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The Dawn of Metallurgy at Chalcolithic Arslantepe: Metal Finds and Other Metallurgical Remains from Level VII

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Keywords

Arslantepe VII, Late Chalcolithic, lead production, slag, copper, copper high in arsenic and nickel, lead isotope analyses

Abstract

This paper deals with archaeometallurgical remains from period VII (Late Chalcolithic 3-4, 3900-3400 BC) of the settlement of Arslantepe (Malatya, Turkey). It aims at compiling early metallurgy (metallurgical artefacts, slags) by means of interdisciplinary scientific analysis. In contrast to later periods, the metallurgy of the Late Chalcolithic has as yet only been investigated to a limited degree. Trace elements and lead isotope analysis of metal artefacts and slags allowed for a reconstruction of provenances of raw materials in order to deal with trade networks. Lead and copper slags were analysed for texture and phase content to reconstruct metallurgical operations. Late Chalcolithic activities from Arslantepe level VII are compared with the metallurgy of the previous level, VIII, as well as of later periods (levels VI A, VI B). Various metal groups could be identified at Arslantepe VII, most notably copper, arsenic copper and arsenic-nickel copper. The slag samples show the smelting and processing of copper and lead within the settlement, while the lead slags may also indicate the extraction of silver. The differing compositions indicate the use of different raw material sources, which is also supported by the lead isotope analysis data.

Introduction

Archaeometallurgical research at Arslantepe dates back to the 1980s and is closely linked to the name of A. Palmieri. The German Mining Museum Bochum was in the fortunate position to collaborate and contribute to the research of metallurgy at Arslantepe. After his untimely death in 2006, further archaeometallurgical research tailed off. The archaeometallurgical investigation of period VII is intended to revive the successful collaboration with Missione archeologica italiana nell'Anatolia orientale (MAIAO), which has been directing the research at Arslantepe since 1961.

Period VII is largely unexplored in terms of metallurgy and metal finds, and scientific analyses of the deposits are lacking. This period does not show the earliest finds of metal objects, but first evidence of metallurgical practice at Arslantepe. This work aims at the chemical characterization and provenance analysis of the find material from period VII by means of lead isotope analysis and tries to shed light on the initial phase of metallurgy at Arslantepe. The question of the provenance of the ores at Arslantepe is an ongoing one and has not yet been answered satisfactorily. With further research, socio-economic and intercultural relations in the wider area of the upper Euphrates could also be addressed.

By way of synthesis, the obtained data will be embedded in the metallurgical tradition of the later phases at Arslantepe, especially with regard to the eventful transition period from the Late Chalcolithic (VI A) to the Early Bronze Age (VI B1-B2).

Geography

A brief geographical analysis of the region around Arslantepe is fundamental for understanding later discussions of the provenance of ores and metals from the south and north and thus for analysing intercultural relationships (Marshall, 2015). In the present case it is the region of the upper Euphrates, which covers its course up to the Taurus Mountains. The course of the Upper Euphrates is formed by the confluence of the Euphrates headwaters Murat and Karasu, which then flows further south towards Mesopotamia.

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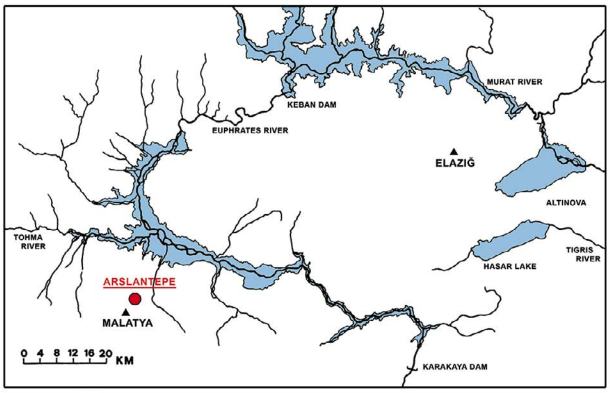


Figure 1. The geographic situation of the settlement of Arslantepe near the upper Euphrates, which is largely banked up today. Modified after Liberotti, et al. (2016).

The valleys of this region are characterised by fertile plains such as the Altınova and the Malatya Plains. The wide river valleys form settlement chambers of the region with several known settlement mounds, which testifies to the importance of these spaces in prehistoric times.

Arslantepe is located on the Malatya plain close to the present city of Malatya, just about 12 km southwest of the banks of the upper Euphrates (Figure 1) in eastern Anatolia. The Malatya plain currently has a semi-arid climate, however a hydrogeological system rich in streams and natural springs makes the area today, as in the past, attractive for agriculture and livestock (Marcolongo and Palmieri, 1983; Masi, et al., 2013). This plain is surrounded by mountains that were forested in prehistoric times. The hill of Arslantepe itself has an extension of 4.5 ha and a height of 30 m. It is the largest settlement mound in the Malatya plain, even though the mound is small compared to contemporaneous Mesopotamian settlements (Frangipane, 2011).

The Malatya plain is demarcated in the south by the Taurus Mountains, which are highlands that in turn separate it from the alluvial plains of northern Syro-Mesopotamia (Figure 2). The mountain range stretches from west to east and extends from south-west Anatolia to the Zagros Mountains. Further east lies the source of the Tigris. South-east of Arslantepe, the mountain range is intersected by both rivers. Both rivers flow through Mesopotamia and meet there before their confluence with the Persian Gulf. The Upper Euphrates is part of so-called Greater Mesopotamia, understood as 'land between rivers' in its broadest sense, that is including all the regions

Figure 2. The physical geography of eastern Anatolia, showing the mountainous regions between the Central and the Southeastern Taurus and the Lesser Caucasus. South of the Southeastern Taurus there follows the plain of upper Mesopotamia. Pointed are some cities mentioned in the text.

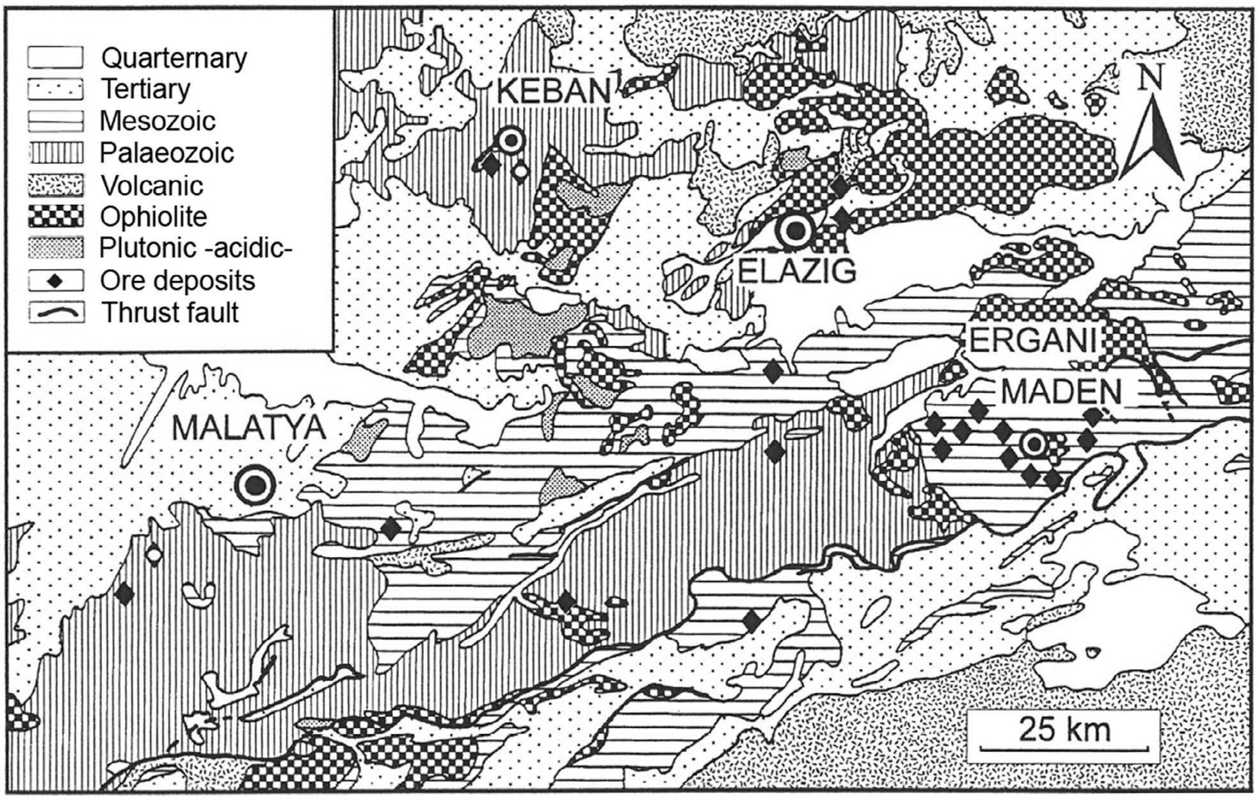
that gravitate to the Tigris and Euphrates and the surrounding mountainous areas (especially those of eastern Anatolia) strongly related to them (Frangipane, 1996). The eastern tributary of the Euphrates is the Murat. It springs in the mountains to the west of Mount Ararat and the south of the Araxes valley. The western headwater stream Karasu has its origin on the Erzurum plateau north of today's provincial capital. Today, the point of confluence of the tributaries and thus the starting point of the Euphrates is the Keban reservoir, which was built as the first dam along the Euphrates and covers an area of 675 km².

Geology and ore deposits

The geology of Anatolia with its unusually numerous, rich ore deposits has been studied intensively (summary in Yiğit, 2009; Piraijno, et al., 2019). Tectonically, Anatolia can be divided into the Pontides, Anatolides, Taurides, and the border folds (Ketin, 1966; Okay, 2008). The upfolding of these units took place during the Alpine orogeny. The Anatolides and Taurides form the southern part of the Anatolian peninsula and border the Pontides to the north. This border is characterised by the Izmir-Ankara-Erzincan Suture. In the south/south-east, the Bitlis-Zagros-Suture is the border to the Arabian Plate. The collision of the two tectonic units occurred in the upper Miocene (11-5 Ma) and the Bitlis-Zagros Suture represents a relic of the southern arm of the Neotethyan Sea.

Anatolia is characterised by a complex geology linked to the development of the Tethys Sea. The formation as a landmass occurred in the Oligocene (34-23 Ma) after the last closure of the Tethys and the collision of the individual terranes. Anatolia is part of the "Tethyan Eurasian Metallogenic Belt" (TEMB; Jankovic, 1977). This is a global metallogenic belt formed by the obduction of ocean crust during the closure of the Tethys. Most of the ore deposits in the TEMB are dominated by copper. The TEMB stretches between the African, Arabian, and Indian Plates on the southern side and the Eurasian Plate on the northern side across about 10,000 km. Throughout the formation processes of the TEMB, terranes of accretions were formed. Remains of ophiolitic suites, mainly from the upper Cretaceous (100-60 Ma), give testimony to former oceans. Ophiolites are associations of mafic and ultramafic rocks, parts of the oceanic incrustation and the upper mantle of the globe (Nebert, Brosch and Morth, 1986). For Anatolia, this created a west-east running ophiolite-bearing belt that extends from Cyprus mainly over parts of the Zagros Mountains to Oman.

Figure 3. A geological map of south-eastern Anatolia, showing the region around Malatya, Elazığ, Keban, and Ergani Maden. The two rhombs with white circles at Keban and south-west of Malatya are lead-silver deposits, the filled rhombs are copper deposits. Note the large number of filled rhombs near the copper district of Ergani Maden. From Hess (1998).



In the region around Arslantepe there are a number of ore deposits, either located in ophiolites or in Palaeozoic-Precambrian metamorphic rocks (Figure 3). Arslantepe is located on the border between the Anatolides/Taurides and the Arabian Plate. The Bitlis-Zagros Suture zone lies about 40 km south of the settlement. North of this border there are three massifs, formed by nappes of Palaeozoic-Precambrian sediments. From west to east, these are the Pütürge, Keban and Bitlis massifs. The Malatya Plateau is composed of Neogene to Quaternary stratified sediments. Northeast of that plateau there are acidic intrusive rocks of Mesozoic plutonic activity.

The ore deposits of the Keban region are located approx. 65 km north-east of Malatya. Mining for lead and silver was conducted in modern times until 1983. Seeliger, et al. (1985) report mining traces from prehistoric times. Comprehensive references are described by Wagner, et al. (1989). They report more than 100 probably ancient adits and finds of stone tools. Based on such archaeological finds and findings, they were able to date activities to the Bronze Age, the Iron Age and the Ottoman period. During another survey, Hess (1998) was able to confirm the dating of prehistoric mining activities at Zeytindağ and Zeryan Dere. The Keban deposit district displays a variety of different mineralisations, including Dere Baca (Pb-Ag-Zn-deposits), Siftil Tepe, Zeryan Dere (Cu-Zn-Pb-deposits) and Zeytindağ (haematite-magnetite-deposits). From today's point of view, many of them are not economically exploitable deposits, but they may well have been important in prehistoric times. Limestone schist and marbles occur as host rocks of mineralisations. The district of ore deposits east of the Euphrates shows clear evidence of karstification (e. g. Zeytindağ; Hanelçi and Çelebi, 2015). At Dere Baca, mineralisations are formed at the boundary between shist and limestone. Here, main minerals are pyrite (FeS_2), sphalerite (ZnS), argentiferous galenite (PbS) and subordinated argentiferous loellingite (FeAsS) and marcasite (FeS_2). At Zeryan Dere and Dere Baca there are area sulfidic stockwork mineralisations with chalcopyrite ($CuFeS_2$) (Hess, 1998).

In the southeast Anatolian ophiolite belt near the Bitlis-Zagros Suture, there are numerous copper deposits. The most well-known and by far the largest one is Ergani Maden. It consists of several single deposits. The main mineralisation is Anayatak, others are Kısabekir, Mızırtepe, Weiss and Hacan. The dominant metal is copper, predominantly bound to pyrite-chalcopyrite mineralisations. The district represents the largest volcanic massive sulphide copper deposit in the eastern Taurus. Geologically speaking, it is of the Cyprus type (summary in Seeliger, et al., 1985). Copper ore was extracted from the main Anayatak mineralisation in the form of open-cast mining from 1930 - 1993; it is now considered exhausted. Tylecote (1970) noted native copper and "ancient shafts" at Ergani-Maden. Çağatay (1968) reports an extensive gossan at the Weiss-mineralisation. He distinguished four mineralogical parageneses: 1. Chalcopyrite, digenite, covellite and limonite; 2. Limonite, cuprite and tenorite; 3. Limonite with siliceous gangue; 4. Limonite, rutile, anatase and spinel. Besides of that, chromite was detected frequently in the Weiss occurrence.

Another Cyprus-type copper deposit is Siirt-Madenköy, east of Ergani Maden, also close to the Bitlis-Zagros Suture. There was no overprinting by modern mining in this area. Copper ores from this deposit contain some nickel as well (Akıncı, 2009).

Southwest of Malatya lies the Görgü deposit (Cafana). It is a lead-zinc (silver) mineralisation in volcanic and metamorphic rocks. Veins in andesitic host rock are common. Main minerals are galenite (PbS), sphalerite (ZnS), pyrite/marcasite (FeS_2) and smithsonite ($ZnCO_3$) (Kalender, et al., 2009). Remains of Pre-Roman mining activities were found during investigations, and Bronze Age settlement and smelting traces were discovered in the vicinity of the deposit (Palmieri, et al., 1996; Palmieri, Hauptmann and Sertok, 1996).

In the region around Arslantepe there are numerous smaller occurrences of ore (Palmieri, Sertok and Chernyk, 1993; Wagner and Öztunalı, 2003). A lead-copper occurrence, which also shows traces of prehistoric exploitation, is located in the Munzur Mountains (East Taurus) near the village of Mamlis in the Tunceli province. This province lies to the north of Elazığ. Here, the mineralisation is a porphyritic copper deposit with chalcopyrite, galenite and fahlore impregnated in granodiorite (Wagner, et al., 1989). Another attempt of a survey was not possible until now.

Two large massive sulphidic deposits are located in the Pontides, one centrally at Küre and the other further east at Murgul. On the southern border of Anatolia, a small copper mineralisation exists near the village of Süğüt in the Amanos Mountains. The outcrop is probably an oxidation zone of a skarn deposit. Pre-modern mining traces there could not be dated (Wagner and Öztunalı, 2003). Two Anatolian tin deposits exploited in prehistoric times were discovered near Kestel and Hisarcık (Yener, 1989; Yener and Vandiver, 1993a; 1993b; Yalçın and Özbal, 2009).

A copper deposit of northern Central Anatolia is Derekutuğun. Mining activities there were traced back to the Early Bronze Age (first half of 3rd millennium BC). The exploitation was focused primarily on native copper, which is widely distributed there (Yalçın and İpek, 2016).

Archaeological background: History of the settlement, chronology

The chronological focus of this work is the Late Chalcolithic development of Arslantepe. This period roughly covers the second half of the 5th to the 2nd half of the 4th millennium BC. In terms of cultural history, it represents the phase after the first urban agglomerations in the Mesopotamian lowlands. During the late 4th and early 3rd millennium BC the formation of urban centres and the development of hierarchically structured societies continued (H. Hauptmann, 2000). In the following, Rothman´s (2001) chronological division of the Late Chalcolithic is applied. For the updated radiocarbon ages of the Arslantepe occupational periods the authors follow the contribution by Vignola, et al. (2019).

The settlement history of Arslantepe most likely begins as early as in the 6th millennium BC (Frangipane, 2011; Balossi Restelli, 2012a, p.236). A detailed sequence for a large area of the mound was provided by Italian research, especially for the prehistoric periods from the Late Chalcolithic (LC) until the Middle Bronze Age (MBA) (end of 5th to beginning of 2nd millennium BC) (Di Nocera, 2000a, p.340; 2000b).

The Late Chalcolithic phase includes periods VIII, VII and VI A, while the Early Bronze Age (EBA) includes layers VI B to VI D (Table 1). The sequence of the relevant layers will be explained here very briefly.

Arslantepe VIII marks the oldest settlement phase that can be identified to date and dates to the end of the 5th millennium (4700-3900 BC, LC 1-2). Traces of period VIII lie in the western area of the settlement mound approx. north and northwest of the known remains of Arslantepe VII. The architectural findings show buildings with a clear domestic character and up to eight construction phases (Balossi Restelli, 2012b, pp.44, Fig.2; 45). The ceramic repertoire of the period represents regional ceramic traditions whose comparisons can be found in settlements of Upper Mesopotamia such as Oylum Höyük, Korucu Tepe, Tell Hammam (Balossi Restelli, 2012a). A clear settlement continuity from Arslantepe VIII to VII cannot be observed, despite individual typological parallels (Balossi Restelli, 2012a, pp.244-245).

Table 1. A chronology of the Late Chalcolithic and Early Bronze Age periods at Arslantepe (modified after Frangipane, 2018; updated chronological data from Vignola, et al., 2019)

Greater Mesopotamia		Arslantepe periods
LC 1-2	4700 – 3900 BC	VIII
LC 3	3900 – 3600 BC	VII
LC 4	3600 – 3400 BC	VII
LC 5	3400 – 3200 BC	VI A
EB I	3200 – 3100 BC	VI B1
EB I	3100 – 2800 BC	VI B2

In period VII (3900-3400 BC, LC 3-4), signs of a first centralisation and hierarchisation of society could be detected. Remains of this phase were discovered in the west and on the north-eastern edge of the mound. The settlement seems to extend over the entire hill, with a clearly structured layout. All in all, a continuous development without major breaks can be seen over the large time span of approx. 500 years (Balossi Restelli, 2019; Frangipane, 2011; 2018, p.26).

During the investigations in the north-east several residential buildings were uncovered, which, with their ovens and floor burials, show parallels to period VIII (Frangipane, 2011, p.972). In the western area, "temple C" (Figure 4), built in the last phase of layer VII, stands out. It was a free standing, three-aisled building whose layout is based on Mesopotamian architecture but also features wall paintings that follow local traditions from the Ubaid period (Balossi Restelli, 2012b, p.45). In the so-called temple, cretulae with seal impressions and mass-produced bowls were found grouped or stacked. Both findings indicate practices of accumulation and (re)distribution and are indicators of a centralised administration (Frangipane, 2018, pp.30-33).

So-called temple D (Figure 4), which directly adjoins "temple C" to the east, already dates from the earlier phase 1b. Despite their earlier construction, both buildings existed in parallel in the final phase of period VII and shared common functions, as suggested by finds of cretulae and bowls (Frangipane, 2018, p.28). Situated to the west, on a higher part of the hill, lie the so-called elite residences. Especially their size and the wall decoration with white plaster and sometimes wall paintings set these buildings apart from the ordinary dwellings (Frangipane, 2011, p.972).

The pottery repertoire of period VII, despite southern influences, continues the local tradition of the Malatya Plain from period VIII (Balossi Restelli, 2019). Connections are found in the Amuq Plain and the rest of the areas west of the Euphrates. Nevertheless, steps of mass production, which follows the Mesopotamian trend, are discernible in the manufacturing process (Guarino, 2014, p.92). The simple bowls at Arslantepe also represent mass-produced goods, but they differ from their Mesopotamian counterparts, the "bevelled rim bowls" (Guarino, 2014, pp.125-126).

Arslantepe VI A (3400-3200 BC, LC 5) represents the last Late Chalcolithic phase and simultaneously the transition to the Early Bronze Age. The settlement devel-

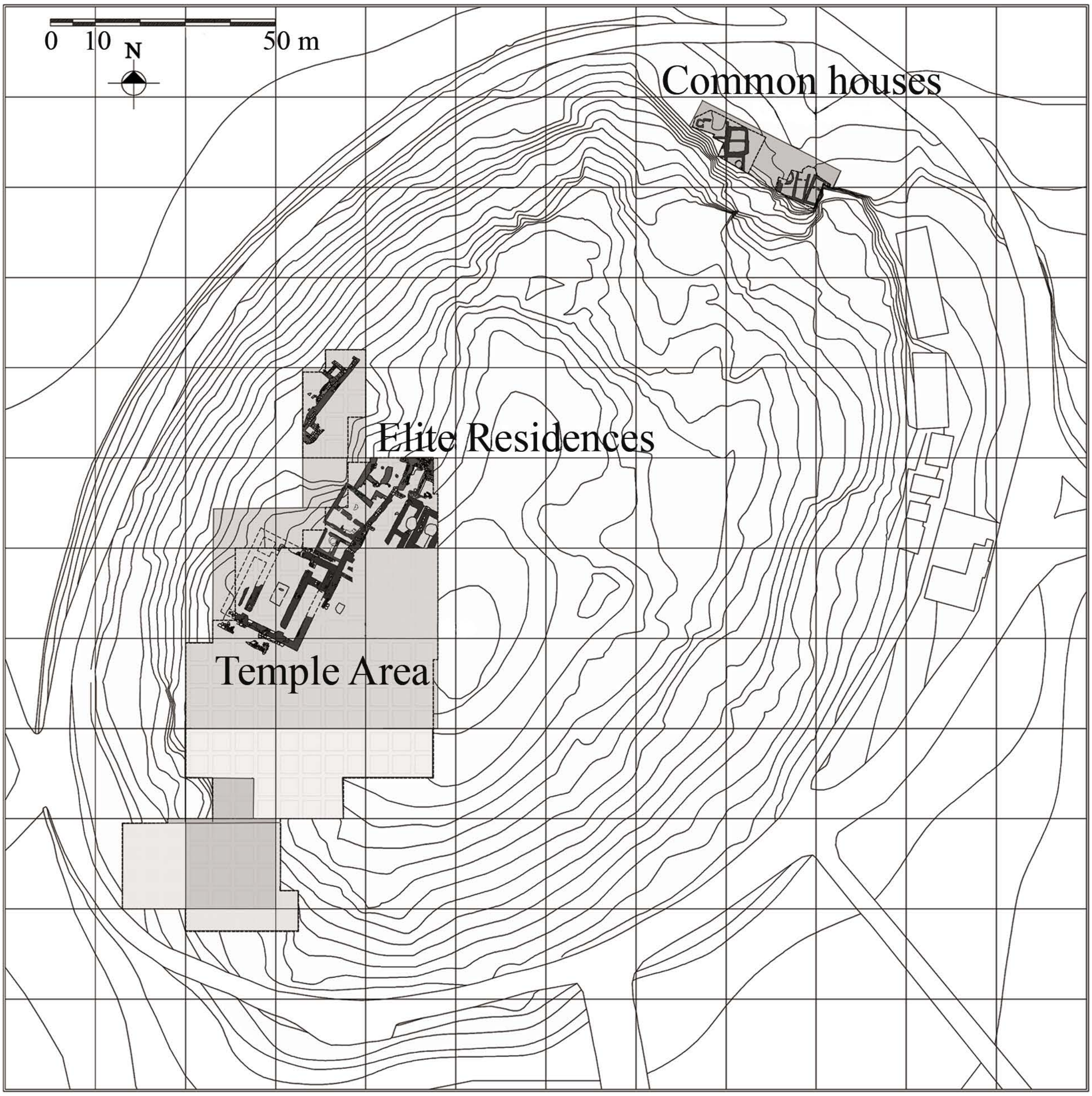


Figure 4. Arslantepe, period VII. Plan of the mound with the excavated areas and building structures from level VII. Modified after MAIAO (Frangipane, 2018, Fig.15a, p.27).

ops into a complex centre with administrative, political and religious/cultic functions. What is likely the most important building complex (palace) is characterised by storage rooms, wall paintings and a so-called audience hall (Frangipane, 2018, pp.33-37). Accumulation and redistribution seemed to be a central task. This is indicated by the numerous finds of cretulae and mass-produced clay bowls. In addition, evidence of a wider range of metallurgical activities and exchange is apparent. This also underlines the unusual finding of the "palace hoard", consisting of swords, spears and a quadruple spiral made of an arsenic-copper alloy (Frangipane and Palmieri, 1983, pp.394-407; Di Nocera, 2013, p.117). The end of this layer is marked by a major fire event that led not only to the destruction of the central structures but also to the abandonment of the settlement's economic and political system (Frangipane, 2012, pp.237-239).

The collapse of the old settlement system is followed by period VI B1 (3200-3100 BC, EB I). Recent investigations show a direct chronological sequence from Arslantepe VI A to VI B1 (Palumbi, et al., 2017, pp.118-119, Fig.30). With its lightweight wattle-and-daub architecture, the settlement has an obvious seasonal character. In addition, a central assembly building (No. 36) in mud-brick architecture has also survived, where common drinking and eating practices have been attested (Palumbi, et al., 2017, p.92). In addition, findings attributed to the Transcaucasus and the Kura-Araxes phenomena are increasing (Di Nocera, 2013, p.121). These influences can also be observed in the so-called Royal Tomb (T1), dating to this period or possibly to the beginning of Arslantepe VI B2 (Frangipane, et al., 2001; Frangipane and Erdal, 2020). Arslantepe VI B1 ended with a fire that destroyed large parts of the settlement.

In the last phase of EBA I, Arslantepe VI B2 (3100-2800 BC, EB I) the hill develops into a dense settlement consisting of small houses, yards and streets. An astonishing feature is a massive mud-brick wall that limits the upper part of the hill, while the actual settlement with various workplaces, including smelting sites, lies on the slope (Di Nocera, 2013, pp.124-129). Due to a fire at the end of VI B2, there is another break in the settlement development (Frangipane, 2011, p.982).

Archaeometallurgical finds

As of today, some metal artefacts have been found in Arslantepe VIII, but there is no evidence of metallurgical activity. In 2017, the archaeometallurgical finds of

Figure 5. Distribution of archaeometallurgical finds of phase 1a in the area of temples C and D, Arslantepe VII. Modified after MAIAO.

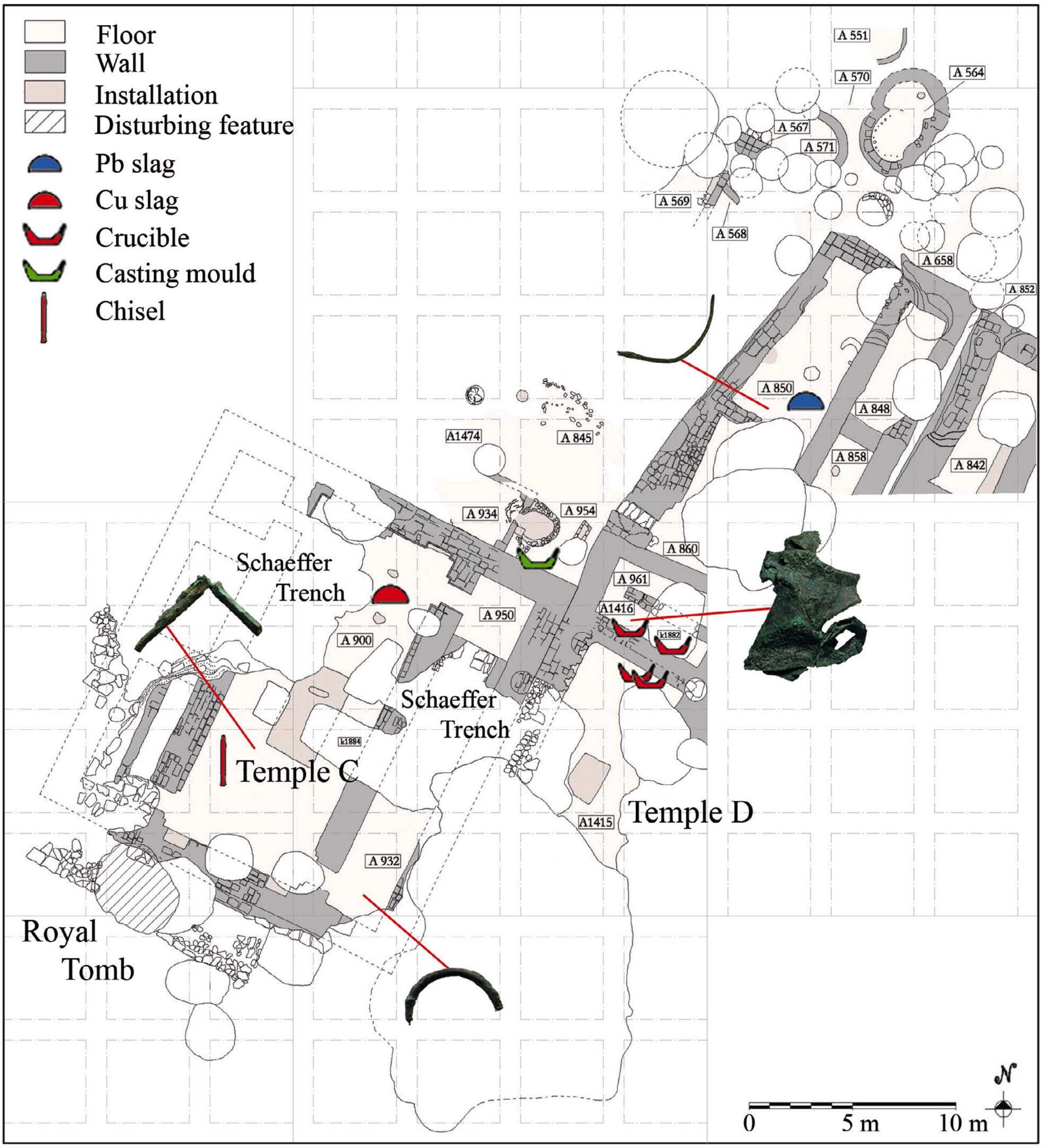


Figure 6. Selection of metal artefacts found in the course of the excavation of Arslantepe VII. Figure a: Needlepoint (146/17); b: Fragment of a ring (1145/11, 149/17); c: Small ingot (154/17); d: Copper awl (20/04); e: Knifepoint (21/04); f: Awl (151/17); g: Needle (835/12); h: Fragment of a deformed vessel (145/17). Details on these metal objects are compiled in Table 2. Photo: Th. Stöllner, Deutsches Bergbau-Museum Bochum/Ruhr-Universität Bochum.

the following period VII included slags, crucibles, casting moulds and ores, as well as some metal artefacts. Of the metal artefacts from Arslantepe VII, only seven awl and needle fragments and one piece of sheet metal have already been published (Di Nocera, 2013, p.113). Archaeometallurgical finds from the later phases VIA - VIB2 have been analysed by Palmieri, et al. (1996; 1999; Palmieri, Hauptmann and Sertok, 1996) and in an unpublished dissertation by Hess (1998).

The distribution of some metal artefacts and other metallurgical remains found in Arslantepe VII are shown in Figure 5. Unfortunately, no metal workshops were discovered, but individual, smaller concentrations of metal objects and slags can be observed in certain spatial contexts. They are distributed from the central buildings (temples C and D) to the elite houses, to the residential houses in the north-east of Arslantepe. At the same time, the stratigraphic distribution reflects a certain temporal depth, ranging from phase 3 ("elite residences") to phase 1 (temple C) of period VII.

Metal artefacts

Of the 22 metal artefacts in the excavation depot at Arslantepe and in the Malatya Museum, 15 finds were sampled. They consist mainly of small tools (awls, needles, chisels). They are compiled in Figure 6 and Table 2.

The only larger metal object is a deformed metal vessel (Figure 6h, sample 145/17) which may have been a funnel. At least a spout and a loop-shaped handle are recognisable. Along the spout, a seam is visible where it was connected to the body of the vessel. The manufac-

Table 2. List of analysed samples of metal artefacts and slags from Arslantepe, level VII.

Sample no.	Object	Find context
Metal artefacts		
20/04	Pricker	D5(7) A924 rM RI 1
21/04	Knife point	D5(3) A968 rM 1 RI 2
215/10	Copper fragment	C3(E4) VII d 2av '70
405/10	Needle	D5(10) K1733 2a
406/10	Needle	D5(10) K1733 2a
1194/11	Awl	C3(e1) VII h
1195/11	Ring fragment	A932 Y2
835/12	Needle	E6(9) A850 rM 4 RI 23
145/17	Deformed, spouted vessel	D7 (3)(4) A1416 5 RI 373/2015
146/17	Needle point	D6(8) 6-9 sotto A562 M1
149/17	Ring fragment	A932 Y2
151/17	Pricker	A900 rP
153/17	Crushed copper	D5(6) K1748 1a R.I. 1
154/17	Copper ingot	D7(13) A828 E1
217/17	Copper fragment	C7(6)
Slag		
38/02	Lead slag	D7(3) A853 1b
39/02	Lead slag	D7(3) A853 2b
40/02	Copper slag	D7(3) A853 2b
44/02	Lead slag	D7(3) A853 rP1
53/03	Lead slag	D7(3) A853 rP1
94/02	Lead slag	D5(3-7) 8e
98/03	Lead slag	D5(3-7) 8e
222/10	Copper slag	C3(E4) VI p. 17/8/1970
1145/11	Copper slag	A900 rM3
874/12	Lead slag	A850 E1 G1
148/15	Crucible slag	D7(7) A1415 2 Ri 29/15
152/17	Lead slag	E6(9) A850 e1 g1
199/17	Copper slag	D7(13) A828 e4 2h
148/17	Crucible slag	D7(8) A1415 3a R.I. 414/15 and A1415 1a R.I. 20/15
156/17	Crucible slag	D7(16) A1424 beta1a R.I. 61

turing could not be examined in detail. The individual parts of the object were made of a copper sheet about 1-2 mm thick. The find, therefore, shows a developed processing of hammered sheet metal.

A tongue-shaped metal fragment (Figure 6e, sample 21/04) is interpreted as a possible piece of a small knife blade. The metal tongue has a symmetrical, elongated shape, and the well-preserved tip is rounded. At the other end of the piece there is a fracture. The dimensions are ca. 4.5 cm in length and approx. 2 cm in width. The surface is partially more heavily corroded and chipped.

One half of a fragmented ring (Figure 6b, sample 149/17 and 1195/11) is preserved. It had a diameter of slightly less than 2 cm. The thickness of the ring is 2-3 mm. No decorations are visible.

One of the finds, referred to as a copper ingot, is a nearly 3 cm long and almost rectangular piece of copper (Figure 6c, sample 154/17), but its function is unclear. It could represent a semi-finished product, a small ingot or a piece of raw copper for making an awl or something similar. The rough shape resembles that of the finished awls or chisels of this period VII, which also show a square cross-section. Furthermore, a function as a weight would be conceivable, but possible markings on the piece are missing.

Awls dominate the metallurgical finds. However, no uniform shape can be discerned; the length and width of the pieces vary. The finds sometimes have one, sometimes two points and sometimes one point and one cutting edge. The design of these working surfaces varies. Thus, these small tools were multifunctional. As mentioned above, there is evidence of shafted use. Some show clear edges and a uniform cross-section, while others are of a more amorphous nature. This could have been caused by corrosion during soil storage, or even by reworking. Especially pure copper is very ductile, the tips or cutting edges blunt with steady work and have to be re-hammered.

An example of a filigree object is a needle (approximately 13 cm, diameter 0.2 cm) with a spiral ornamentation (sample 228/90, unpublished data of Palmieri). The needle is divided into three sections. The tip is made in a round, the upper half in a square cross-section. Here there is a spiral with 6 loops, which in turn is made of a round wire.

One of the largest and at the same time heaviest objects among the tools is a chisel (sample 169/98, unpublished data of Palmieri) (Length approximately 8 cm, width 0.5-1 cm, weight 21 g). On one side there is a slightly widened cutting edge (1 cm), on the other side the end piece tapers to 0.5 cm. The approximately square cross-section is 0.2 x 1 cm at the cutting edge and approx. 0.5 x 0.7 cm in the back third. A striking burr ("Schlagbart") is visible at the end of the chisel, which indicates that it was used with percussive tools, probably made of wood or stone.

Slags

Twelve slags and three samples from slagged crucibles were analysed. The nut-sized slag fragments were found distributed in the area of temples C and D and the residential buildings in the north-east of the mound. Earlier, Hess (1998) analysed a few slags from level VII for their mineralogical composition. Interestingly, among them there was also a complete palm-sized slag cake (TR8/1; Hess, 1998, pp.105-106). Such slag cakes had not been known to us until then.

Five slag fragments were found in room A853, dated to phase 2, level VII. A kiln with a diameter of 2 m is known to have existed in the immediate vicinity. A hearth or oven is again known from the more recent phase 1. For both contexts, no evidence of high-temperature processes or other indications of metallurgical activity is described. The features can rather be attributed to food preparation in a domestic context or later in the context of redistribution in temple C or D (Guarino, 2014, pp.210-211, 240). The distribution of the slag finds does not suggest intensively used workplaces, but possibly a nearby smelting of ores and further processing of metal. As the small pieces of slag were not securely in situ, no definitive conclusions can be drawn.

Crucibles

A total of seven crucible fragments were recovered in period VII. Five of them were selected for later sampling. They were recovered in the context of temple D but originate from backfill layers and were not found in situ (Frangipane, et al., 2019, pp.26-27). Probably all crucibles are of a tall cup shape with a rounded base, the inner diameter is 12 cm. The wall thickness of the crucibles varies; at their base it can reach several centimetres. Hole-like mouths in the lower parts of the vessels (diameter 3-4 cm) could have been used either for air supply, for casting or for handling when hot, using a wooden dowel. They thus resemble the crucibles of the Sialk III.5-6 period from Arisman and the surrounding area as well as the older crucibles from the known workshop of Ghabristan (see Pernicka, et al., 2011; Nezafati, et al., 2021; Stöllner, 2021, Fig.3, 1-2). Due to the size and shape of the crucibles, any pouring from the crucible seems difficult to accomplish. Hauptmann, Frangipane and Di Nocera (in prep.) are working on the thermodynamics and composition of the material of these crucibles to clarify their function.

Two more crucibles from Arslantepe, level VII, were presented by Hess (1998). Base and wall are preserved on one. Compared to the crucibles just mentioned above, this find is rather small. The other fragment is a flat sherd. The first fragment was covered by a thin slag line from which a metal droplet of a CuAsNi alloy was recovered. This feature is attributed to the (re-)melting and fusing of metal (Hess, 1998, pp.106-107, 118).

Figure 7. "Casting mould". Arslantepe, room D7(3) A 954 (see Figure 5). More probably, this trough-shaped crucible without handles was made for the re-melting of tiny copper prills to larger units, as suggested by Hauptmann (2020) for the chalcolithic copper production in Switzerland. Photo: Th. Stöllner, Deutsches Bergbau-Museum Bochum/Ruhr-Universität Bochum.

„Casting moulds"

Fragments of three open, single-shell ceramic vessels were found. Two are from around the elite residences, while one other fragment was found in the area near temples C and D (Figure 7). But no metal residues or slag incrustations could be observed. However, their size clearly exceeds that of the metal artefacts found. So far, no larger, massive metal artefacts, such as axes, are known from this period. It is not certain whether the vessel shown in Figure 7 was not also intended for the re-melting of tiny copper prills. Such a process appears sensible to be useful for recovering mechanically extracted metal residues from semi-liquefied slags in order to fuse them into larger units. Even though direct evidence for the mechanical processing of slag, as documented in phase VI B2, is lacking in period VII (Hauptmann, et al., 2022).

Based on macroscopic observation, the three "moulds" differ in clay composition and firing. In addition, a coating of slurry is partly visible. Like in the case of other archaeometallurgical finds, there are no indications of any associated workshops.

Ores

Some pieces of lead ores (galenite, PbS) and their oxidation products cerussite, $Pb(CO_3)$ and anglesite ($Pb(SO_4)$), copper ores (cuprite, Cu_2O, malachite ($Cu_2(CO_3)(OH)_2$), and polymetallic copper ores (fahlores, e.g. tetrahedrite, $Cu_{12}Sb_4S_{13}$, or tennantite, $Cu_{12}As_4S_{13}$) were found. Some of them were discussed in Hauptmann, et al. (2002) and Hess (1998).

Analytical investigations methods

The samples of metal objects and slags examined in this study were sporadically taken by archaeologists at different times. In some cases, samples were taken in only very small quantities, so that chemical and isotope analyses could not be carried out. Pieces of slag could be transported to the German Mining Museum Bochum (DBM) for further processing. Analytical investigations were carried out at the research laboratory of the Museum.

Mineral phases were determined by X-ray diffractometry and by optical and scanning electron microscopy on thin and polished sections. Bulk chemical analyses (Tables 3 and 4: Main, minor and trace elements) of metal and slag samples were carried out by mass spectroscopy (ICP-SFMS Thermo Fisher Scientific Element XR). Detection limit of the spectrometer is within the ultratrace range (<1 ppm).

Analyses of lead isotope ratios (abbreviated as LIA) were carried out at the Frankfurt Isotope & Element Research Center of Goethe University Frankfurt (FIERCE) by help of a Multicollector-ICPMS (Neptune Plus, Thermo Fisher Scientific). LIA measures ^{204}Pb and the three radiogenic isotopes ^{206}Pb, ^{207}Pb and ^{208}Pb. For our samples, standard errors and standard deviations are shown in the results in Table 5. Older data from the literature were partly measured by thermionic mass spectrometer (TIMS) and have a larger error. These errors of 0.1 % are also indicated in the diagrams by an error cross.

Metal artefacts

Chemical analyses

The chemical composition of 15 metal samples was measured (Tables 3 and 4). The main elements show that all objects are either copper or copper-based alloys with arsenic, nickel, lead, sulphur, and tin. The alloy components are all in the lower percentage range. The element contents do not add up to 100 wt.%, due to corrosion.

The As/Ni correlation diagram (Figure 8) shows possibly three to four different material groups, assuming that such a grouping is possible at all using this limited sample number, especially since there is only one sample for CuPb. Analytically determined copper varieties based on trace elements:

1. Pure copper low in arsenic (0.01 – 0.18 wt.%) and nickel (0.001 – 0.47 wt.%). The sum total of the minor elements (As, Ni, Fe, S) is below 1 wt.% for each of the five samples. Samples 1195/11 and 149/17 are samples from the same ring fragment. The distribution of the elements Pb, Ag, Sb, As, Bi, Co and Ni reveals a uniform compositional trend despite their

Table 3. Bulk composition, main and minor element analyses of metal artefacts from Arslantepe VII. Values are given in weight-percent (wt.%). For an explanation of the samples, see Table 2.

Sample no.	Cu	As	Ni	Pb	Sn	Total
20/04	83.9	1.39	2.50	0.08	0.002	89
21/04	93.2	3.53	0.09	0.001	0.002	97
215/10	83.2	1.03	0.85	0.15	0.005	85
405/10	92.6	2.12	0.02	0.02	0.003	95
406/10	92.2	2.21	0.02	0.03	0.002	94
1194/11	97.3	0.05	0.06	0.04	<0.001	97
1195/11	79.6	0.02	0.02	0.001	0.003	80
835/12	67.6	1.82	0.87	1.07	<0.001	75
145/17	67.8	1.18	0.54	0.07	0.003	70
146/17	67.9	0.18	0.47	0.003	0.002	69
149/17	83.6	0.02	0.02	0.002	0.021	84
151/17	91.8	0.01	0.001	0.001	0.002	92
153/17	58.7	0.93	0.37	0.09	<0.001	60
154/17	91.2	3.14	1.66	0.20	0.009	96
217/17	75.0	0.02	0.00	1.05	<0.001	76

Table 4. Bulk composition, trace element analyses of metal artefacts from Arslantepe VII. Values are given in parts per million (ppm). For an explanation of the samples, see Table 2.

Sample no.	Ag	Sb	Te	Bi	U	P	Fe	Co	Zn	Se
20/04	580	3700	18	40	<0.2	100	670	200	15	35
21/04	80	20	5.6	2.4	2.1	25	160	0.4	180	30
215/10	260	650	30	45	5.2	310	80	65	15	150
405/10	1100	100	8.3	10	0.5	65	85	0.9	20	80
406/10	1000	120	8.2	20	0.8	80	110	1.1	20	30
1194/11	35	50	11	9.0	<0.2	3.4	50	6.5	35	460
1195/11	250	15	20	<2	0.7	200	1500	9.4	25	250
835/12	490	1900	32	940	1.2	280	410	25	15	30
145/17	390	40	22	2.4	0.9	320	310	8.4	10	100
146/17	100	610	30	20	1.8	330	110	120	35	35
149/17	310	25	17	3.1	1.1	130	1200	10	50	240
151/17	320	15	13	2.2	0.8	80	140	<0.3	20	<10
153/17	330	780	12	95	2.9	1200	2200	6.0	25	35
154/17	480	1200	76	50	<0.2	10	120	250	45	40
217/17	550	420	12	95	6	345	1100	0.9	13	33

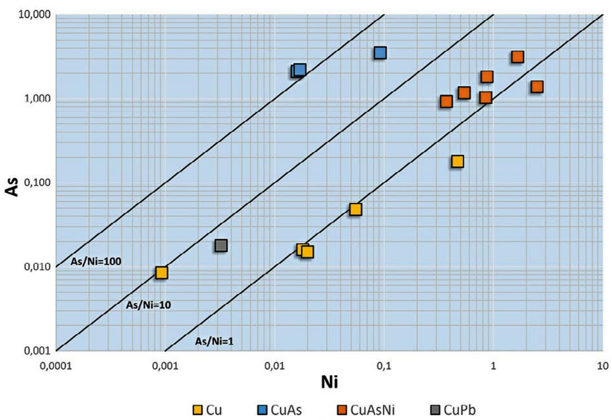


Figure 8. Correlation diagram for arsenic and nickel (As/Ni), the mostly discussed minor impurities in prehistoric copper from eastern Anatolia, so in 15 metal objects from Arslantepe VII. Note the division of possibly four different groups of metals (see text). Values are given in weight-percent (wt.%).

variations. Ni shows the largest range, Bi and Co are partly below the detection limits of 2 ppm (Bi) resp. 0.3 ppm (Co). Sample 146/17 stands out due to its high nickel and antimony contents.

2. Arsenical copper high in arsenic (> 2 wt.%) and low in nickel (c. 0.02 wt.%). The samples are a point of a knife (21/04) and two fragments of a needle (405/10; 406/10) which were found in a pit. The fragments of the needles are very homogeneous. The two fragments of a needle contain 1000 and 1100 ppm of silver. The knife contains only 80 ppm of silver, and it is also much lower in antimony and lead. In contrast to this, the contents of nickel and zinc are higher than with the needle fragments (only 920 ppm, resp. 20 ppm). The As/Ni ratio of sample 21/04 is around 40, while it is approx. 100 for the two needle fragments (405/10; 406/10) (Figure 8). Arsenical copper is the most widespread alloy found all over eastern Anatolia, in the Caucasus, in Azerbaijan, in the Transcaucasus and on the north-western Iranian Plateau (Courcier, Kuparadze and Pataridze, 2008; Helwing, 2012; Stöllner, 2021, p.452, Tab.1, Fig.4).
3. Arsenical copper high in nickel. Analyses of six samples show similar contents of nickel, arsenic, zinc and silver. Arsenic and nickel reach the lower percentage level. In the arsenic vs. nickel plot (Figure 8), they show a cumulus at a one to two ratio and a positive correlation of both elements. This cluster is separated from the arsenical copper low in nickel and suggests different ore provenances. Similar results are known for arsenical copper alloys from the "Royal Tomb" (Hauptmann, et al., 2002). Antimony varies from 40 to 3700 ppm. Worth mentioning is one needle (835/12) which additionally contains > 1 wt.% Pb and 3.5 wt.% S. It is the only sample containing > 3 wt.% sulfur, 0.19 wt.% antimony and almost 0.1 wt.% bismuth. Only two other samples (20/4 and 154/17) are very high in arsenic (> 1 wt.%) and antimony (> 0.12 wt.%). Arsenical copper high in nickel is less widely distributed and geographically confined to areas of western Asia, including Anatolia, Mesopotamia and

the Levant. Its origin is often closely associated with ophiolite ore deposits (Hauptmann, et al., 2002; Salzmann, 2019; see Pernicka, 1995).

4. One lead-containing piece of copper (217/17) is an outlier. It is a copper fragment with a lead content of just over 1 wt.%. Unlike 835/12, it contains only traces of nickel and arsenic, and apart from lead, no other element reaches the percentage level.

Lead isotope analyses

Six metal artefacts were analysed for their lead isotope ratios (Table 5). In the binary diagrams, the isotope ratios $^{208}Pb/^{206}Pb$ vs. $^{207}Pb/^{206}Pb$ and $^{208}Pb/^{204}Pb$ vs. $^{206}Pb/^{204}Pb$ are plotted (Figure 9). To find out from which ores these metal artefacts were smelted, the diagrams were compared with isotope data from ore deposits in eastern Anatolia. The diagrams show that five arsenic-nickel-containing copper samples plot in a relative narrow field in close proximity to some ore samples from the neighbouring district of the ore deposit of Ergani Maden. Also ores from the term "Taurus 2B" (Yener, et al., 1991) plot in the same field. This field includes ore deposits of the eastern Tauride block. Mamlis (Tunceli) and Keban are located there, among others, where (pre-) historical metal extraction has been proven (Wagner, et al., 1989). However, no suitable CuAsNi ores have as yet been described from these regions, so that it might be ruled out as a supply areas for these metal artefacts for the time being.

The ring fragment 149/17 plots separately from this cluster. It consists of pure copper (Group 1) and it is identical with ores from the copper deposit of Trabzon in all its lead isotope ratios. The ring fragment shows significantly higher isotopic signatures of $^{208}Pb/^{206}Pb$ vs. $^{207}Pb/^{206}Pb$. It plots to > 2.08 and 0.845. Hence it is identical with ores from copper deposits located far away from Arslantepe in the north, near the Black Sea coast. In the $^{208}Pb/^{204}Pb$ vs. $^{206}Pb/^{204}Pb$-diagram it displays a good match with the Artvin/Murgul-mining district. Both ore deposits are of the volcanic massive sulphide type and lie far away in north Anatolia. This means that metal was either imported from these northern regions or that ores from there were smelted at Arslantepe. This generally indicates the importance of the ore-districts of NE-Anatolia and the Transcaucasus as possible provenance for some of the period VII ore concentrates and metals.

For a diachronic comparison of the metal finds, data from Hauptmann, et al. (2002) were also included. A sample of CuAsNi as well as a polymetallic ore (CuAsSb) from the settlement fit into the cluster of new measurements. Comparisons from period VI A of the same material group show overlapping, but overall more variance

Table 5. Lead isotope ratios of metal artefacts and slag samples from Arslantepe VII. Values of standard error (StdErr) and standard deviation (StdDev) in percent.

Sample no.	206Pb/ 204Pb	StdErr	StdDev	208Pb/ 204Pb	StdErr	StdDev	207Pb/ 206Pb	StdErr	StdDev	208Pb/ 206Pb	StdErr	StdDev
Metal artefacts												
20/04	18.974	0.0143	0.0856	39.109	0.0141	0.0843	0.827	0.0017	0.0104	2.061	0.0017	0.0105
215/10	18.893	0.0167	0.1016	39.060	0.0172	0.1049	0.831	0.0011	0.0065	2.067	0.0014	0.0083
835/12	18.924	0.0155	0.0932	39.132	0.0161	0.0965	0.829	0.0011	0.0065	2.068	0.0018	0.0108
145/17	18.916	0.0133	0.0807	39.089	0.0134	0.0813	0.829	0.0010	0.0058	2.066	0.0012	0.0075
149/17	18.522	0.0128	0.0776	38.574	0.0132	0.0800	0.845	0.0017	0.0010	2.083	0.0022	0.0134
153/17	18.906	0.0122	0.0734	38.972	0.0136	0.0826	0.831	0.0008	0.0047	2.061	0.0016	0.0094
Slag												
38/02	18.916	0.0052	0.0312	39.112	0.0065	0.0395	0.830	0.0009	0.0053	2.068	0.0018	0.0106
39/02	18.906	0.0076	0.0470	39.137	0.0085	0.0525	0.830	0.0017	0.0104	2.070	0.0022	0.0132
44/02	18.928	0.0059	0.0355	39.106	0.0075	0.0454	0.829	0.0010	0.0062	2.066	0.0020	0.0119
53/02	18.925	0.0070	0.0436	39.134	0.0079	0.0489	0.829	0.0010	0.0061	2.068	0.0020	0.0119
94/03	18.933	0.0055	0.0336	39.144	0.0068	0.0412	0.829	0.0011	0.0069	2.068	0.0021	0.0128
98/03	18.948	0.0064	0.0385	39.178	0.0080	0.0486	0.829	0.0011	0.0067	2.068	0.0019	0.0116
874/12	18.909	0.0056	0.0338	39.113	0.0063	0.0381	0.830	0.0013	0.0080	2.068	0.0022	0.0129
148/15	18.818	0.0052	0.0320	39.019	0.0063	0.0388	0.834	0.0012	0.0071	2.074	0.0023	0.0138
152/17	18.910	0.0041	0.0245	39.116	0.0043	0.0260	0.830	0.0013	0.0077	2.069	0.0019	0.0115
156/17	18.896	0.0063	0.0384	39.099	0.0071	0.0434	0.830	0.0008	0.0050	2.069	0.0017	0.0104

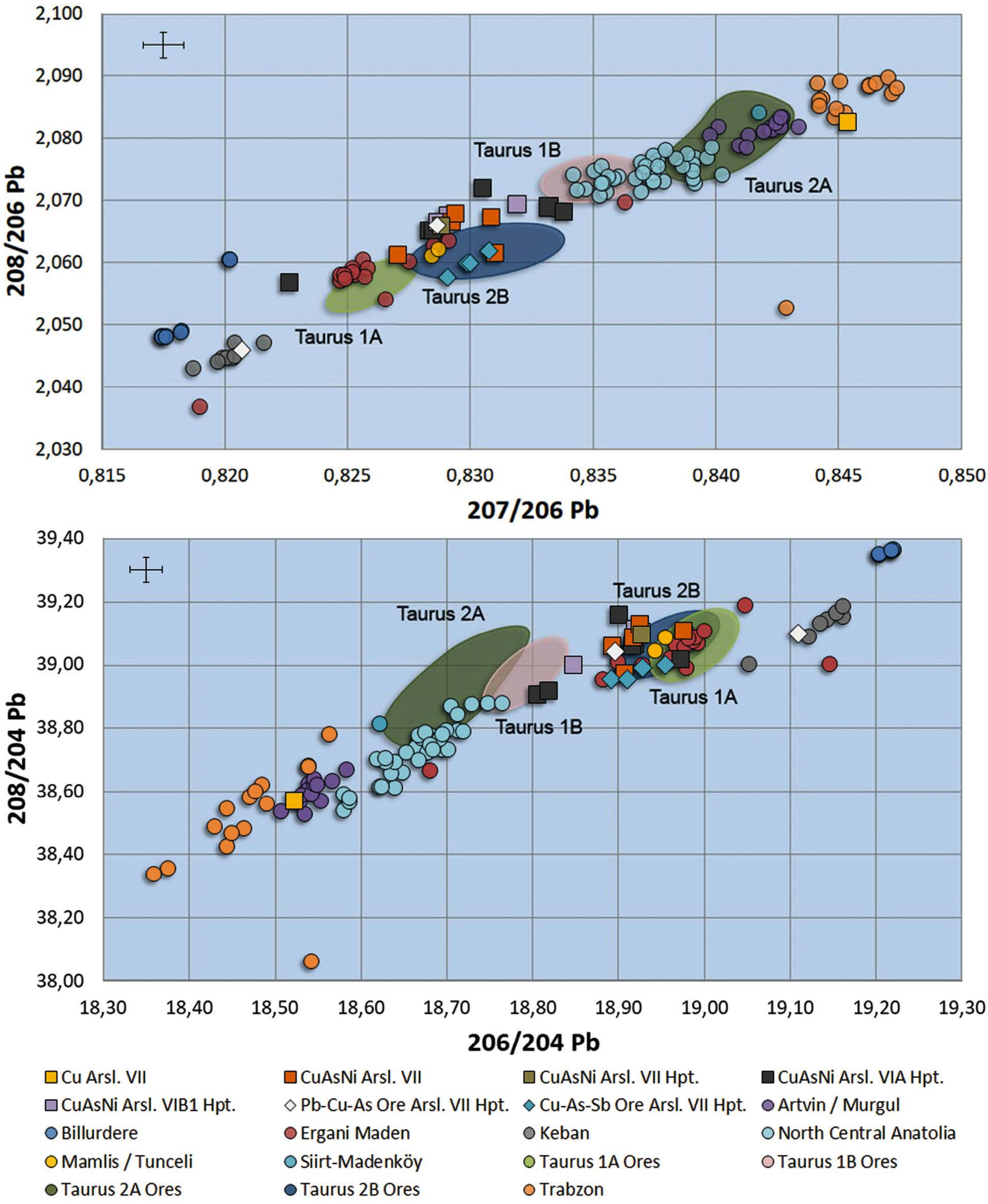


Figure 9. Lead isotope ratios of metal objects from Arslantepe VII in comparison to copper ores of eastern Anatolia. These data are taken from Seeliger, et al. (1985); Wagner, et al. (1986); Wagner, et al. (1989); Yener, et al. (1991) and Sayre, et al. (2001), and from the OXALID database (http://oxalid.arch.ox.ac.uk). The grouping of ore deposits in the central Taurus and the Pontides is taken from Yener, et al. (1991). The data were later re-evaluated by Sayre, et al. (2001).

than in period VII. Three further samples are available from VI B1 and also fit into the picture. Only from period VI B2 onwards there are indications of a different development. No isotope data of CuAsNi artefacts are available from this period so far. In general, this phase is characterised by the increased occurrence of pure copper. Two of these slightly younger copper artefacts partially overlap with the field of CuAsNi finds from Arslantepe VII. A single sample of CuAsNi comes from phase VI C and plots slightly outside the concentrated range of CuAsNi finds. Looking only at the lead isotope ratios, no significant changes are visible in the time frame of Arslantepe VII-VI B1. Based on the isotope data, there were no clear changes in the source of raw materials here, or at least they were mineralisations of a similar age.

Metallurgical debris: Slags

Macroscopic features

The 15 slag samples are crushed pieces of the size of a nut or even smaller (1-5 cm). Based on their outward typology (texture, colour), several sorts of slag can be distinguished.

One type is a dark-grey to black coloured, compact type of slag, with numerous light coloured angular inclusions and dots of reddish-yellow, sometimes green corrosion. They are partly ferromagnetic, caused possibly by high concentrations of magnetite (Fe_3O_4). These are copper slags.

Other slags are remarkably porous, high in gas bubbles, and built up in breccial textures (Figure 10). In terms of colour, they vary between light grey and light brown with light traces of weathering. Ferromagnetism is very low. These are lead slags that were formed by lead smelting.

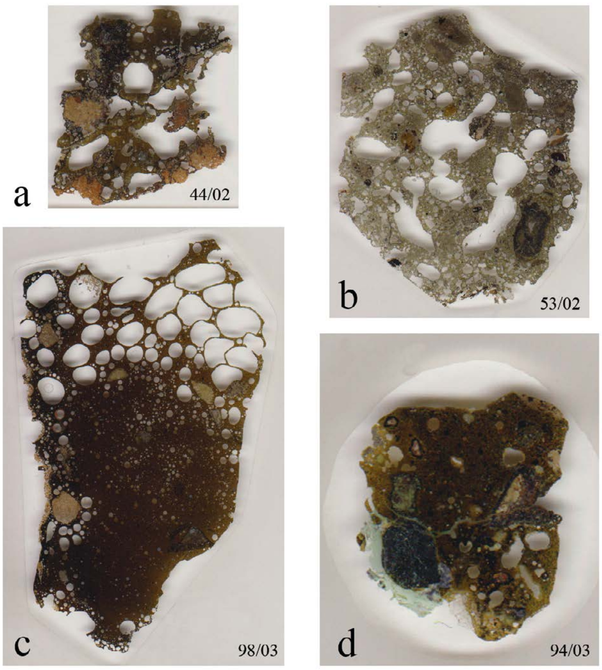


Figure 10. Macroscopic views of thin sections of some lead slags from Arslantepe VII (LC 3-4). Widths of the pictures each 28 mm. Figure a: Sample 44/02; D7(3) A853 2e. Partly liquefied, very porous and foamy light brown slag. Inclusions of granular carbonate host rock; b: Sample 53/02; D7(3) A853 rP1. Foam-like piece with vesicular cavities up to millimetre range; c: Sample 98/03; D5(3-7) 8e. Note the partly foamy, porous slag with a blister-like surface that developed from the intergranular lead rich part of the partly liquefied ore; d: Sample 94/03; D5(3-7) 8e. Partly more dense brown slag showing still solid inclusion of host rock. Photos: N. Heil.

Another type of slags are those that originally adhered to ceramic crucibles. These are crucible slags. Some of the crucibles showed only slight slagging. The sampled material was of a dark to light green colour due to corrosion, and the microstructure could not be further described macroscopically.

Chemical and phase composition

Generally, chemical composition and phase content of slag is regulated by the mixture of the charge (ore, gangue, host rock, possible fluxes and fuel). In addition, physical conditions (temperature, air supply, redox conditions) and the duration of metallurgical operations are crucial for slag formation, as well as furnace design and technical ceramics (tuyères, crucibles, furnace material). Ceramics may react with slag phases or liquids and influence slag formation with an input of alkaline earth metals and SiO_2.

Copper slags

Four pieces are iron-rich silica slags as they can be found worldwide at many ancient smelting sites. In this study, the chemical concentrations of iron in the slags are calculated as FeO. They are between approx. 40 and 60 wt.% (Table 6). The SiO_2-contents vary between 10 and 20 wt.%, and in one case 36 wt.%. Cu-concentrations are between c. 2 and 11 wt.%.

Bulk chemistry normalised to 100 % plotted into the ternary FeO-Al_2O_3-SiO_2-system (Figure 11) resulted in the following observations:

Two of the copper slags fall into the eutectic field of wuestite-fayalite. Slag 199/17 with higher SiO_2, and Al_2O_3 is at the upper limit of the fayalite field. This is due to an over-abundance of quartz and host rock, resulting in some unliquefied inclusions. The eutectic field is around 1200 °C, i.e., the slags were heated up at least to some 1250-1300 °C. It can be assumed that in prehistoric pyrotechnology these temperatures were reached very early on without any problems. Nevertheless here, as at many other prehistoric localities, refractory oxidic and siliceous inclusions such as quartz, feldspar and other minerals phases can be observed in slags. E.g. Hess (1998) described widespread siliceous inclusions in slags from Arslantepe VI B2. This may be explained by short-term smelting times, which hamper the complete liquefaction of the charged material. It is debatable whether this was not intentional and whether one was satisfied

Table 6. Bulk composition, main and minor element analyses of copper slags, lead slags and crucible slags from Arslantepe VII. Values are given in weight-percent (wt.%). For an explanation of the samples see Table 2.

no.	Na_2O	MgO	Al_2O_3	SiO_2	S	CaO	TiO_2	FeO	Cu	K_2O	MnO	ZnO	Pb	Total
Copper slag														
40/02	0.12	1.01	2.18	12.1	1.45	4.87	0.07	58.6	6.36	0.39	0.03	0.03	0.03	87
222/10	0.08	0.57	1.85	9.90	2.15	1.07	0.05	57.3	11.4	0.23	0.02	0.02	0.01	85
1145/11	0.11	1.05	3.32	19.0	0.31	2.15	0.15	60.9	2.06	0.39	0.06	0.08	0.01	90
199/17	0.05	2.98	7.75	36.0	0.14	1.75	0.27	41.2	1.84	0.50	0.24	0.05	0.05	93
Lead slag														
38/02	0.10	1.82	2.05	21.8	0.06	8.19	0.41	16.4	0.57	0.34	0.51	2.07	33.2	88
39/02	0.03	0.38	0.74	4.91	0.15	4.77	0.22	3.74	0.29	0.13	0.12	0.99	49.4	66
44/02	0.08	0.59	3.80	15.0	0.12	11.4	0.36	3.05	0.40	0.81	0.04	0.47	53.8	90
53/02	0.04	0.14	0.34	8.86	0.15	4.07	0.01	2.95	0.27	0.19	0.08	9.94	52.5	80
94/03	0.08	4.09	0.94	26.4	0.06	9.37	0.11	11.4	0.64	0.31	0.33	3.62	29.0	86
98/03	0.02	0.07	0.27	5.66	0.14	2.27	0.02	4.40	0.32	0.19	0.01	0.48	53.2	67
152/17	0.09	1.64	1.09	15.5	1.23	6.20	0.13	9.3	10.5	0.34	0.19	1.55	36.1	84
874/12	0.07	1.68	1.15	16.2	0.42	6.28	0.14	9.5	5.90	0.28	0.22	1.88	40.9	85
Crucible slag														
148/15	0.06	0.06	0.27	1.85	0.12	0.54	0.02	0.10	69.6	0.10	0.002	0.002	0.39	73
148/17	0.27	0.47	1.27	8.01	0.08	4.28	0.12	0.83	59.7	0.52	0.01	0.01	0.07	76
156/17	0.46	2.06	4.99	21.7	0.09	13.2	0.39	2.28	32.3	2.61	0.04	0.01	0.49	81

Table 7. Bulk composition, minor and trace element analyses of copper slags, lead slags and crucible slags from Arslantepe VII. Values are given in parts per million (ppm). For an explanation of the samples see Table 2.

no.	Ag	Co	Ni	Cr	As	Sb	Bi	P	Sn	Te	Se
Copper slag											
40/02	5.0	273	116	174	284	67	1.0	622	3.7	9.1	172
222/10	19	115	37	98	47	26	0.5	556	3.3	6.2	298
1145/11	8.4	383	314	233	47	43	0.5	684	3.3	4.7	22
199/17	2.8	320	59	197	352	61	14	599	5.3	4.6	7.0
Lead slag											
38/02	22	1440	7312	1304	43502	26593	39	4269	24	11	2.0
39/02	69	4.7	24	301	110552	29310	4.3	3814	8.2	7.4	49
44/02	94	8.7	55	99	52207	3202	5.5	4401	12	6.3	3.2
53/02	206	0.8	5.5	106	61089	68881	7.5	3431	9.0	28	25
94/03	86	618	24333	815	20263	48153	78	3596	15	6.1	4.6
98/03	178	1.1	51	80	46639	210867	4.9	1304	42	18	7.0
152/17	402	768	3370	626	61534	48231	760	2343	45	5.3	38
874/12	473	966	4179	730	58895	46815	561	2675	50	5.4	11
Crucible slag											
148/15	487	50	2796	60	11066	755	31	384	3.1	18	39
148/17	49	97	19983	123	38507	184	7.9	1435	4.8	17	36
156/17	412	36	861	169	1990	241	6.5	3184	3.2	25	8.2

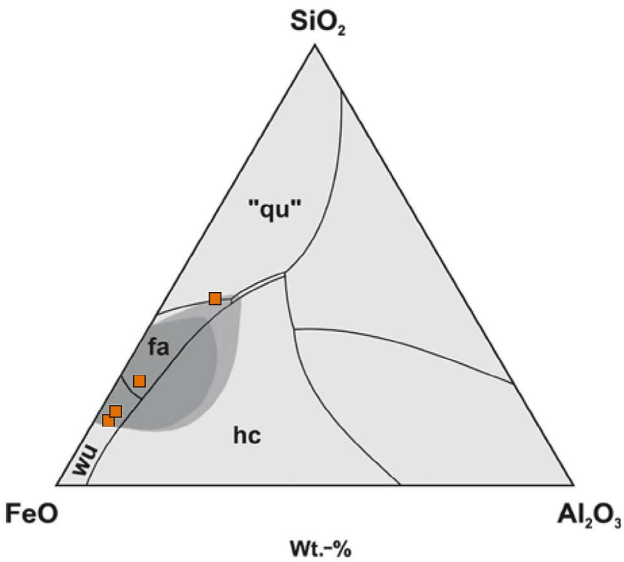


Figure 11. Composition of copper slags from Arslantepe VII plotted in the ternary system FeO-SiO_2-Al_2O_3 (Muan and Osborn, 1965). With one exception, these were small samples of well-liquefied slags in or around the eutectic field of the system. Iron oxides are calculated as FeO. Abbreviations: Wt.% = weight percent, "qu" = quartz; fa = fayalite; wu = wuestite; hc = hercynite.

with producing small metal droplets and easily flowing CuFe sulphides.

The copper slags consist mainly of fayalite (Fe_2SiO_4), magnetite (Fe_3O_4), possibly wuestite (FeO) and, as a minor constituent, Fe-rich clinopyroxene ($CaFeSi_2O_6$). Typical is a dendritic, partly massive structure of sometimes Mg-containing iron oxides (spinel) in between glass and fayalite in the second crystallisation phase. Especially with slag no. 222/10 (but not only there), globules of copper sulphide inclusions (Figure 12) occur next

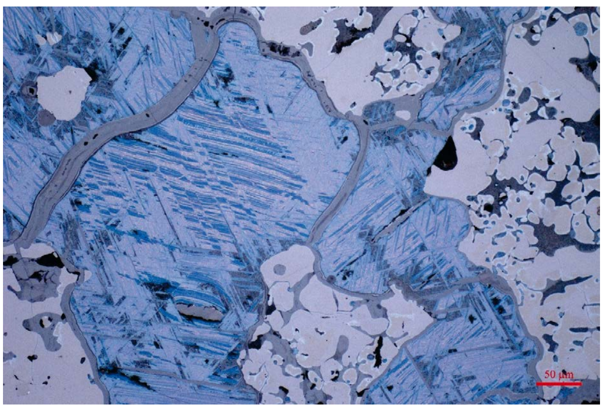


Figure 12. Arslantepe VII. Inclusion of copper sulphide (covellite, CuS, blue laminated texture) intergrown with semi-liquefied grains of magnetite (grey-white) characteristic for copper slags. Sample 222/10, Polished section and reflected light. Photo: N. Heil.

to tiny droplets of copper. The sulphides consist mainly of covellite (CuS) and chalcocite (Cu_2S). They are very low in iron, i.e. they are formed during smelting from high-grade sulfidic copper ore.

Lead slags

Eight lead slags are mostly from phase 2 of period VII, room A853. In the adjoining room, a hearth was discovered.

The chemical main component is lead (29-54 wt.%), although it is not clear how this metal should be calculated in the slag due to the phase/mineral composition, as lead may occur as Pb0, Pb2+ (oxidic or sulphidic) or Pb4+ (siliceous). With the main components of SiO_2 (5-26 wt.%), FeO (3-16 wt.%), and CaO (3-11 wt.%), lead slags differ fundamentally from slags from other metal extraction processes. It is not clear whether these slags were only connected with lead extraction or whether they were not perhaps a precursor to silver production. The concentration of silver may reach more than 400 ppm.

The significantly high lead contents (up to 54 wt.%) mean that these slags were formed by varying amounts of partial melts, altogether within a significantly low temperature range, at 700-800 °C.

This is far below the temperature range of copper slags. High CaO contents point to calcareous host rocks of the ores. Most interesting is that lead slags are much higher in arsenic (5-11 wt.%) and antimony (3-7 wt.%, in one case 21 wt.%), and higher in ZnO than copper slags (Table 6), while copper itself ranges in the lower percentage level of the slags.

The macroscopically determined heterogeneous texture of the lead slags continues down to the micro-level, and unmelted inclusions of quartz, calcite and technical ceramics (crucibles or furnaces) were identified.

In addition, in contrast to copper slags, no inclusions of sulphidic minerals were identified. By X-ray diffraction and by microscopic methods, only oxidic lead minerals were detected regularly and in significant proportions in all lead slags, as they typically occur in the oxidation zone of Pb/Zn deposits: lead-arsenate mimetesite ($Pb_5(Cl|AsO_4)_3$), pyromorphite ($Pb_5(Cl|PO_4)_3$), and cerussite ($Pb(CO_3)$). The abundance of these minerals is an indication of the use of oxide ores, as they occur in carbonate-hosted non-sulphidic lead-zinc-(silver) deposits in the south-western Tauride Block in the Aladağ Mountains. Globules of metallic lead very high in arsenic and antimony occur very frequently, which are dissolved as tiny dots in the metal (Figure 13).

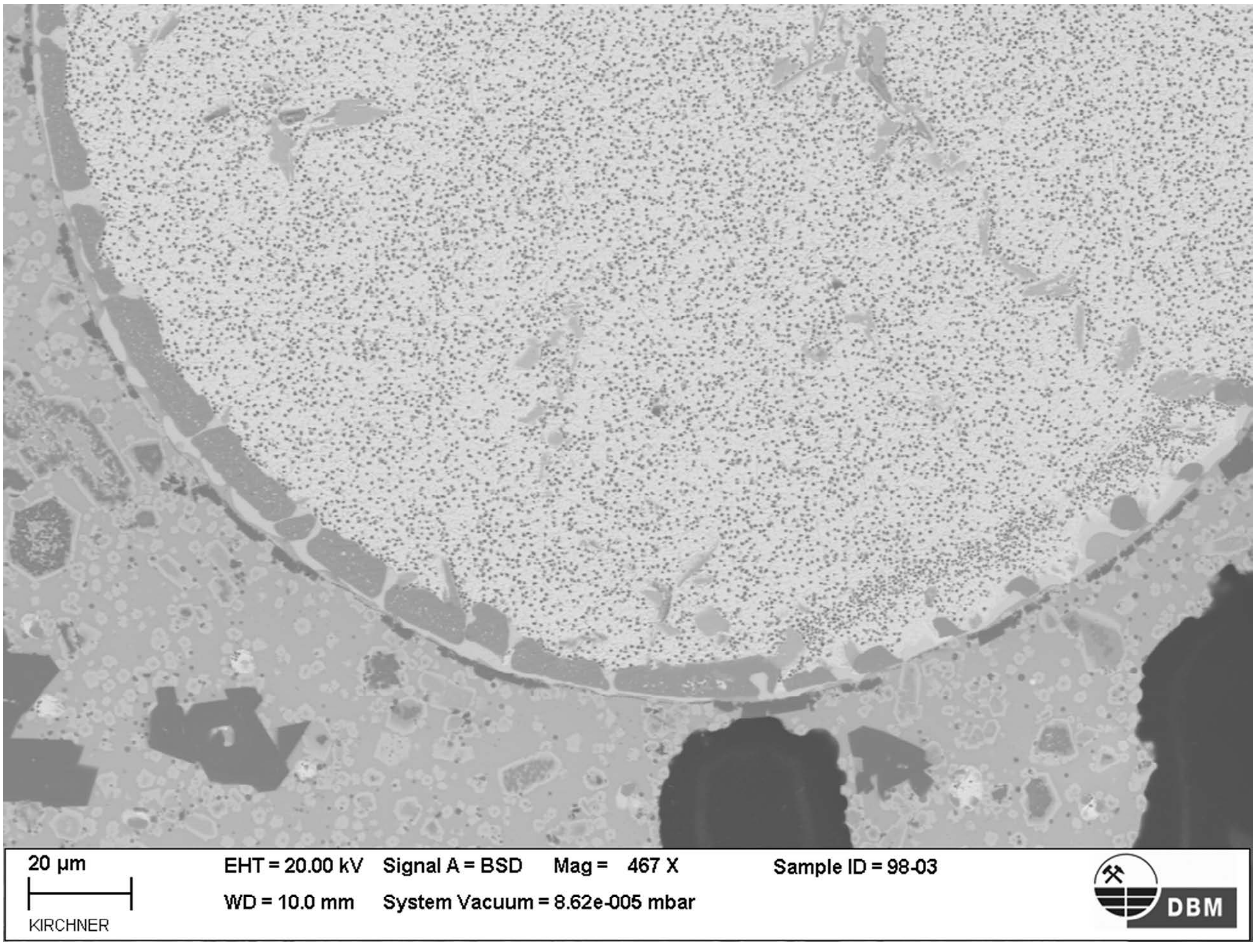


Figure 13. Arslantepe VII. Inclusion (tiny dots) of a lead-antimony-arsenic droplet in lead slag 98/03 (82 wt.% Pb; 9 wt.% Sb; 6 wt.% As). Polished section, scanning electron microscopy, backscattered mode. Photo: S. Merkel, Deutsches Bergbau-Museum Bochum.

Siliceous phases are clinopyroxene (diopside, hedenbergite, $Ca(Mg,Fe)Si_2O_6$), anorthite ($Ca(Al_2Si_2O_8)$), åkermanite ($Ca_2Mg(Si_2O_7)$ and larsenite ($PbZn(SiO_4)$). The latter is a rare phase that can form with or instead of lead silicates. Fe-contents in the ore batch crystallised to magnetite (Fe_3O_4) resp. magnesioferrite ($MgFe_2O_4$). Quartz was found, which was sometimes transformed to its high-temperature modification cristobalite (α-SiO_2).

Of particular interest are the conspicuously high levels of antimony. With but two exceptions (sample 44/02: < 1 wt.% and sample 98/03: 21 wt.%), these range in the lower percentage level (2.7-6.9 wt.%). Arsenic is also unusually high (2-11 wt.%). These concentrations are probably due to inclusions of fahlores, especially of tetrahedrite-rich composition ($Cu_{12}Sb_4S_{13}$).

Nevertheless, some Cu-Fe-containing alloys with As, Sb or also Ni should also be mentioned, which can be described as speiss. Speiss crystallises from liquids high in copper, iron, arsenic and/or antimony. In addition, speiss may contain lead, tin, zinc, bismuth, and cobalt. During the smelting of complex ores, such as fahlores, speiss can be produced as an unwanted waste product, but also as a targeted intermediate product (Bachmann, 1982; Hess, 1998; Hauptmann, 2020). In Arisman (Iran), the targeted extraction and use of speiss was postulated as early as for the 3rd millennium (Thornton, Rehren and Pigott, 2009; Rehren, Boscher and Pernicka, 2012; Boscher, 2016).

Also, a thin incrustation of a corroded speiss-like material was found in one of the crucibles at Arslantape VII (Hauptmann, Frangipane and Di Nocera, in prep.).

Crucible slags

Three slag samples were taken from slag layers inside crucibles. During sampling, the slagged material was mechanically detached from the crucible wall and parts of the crucible wall were also integrated. The slags show enormously high copper contents (32-70 wt.%) (Table 6), which make them easily identifiable as slags from a metallurgical copper operation. At the same time, however, they also contain arsenic and nickel concentrations in the lower percentage level. According to their Ni/As ratio, they correspond to the typical ratio of As-Ni containing copper (see above). Moderate SiO_2- and FeO-, CaO-and Al_2O_3-contents may indicate only few siliceous slag phases, which are more indicative for contamination by the ceramic crucibles. Minerals and phases such as muscovite, gehlenite, clinochlore, and even calcite were determined by X-ray diffraction.

Due to the material composition of the sample (corrosion), neither any exact identification of slag phases nor any petrographic analysis was possible. Perhaps the samples might be slagged copper remains, i.e. remains of copper formed by re-melting.

Lead isotope analyses

Eight lead slags and two crucible slags were analysed for their lead isotope ratios (Figure 14). In the $^{208}Pb/^{206}Pb$ vs. $^{207}Pb/^{206}Pb$-diagram they plot in the field between

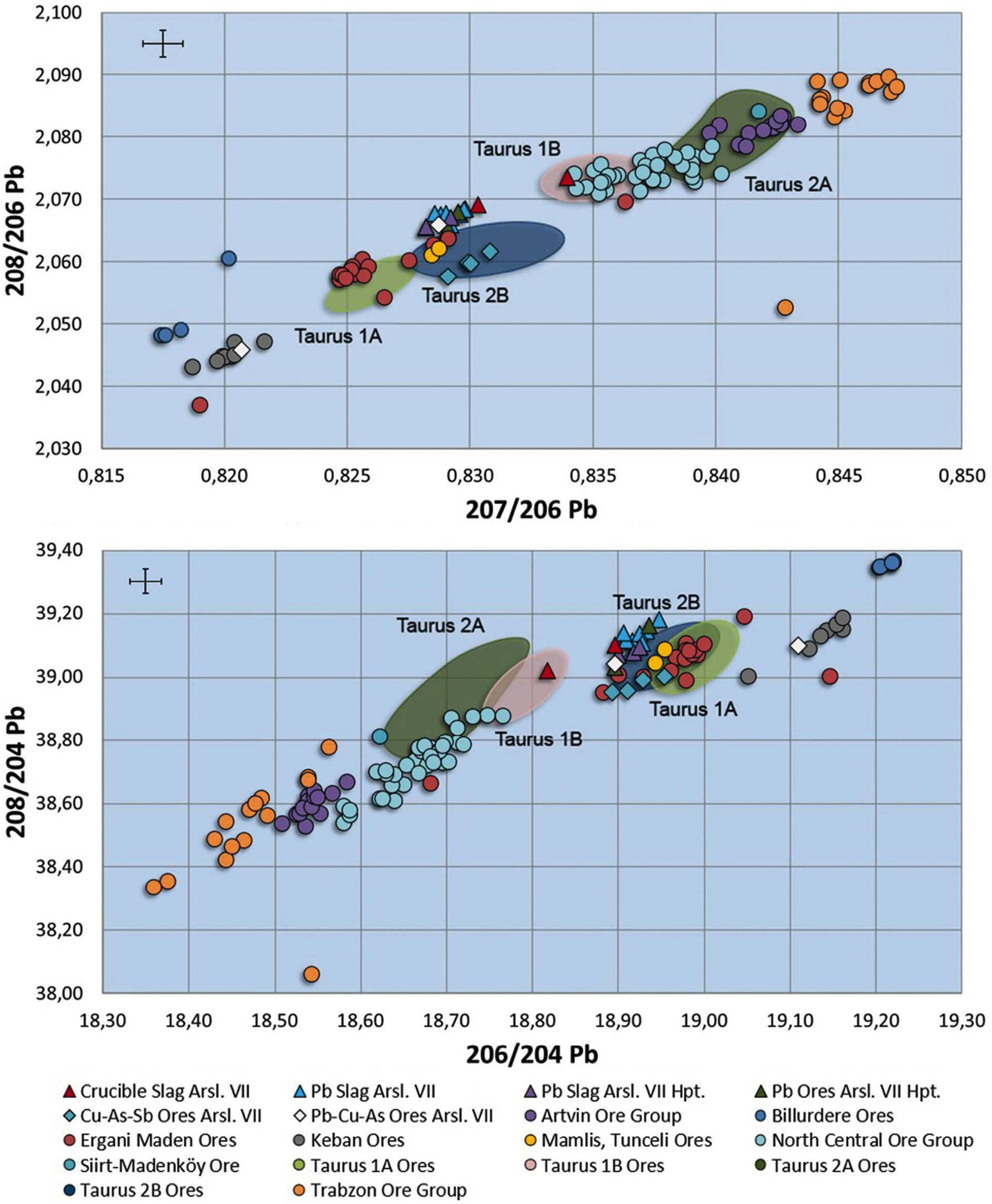


Figure 14. Lead isotope ratios of slag samples from Arslantepe VII in comparison to various ores from Anatolia (see also Figure 9).

2.066 to 2.069 and 0.828 to 0.830. Thus, they are partly overlapping with those of the copper artefacts high in arsenic and nickel and are of similar geological age (see also Figure 9).

Further lead isotope ratios of metals, slags and ores from period VII, and also from the Early Bronze Age periods (VIB-VID), have already been published by Hauptmann, et al. (2002). A comparison of these data with the new results shows a good match with their cluster (Figures 9 and 14). From the later periods, however, two lead metal objects from VI A and two others from VI D3 and VA show a clearly older signature. These data plot in close proximity to the argentiferous Pb-Zn-deposit of Görgü (Cafana) near Malatya, only a few kilometres from Arslantepe (Hauptmann, et al., 2002, p.64). In the $^{208}Pb/^{206}Pb$ vs. $^{207}Pb/^{206}Pb$-diagram, the ores with a high abundance ratio lie outside of the shown area at approx. 2.112 and 0.865. Surprisingly, there is no archaeological evidence from Arslantepe VII for the use of the deposit at Görgü (Cafana) so far. However, prehistoric lead-silver mining was already suspected by Wagner, et al. (1985) after a field survey.

So far, no evidence of early lead-silver exploitation has been found at the Billurdere site in the present-day province of Elazığ, either. The four galena samples from this deposit by Kunç were published in OXALID (http://oxalid.arch.ox.ac.uk). The analyses plot near the deposits of Keban (Figure 14). The mining district of Keban, which is also located in the province of Elazığ, is characterised by a large number of smaller mineralisations (see Hanelçi and Çelebi, 2015). Two analyses of galenite, which also contains Cu and Ag, are available. Further samples from slags and primary and secondary ores from different mineralisations in the Keban district exist as well. A Pb-Cu-As ore find (TR-8/68) from Arslantepe VII belongs to the field of these samples (Hauptmann, et al., 2002, p.56, Tab.9). Unfortunately, it does not correspond with slag or metal finds from this period. However, the samples from the two deposits, Billurdere and Keban, are of a younger geological age than the finds from Arslantepe VII.

Comparisons with ores from the Central Taurus ("Taurus 2B"; Yener, et al., 1991; Sayre, et al., 2001) show that these belong to the marginal field of the cluster of lead slags from Arslantepe VII (Figure 14). In the diagram $^{208}Pb/^{204}Pb$ vs. $^{206}Pb/^{204}Pb$, a slight overlapping can be seen. "Taurus 2B" is characterised by geological samples of the Aladağ Mountains, the Yahyalı area and the Niğde Massif.

The lead isotopic compositions of the crucible slags are only in the vicinity of the samples of AsNi-copper. Sample 148/15 lies in the marginal field, whereas sample 156/17 plots outside the finds of Arslantepe VII. For 156/17 there is an overlapping of the isotope signature of the ore region "Taurus 1B", which includes e.g. the deposits around Çamardı/Kestel (Yener, et al., 1991).

Discussion of results

Metallurgy at Arslantepe VII

At Arslantepe, the production and processing of several metallic raw materials like copper, lead and possibly silver is attested for the Late Chalcolithic period VII (LC 3-4). The analysed lead slags are products of the smelting of polymetallic PbAsSb-ores, mostly of oxidic compositions. They show varying contents of zinc, nickel, iron, and copper. In contrast, copper slags show inclusions of relatively pure Cu-sulfides ("matte"), indicating the smelting of high-grade sulfidic ores such as covellite, digenite with portions of chalcopyrite or bornite, perhaps with some oxidic ores. This is also reflected by evidence of ores from this time. Hess (1998) investigated oxidic and sulfidic copper ores, lead ores, and polymetallic ores (fahlores, Cu-arsenates).

On the basis of the crucible finds, further steps in metal production can also be seen: the slagged crucible lines indicate the re-melting of copper. In addition, finds of possible casting moulds indicate the process of casting within the settlement, though workplaces and casting spills are still missing. In addition to copper production, the extraction of lead is shown, which could perhaps also be linked to that of silver.

Several alloys are found at Arslantepe, level VII. Due to their large variations of compositions, it can be assumed that most of them are natural alloys, i.e. that they were probably produced by polymetallic ores, and that they were not intentionally made. However, there is also a theoretical metallurgical model that explains the perhaps intentional production of As- or AsNi-copper by way of so-called "co-smelting" (Boscher, 2016; Hess, 1998; Rostoker and Dvorak, 1991). This is based on the colour of minerals: While secondary copper minerals like secondary nickel ores show striking green-blue colours and are easily identified as distinguished materials, arsenical minerals are very inconspicuous. Their identification by the old metallurgists is questionable, even though their knowledge and skills should not be underestimated.

Another proposition sees the targeted alloying of arsenic with copper by making speiss as a master alloy in an intermediate production (see discussion at Hauptmann, Frangipane and Di Nocera, in prep.). The speiss, in a second step, would be co-melted with (pure) copper. This is also being discussed, for example, for metal extraction in Arisman (Iran) (Thornton, Rehren and Pigott, 2009; Rehren, Boscher and Pernicka, 2012).

Other chalcolithic finds from eastern Anatolia also prove sophisticated metallurgical processes, such as the production of silver by smelting lead minerals and cupellation at Fatmalı Kalecik and Habuba Kabira (Hess, et al., 1998; Pernicka, Rehren and Schmitt-Strecker, 1998). The lead slags from Arslantepe layer VII may also have elevated silver contents, but it is not clear whether these could also be evidence of silver extraction or are merely associated with the production of lead. Unfortunately, there are no finds of silver or litharge as evidence of a possible cupellation.

In general, it can be observed that at Arslantepe VII there are remains of metallurgical activities from both domestic and "public" contexts. This means that metallurgical craftsmanship does not seem to have been regulated by the central administration, as it is known from the redistribution of food, and metallurgical practice was not spatially limited to a single defined area. To what extent it was embedded in social or religious practices, however, is still unclear. The variety of the metal artefacts indicates the ubiquity of jewellery or small tools, which are predominant. Larger objects (axes) are missing from the finds inventory. They are only indirectly likely because of the existence of possible casting moulds. The situation is similar for possible lead or silver objects. It is possible that the high symbolic value of some objects constituted a kind of transmission filter, preventing them from finding their way into the archaeological context. Damage to an object does not necessarily lead to a complete "loss of value". In addition, possible recycling must be taken into account.

The concentration of metallurgical finds in grid square D7(3) could potentially yield a better picture of a possible work site in phase 2 of level VII during further excavations of the Late Chalcolithic period. So far, the few slags investigated in this study represent the only evidence of metallurgy. Surrounding hearths or ovens appear to have rather served for everyday use (Guarino, 2014).

Provenance studies

Metal artefacts from Arslantepe VII include pure copper, arsenic copper and arsenic-nickel copper. There exist only one copper artefact with increased lead content that was probably a natural alloy.

Due to their geographical proximity to ophiolite copper deposits and the concentrated occurrence of AsNi-copper in the 4th and 3rd millennium, their origin is assumed to be from the nearby ore deposits in south-eastern Anatolia. Indeed, arsenic and nickel contents in metal artefacts as well as inclusions of chromite in ores and slags found nearby are useful indicators for the origin of the material from copper deposits in an ophiolite context, i.e. in ultramafic host rock or ore (Hauptmann, et al., 1993). Examples are Ergani-Maden and Siirt-Madenköy near the Bitlis-Zagros Suture in south-eastern Anatolia (Figure 3).

This copper occurred in the south along the Rift Valley and the Jordan Valley, and also in Mesopotamia (Hauptmann, et al., 2002; Salzmann, 2019). There are no corresponding ore deposits in these regions, so that artefacts of these copper varieties were probably traded from there. On the other hand, Salzmann (2019) was able to assign a provenance from the ophiolite copper deposits of Oman (Makan) at least for later, Early Dynastic artefacts made of AsNi-copper from the Royal Tomb of Ur. In her opinion, only a few objects of the grave may have come from Ergani-Maden.

As far as the mineralogical composition of the ores is concerned, most probably secondary ores were used. The example of Ergani-Maden shows the significant enrichment of arsenic, nickel, cobalt, antimony and bismuth especially in the lower part of the oxidation zone compared to the primary ore. Although only ores low in arsenic and nickel were extracted from the Ergani-Maden open pit mine a few years ago (Seeliger, et al., 1985; Wagner, et al., 1986; Hauptmann, et al., 2002), Hess (1998) found ores enriched with these two trace elements (several hundred ppm). He concluded that the sulphide-rich ore would only have produced a metal low in Ni and As, while the smelting of the oxidation ores would have produced a metal rich in As and Ni. According to the investigations of the lead isotope ratios, the Siirt-Madenköy occurrence can be ruled out for the AsNi-copper group.

On the other hand, the lead isotope measurement of a pure copper ring from Arslantepe shows a possible origin from far to the north. It is separated from the data discussed so far. It lies close to the compositions of ores from Trabzon, alternatively from Artvin/Murgul. These ore districts are located in the north-eastern part of Anatolia (Figure 9), and they are partly of the same type of ore deposits (Yiğit, 2009). It is striking to observe connections to the northern fringes in general, for instance also other connections to northern central Anatolia, as evidenced by handmade burnished ware which possibly indicate small scale contacts and individuals that were present at and near Arslantepe (Frangipane, 2011; Balossi Restelli, 2019).

The search for the raw material sources of other arsenic-, antimony- and nickel-containing objects of the hoard from Nahal Mishmar from the mid-4th millennium has been sadly unsuccessful so far. Their provenance from eastern Anatolia has repeatedly been suspected (Tadmor, et al., 1995; Shugar, 2000). However, the contemporaneous metal artefacts from Arslantepe are only low in arsenic and antimony. According to the present state of knowledge, also the previously recorded surrounding ore deposits can probably be excluded from further considerations.

Conclusion

Arslantepe VII belongs to the early epochs in which metallurgical activities can be traced at this site. However, this period is already preceded by a phase of metallurgical activity: metallurgy thus seems to be following local traditions. The earliest evidence consists of some functional small objects from period Arslantepe VIII: they are made of copper containing arsenic and nickel (Di Nocera, 2013). Due to the limited number of finds, however, they could only be characterised inadequately.

For period VII it is now possible to prove the use of several, geographically different, metalliferous raw sources. This is evident from the different compositions of metals and copper alloys. As a minimum, the use of copper and lead must be assumed. Evidence of silver extraction is currently lacking. Even if metallurgical installations are not present in the archaeological record, a number of finds roughly represent the whole range of the metallurgical chain, from smelting to melting to casting.

So far, copper containing arsenic and nickel seems to have been the primary choice. Perhaps this was a question of access and/or political control. The role of metallurgy in the context of redistribution, which is considered confirmed for food (Frangipane, 2011, p.974, 978), cannot be assessed so far. Spatial concentrations like the so-called "palace hoard" from VI A do not emerge for period VII.

Social centralisation on the Arslantepe reaches its peak in period VI A and is evidenced by thousands of cretulae (Frangipane, 2011, p.974). The metals of this phase include copper, lead, silver and gold. As for the previous period, no metallurgical furnaces or similar constructions are known. However, the spatial distribution of the slags and ores is limited to the east of the so-called palace, which is a tentative indication of a specialised workshop area. In general, the metallurgy of period VI A seems to have made a significant step forward in development.

In the palace hoard, new types and forms of objects were found for the first time at Arslantepe. The implements found there, which are described as weapons (cf. discussion in Piller, 2009), are typologically significant objects and, due to their size, require a sophisticated production technique. In addition to the casting of large pieces of metal, the filigree inlaid silver work on the spear and swords is also a novelty. In addition, the use of clamshell moulds is likely (Di Nocera, 2013, p.117). Despite the novelties, the picture is similar to that of phase VII when looking at the ores used. Polymetallic ores are predominant, and the lead isotope data of the metal finds show a greater variance but largely overlap with the AsNi copper objects from Arslantepe VII.

With the collapse of the palace in VI A, the appearance of the settlement changes significantly. Besides a large mud-brick house, wooden huts shape the image of the settlement in VI B1 (Frangipane, 2014, pp.171-173; Palumbi, et al., 2017, pp.90-92). This striking break in social and political organisation is also expressed in metallurgy. There is a decline in the scale of metallurgy in VI B1. However, it is only with the resumption of metallurgy in Arslantepe VI B2 that a shift becomes visible both chemically and isotopically. The first metal workshop and increased metallurgical activity are recorded from this phase. The metal finds are characterised only by low arsenic contents. Polymetallic ores, as they were processed in the periods of the Late Chalcolithic at Arslantepe, are now completely absent. Pure copper ores occur instead (Di Nocera, 2013). Now lead isotope data also indicate an origin from the Pontic and Transcaucasian region as well as from Central Anatolia (Hauptmann, et al., 2002). Thus, period VI B2 represents the first major break in the metallurgical tradition.

A particular exception from the metallurgical tradition in the settlement is the "Royal Tomb" (VI B1/2) with its exceptionally rich metal finds, which indicate a variety of cultural contacts (Hauptmann and Palmieri, 2000).

The metallurgical tradition observed with Arslantepe period VII seems to be continuously developing, despite various changes. If phase VI B1 is to be included in this development, it covers a period of almost 1000 years. Works on period VIII could provide further insight into the possible beginnings of metallurgy at Arslantepe. This continuity of metallurgy despite socio-political changes suggests that it is a locally or regionally consolidated tradition that is not tied to a social or political system ("palace"). AsNi copper seems to be a constant in metallurgical development. Its use is evident over centuries and indicates the long-standing use of similar or possibly even the same deposits. Only with period VI B2 there happens a break in the use of AsNi-copper.

The results of the analyses provided evidence for a rather complex and differentiated metallurgy as early as period VII at Arslantepe. The evidence adds to the concept of Arslantepe as a hub on the Upper Euphrates inter alia, for the Transcaucasian and Mesopotamian region, especially from an archaeometallurgical perspective. In the future, further investigations will hopefully provide more detailed insights into the metallurgical processes and operations as well as the possible use of silver at Arslantepe.

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