Metallurgy of prehistoric Armenia

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Titelbild

Alacahöyük gehört zu den wichtigsten prähistorischen Städten in Anatolien. Besonders berühmt sind die frühbronzezeitlichen Fürstengräber mit ihren zahlreichen Grabbeigaben aus Gold, Silber und Bronze, darunter die frühesten Eisenfunde Anatoliens. Zum Grabinventar zählten auch zahlreiche bronzene Sonnenstandarten und Tierfiguren. Im Vordergrund ist eine dieser Sonnenstandarten zu sehen. Sie dient heute als Symbol des Kultur- und Tourismusministeriums der Türkei.

Im Hintergrund ist eine schroffe Landschaft bei Derekutuğun, Kreis Bayat, Provinz Çorum zu sehen. In Derekutuğun wurde seit dem 5. Jt. v. Chr. gediegenes Kupfer bergmännisch gewonnen. Im Vordergrund ist eine der prähistorischen Strecken abgebildet. Fotos stammen von Herausgeber.

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Inhaltsverzeichnis

Vorwort 9

Grußwort 11

Rainer Slotta & Andreas Hauptmann
Robert Maddin and the Deutsches Bergbau-Museum Bochum 13

James D. Muhly
Robert Maddin: An Appreciation 17

Mehmet Özdoğan
The Dynamics of Cultural Change in Anatolia 21

H. Gönül Yalçın
Die Karaz-Kultur in Ostanatolien 31

Ulf-Dietrich Schoop
Çamlıbel Tarlası, ein metallverarbeitender Fundplatz des vierten Jahrtausends v. Chr. im nördlichen Zentralanatolien 53

Horst Klengel
Handel mit Lapislazuli, Türkis und Karneol im alten Vorderen Orient 69

Metin Alparslan & Meltem Doğan-Alparslan
Symbol der ewigen Herrschaft: Metall als Grundlage des hethitischen Reiches 79

Ünsal Yalçın & Hüseyin Cevizoğlu
Eine Archaische Schmiedewerkstatt in Klazomenai 85

Martin Bartelheim, Sonja Behrendt, Bülent Kızılduman, Uwe Müller & Ernst Pernicka
Der Schatz auf dem Königshügel, Kaleburnu/Galinoporni, Zypern 91

Hristo Popov, Albrecht Jockenhövel & Christian Groer
Ada Tepe (Ost-Rhodopen, Bulgarien): Spätbronzezeitlicher – ältereisenzeitlicher Goldbergbau 111

Tobias L. Kienlin
Aspects of the Development of Casting and Forging Techniques from the Copper Age to the Early Bronze Age of Eastern Central Europe and the Carpathian Basin 127

Svend Hansen
Metal in South-Eastern and Central Europe between 4500 and 2900 BCE 137

Evgeny N. Chernykh
Eurasian Steppe Belt: Radiocarbon Chronology and Metallurgical Provinces 151

Andreas Hauptmann
Gold in Georgia I: Scientific Investigations into the Composition of Gold 173

Thomas Stöllner & Irina Gambashidze
Gold in Georgia II: The Oldest Gold Mine in the World 187

Khachatur Meliksetian, Steffen Kraus, Ernst Pernicka Pavel Avetissyan, Seda Devejian & Levron Petrosyan
Metallurgy of Prehistoric Armenia 201

Nima Nezafati, Ernst Pernicka & Morteza Momenzadeh
Early Tin-Copper Ore from Iran, a Posssible Clue for the Enigma of Bronze Age Tin 211

Thomas Stöllner, Zeinolla Samaschev, Sergej Berdenov †, Jan Cierny †, Monika Doll, Jennifer Garner, Anton Gontscharov, Alexander Gorelik, Andreas Hauptmann, Rainer Herd, Galina A. Kusch, Viktor Merz, Torsten Riese, Beate Sikorski & Benno Zickgraf
Tin from Kazakhstan – Steppe Tin for the West? 231

Autorenliste 253

Khachatur Meliksetian, Steffen Kraus, Ernst Pernicka
Pavel Avetissyan, Seda Devejian & Levron Petrosyan

Metallurgy of Prehistoric Armenia

Abstract

This paper is focussed on the chemical compositions and lead isotope ratios of archaeological metal artefacts related to the early stages of metallurgy in Armenia – from the Neolithic until the Late Bronze Age. Possible relationships between artefacts and Armenian and other copper ores in the region are also discussed.

Introduction

In the last century the archaeological and geological research revealed numerous new sites of prehistoric metallurgical activities in Armenia and the Caucasus. The early appearance of metallurgy in South Caucasia and the abundance of copper and polymetallic ores make this region particularly important for archaeometallurgical studies (Fig. 1). Few recent finds of copper objects dating back to the early 4th and late 5th / early 6th millennium BC strongly support this assumption.

The archaeological metal finds in the territory of Armenia cover all significant phases of the early stages of metallurgy (Fig. 2): Neolithic use of native copper, the transition to extractive metallurgy in the Eneolithic period, the extensive use of copper, arsenical copper, some other alloys and the early appearance of tin bronzes in the Early Bronze Age and the transition to more advanced metallurgy and alloying in the Middle and Late Bronze Age.

Early Metal Use: Neolithic and Eneolithic

The earliest metal items made of copper – beads and small objects – were discovered in eastern Anatolia and northern Mesopotamia and date back to the end of the 8th millennium BC (Braidwood *et al.* 1981; Pernicka 1990). It is usually assumed and in fact it has been demonstrated (Yalçin & Pernicka 1999) that native copper was used during these times. Native copper occurs as rare component in some copper ore deposits and it was utilised by man for making metal objects before the 5th millennium BC. The earliest reliable evidence for copper smelting is dated to the 5th millennium BC and is found at sites related to the Late Ubaid period in Mesopotamia (Zwicker 1977; Hauptmann 1982; Seeliger *et al.* 1985) and Late Neolithic and Chalcolithic sites in Iran (Pigott 1999).

Recently, copper beads and their fragments as well as some pieces of copper ore minerals - malachite and azurite - were excavated in Armenia in the Neolithic settlement of Aratashen. They date to the early 6th millennium BC and thus represent as yet the earliest appearance of copper in the southern Caucasus (Fig. 3). These beads and fragments weighing from 0.37 to 0.72 g each (total weight 12.55 g) were made of a copper sheet rolled up around a stem. These objects are similar to the rare metal finds known from Neolithic sites in northern Mesopotamia and eastern Anatolia from the beginning of the 8th millennium to the end of the 6th millennium BC. The beads of Aratashen turned out to be pure copper and we assume that native copper was used for making these objects, as smelted copper usually contains higher concentrations of impurities like arsenic, antimony and cobalt. This point of view is in good agreement with other contemporary copper objects in the Near East (Yalçin & Pernicka 1999; Schoop 1995).

The transition to extractive metallurgy between the 5th and 4th millennium BC is also evident in the territory of Armenia. The metal objects from the Eneolithic settlement of Teghut, analysed in 1980 (Gevorgyan 1980) turned out to be made of arsenical copper (3 – 4.6 % As) and date back to the late 5th millennium BC. Several small pins made of arsenical copper originate from the recently excavated Late Eneolithic settlement in Nerqin Godedsor and date to the early 4th millennium BC (Fig. 4). The composition of these pins is similar to the objects from Teghut, arsenical copper with an arsenic content ranging from 4 to 5 %.

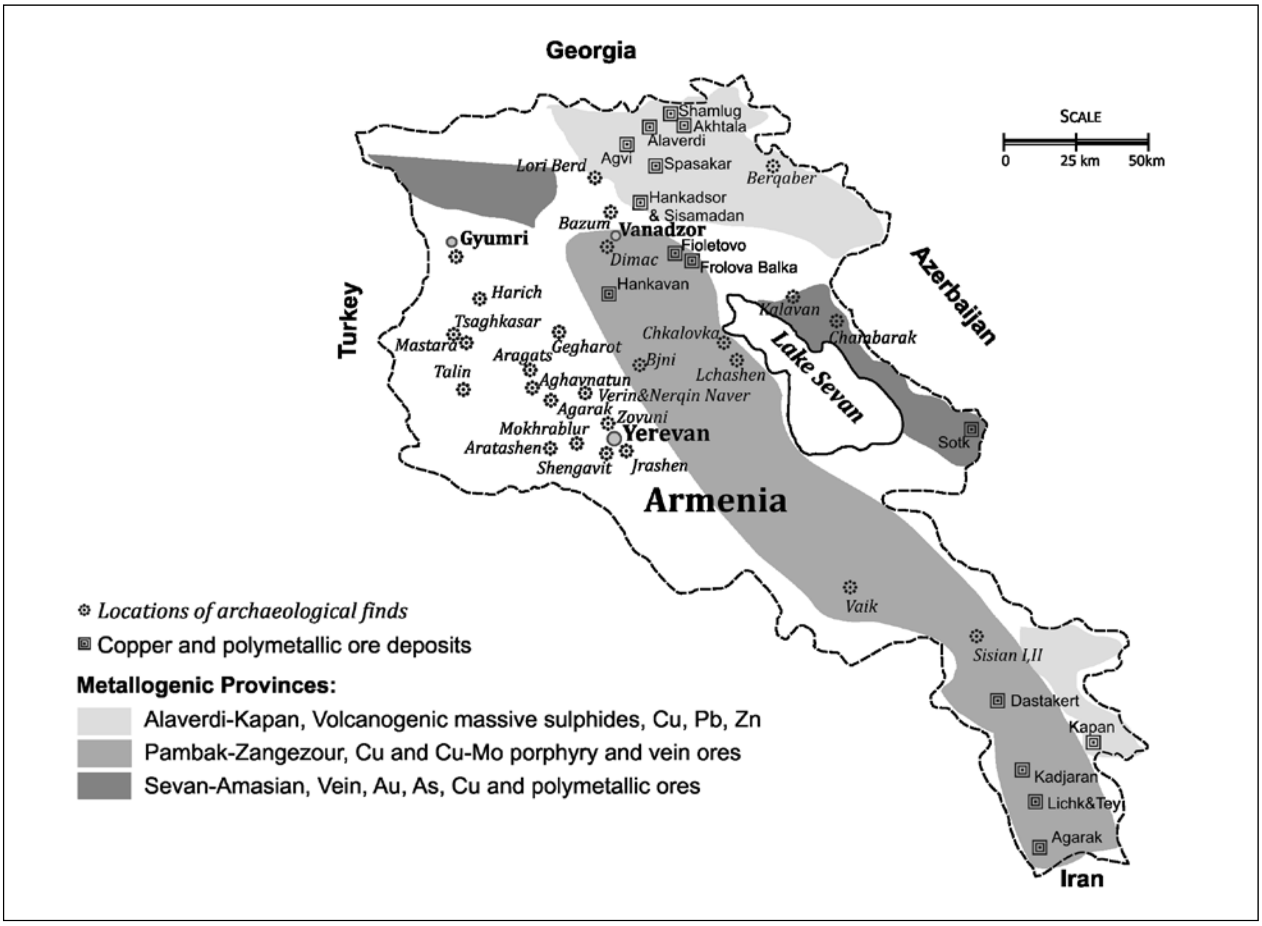

Fig. 1: A schematic map of the Republic of Armenia, showing the metallogenic provinces, the origin of the archaeological artefacts analyzed in this study and the locations of the main copper deposits.

Fig. 2: List of periods and related chemical groups of studied artefacts.

Periodisation		Key Sites	Chemical groups of studied artefacts
Neolithic	Early 6th millenium BC	Aratashen, Aknashen	Pure Cu (native Cu), Malachite and azurite finds
Eneolithic	Middle 5th - Early 4th millenium BC	Teghut, Nerqin Godedsor	Cu+As alloys
Early Bronze Age	Middle 4th - Late 3th millenium BC	Shengavit, Talin, Mokhrablur, Tsaghkasar, Harich, etc.	Cu, Cu+As, Cu+As+Pb, Cu+Sn, Cu+Sn+As
Middle Bronze Age	Late 3th - Middle 2nd millenium BC	Verin Naver, Lchashen, Lori - Berd, Aghavnatun, Sisian I, etc.	Cu, Cu+As, Cu+As+Pb, Cu+Sn, Cu+Sn+As, Cu+Sn+Pb, Cu+Zn
Late Bronze Age	Middle 2nd - Late 2nd millenium BC	Lori - Berd, Gegharot II, Talin II, Bjni, Sisian II, Mastara, Lchashen, etc.	Cu, Cu+As, Cu+As+Pb, Cu+Sn, Cu+Sn+As, Cu+Sn+Pb, Cu+Sb, Cu+Sb+Pb, Sn, Pb, Sb, Sb+Pb

Fig. 4: Pins from the Late Eneolithic site of Nerqin Godedsor, arsenical copper, early 4th millennium BC.

Fig. 3: Fragments of the bracelet of Aratashen, Neolithic, early 6th millennium BC.

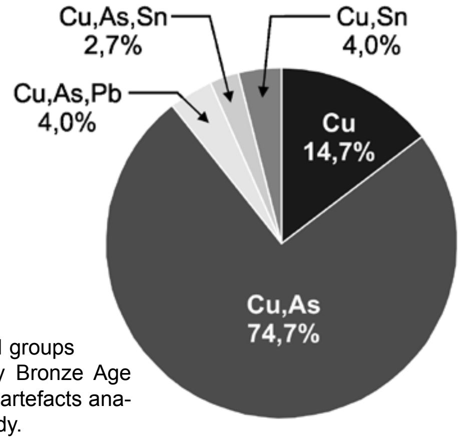

Fig. 5: Chemical groups among 80 Early Bronze Age copper – based artefacts analyzed in this study.

Widespread Use of Copper and the Appearance of Tin Bronzes: The Early Bronze Age

In the Early Bronze Age the use of copper becomes more regular in the Kura-Araxes culture that lasted from the mid 4th until the last quarter of the 3rd millennium BC (Chernykh 1992: 57; Dshaparidse 1995a: 57). It covers wide territories of the Armenian highlands, the Caucasus and Anatolia, reaching the Levant and the west-central Zagros.

Armenian Early Bronze Age copper based artefacts can be classified into five main groups according to their chemical compositions (Fig. 5): Unalloyed copper, arsenical copper, copper-arsenic-tin alloys, copper-arsenic-lead alloys and tin bronze. In addition to the five main groups the trace element concentrations of the artefacts helped to reveal a group characterised by high nickel and silver contents (Fig. 6). Artefacts with similar characteristics are also known from the “Royal Tomb” of Arslantepe. (Hauptmann *et al.* 2002).

High As/Sb ratios are generally characteristic of most of the analysed artefacts (Fig. 7). The majority of the analysed Early Bronze Age artefacts consist of arsenical copper and this is one of the most important features of artefacts from Armenia and the southern Caucasus as has been noted by many scholars who studied prehistoric metallurgy of the region in 20th century, namely E. Chernykh, I. Selimkhanov, T. Abesadze, A. Gevorgyan and others.

Fig. 6: Plot of Ag versus Ni for Armenian copper ores and copper based artefacts. Some Early and Middle Bronze Age artefacts exhibit high concentrations of nickel and silver. In the Late Bronze Age the High Ni and Ag group disappears.

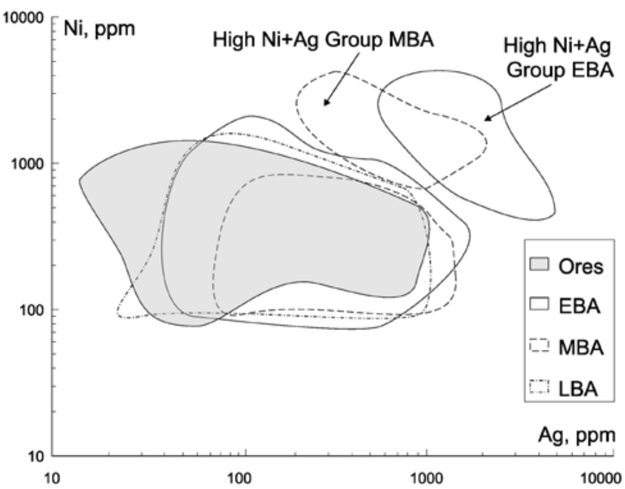

Fig. 7: Plot of As versus As/Sb ratios in Armenian copper ores and copper-based artefacts. Most Early Bronze Age artefacts seem to be enriched in arsenich compared with the copper ores. Also Middle Bronze Age artefacts show some enrichment of arsenic and antimony but it is less pronounced than in the Early Bronze Age. In Late Bronze Age very low As/Sb ratios are generally characteristic for most of the analysed artefacts.

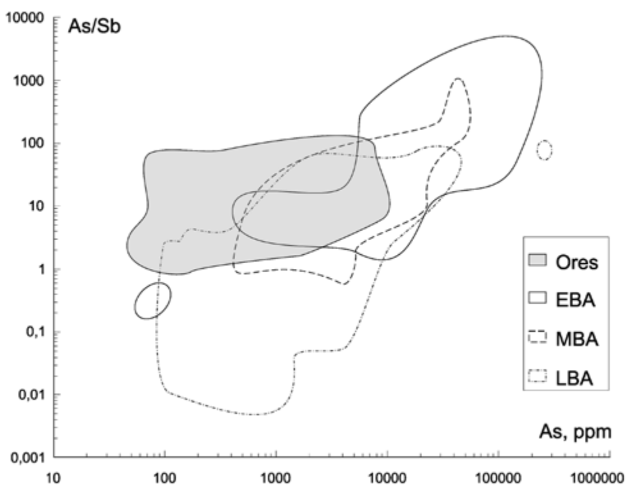

Fig. 8: The necklace of Gegharot, Early Bronze Age, Kura-Araxes culture.

Some alloys exhibit extremely high arsenic contents ranging from 15 to 20 % as revealed by analyses of the metal parts of the Gegharot necklace (Fig. 8). It turned out that different beads are not identical in their chemical compositions. Therefore it can be assumed that various metal parts of the necklace were intentionally made of different alloys. Presumably the ancient craftsman used alloys with different colours to give the necklace an extraordinary, "precious" appearance and emphasize its high artistic value.

Of particular interest is an object consisting of tin bronze from an Early Bronze Age tomb from Talin, a spiral ring containing 11 % Sn. This find is dated to Early Kura-Araxes culture and belongs to the earliest known tin bronzes in the Caucasus, Anatolia and the Aegean. Two spearheads from Talin contain 0.71 and 1.5 % Sn, respectively.

Another interesting and unusual type of metal used in Early Bronze Age Armenia is a lead-arsenic-copper alloy. Some artefacts from Harich and Gegharot exhibit lead concentrations ranging from 3.7 to 9.1 % and the arsenic contents range from 3.2 to 13 %. Lead is often associated with copper ores and is a quite common trace element in copper based artefacts. But as the lead concentrations in the objects from Harich and Gegharot are relatively high, we assume that lead was intentionally added. In the Early Bronze Age alloying with lead is rare but has been assumed for some artefacts in the Aegean and other regions of Mediterranean, however, Aegean copper-lead alloys are not high in arsenic (Pernicka *et al.* 1990).

Advanced Metallurgy and Alloying: Middle and Late Bronze Age

The beginning of the Middle Bronze Age (3rd millennium BC) in Armenia is marked by an increase of individual burials in kurgans. This period is called "Early Kurgan Culture" (Dshaparidse 1995b: 81; Avetisyan *et al.* 2000: 23) and in northern Armenia it was replaced by the Trialeti-Vanadzor culture (Chernykh 1992: 110; Kushnareva 1997: 92), which lasted until the end of the Middle Bronze Age. At the beginning of the 2nd millennium BC the Sevan-Artsakh culture appeared in southern Armenia (Kushnareva 1997: 114; Avetisyan *et al.* 2000: 23). While the deceased of the Trialeti-Vanadzor culture were buried in kurgans with rich grave goods, the burials of the Sevan-Artsakh culture were in flat graves with few metal objects, mostly spearheads, daggers, bracelets and pins.

The Late Bronze Age began in the middle of the 2nd millennium BC with the appearance of the Lchashen-Metsamor culture (Avetisyan *et al.* 2000: 22), which lasted until the rise of the Urartian empire in the 8th century BC. It covered wide territories of the Armenian highland and the southern Caucasus. Typical metal finds with significant parallels in the Caucasus, Anatolia and the Near East are Caucasian axes, flat axes, daggers and swords, spearheads and decorative objects like amulets, bracelets and pendants.

Based on their alloy types (Fig. 9), the Middle Bronze Age artefacts can be divided into six groups: unalloyed copper, arsenic copper, tin bronzes, copper-tin-arsenic alloys, copper-tin-lead alloys and one copper-zinc-lead object. Unalloyed copper and arsenic copper were still in use, but it is obvious that tin became more common

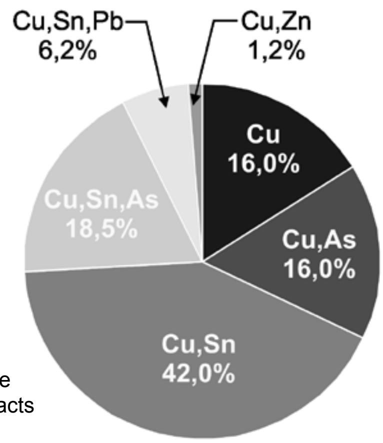

Fig. 9: Chemical groups among 81 Middle Bronze Age copper based artefacts analyzed in this study.

Fig. 10: Pin from Vaik containing 7,9 wt-% Zn, Middle Bronze Age.

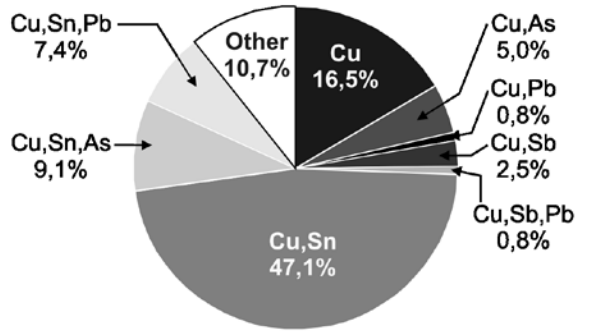

Fig. 11: Chemical groups among 121 Middle Bronze Age copper based artefacts anlyzed in this study.

as alloying metal for copper (Fig. 10). An object consisting of copper-zinc-lead, a pin containing 7.9 % Zn and 1.3 % Pb, was found in a tomb in the cemetery of Vaik together with a dagger and several pins of tin bronze. The tomb dates to the 20th century BC. There are only few brass objects known in the Near East before the first millennium BC, apart from Tepe Yahya (1700 BC, S-Iran), Nuzi (1700 BC, Iraq, Euphrat), Ugarit (1400 BC, Syria) (Thornton & Ehlers 2003: 4) and Thermi on Lesbos (Begemann *et al.* 1995).

In the Late Bronze Age the number of alloys used increases (Fig. 11). Antimony appears as alloying metal. According to their chemical compositions, Late Bronze Age copper-based artefacts can be classified into eight main groups: unalloyed copper, arsenical copper, tin bronzes, copper-tin-arsenic alloys, copper-tin-lead alloys, copper-lead alloys, copper-antimony alloys and copper-antimony-lead alloys. Although unalloyed copper and arsenical copper are still in use, the majority of the Late Bronze Age artefacts consist of copper-tin alloys. It is obvious that tin replaces arsenic as majoralloying metal for copper during Middle and Late Bronze Age. In the Early Bronze Age, alloys with a high arsenic content were often used. In comparison, the number of artefacts with a high arsenic content decreases in Middle and Late Bronze Age. It is noteworthy that most of the Middle and Late Bronze Age copper-based artefacts with high arsenic contents are decorative objects as it was observed also for the Early Bronze Age (Fig. 12).

Apart from the copper-based alloys, also unalloyed lead, unalloyed antimony, antimony-lead alloys and unalloyed tin were used in the Late Bronze and the Early Iron Age. Antimony objects are quite abundant in the southern Caucasus. For example five antimony buttons were discovered in a burial of the 9th century BC in Chambarak, on the north-west coast of Lake Sevan (Meliksetian *et al.* 2003b: 311). Three of them consist of unalloyed antimony and two are antimony-lead alloys. A bracelet from Late Bronze Age Talin consists of unalloyed lead.

In a tomb of the Late Bronze Age cemetery of Lchashen four small pendants were discovered (Fig. 13). It turned out that these four objects consist of unalloyed tin (with only 0.5 % impurities).

Similar to the Early Bronze Age, two groups can be identified in the Ni/Ag-diagram (Fig. 6) for the Middle Bronze Age. The first group is generally matching with Armenian ores and the other group shows higher nickel and higher silver contents than the Armenian ores. In comparison with the Early Bronze Age the Middle Bronze Age artefacts of the High Ni/Ag group exhibit a lower Ag content. In the Late Bronze Age the High Ni/Ag group disappears. A possible explanations of this remarkable feature is that most probably the role of local ores for metal production increased during the Late Bronze Age.

Middle Bronze Age artefacts show some enrichment of arsenic and antimony compared to the Armenian ores but it is less pronounced than in the Early Bronze Age (Fig. 7). The As/Sb ratios of most of the Middle Bronze Age artefacts are mostly lower than those of the Early Bronze Age artefacts and match partly with Armenian copper ores. In Late Bronze Age very low As/Sb ratios are generally characteristic for most of the analysed artefacts. The exception is a special copper-arsenic alloy, which was used for bi-metallic objects. These three objects were found in a royal tomb of the Lori Berd cemetery and date back to the 12th century BC (Fig. 14). The black shining metal consists of a copper-arsenic alloy with an arsenic content between 24.2 and 27.6 %. The bronze coloured metal is classic tin bronze with 9.2 to 10.2 % Sn. Considering the fact that arsenic is highly volatile at the temperatures of copper smelting, it is obvious, that ancient craftsmen had an advanced special knowledge to produce copper-arsenic alloys with such a high As-content.

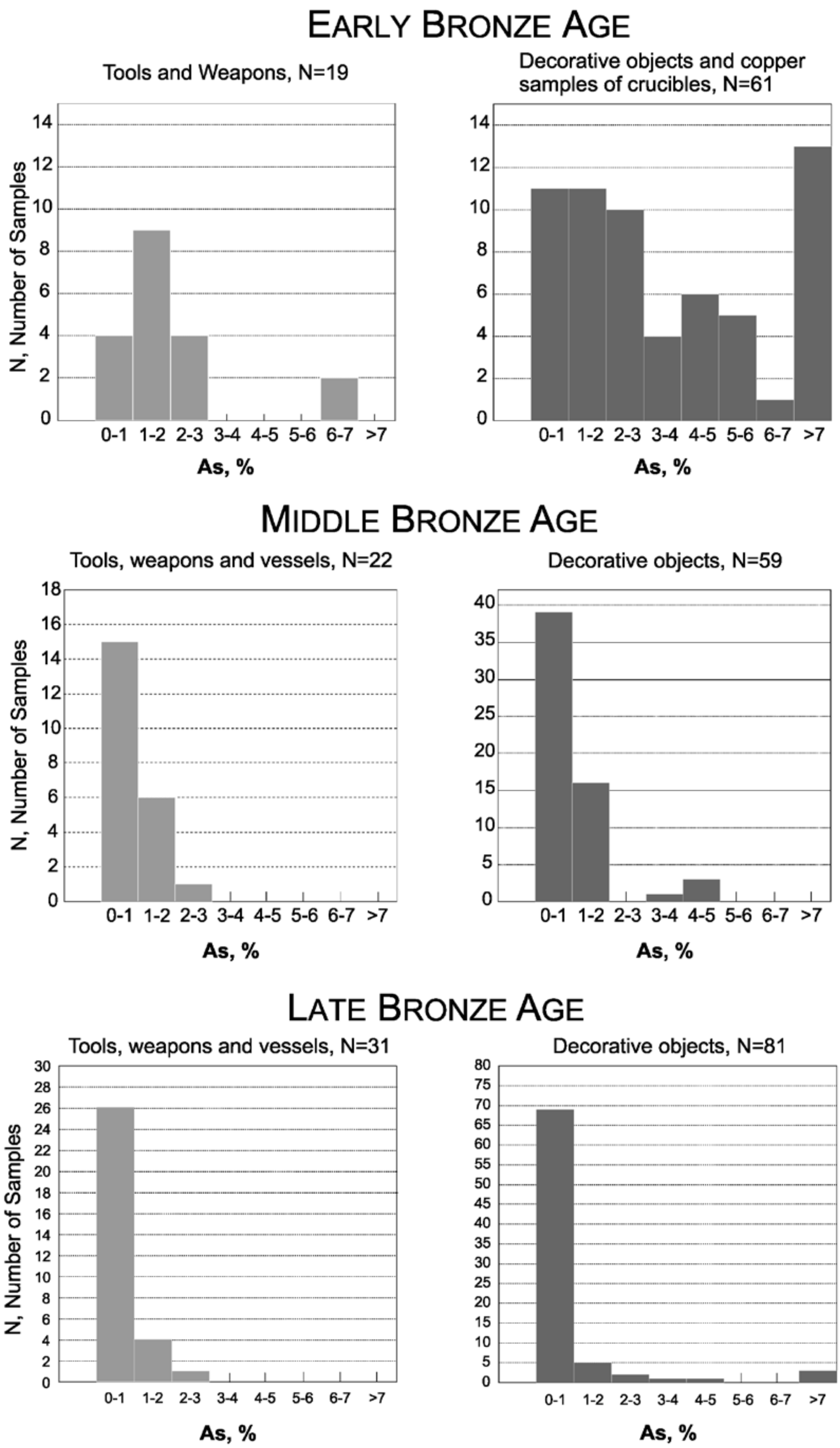

Fig. 12: Statistical distribution of arsenic concentrations in Armenian artefacts. In general, high arsenic contents are typical for decorative objects, rather than for tools and weapons.

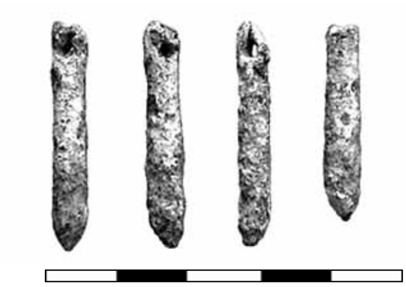

Fig. 13: Pendants from Lchashen, unalloyed tin, Late Bronze Age.

Fig. 14: Metal artefacts from tomb 29, Lori Berd, 12th century BC. Three objects are bimetallic. The black shining metal is made of a copper-arsenic alloy with an arsenic content up to 27,6 wt.%.

Lead Isotope Compositions of Ores and Analysed Artefacts

It should be mentioned that many of the large copper deposits in Armenia were exploited from antiquity until the 19th century (Goginyan 1964). Some of the deposits are still in operation today. In most cases the intense exploitation of the copper ores in Armenia destroyed the oxidation zones of the deposits as well as any remains of prehistoric mining. Under these circumstances geochemical investigations, particularly the lead isotope analysis of copper-based artefacts and their comparison with copper ores, may be helpful to reveal the ore sources for the early stages of metallurgy on the territory of Armenia.

It turns out that a large variation of lead isotope ratios is typical for Armenian copper ore deposits. Presumably the complex geological structure of the territory of Armenia with different tectonic units (fragments of continental plates, ocean crust and island arcs) affects the lead isotope signature of regional ores (Meliksetian *et al.* 2007). Some of the copper deposits also contain uranium minerals. Of course this makes the interpretation of lead isotope ratios of artefacts and their comparison with ores more complex and precludes definitive statements about the sources of copper in the early stages of metallurgy in Armenia. Nevertheless, some relationships between isotope compositions of Armenian ores and some artefacts can be observed. Three groups of Armenian ores have been defined based on their lead isotope ratios: “radiogenic”, “ordinary” and “old” lead (Meliksetian *et al.* 2003: 603). The “ordinary” lead overlaps with the Anatolian lead isotope field, generally matching the isotope composition of the ores of the Pontides. Parts of the “radiogenic” and “old” lead groups are located outside the “Anatolian” field demonstrating wider variations of lead isotope compositions in Armenian ores.

The comparison of artefacts and ores (Fig. 15) suggests a significant probability that north Armenian copper ores and/or isotopically similar ores from eastern Turkey could be related to some of the analysed artefacts from the Early Bronze Age settlements in the Ararat valley, the Shirak plateau and northern Armenia. This conclusion is based on the isotope composition of artefacts matching the “ordinary” and “radiogenic” lead groups of Armenian ores as well as on their trace element composition which generally fits Armenian ores. These artefacts match the “ordinary”, “radiogenic” and “Anatolian” compositional fields and it can be assumed that they were produced from local copper ores. On the other hand, some artefacts do not match Armenian and Anatolian ores isotopically and were most likely imported. Three axes from Gyumri with high concentrations of nickel and silver also exhibit unusually high $^{208}Pb/^{206}Pb$ and $^{207}Pb/^{206}Pb$ and low $^{206}Pb/^{204}Pb$ isotope ratios signifying the import of copper, possibly from sources in Jordan or Oman with generally fitting $^{208}Pb/^{206}Pb$ and $^{207}Pb/^{206}Pb$ ratios.

The lead isotope signatures of copper based alloys used in the Middle Bronze Age show similar wide variations like the Early Bronze Age artefacts. The lead isotope signatures of the majority of the Middle and Late Bronze Age artefacts generally match local ores – e.g. Armenian and Anatolian ores compositional fields. It is noteworthy, that in comparison to the Middle Bronze Age the Late Bronze Age artefacts show a little (bit) lower

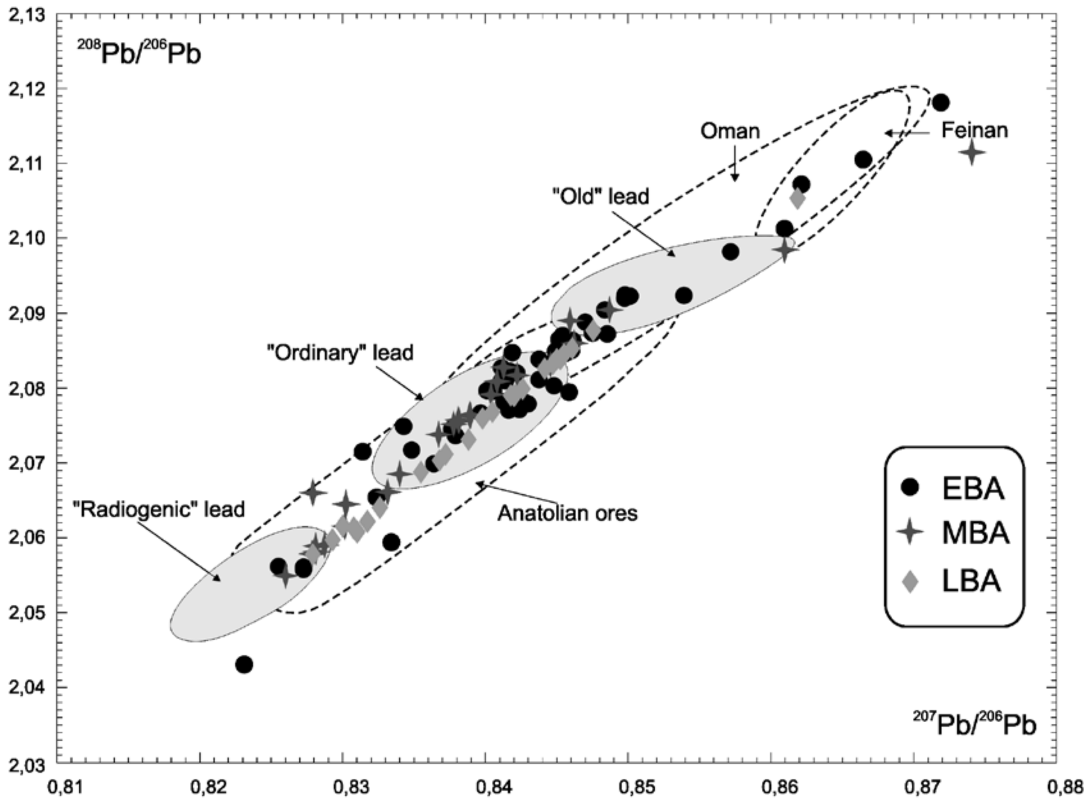

Fig. 15: Diagram of $^{207}Pb/^{206}Pb$ vs. $^{208}Pb/^{206}Pb$ in analysed copper based artefacts combined with lead isotope fields of Armenian ores: 1. "radiogenic" lead group, 2. "ordinary" lead group, 3. "old" lead group. Dotted lines outline the lead isotope fields of ores from Anatolia, Oman and Feinan, Jordan.

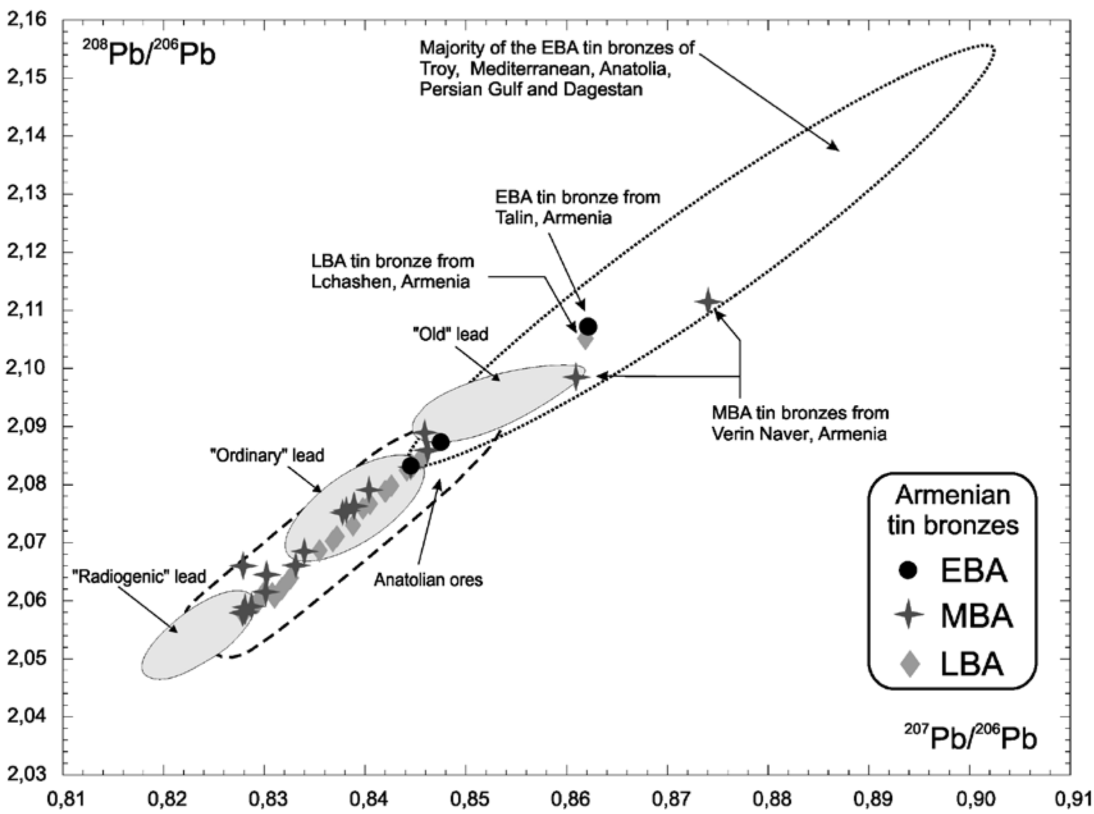

Fig. 16: Diagram of $^{207}Pb/^{206}Pb$ vs. $^{208}Pb/^{206}Pb$ in the analysed Early, Middle and Late Bronze Age tin bronze artefacts combined with isotope fields of Armenian ores. The isotope signatures of the Early Bronze Age tin bronze from Talin, of the two Middle Bronze Age tin bronzes from Verin Naver and of one Late Bronze Age tin bronze from Lchashen are clearly separated from the rest of the Middle and Late Bronze Age tin bronzes.

$^{208}Pb/^{206}Pb$ ratios. This indicates a change of metal sources in the Late Bronze Age.

The Early Bronze Age tin bronze from Talin is characterised by high silver and nickel contents and unusual lead isotope ratios. The lead isotope ratio of the tin bronze from Talin is comparable to most contemporaneous tin bronzes from Troy, other sites in the Aegean the Persian Gulf and Dagestan (Fig. 16). However, the provenance of these tin-copper alloys remains so far unknown. It is noteworthy, that only two Middle Bronze Age tin bronzes from Verin Naver and one Late Bronze Age tin bronze from Lchashen artefacts exhibit lead isotope signature similar to these Early Bronze Age tin bronzes. They might have been imported from rather distant regions. For the most Middle and Late Bronze Age tin bronzes there are several isotopically matching copper ore sources in Armenia. Considering lead isotope compositions of artefacts so far analysed, it can be assumed that an unknown source of tin bronze in Early Bronze Age was present, but in Middle and Late Bronze Age locally produced copper was alloyed with imported tin.

Conclusions

The early use of native copper (early 6th millennium BC) and early extractive metallurgy (late 5th millennium BC) are evident in the territory of Armenia. The Early Bronze Age is marked by a widespread use of arsenical copper, unalloyed copper, rare tin bronzes and some other alloys. Some objects, extremely high in arsenic (15 – 20 %), provide evidence of an advanced alloying technology. The majority of the analysed artefacts could derive from local ore sources, abundant in the territories of Armenia and eastern Turkey. In the Middle and Late Bronze Age the number of alloys used increased. Early brass appeared in the Middle Bronze Age, unalloyed tin, lead and antimony were used in the Late Bronze Age. Alloys, extremely high in arsenic (24.2 – 27.6 %) are still present in some extraordinary artefacts. Some of the artefacts high in nickel and silver exhibit unusual lead isotope signatures and were most likely imported, possibly from Oman. The Early Bronze Age tin bronze from Talin has lead isotope ratios typical for most of the 3rd millennium BC bronzes, signifying an origin of the early tin bronzes from an unknown copper/tin ore source, presumably of Proterozoic – Early Palaeozoic age.

In the Middle and Late Bronze Age tin bronzes exhibit lead isotope fingerprints typical for local ores with just few exceptions, suggesting that in Middle and Late Bronze Age local copper was alloyed with imported tin as lead isotopes of bronzes inherit those of copper sources rather than tin.

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