind as a by-product a shallow bowl of litharge has not been fully clarified yet, but similar examples are known from a number of sites from the end of the Final Neolithic/Early Bronze Age I (second half of the 4th millennium BC) in Mesogeia, Attica (*Kakavogianni et al. 2006.79; 2008; 2016.446–447, Fig. 15*), or neighbouring Eastern Mediterranean regions (e.g., East Anatolia – *Hess et al. 1998*; Syria – *Pernicka et al. 1998*). The dating of the litharge from Limenaria is supported by both the relative and absolute chronology (calibrated date between 3977 and 3789 BC), which makes it one of the earliest, if not the earliest piece, of firm evidence for the process of silver production in the Aegean.

Gold

Dimitra

Three golden artefacts were collected during the excavation in Dimitra, all from Late Neolithic contexts, more or less securely dated to the advanced 5th millennium BC. Two of them are beads: one collected from the upper part of the Neolithic deposits in trench I, disturbed by the LBA wall (*Grammenos 1997.49, Pl. 35.8*), the other from a lower level in the same trench (*ibid. Pl. II*). The third artefact, also from trench I, is described as a fishhook (*Fig. 12*) (*ibid. 51, Pl. 36.1*). The two beads have been examined with different methods (*Mirtsou et al. 1997*; see above). Analysis showed that they were made from gold (both contained 10% silver and 0.05% copper), which was cold hammered and annealed to produce ring-pearls with a wall thickness of 0.4mm and an inner diameter of 2 and 5mm (*ibid. 93*). The polished but unetched structure of both gold samples exhibited a number of quartz inclusions, so the artefacts were most probably made from native gold without melting. The etching with *aqua regia* solution revealed a single-phase structure with few twin-

ned crystals, indicating hammering followed by a reheating process.

Sitagroi

One gold bead was retrieved from the sieving of sediments in a small sounding (ZB) adjacent to the main stratigraphic sounding ZA, and it is assigned to phase III, i.e. the 5th millennium BC (*Renfrew, Slater 2003.319–320, Fig. 8.1.e, Pl. 8.4*). It belongs to the narrow cylindrical type. Examination under a microscope showed that the bead was made by beating a flat piece of metal around an inner core (which was not preserved). The ends were cut diagonally and had tooling marks near their edges, indicating that an attempt was made to weld them together. No further analysis has been conducted.

Dikili Tash

A small gold bead was found in the sector excavated by Theocharis in 1961 on the east slope of the tell (sector I), but was not included in the relevant preliminary report (*Koukouli, Rhomiopoulou 1992*). Comparison with artefacts from other sites and from the more recent excavations at Dikili Tash itself allows its dating to the late 5th millennium BC (*Tsirtsoni 2018.1276–1279, Fig. 2*). The piece of gold M206 kept at the Philippi Museum is probably the one mentioned by Michel L. Sfériadès (*1992. 113*) as coming from a coeval context from the excavations by Deshayes.

During the 2012–2013 excavations at Dikili Tash, four new gold objects were recovered, but this time from a perfectly secure context: a rich group of finished and half-finished ornaments, tools and raw materials found in the northern part of House 1, in sector 6 (see description and chronology above). Two of the gold ornaments, conventionally described as cylindrical beads, were flat strips of gold rolled around smaller beads from stone, while the third, which was smaller in size, was made with the same technique but contained no beads from a different material. The fourth was a twisted band forming a ring at one end (*Fig. 13*). The last type had not been attested yet in Northern Greece, but is known from a number of sites (mostly cemeteries) in the Balkans (e.g., at Varna, *Le premier or de l'humanité* 118–

119 (grave 4), 136–137 (grave 41), 149 (grave 97); see also *Tsirtsoni 2018.1280*). An SEM Examination by Michael Vavelidis (Department of Geology, Aristotle University of Thessaloniki) showed that all four are



Fig. 10. Limenaria, silver pin (© Ephorate of Antiquities of Kavala).

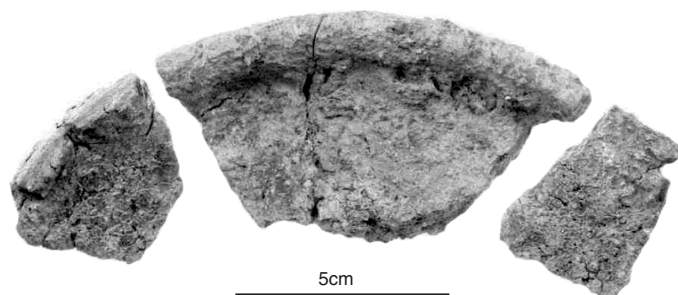


Fig. 11. Limenaria, litharge fragments of the 'shallow bowl' type (© Ephorate of Antiquities of Kavala).

made of gold with a low percentage of silver (4–9%). This points to native gold as a raw material. The surfaces show traces of hammering and rubbing (Fig. 14).

Although no direct connection has been made yet between the traces observed on the gold ornaments, or the other associated artefacts, and the tools found near them (one copper and one bone awl, one stone hammer/polisher, and several chipped-stone tools), it is clear that we see here an on-going process of manufacturing of jewels and decorative elements. Other craft activities were taking place in House 1 (e.g., decorating of pottery) together with more – and less – ordinary activities, such as storage, cooking and wine-making (Darcque et al. 2012–2013. 752–758; 2014.607–610; 2020.298–303).

The same house yielded another unexpected find connected with gold: a complete clay vessel, a bowl decorated with brown-on-cream painted decoration, which proved to display gold 'stains' at several spots of its exterior surface and in the inner part of the rim (Tsirtsoni 2018. 1313, Fig. 8; originally illustrated in Darcque et al. 2015.408, Fig. 7; Tsirtsoni 2016.285, Fig. 14) (Fig. 15). The bowl was standing on the floor a few meters to the south of the group of ornaments discussed above, and at short distance from the other group of ornaments that contained the copped beads (see above). The gold material seems to have been added after the original firing of the vessel, and indeed one wonders if this was done intentionally, as part of a restoration, or accidental-



Fig. 12. Dimitra, gold 'fish-hook' (© Ephorate of Antiquities of Serres).

ly, when someone working with gold paint on another object would have touched the vessel with his/her gold-stained fingers or tools.

However, intentional gold-painting on pottery is also attested at Dikili Tash. Evidence comes from a unique sherd found in sector 2, at the southern periphery of the tell: it is a fragment from the rim and handle of a small amphora, decorated with parallel oblique lines (Fig. 16). Its stratigraphical position is not secure, but according to its typological characteristics (shape, fabric, firing, surface finishing and decorative motifs) it can be dated with certainty to the years between 4800–4200 BC, and most probably after 4500 BC. This vessel type is indeed very common for this period both at Dikili Tash and the wider area, with decoration typically executed with graphite paint. Here, graphite has been replaced by gold, applied before firing (Tsirtsoni 2018. 1285–1288). Examples of such substitution of an ordinary colouring material (graphite) for a less ordinary, and presumably precious one (gold) are known so far with certainty only from the Varna necropolis (*Le premier or de l'humanité* 118–125: two vessels from grave 4). Two sherds from the site of Bubanj in Serbia (Stojić, Jocić 2006.154–155, Pl. 41.b; Bu-



Fig. 13. Dikili Tash, gold ornament from House 1 (© Dikili Tash Project-EFA).

latović et al. 2020.46, 53, Pl. 6/1) and two more from Krivodol and Chirpan in Bulgaria (personal communication of our colleagues Nadezhda Todorova and Petur Leshtakov) could be the only other candidates, if their dating was confirmed.

Discussion

Metal objects, albeit in small numbers, are attested in most sites investigated to some extent (Tab. 1). They are all either small tools (awls, pins, fishhooks, wires) or ornaments (beads, twisted bands, pen-

dants), and no objects with any substantial thickness of metal (like flat axes, shaft-hole axes or axe-adzes) have been found. Few of the metal finds came from a recognisable in situ context of use, with the exception of Dikili Tash, where a copper awl was found in House 1, sector 6, together with golden beads and a group of other finished and semi-finished elements of jewellery.

Although a complete metallurgical chain of production (*chaîne opératoire*), beginning with the raw material and ending with the finished product, was not successfully proven for any of the excavated sites, extraction from ores was attested for copper and silver. Metallurgical installations are not well documented in general either in the Balkans or Aegean. The recovery of a significant number of sherds or crucible fragments and associated copper deposits from phase III levels at Sitagroi is of considerable interest. They offer evidence of, at the least, copper melting on the site. Smelting in small shallow pit-shaped hearths opened in the clay-rich ground is probably also attested at Dikili Tash.

Copper production at Promachon-Topolnitsa is among the earliest in the Balkans, dating from the end of the 6th and the early 5th millennium BC, and is documented both by stratigraphy and absolute dating. There, the production is based on simple smelting of very pure malachite, producing no slag

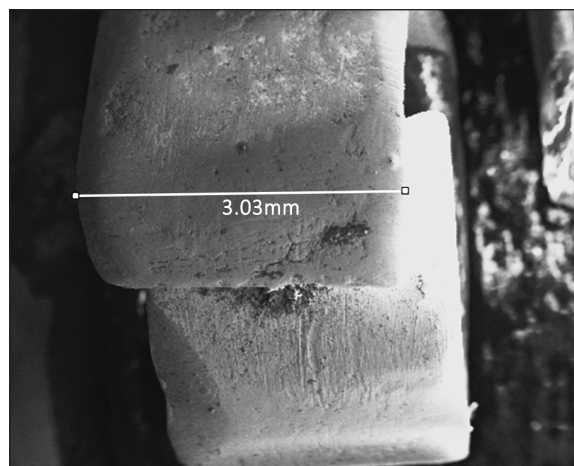


Fig. 14. Dikili Tash, SEM microphotograph of a gold bead from House 1 (© M. Vavelidis-Dikili Tash Project).

(Koukouli, Bassiakos 2002), a method consistent with what we know of the main metal-producing techniques of the Balkan hinterland, described as ‘slagless’ or ‘nearly slagless’ (Pernicka et al. 1997; Radivojević, Rehren 2016; Rehren et al. 2020; Radivojević, Roberts 2021).

The finds from Limenaria provide a more diversified picture. Items like the malachite bead show that the properties of the ores, which were collected from sources not far from the settlement, were already understood with regard to their use as pigments and

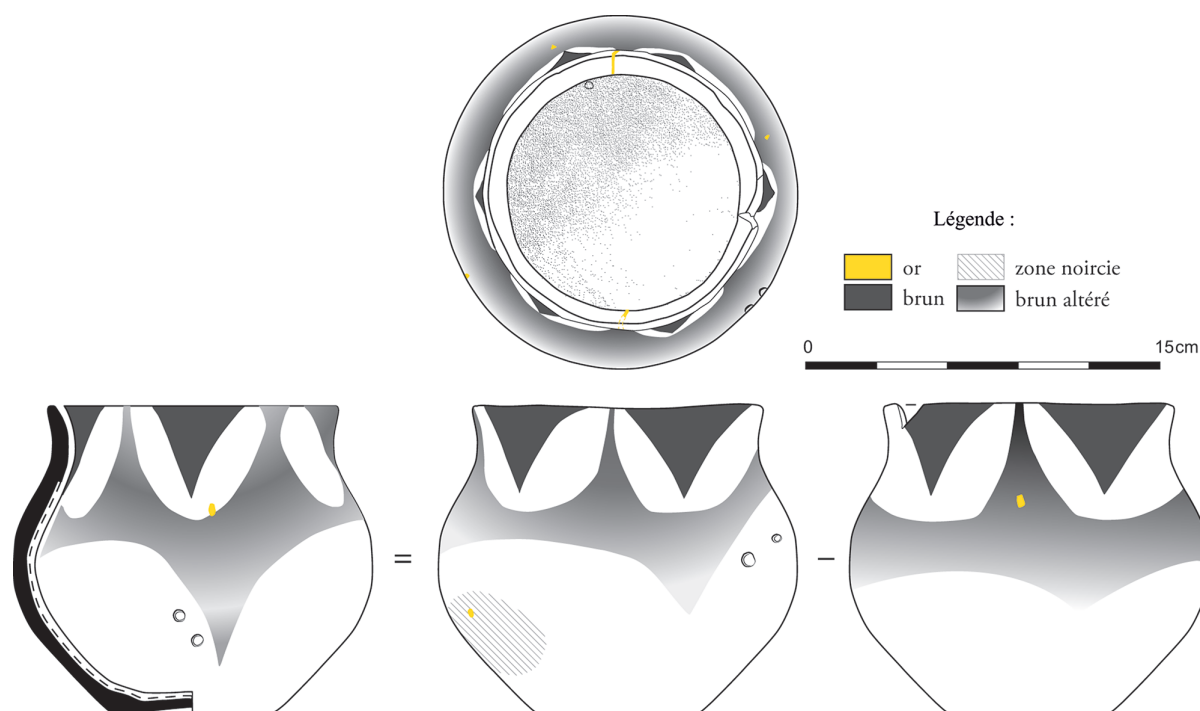


Fig. 15. Dikili Tash, brown-on-cream painted vessel from House 1 with gold ‘stains’ (drawing A. Ilioglou) (© Dikili Tash Project).

raw materials for other purposes. Given the long tradition among prehistoric communities on Thasos in the exploration of local minerals, it is not surprising that as early as the 5th millennium BC the inhabitants of Limenaria were trying to extract copper from the Thasian polymetallic ores (Nerantzis, Papadopoulos 2013; Bassiakos et al. 2019). The gaining of copper from such polymetallic raw materials required, apart from enriching the ore with copper minerals, a considerable level of skills and experience, which recalls pyro-metallurgical practices that are also attested later in the Aegean (Bassiakos, Catapotis 2006). On the basis of this evidence, Thasos seemed to be part of a technological tradition with ties to the Aegean and Asia Minor, rather than to the Balkans (Bassiakos et al. 2019:2754). The recent discovery of similar remains at the late 5th millennium BC Akladi Cheiri, in South-East Bulgaria (Rehren et al. 2020), invites us to reconsider the evolution patterns of early small-scale metallurgy in the wider area.

The fact that Limenaria, Thasos, provided firm evidence for extraction of silver in the early 4th millennium, and possibly earlier, is of great interest. Silver objects are not known in the Balkans before the end of 4th millennium (Nikolova 1999:303–308; Alexandrov 2009; 2018), whereas they appear earlier and are more common in the Aegean and southern Greece (e.g., Cyclades, Attica, Dimakopoulou 1998:64–65; Zachos 2010:89; see also Tsirtsoni 2014:295). Silver is rarely found as a native mineral element. Unlike gold, it is rarely found in significant amounts in placer deposits. The principal sources of silver are the ores of copper, copper-nickel, lead, and lead-zinc. Lead and silver extraction was attested through compositional data and microstructural examination of fragments of litharge and the silver pin found at Limenaria. The availability of resources locally was an important factor for the early exploitation of silver ores by Neolithic people. Nevertheless, given the fact that Thasos is open to southern influences in many regards, sharing of Aegean technological traditions must also have played an important role.

Gold objects are found in mainland sites of Eastern Macedonia only as beads in the form of hammered sheets rolled in a cylindrical shape, while hammered sheets of other shapes and ring-shaped pendants are absent, perhaps because objects of this type are more common in tombs (e.g., Aravissos, central Macedonia, Zachos 2010:89; 1996b:167, 339–340; Thessaly, *ibid.* 167, 339). All the above have close typological



Fig. 16. Dikili Tash, fragment of a gold-painted vessel from sector 2 (© Dikili Tash Project-EFA).

similarities with the ones from southeast Europe, which were found in large quantities in tombs, mainly in the extensive cemeteries of Bulgaria.

Gold paints in Dikili Tash show that the Late Neolithic inhabitants of the site were familiar with this metal. They were using it not only as precious material for jewellery making, but also in the *chaîne opératoire* of decorating pottery, in the same way as graphite and other minerals. This fact provides evidence for close relations with the pyrotechnology traditions of the Balkans.

Evidence for the extraction of metals from mining sites is lacking at the moment. Placer gold areas and alluvial gold washeries are difficult to distinguish and locate after centuries of erosional processes. Nevertheless, bearing in mind the abundance of mineralization in the area, one can assume that some prehistoric hard rock mining locations, which survived later mining activities, must exist. Therefore, and despite difficulties, locating traces of prehistoric mining and identifying the origin of the various raw materials and their relationship to the finished objects should be targeted, and ongoing research (Vaxevanopoulos 2017a; Vaxevanopoulos et al. 2021) is engaged in this.

Conclusion

Despite the relatively modest nature of the individual metal objects retrieved from Eastern Macedonian settlements, their widespread presence and coherent picture add significant data about the development of metal production in the North Aegean and more broadly in the Balkans. The fact that they come from secure stratigraphic contexts allows their dating to be firmly established, whereas their integration in considerably long sequences such as those

Settlement	Copper Artefacts	Silver Artefacts	Gold Artefacts	Ore and Metallurgical Findings	Dating	References
Promachon-Topolnitsa	malachite beads, 1 pendant, fragments			1 crucible, hollows with remains of burnt clay, slags	First half of the 5 th mill. BC	Koukoul-Chrysanthaki et al. 2007
Dimitra	21 beads, 1 fishhook, 5 parts of pins or wires, 11 undefined fragments		2 beads, 1 fishhook	polymetallic ore fragments	Second half of 6 th to 5 th mill. BC	Grammenos 1997; Mirtsou et al. 1997
Kryoneri	3 pins/awls, 2 fragments of rings			copper ore fragments	Mid-5 th mill. BC	Malamidou 1997; 2007; 2016
Sitagroi	1 roll-headed pin, 1 awl, five beads		1 bead	oval crucible, copper fragments of lumps, sherds with copper incrustation	Late 6 th to 5 th mill. BC	Renfrew, Slater 2003
Dikili Tash	4 beads, 10 awls or pins, 2 unstratified roll-headed pins, parts of 5 more pins or awls, 1 flat sheet		4 beads, 1 twisted band, 1 unidentified, 1 ceramic vessel with gold stains, 1 gold-painted sherd	copper oxides and copper fragments, 1 burnt sherd with copper traces, 1 sherd with copper incrustation, 1 clay mould, 1 clay-plastered pit	End of 6 th to late 5 th mill. BC	Séfériadès 1992; Koukoul, Rhomiopoulou 1992; Darque et al. 2015; 2020; Tsirtsoni 2016; 2018
Paradeisos	2 needles and fragments				Late 5 th mill. BC	Hellström 1987
Kastri Theologos (Thasos)	2 pins				Mid-5 th mill. BC	Koukoul 1992
Limenaria (Thasos)	1 malachite bead	1 pin		20 slags and fragments of hematite/limonite lumps containing malachite and azurite, 3 litharge fragments	End of 6 th to early 4 th mill. BC	Papadopoulos, Malamidou 1997; 2008; Papadopoulos 2008; 2012; Bassiakos 2012; Bassiakos et al. 2019

Tab. 1. Summary of the evidence of metal use in Greek Eastern Macedonia during the Neolithic period.

of Sitagroi and Dikili Tash enables us to follow their evolution.

The identification of smelting activities in several sites provides documented proof of pyrometallurgical copper extraction at the turn of the 6th to 5th millennium BC, while silver was definitely extracted at Limenaria since the 4th millennium BC. Attested are both the main techniques of production of metal items: ‘cold’ bead-making using established Neolithic technologies, and ‘hot’ copper smelting using developed metallurgical skills. Indeed, there is sound evidence for copper smelting being carried out using simple holes in the ground, followed by melting and casting elsewhere in the settlement.

Metal objects have not been found in significant numbers, and most tools and ornaments were still made of stone, bone, shell, or other organic materials. So, even though they might have been significant as prestige objects, metal artefacts do not seem to have been an essential component of the local material culture during the Neolithic. According to the data presented here, metal-working was only supplementary to other more important economic activities like agriculture and pastoralism, and second to other crafts, such as pottery or bone-tool manufacture. Considering the scale and importance of metallurgy during the 5th millennium BC, metal-working should be seen more as part of a bigger techno-complex and less as a main incentive for cultural development.

Nevertheless, new research programs should address the question of the geological origin of the various minerals in use, by identifying ancient mining relics and locations (*cf. Kunze et al. 2018; Krauss et al. 2020; esp. Kunze, Pernicka 2020b*). New analytical approaches, such as the LA ICP-MS method performed on metal objects, should play a more critical role in gai-

ning additional information about the trace elements included and their relation to ore mineralization. Moreover, coherent and comparable analytical results for Late Neolithic finds could further contribute to our interpretations about the technical and social dynamics of appropriation and early use of metals in the Balkans.

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