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Prehistoric copper in Bulgaria

Its composition and provenance

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Introduction

South-eastern Europe has yielded a substantial number of early copper and gold artifacts that quantitatively exceeds by far all contemporaneous inventories of other regions. Most of these metal objects date from around the middle of the 5th until the first half of the 4th millennium B.C.[1] Altogether, the finds known today from this region, including Hungary, add up to about 4,700 kg copper and more than 6 kg of gold. Already last century, this immense abundance of metal from periods "before the Bronze Age" has led to the suggestion that a separate period be introduced between the Neolithic and the Bronze Age, namely the "Chalcolithic" or "Copper Age"[2]. Due to the lack of equivalent copper finds this concept has never received full acceptance in western and northern Europe, however, where indeed it is in dispute even today[3]. Possibly, this is the reason why, in the prehistory of south eastern Europe, "Eneolithic" is nowadays the much more common term. It would appear, however, that "Eneolithic" and "Chalcolithic" are used in the same way and can thus be considered to be synonymous. From a minimalist point of view they simply denote a chronological stage between the Neolithic and the Bronze Age that is characterized by the more or less regular use of copper[4]. There seems to be a growing consensus, however, that this period actually represents a specific era of cultural history between the Neolithic and the Bronze Age, with specific modes of food production[5], social structure as indicated by settlement patterns, burial customs, and the exchange systems of material goods.

The reconstruction of production and exchange systems in prehistory is essentially based on typology. Sources of raw materials are usually sought within the distribution areas of certain types of objects and it is often implicitly assumed that the geographically nearest occurrences should also have been utilized as sources of these raw materials. This basic model can relatively easily be tested with scientific methods by comparing the chemical and/or isotopic composition of the artifacts with those of the supposed source materials. So far this approach has resulted more often in a rejection than a confirmation of the simplistic standard geographical-proximity paradigm and it has proved necessary to modify existing archaeological models. A typical example was the finding that the majority of north-east Aegean Early Bronze Age copper and bronze objects do not derive

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[1] All dates are calendar dates, i.e. radiocarbon dates are dendrochronologically corrected.

[2] von Pulszky 1884, Much 1886.

[3] Lichardus 1991.

[4] There exist, however, considerable differences as to which cultures should be summarized under this term. In this article we largely follow the usage in Bulgaria where the Eneolithic is considered to begin with the first appearance of heavy copper implements, while researchers in former Yugoslavia prefer to speak of the Eneolithic only when copper objects become abundant (e.g. Tasič 1995).

[5] Sherrat 1983.

from presently known Aegean ore sources[6]. This made much less convincing all suggestions that the remarkable cultural development of the Aegean during the early third millenium B.C. should have been owing to the invention of tin bronze in this region[7]. Actually, for some bronze objects there was, and still is, no ore deposit at all known in Anatolia and Southeastern Europe that could possibly have yielded their copper; this metal may well derive from very distant regions.

The same approach has been used to investigate the role of the Eneolithic copper mine at Rudna Glava in eastern Serbia as a possible source of copper in the late Vinča period[8]. Again the result was surprising in that not a single one of the Serbian metal objects studied could be ascribed to Rudna Glava. In this case, however, the consequences for the preferred archaeological model were less severe than in the case of the Aegean Early Bronze Age because most artifacts could be related with high probability to another, actually much larger, copper ore deposit within the same region (Majdanpek). Thus, there was no need to modify the widely accepted model that much of the copper used in the time of late Vinča and Bodrogkeresztúr in the central Balkans was supplied by ore deposits from eastern Serbia.

An extension of this study to Bulgaria appeared desirable for several reasons. First, in Bulgaria most Eneolithic metal finds derive from controlled excavations so that their chronological context is rather well known. This was not so for the majority of the Serbian artifacts because they were mostly surface finds for which little detailed information on the actual find context existed and which were only roughly dated by typology. Secondly, some Eneolithic cultural complexes (Vinča, Krivodol-Sălcuţa-Bubanj Hum) extended over parts of present-day Bulgaria so that a complete evaluation of Rudna Glava's role is only possible when the Bulgarian finds are included. Finally, an important motivation for the present investigation was, of course, the existence at Ai Bunar near Stara Zagora[9] of a further Eneolithic copper mining region that actually appears to have been much larger than Rudna Glava. The total amount of copper ore extracted from Ai Bunar in the Eneolithic has been estimated to range between 2,000 and 3,000 tons which may have yielded around 500 tons of copper[10]. There are no such estimates for Rudna Glava by the excavators, but judging from published plan sketches and visual appearance the ore tonnage seems to have been at least an order of magnitude smaller there. For Majdanpek such an estimate is impossible because any potential prehistoric exploitation has been completely obliterated by modern open pit mining; its possible role as supplier of Eneolithic copper is inferred from the agreement of the lead isotope signatures[11].

Various types of Eneolithic heavy implements exhibit distinctly different geographic distributions[12]. According to Černych[13] differences in typology are accompanied by differences in material properties such as the concentrations in the implements of trace elements like, e.g., arsenic, antimony, nickel, silver, and gold. Based on results obtained by optical emission spectroscopy of copper-based objects Černych defined his Balkano-Carpathian metallurgical province, with several sub-provinces[14]. This concept was adop-

[6] Pernicka u. a. 1984; Pernicka u. a. 1990; Begemann u. a. 1992.
[7] Renfrew 1967; Branigan 1974.
[8] Jovanovič 1976, Pernicka u. a. 1993.
[9] Черных 1978, 56, 76.
[10] Černych 1988.
[11] Pernicka u. a. 1993.
[12] F. Schubert 1967; Todorova 1981 a.
[13] Черных 1978.
[14] Chernykh 1992, 48–53.

ted by Todorova[15] who complemented the previous work with a detailed typological study and defined three metallurgical sub-provinces in southeastern Europe, namely Transylvania, Serbia with Bosnia and parts of Dalmatia, and Thrace. Of these, Thrace was thought of as the distribution area of ores from the Sărnena Gora (with Ai Bunar) while Rudna Glava and other deposits in eastern Serbia were to have yielded the metal of the objects in Serbia, Bosnia and the coastal parts of Dalmatia. The presumed border line was to have run roughly north-south somewhere east of Sofia. It was one of the aims of the present study to test the hypothesis of this presumed different regional and metallurgical orientation of western Bulgaria versus the central and eastern parts of the country.

Finally, due to its geographical location one expects an intense cultural interaction to have occurred between the territory of present Bulgaria, on the one hand, and Anatolia or the Aegean on the other that might be identifiable in the composition of metal artifacts, although the postulated autonomy of the southeast European Copper Age was specifically based on evidence from metal artifacts from Bulgaria. Renfrew[16] has argued that a continuous cultural and technological development was recognizable that suggested a largely independent development based on the use of local resources. But this, of course, does not preclude external contacts and, indeed, these have been demonstrated by the same author by the use of oxygen isotope analyses of dentalium shells from Goljamo Delčevo and Gradešnica[17], two sites that are included in the present study. Recent evidence based on trace element analyses of obsidian suggests, in addition, contacts between central Europe and the Aegean in the Eneolithic period[18]. Thus, in the late fifth millennium B.C. some material exchange across southeastern Europe obviously took place in both directions and it is only reasonable to assume that metal should have been part of this exchange system. It is a fortunate situation that analyses of metal artifacts from Serbia, Greece, and Anatolia are already available[19] that can be used as the basis for an interregional comparison.

Chronology of the use of copper in Bulgaria

Based on numerous stratigraphic sequences the relative chronologies of the Bulgarian Neolithic and Chalcolithic[20] are rather well established *(Fig. 1)*. While there are many connections with prehistoric cultural groups in south-eastern Europe *(Fig. 2)*, those with Anatolia are much more evasive due to the small number of contemporaneous sites in western Anatolia. Only for the Early Neolithic and the Early and the Late Bronze Age have relations been established that are generally accepted. The absolute chronology of the periods central to this study, namely the sequence from the Late Neolithic to the Early Bronze Age, is well defined by large series of radiocarbon dates of samples from stratigraphically clear contexts. Calibrated dates range from the end of the sixth to the end of the fourth millennium B.C.[21] as summarized in *Fig. 3*.

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15 Todorova 1981 a; Тодорова 1986, 144–150; Тодорова 1994.

16 Renfrew 1969.

17 Renfrew/Shackleton 1970.

18 Kilikoglou u. a. 1996.

19 Bilgi 1984; Bilgi 1990; Gale u. a. 1991; Pernicka u. a. 1990; Begemann u. a. 1992; Begemann u. a. 1994.

20 Vajsova 1966; Todorova 1981 b; Тодорова 1986, 26–40; Тодорова /Вайсов 1993, 64–92.

21 Bojadžiev 1986; Bojadžiev 1992; Weninger 1992; Bojadžiev 1995; Mantu 1995; Görsdorf/Bojadžiev 1997.

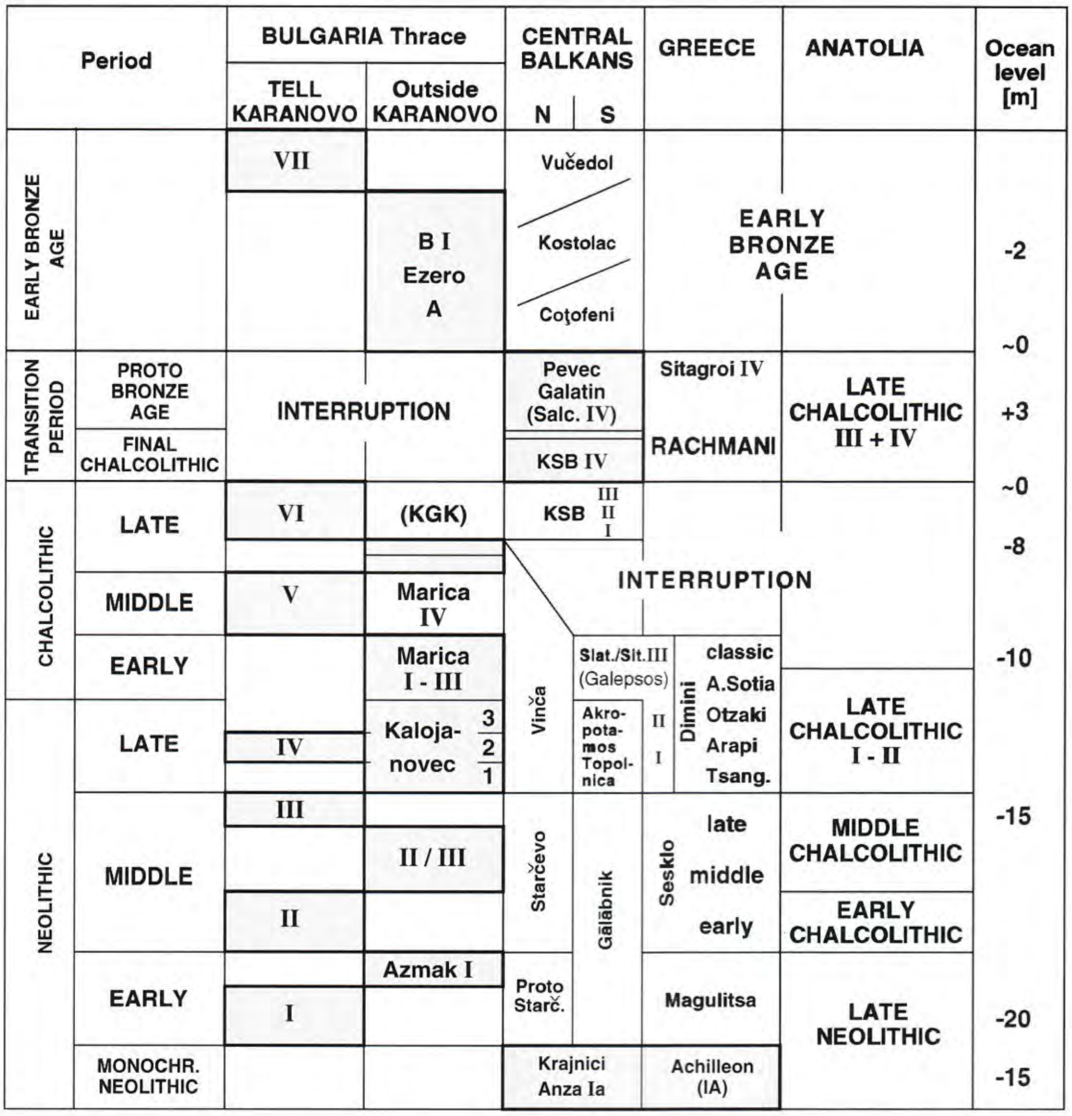

Fig. 1. Relative chronology, from the Neolithic to the Early Bronze Age, within present-day Bulgaria and neighbouring regions.

The earliest finds of malachite and copper from Bulgaria date to the latest stages of the Neolithic. Most of them are beads but also a small malachite pendant and several very small unidentified, but worked objects of malachite were found[22]. Two sets of artifacts belonging to Hamangia IIb derive from the cemetery at Durankulak (see *Fig. 4* for location of sites and regions mentioned in the text). In grave no. 626 a young woman of an age at death of about 20 to 25 years was buried with her ornaments and together with four clay idols[23]. Her splendid necklace of malachite (possibly also copper) beads is the earliest ornament of this type in southeastern Europe. A further metal find from this period is

[22] Тодорова 1973, 29 Fig. 11,1.
[23] Vajsov 1992a, 97 Pl. 1,2.4.5.

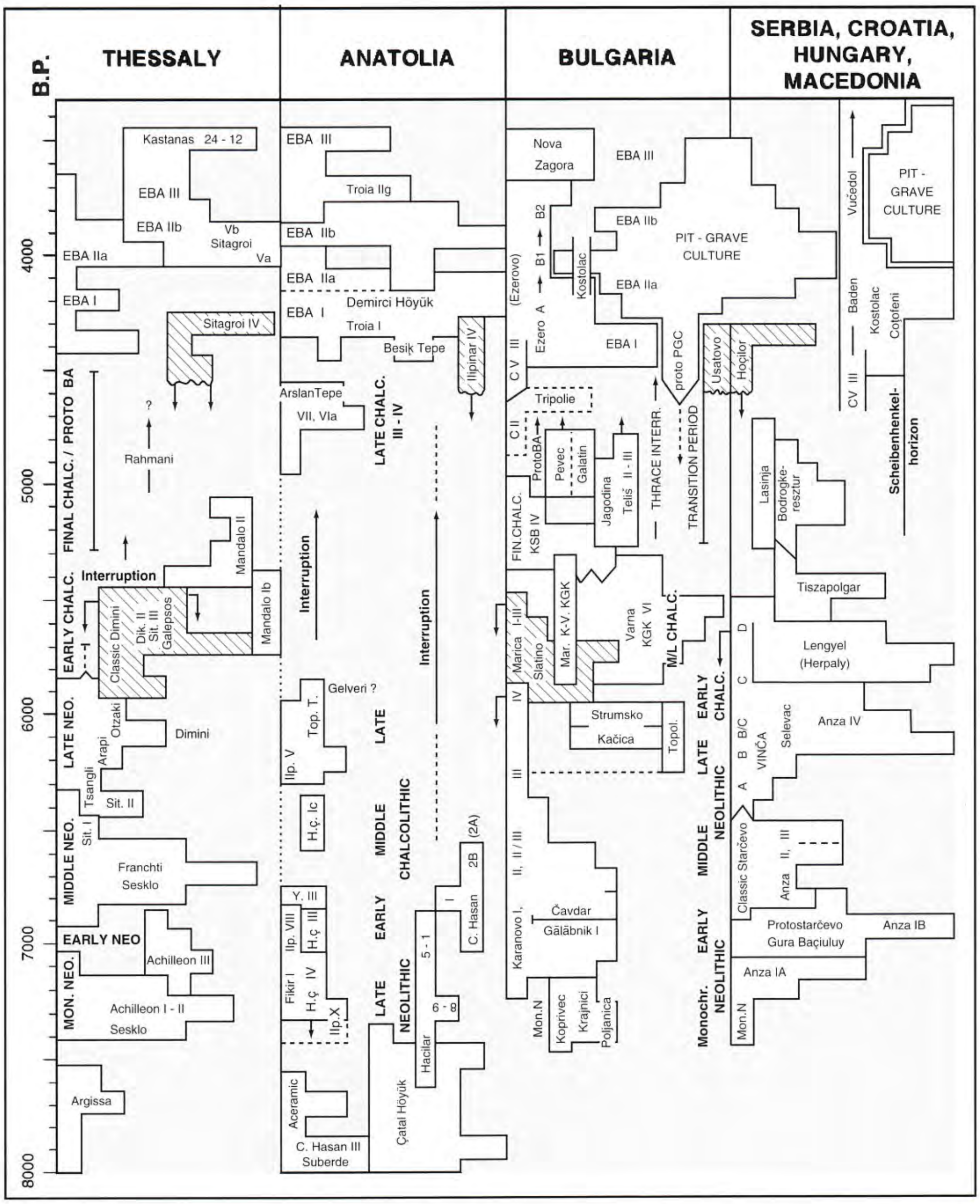

Fig. 2. Summary of uncalibrated radiocarbon dates from southeastern Europe and Anatolia showing the chronological relationships between the two regions. Each radiocarbon date is represented by a white square. The following abbreviations are used: Dik. = Dikili Tash, Sit. = Sitagroi, EBA = Early Bronze Age, Ilp = Ilıpınar, Top. T. = Toprakkale Tepe, H.ç. = Haçilar, C. Hasan = Can Hasan, PGC = Pit Grave Culture; KSB = Krividol-Sălcuţa-Bubanj complex, KGH = Kodžadermen-Gumelniţa-Karanovo complex, K. V. = Karanovo V, Mar. = Marica; Mon. N. = Monochrome Neolithic, Topol. = Topolnica culture.

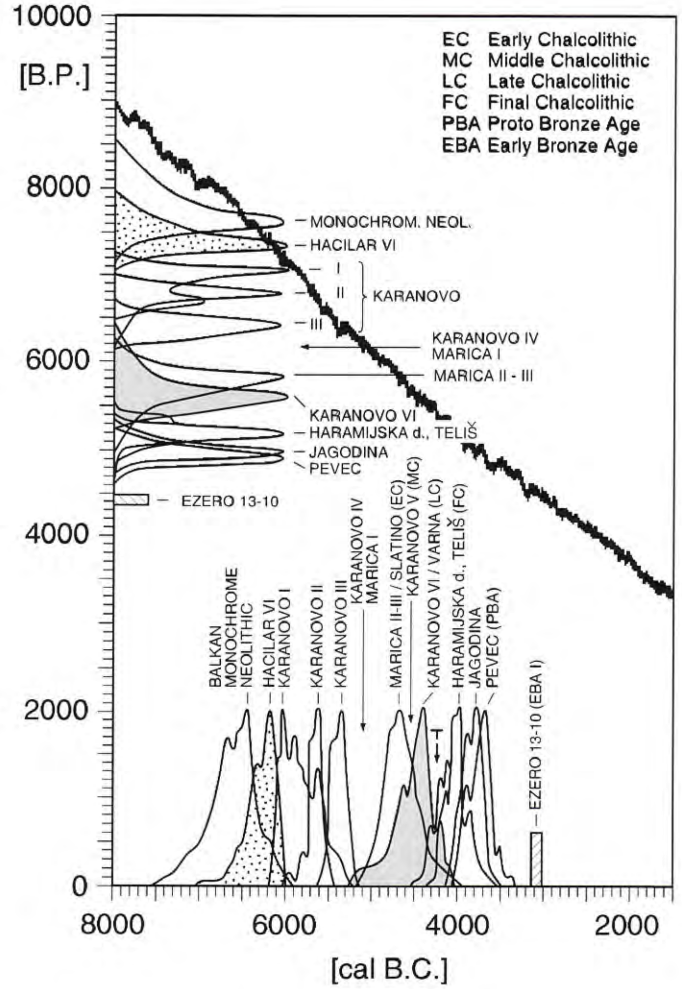

Fig. 3. Calibration of the presently available radiocarbon dates of the Bulgarian Neolithic and Chalcolithic. After B. Weninger, utilizing radiocarbon calibration data of Stuiver u. a. 1993. For a detailed catalogue of many of the radiocarbon dates used here see Görsdorf/Bojadžiev 1997.

a corroded bead from the plateau settlement of Usoe near Asparuchovo, Varna district[24]. Unfortunately, this object is now no more available; it has been used up in the course of Černych's analyses in Moscow. These few metal objects together with similar contemporaneous finds on the Balkan peninsula[25] mark the beginnings of the use of metal in southeastern Europe. Although the dates of several objects of this corpus are not beyond doubt it seems indubitable that with the end of the Neolithic (late Starčevo and Criş, around 5500 B.C.) malachite and copper began to be used for the production of ornaments[26].

In southwest Asia this development took place much earlier, in the Pre-Pottery Neolithic (PPN) of the ninth and eighth millennium B.C. What started this decisive cultural change is not known but there is general agreement that its origin has to be sought in the Fertile Crescent from where it then spread out into other regions. As far as Europe is con-

[24] Тодорова 1973, 27 Anm. 18.
[25] Comşa 1991, 77–79.
[26] Vlassa 1967, 407; Srejović 1969, 173, Niveau III A.

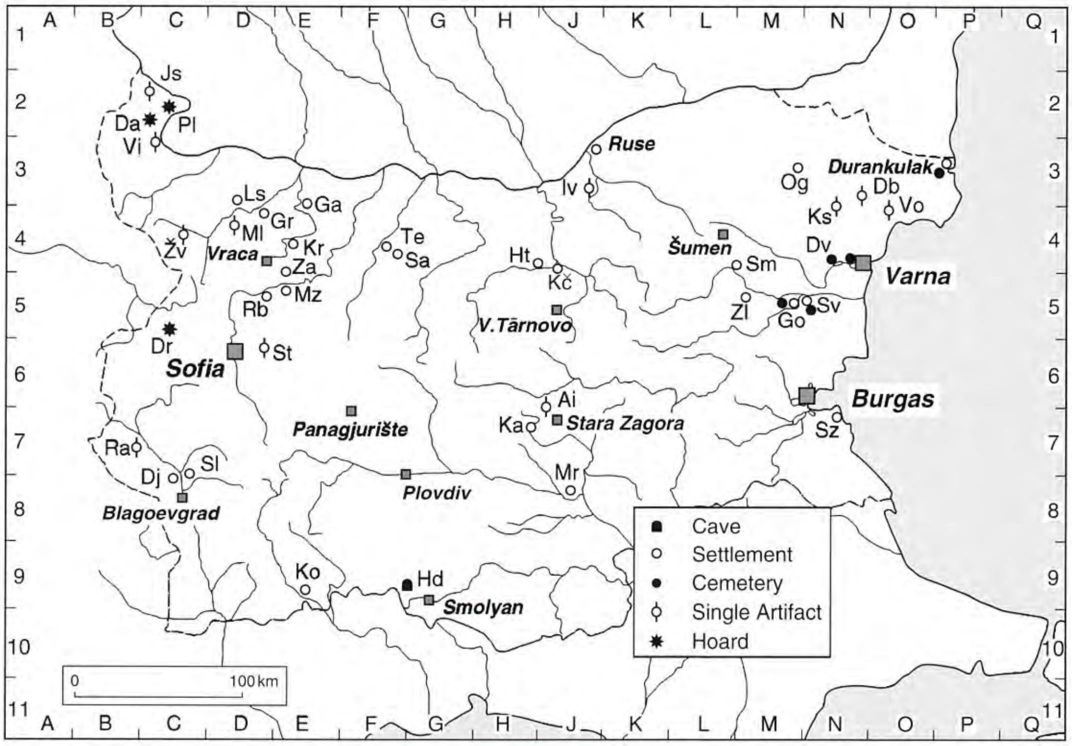

Fig. 4. Find localities of the analyzed Bulgarian artifacts. Abbreviations are as follows (squares on the map denoted by a letter and a number are also given for easy reference):

Ai Ai Bunar (Stara Zagora) J6.
Da Dăržanica (Vidin) C2.
Db Debrene/Prilep (Dobrič) N3.
Dj Djakovo (Kjustendil) C8.
Dr Dragoman (Slivnica) C5.
Dv Devnja (Varna) N4.
- Durankulak P3.
Ga Galiče (Vraca) E3.
Go Goljamo Delčevo (Varna) M5.
Gr Gradešnica (Vraca) D4.
Hd Haramijska Dupka (Smoljan) F/G9.
Ht Hotnica (Veliko Tărnovo) H4.
Iv Ivanovo (Ruse) J3.
Js Jasen (Vidin) C2.
Ka Karanovo (Sliven) H7.
Kč Kačica (Veliko Tărnovo) J4.
Ko Kočan (Goce Delčevo) E9.
Kr Krivodol (Vraca) E4.
Ks Kozlodujci (Dobrič) N3.
Ls Lesura (Vraca) D3.
Ml Malorad (Vraca) D4.
Mr Marica (Haskovo) J8.
Mz Mezdra (Vraca) E5.
Og Onogur (Dobrič) M3.
Pl Plakuder (Vidin) C2.
Ra Radlovci (Kjustendil) C7.
Rb Rebărkovo (Vraca) D5.
- Ruse J3.
Sa Sadovec (Pleven) F4.
Sl Slatino (Kjustendil) C7.
Sm Smjadovo (Šumen) L/M4.
St Stolnik (Sofia) D6.
Sv Sava (Varna) N5.
Sz Sozopol (Burgas) N7.
Te Teliš (Pleven) F4.
- Varna N4.
Vi Vidbol (Vidin) C2/3.
Vo Vodnjanci (Dobrič) O4.
Za Zaminec (Vraca) E4/5.
Zl Zlatarevo (Šumen) M5.
Žv Živlovci (Montana) C4.

cerned the present find situation suggests that this did not coincide with the introduction into Europe of the domestication of cereals and animals at around 6400–6300 B.C. but occurred only a millenium or so later. Indeed, it was this fact together with an apparent large geographical and chronological gap between the Fertile Crescent and the Balkans of the occurrence of early metal finds that led Renfrew to postulate an independent development of metallurgy in southeast Europe[27]. The second argument, however, does not hold any more. New abundant finds of copper objects from the PPN at Asıklı Höyük in central Anatolia and from the Middle Neolithic (Karanovo III) at Asağı Pınar in Turkish Thrace are beginning to fill this seeming gap so that a common origin in the Fertile Crescent of both agriculture and metallurgy can no longer be ruled out. In fact, the spread of metallurgy followed the same pattern and pathways[28] as agriculture, possibly with a certain delay in time. It has also been noted that although copper minerals are bound to have been known and available to early Man they came into use only with the beginning of the Neolithic[29]. Possibly, this has something to do with the symbolic value of the colours green and blue that apparently have played no role in the Palaeolithic but gained their meaning during the Chalcolithic. Even today the blue colour in particular is often connected with magic and religious symbolism and it is certainly no coincidence that one of the hall-marks of the Early Neolithic in the Near East as well as in southeast Europe is the extensive use of green and blue stones for ornaments. The assumption of a common origin of this custom in the Fertile Crescent does, of course, not rule out the use of local resources in southeast Europe. But it would appear somewhat illogical to assume totally different ways of origin and distribution for two cultural achievements like the domestication of plants and metallurgy that have so much in common in the archaeological record.

The beginnings of the south-east European Copper Age proper are best identified with the appearance of the characteristic heavy implements, i.e. with the evidence for casting and a more or less regular metal supply. Accordingly, this period comprises cultures that have so far been regarded as Late Neolithic such as classic Dimini in Thessaly, Maliq Ia - Kamnik in southern Albania, Vinča C in the central Balkans together with the Rast group in the Banat, Gradešnica in northwestern Bulgaria, Marica I–III in northern Thrace, Dikili Tash-Slatino (Sitagroi III) along the lower Strymon river and in Aegean Thrace, Sava I–III along the southwestern Black Sea coast, Hamangia III in the Dobrugea, Boian-Vidra in Muntenia, Poljanica I–III in northeastern Bulgaria, Pre-Cucuteni II in Moldavia etc. All these culture groups have yielded a small number of copper finds, mostly awls, borers, and beads, as well as ornaments of malachite. The earliest heavy axes appear at the end of this period[30] that comprises roughly the second quarter of the fifth millennium B.C.[31] Typologically they seem to follow the earlier prototypes, made of stone, but they were probably cast using very pure copper[32]. The small chisel from Drama that may have been produced by a very peculiar technique[33] seems to date somewhat later.

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[27] Renfrew 1969; Renfrew 1970.

[28] Chernykh 1992, 1–25 Fig. 1.

[29] Weisgerber/Pernicka 1995.

[30] Among others Slatino (HDM 1904), Kamnik (HDM 1455), and Marica (HDM 1912); For Sesklo see Tsountas 1908, 351–352 Fig. 292–293; Todorova 1997.

[31] In this context one has to take into account that the radiocarbon dates of this period overlap with those of the Late Chalcolithic, especially since the ^{14}C content of atmospheric CO_2 varied significantly in the period between 6000 and 4000 B.C. *(Fig. 3)*. Some false chronological associations drawn from these data have survived in the archaeological literature, such as the parallelization of Marica with Gumelniţa, although numerous stratigraphic contexts (Karanovo, Azmak, Kirilovo, Mădrec etc.) clearly contradict such a view. The ^{14}C dates of the two periods can be statistically differentiated as shown by Weninger 1992. In relative chronological terms this period (Early Chalcolithic) ranges between the Late Neolithic and the Middle Chalcolithic (Karanovo V, Fig. 1).

[32] Рындина 1994, 140–142.

[33] Moesta 1989; Moesta 1991.

In south-eastern Europe there are at present altogether about two dozen metal finds known from this period that we shall refer to as *Early Chalcolithic*. The objects have been proposed to be made of native copper that even today can be found in substantial amounts at the surface of some locations like, e.g., at Plakalnica near Vraca, at Boboševo near Kjustendil and others[34]. On the other hand, copper mines have been in operation at Rudna Glava and Ai Bunar during the Early Chalcolithic in the second quarter of the fifth millennium B.C. so that to the extent that mining of copper ores marks the beginning of extractive metallurgy smelted copper will have been available also.

It is remarkable that the Early Chalcolithic is not present at Tell Karanovo in Thrace *(Fig. 1)*. Its relative chronological position between Karanovo IV und Karanovo V is rather defined by the stratigraphy of Kirilovo where Karanovo IV is situated below Marica III, while at the tells of Mădrec, Marica, Kukuva, Račeva, Azmak and others Marica I–III layers are below Karanovo V or Karanovo VI. Therefore, Karanovo V can be regarded as equivalent to the fourth - Middle Eneolithic - sub-phase of the Marica culture.

In southwest Bulgaria the Late Neolithic Akropotamos-Topolnica culture (equivalent to Sitagroi II) in the settlement of Topolnica near Petrič[35] is overlain by a destruction layer of the first phase of the Dikili Tash-Slatino culture (equivalent to Sitagroi III). The same layer was encountered without any destructions at Strumsko near Blagoevgrad[36] while the second and third phases of the Dikili Tash/Slatino culture are known from Slatino[37] with Galepsos and Pseudodimini-Classic ware, from Sitagroi III with graphite-painted Slatino and Galepsos ware[38], from Pevkakia I with Galepsos, Slatino und classic Dimini ware[39]. Furthermore, the typical graphite-painted Slatino ware is identical with that of Marica II–III in Thrace. Pottery exactly matching the scratch-decorated ware from Slatino and Sitagroi III with white incrustations has been found at Gradešnica (northwest Bulgaria), Rast (Banat), Hamangia (Dobrugea), Sava (Kamčija region), Marica I–III (Thrace) and Kableškovo (southwestern Black Sea coast). All these culture groups are known to have used metal, at least to a limited extent. Thus, they form the earliest uniform metal-using horizon with a secure relative chronological position before Karanovo V and we suggest to summarize them under the term Early Chalcolithic of the Balkan peninsula. From this period we had four samples available, one each from Slatino and Marica and two from Durankulak.

The following period, the *Middle Chalcolithic*, is characterized by a substantial increase in metal production. A large number of copper ornaments, the earliest gold ornaments, several heavy axes[40] as well as the earliest hammer axes appear in this horizon. Possibly initiated by intensified contacts a process of integration and unification of the material culture over a large area is observed. The typological uniformity of the finds suggests the formation of large cultural complexes by the end of the Middle Chalcolithic[41].

Climatically this was a period of relatively high average temperatures, especially south of 41° degrees north, the latitude of Istanbul. Possibly, in Anatolia and also more to

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[34] Gaul 1942.
[35] Тодорова 1984a, 19.
[36] Перничева/Кулов 1979, 28.
[37] Čochadžiev 1986a; Čochadžiev 1986b, Čochadžiev 1992.
[38] Keighley 1986, 381, 383, 384, 396 Pl. 80–82; Renfrew 1986b, 482–484.
[39] Weißhaar 1989, Pl. 1–4; 12,4.5.16.23; 35,1; 37; see also Todorova 1989c; Weißhaar 1991, 237 ff.; Čochadžiev 1992.
[40] Novotná 1970; Vulpe 1970; 1975; Kuna 1981; Todorova 1981a; Patay 1984; Žeravica 1993.
[41] Тодорова 1986, 96–123; Todorova 1990a, 164.

the north on the Balkan peninsula this gave rise to long lasting droughts, forest fires and intensive erosion processes which undermined the subsistence of the prehistoric population so that breaks in the cultural development occurred, maybe accompanied by a noteable decrease in population size. In any case, interruptions in the settlement history are well attested by pollen analyses from Thessaly, Aegean Thrace, the Strymon valley[42], and from south Albania. In these regions there are acknowledged gaps between the culture groups Dimini and Rachmani, Dikili Tash-Slatino (Sitagroi III) and the Krivodol-Sălcuţa-Bubanj complex, as well as between Maliq Ia and Ib. After the vanishing of the so-called Larissa culture[43] the contemporaneous gap in the prehistory of Thessaly is often overlooked and Dimini und Rachmani are falsely considered as successive cultures instead. From the relative chronological stand point such a view would leave no room for the climax of the Chalcolithic in the Carpathian basin.

In the north and the center of the Balkan peninsula the Middle Chalcolithic comprises the following cultures: Vinča D, including Poduene in the Sofia basin, Marica IV, Karanovo V in northern Thrace, Sava IV on the Black Sea coast, Hamangia IV in the Dobrugea, Boian-Spanţov in Muntenia, Poljanica IV in northeast Bulgaria, and early Pre-Cucuteni III in Moldavia together with Tripol'e A in the southern Ukraine. The earliest metal hoards date to the end of this period, e.g. Ustra[44], Dragoman, Dăržanica, Pločnik I[45] and Pločnik II, and Karbuna[46]. They contain classic Pločnik types such as massive chisels and hammer axes with a short neck and a relatively plump appearance[47], which distinguishes them from similar but later and more elegant hammer axes of the types Varna[48] and Čoka[49].The typological range of the metal objects from this period is more varied with an emphasis on bracelets, finger rings and pendants. It has been suggested that there should be at least seven different regions discernible with different metal types: Eastern Serbia, west Bulgaria, north central Bulgaria, northeast Bulgaria, Muntenia, Black Sea coast, and Thrace.

In terms of absolute age the Middle Chalcolithic is firmly established in the third quarter of the fifth millennium B.C. Radiocarbon dates of this period are more abundant than in the preceding and the following one, which may be due to the frequently encountered destruction horizons by fire. The relative chronological position of the Middle Chalcolithic (called "Transition Period" by E. Comşa[50]), is documented in several stratigraphies in Muntenia (Radovanu), Tangiru[51], northeastern Bulgaria (Ovčarovo[52], Goljamo Delčevo[53], Poljanica[54], Radingrad[55]) and northern Thrace (Karanovo[56], Mădrec, and Ezero[57]).

42 Tonkov/Božilova 1995.
43 Gallis 1987.
44 Черных 1978, analyses nos. 12689-12691; Todorova 1981 a, 96-98 Pl. 6.
45 Kuna 1981, 13-81 Pl. 2,7/2.
46 Сергеев 1963.
47 Тодорова 1986, 35-37.
48 Тодорова 1986, 39-41.
49 Sometimes these axes have been classified as Pločnik type, e.g. Novotná 1970, No. 73, 74, 77, 80.
50 Comşa 1974, 25-27, 245-247.
51 Berciu 1961, 367-368, 394-410.
52 Todorova 1982, 111-143; Тодорова u. a. 1983.
53 Тодорова u. a. 1975; Тодорова u. a. 1977; Todorova 1982, 80-110.
54 Todorova 1982, 144-165.
55 T. Ivanov 1982, 175-180.
56 Georgiev 1961, 57-80; Todorova 1981 b, 203 ff.
57 Георгиев u. a. 1979, 16, 24-32 Fig. 18.

Of the Pločnik type hammer axes there are altogether twelve pieces known from Bulgaria, ten from the area of former Yugoslavia, five from Hungary, twelve from Romania, and one from the metal hoard of Karbuna in Moldavia[58]. With the exception of one piece from Varna I, burial no. 43, this type is not encountered in later contexts. In Bulgaria three examples each derive from the hoards of Dragoman (Slivnica) and from Ustra (Servikija). According to the analyses by Černych[59] the concentrations of some trace elements seem to be different in objects from different regions. He reports the mean silver content in four objects from western Bulgaria to be about ten times lower, and the iron content to be fourty times higher, than those of three objects from Ustra in the Rhodope mountains.

Two of the axes from Dragoman and a new find, a Pločnik type from Radlovci near Kjustendil have been sampled by us as well as several other objects from the end of the Middle Chalcolithic period.

The period of the *Late Chalcolithic* is the richest one with respect to prehistoric metal finds. The increase in metal production is so large that Černych[60] is speaking of a "metal boom". The increased number of finds now also includes gold objects. There is no more the tendency to imitate earlier types made of stone but the specific properties of metal are fully exploited, which results in a larger typological variety. The geographical distribution of certain types of axes seems to suggest that several different production centers were in operation that may have been part of their respective cultural complexes[61]. The expansion in metal production goes parallel with a demographic explosion as has been deduced from the great density of Late Chalcolithic settlements in Bulgaria *(Fig. 5)*. Actually, this may well account for a large part of the "metal boom"[62] so that the increase of the *per capita* metal production during this period may not have been all that dramatic.

As mentioned above, the integration processes of the Middle Chalcolithic have resulted in extended cultural complexes with rather uniform typology of the material culture. In the Late Chalcolithic the cultural complex Kodžadermen-Gumelniţa-Karanovo VI (KGK VI) comprises the area between the Carpathian and the Rhodope mountains including Muntenia, northeast Bulgaria, and northern Thrace. The so-called Krivodol-Sălcuţa-Bubanj (KSB) complex further to the west (the central Balkans comprising Oltenia, eastern Serbia, west Bulgaria with Pleven as the eastern limit, the western Rhodope mountains, and the lower Strymon valley including the island of Thasos) formed a mediating position between the Aegean and the central Danubian region while the KGK VI complex is separated from the Aegean by the Rhodope mountains. The Late Chalcolithic Varna culture extends from the Dobrugea along the western Black Sea coast, with close connections with the Alteni and Cucuteni A_3-Tripol'e BI_3 complex in the southern Ukraine and in Moldavia, as well as with the KGK VI complex. Many scholars suppose that all three cultural phenomena had their own metal production centers that can be distinguished by specific types of objects[63]. This is most pronounced with hammer axes that are the most frequently encountered heavy tools of this period. Type Ploč-

58 Сергеев 1963.

59 Черных 1978, analyses 12690 and 12691 versus analyses 20417a and 20418.

60 Черных 1978, 274 Fig. 118; Todorova 1981a, 7 Fig. 2.

61 Todorova 1981a, 20.

62 Черных 1978, 274 Fig. 118; Todorova 1981a, 7 Fig. 2.

63 Axes of the types Varna, Čoka, Čoka-Varna, and Devnja, as well as bracelets with oblong cross section.

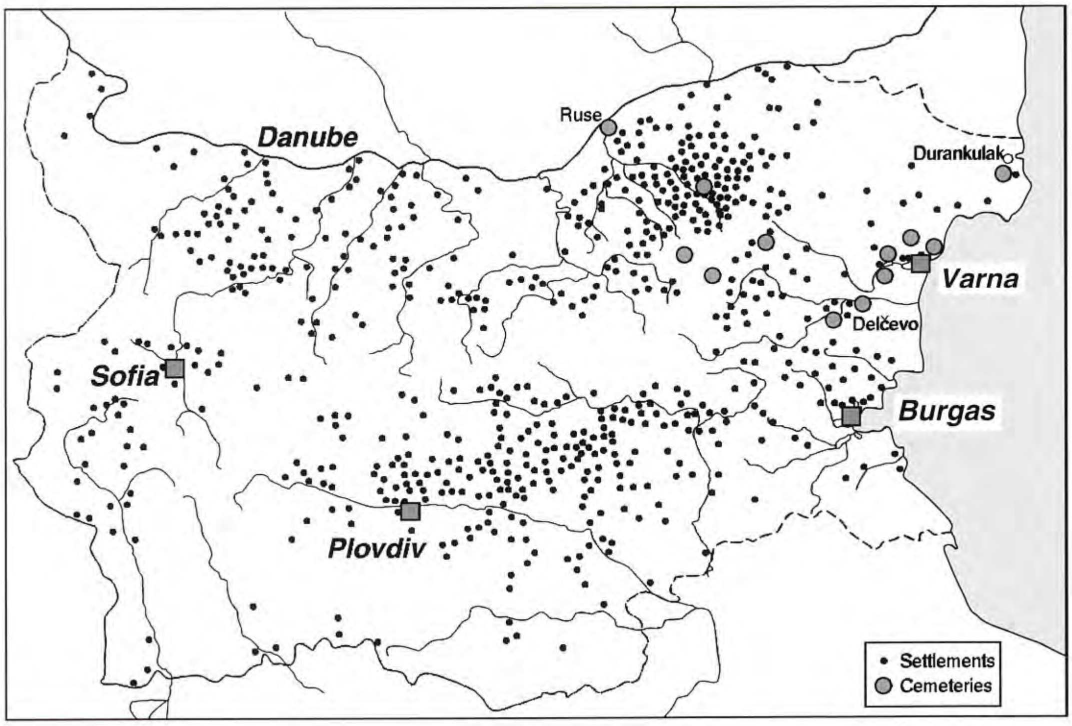

Fig. 5. Late Chalcolithic settlements and cemeteries in Bulgaria.

nik hammer axes occur only occasionally, e.g. in grave no. 43, at Varna I[64]. The dominating types are Vidra, Čoka, Varna, Čoka-Varna, Devnja[65] and Devnja-Brad, elegant cast shapes that may have been tools, weapons, and status symbols at the same time. Metallographic analyses have confirmed their use, e.g. for cutting wood. In graves their occurrence is restricted to grown up males and boys[66]. The so-called "symbolic graves" nos. 1 and 4 at Varna I that most probably represent burials of ceremonial idols[67] contain the whole typological spectrum of Varna without any traces of wear. If Varna I was a sanctuary rather than an ordinary settlement then one may conclude that it, in addition, comprised metallurgical workshops that provided the most important male idols as well as the locally used metal implements, like axes, chisels, and hammer axes of the types Čoka-Varna, Varna and Devnja. It is also possible that the idols represent protective symbols for metallurgists. The prominent position within the Late Chalcolithic of the area around Lake Varna and especially that of its sanctuaries is further documented by the unequalled abundance of gold objects from the cemetery Varna I, which is paralleled by abundant copper finds that represent the earliest known indications for social differentiation[68].

During the years a further important site of the Varna culture has been exposed some 100 km north of Varna, near Durankulak[69], in the district of Dobrič *(Fig. 6)*. A tell set-

[64] I. Ivanov 1991, 143–144.
[65] Todorova 1981 a, 37–41.
[66] Тодорова-Симеонова 1971, 5–6 Pl. 13, 29 grave 4.
[67] Todorova 1992 a.
[68] Todorova 1990 b, 233–238; Todorova 1992 a.
[69] Todorova 1989 d.

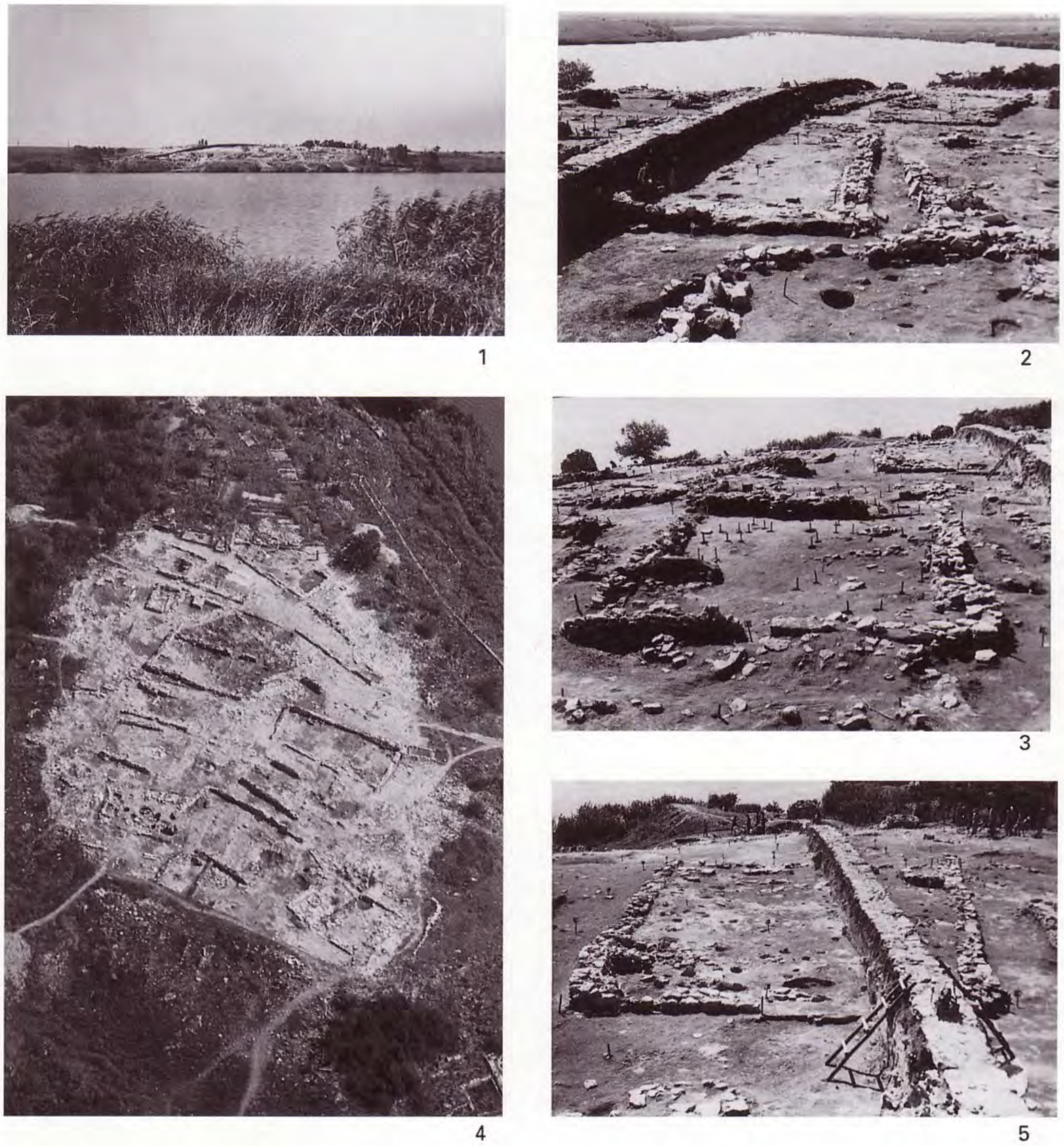

Fig. 6. Durankulak. 1 The Great Island in the lagoon of Durankulak; 2.3.5 building horizon IV B, phase III of the Varna culture: 2 house 1; 3 house 2; 5 house 9 ("palace"); 4 building horizon V, phase II of the Varna culture.

tlement with stone architecture – the earliest yet known in Europe – is situated on an island within a lagoon near Durankulak, while a large cemetery with more than 1200 graves of the Hamangia and Varna cultures extends along the shoreline of the lagoon. All metal finds from that site are included in this study.

Adjacent to the area dating to the Varna culture in the west is the KGK VI complex that is mainly represented by tell settlements with pile architecture, symmetrical and well fortified settlements, and cemeteries *extra muros*. It has not yielded as many metal finds as the Varna culture but the types are considered characteristic. Especially the hammer axes of the Vidra type with three variants (Vidra A, B, and C) are dominant in this area. Heavy axes of the types Gumelniţa, Sălcuţa, and Coţeana are also common. In contrast to the area along the Danube no Chalcolithic cemeteries have so far been detected in northern Thrace. As a result, the number of metal finds from this region is significantly lower than in north-east Bulgaria. The presently somewhat contradictory situation that the number of metal finds is particularly low in the vicinity of the Chalcolithic mines of Ai Bunar is probably due to a reduced excavation activity in this region. It should thus not form the basis of any far-reaching archaeological conclusions.

In western Bulgaria our knowledge of the Late Chalcolithic is comparatively limited. Only a few settlements of the KSB complex are known and even those have not been investigated in great detail, e.g. Krivodol[70], Galatin[71], and Djakovo[72]. Thus far no cemetery has come to light in this region so that, accordingly, there are only a few metal objects available, mostly chance finds and predominantly heavy axes of the Sălcuţa type. North-central Bulgaria is not only geographically but also typologically intermediate between the clearly distinguishable regions in eastern Bulgaria and in the central Balkans, because it is here that the metal types of both overlap, like the Vidra-type hammer axe from Sadovec[73].

The first phase of the KSB complex is only known from the northern parts of its later extension area. Djakovo is the only known site so far, while the second phase is represented by six sites and the third by twelve. During this last phase the KSB cultural complex extends further south and even includes the islands of Thasos and Samothrace. In addition, close connections with the classic Rachmani culture in Thessaly and with Maliq Ib-Mandalo-Mikro Vouni in southern Albania and Macedonia can be recognized from the similarity of their ceramic styles.

Originally, the fundamental changes in the structure of the southeast European prehistoric societies in the fourth millennium B.C., between the Chalcolithic and the (Aegean) Bronze Age[74], were supposed to have occurred rather abruptly. Now we prefer to distinguish two sub-phases, with smooth transitions, namely the *Final Chalcolithic* from around 4000 to ca. 3700 B.C., and the subsequent *Proto Bronze Age (Fig. 1)*. These two sub-phases are, however, not represented in all regions. In northeastern Bulgaria there seems to be a gap in the succession of cultures and the Proto Bronze Age directly overlies the Late Chalcolithic while in northern Thrace both subphases are absent. This gap has been cause of much speculation. The most popular explanation is perhaps that a "steppe invasion" resulted in widespread destruction of settlements and a significant decrease in population[75], possibly because environmental changes had weakened the formerly stable societies and had made them vulnerable to attack from the outside[76]. At any rate, it seems evident that during this time large areas of southeastern Europe have suffered environmental changes that can be seen as a continuation of the climatic development that had affected the regions south of the 41st degree northern latitude a few centuries earlier. While the rise in average annual temperatures brought relative pros-

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70 Миков 1948; Николов 1984.

71 Georgieva 1988.

72 Čochadžiev 1984.

73 Todorova 1992b, 361–383 Fig. 5.

74 Todorova 1989a, 26–27; Todorova 1989b, 700–701; Георгиева 1992.

75 This is not the place to discuss this model in any detail. Generally, a lengthy period of migrations is recorded, often assumed to be connected with the advent of an Indo-European population (e.g. Childe 1926; Gimbutas 1960; Dumitrescu 1963; Scherer 1968; Gimbutas 1970; Gimbutas 1980; Tasić 1995). However, other explanations have also been suggested (Bosch-Gimpera 1961; Häusler 1981).

76 Todorova 1989a; Todorova 1993.

perity to the regions north of the Alps and the Carpathian mountains in the fourth millennium B.C. (e.g. the rise of the Pfyn and Funnel Beaker cultures), it was apparently disastrous for the regions further south like Muntenia, the Dobrugea, northeast Bulgaria, and Thrace. A decrease of the annual precipitation may have severely hampered subsistence and, accordingly, have led to large-scale migrations that could explain the lack of settlements from this period. Areas at higher altitude, like in the central Balkans and the Rhodope mountains were probably not as much affected. The Mediterranean Sea seems to have reached its largest extension in this period so that the climate probably became more humid and the conditions for subsistence in the Aegean improved again.

The consequences of the climatic effects, quite different in time and space, are clearly mirrored in the archaeological record and can thus be dated[77]. Entire societies of the Varna culture and the KGK VI complex vanish that had comprised more than 600 known settlements in the last quarter of the fifth millennium B.C. There is not a single settlement from the Final Chalcolithic known in this area! The burial of idols in the Varna I cemetery dates exactly at the end of the Late Chalcolithic and may symbolize the decline of this culture that abandons its sanctuaries and buries its wealth.

Needless to say that with the decline of the Varna culture and the KGK VI complex (Phase IIIb and IIIc) also the metal production comes to an end. There are only very few metal finds from that area that belong to the Final Chalcolithic, usually from unclear contexts. At the same time there is a distinct cultural rise in western Bulgaria at the end of the third and the beginning of the fourth phase of the KSB complex. This horizon is contemporaneous with Bodrogkeresztúr, Cucuteni AB/Tripol'e B II and classic Rachmani. Calibrated radiocarbon dates place all these cultures into the first centuries of the fourth millennium B.C.

In this period the center of metal production seems to move towards the central Balkans, with the mining areas of Majdanpek and Rudnik[78], and possibly also towards the Carpathian basin as has recently been proposed by Parzinger[79]. Now, the dominant type of implement is the axe-adze of the Jászladány type[80] but hammer axes type Handlova[81], Mezökeresztes[82] and several types of chisels and flat axes belong also to this period. In addition, the first copper knives, type Bodrogkeresztúr[83], appear. In Bulgaria the equivalent types are small curved knives, type Teliš, that have provisionally been regarded as "harpoon tips" for lack of a better explanation[84]. The amount of metal in circulation is considerable; a single Mezökeresztes hammer axe, e.g., weighs 4,000 g[85]!

Present-day Bulgaria is situated at the periphery of this "metal boom" area and was largely depopulated as mentioned above. The southernmost occurrence of a Mezökeresztes hammer axe at Kočan, in the Nestos valley (here published for the first time) differs

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77 Several Late Chalcolithic burials of the Varna culture are 0.50–1 m below the ground water level that is equivalent to the present-day sea level. This suggests that the sea level most likely was lower around 4200–4100 B.C. than it is today. The following ingression has formed a cliff of about 2 m height on the shore in the first half of the fourth millennium B.C. Both features mark the peak of the annual mean temperatures between 4200 and 3500 B.C.

78 Pernicka u. a. 1993, 25, 29 Fig. 10–14.

79 Parzinger 1993, Pl. 228, 229.

80 Dimitrov 1993.

81 E. Schubert/F. Schubert in print.

82 Vulpe 1970, 29; Vulpe 1975, 29; Todorova 1981 a, 42; Patay 1984.

83 Vajsov 1992b; Vajsov 1993, 128.

84 Todorova 1981 a, 54; Гергов 1987, 45.

85 Patay 1984, 57.

distinctly from the local typological spectrum so that it presumably can be identified as an imported piece.

All Jászladány axes must have been cast in two or three-piece moulds[86]. The moulds themselves are very rare[87]. This may be due to the fact that most axes themselves are chance finds, mostly from ploughing or from hoards, and that very few sites of this period have been investigated according to modern standards. In this respect there is a certain imbalance between metal finds and number of settlements that could be interpreted as an indication of a semi-mobile character of the societies involved. Such an interpretation would fit well with the observation that caves and sites at higher altitudes were increasingly inhabitated or at least used seasonally (e.g. the caves at Jagodina, Haramijska dupka, Maronea and others in the Rhodope mountains, Devetaki near Lovеč, Magura, Zlotska pečina and others in the central Balkans).

The distribution map[88] clearly shows a further dicrepancy: The distribution of known metal finds of the Late and Final Chalcolithic does not extend into the south of the Balkan peninsula and western Anatolia. As already mentioned, this observation has led Renfrew to suggest the independence of the southeast European Chalcolithic. In our view this scarcity of finds in the southern regions could as well be explained by the general cultural decline, possibly initiated by climatic changes that first affected the southern regions and then the ones further north.

Chalcolithic phenomena fade out in the transition period between the Chalcolithic and the Bronze Age – a period we suggest to call *Proto Bronze Age*[89] – and vanish completely towards the end of this period. Further changes are attested by the appearance of new ceramic technologies (shell temper) and new vessel shapes (jugs with handles, disc-shaped handles, etc.). Metal production seems to have been greatly reduced and artefacts made of low impurity copper became rare. This also marks the end of the so-called Balkano-Carpathian metallurgical complex[90] as well as the end of the Chalcolithic in general that is accompanied by a reduction in population as suggested by the scarcity of settlements[91].

In terms of absolute age this period dates roughly in the first half of the fourth millennium. In western Bulgaria the Galatin culture[92] forms that is synonymous with "Sălcuţa IV". It forms the southern part of a very large cultural complex along the middle and lower Danube that is characterized by the use of pottery with disc-shaped handles. The formation of Cernavoda I[93] and Pevec[94] in the northeast of the Balkan peninsula are generally interpreted as a result of the immigration of remnants of the late Tripolye and pre-Jamnaja population from the north Pontic steppes[95]. At Durankulak one burial with finds typical for those cultures has been excavated (grave 982, see below).

Five settlements from this period are known in northeast Bulgaria, but none from Thrace, with altogether 29 pit houses: Ovčarovo-Plateau, Hotnica-Vodopada[96], Šemševo,

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[86] Рындина/Орловская 1978, 316.
[87] Vulpe 1970, 19.
[88] Parzinger 1993, Pl. 227, 228, 229.
[89] Todorova 1993.
[90] Černych 1978.
[91] Kalicz 1992, 327–329.
[92] Георгиева 1992, 339–348.
[93] Roman 1971; Dodd-Opriţescu 1992, 38–43.
[94] Тодорова u. a. 1983, 12–13.
[95] Unpublished Kurgan from Durankulak, excavations conducted by H. Todorova; for the primary burial see Vajsov 1992b, 61–65.
[96] Ilčeva 1986; Ilčeva 1996.

Kačica[97], Borovo und Durankulak II. All settlements are small and did not last very long. Compared with the Chalcolithic there is a clear cultural decline to be observed. It is in this period that arsenical copper makes its first appearance (see also below).

Some late types of axe-adzes like Tîrgu-Okna and the flat axe types Altheim, of the Ete variant, and some small axe-adzes of the Jászladány type belong to this period. The total inventory of Proto Bronze Age metal finds from present-day Bulgaria is very small.

Around the middle of the fourth millennium B.C. the formation of a new metallurgical complex can be recognized. It is characterized by the appearance of new elements in the typological spectrum, e.g. daggers, flat axes, and double-spiral pendants, but also by a different metal composition as we shall see below. The earliest sites belonging to this complex are Usatovo[98] in the northern Pontic area, Ilıpınar IV in western Anatolia[99], Sitagroi IV near Drama[100], Arslantepe VII and VIA near Malatya[101], Nahal Mishmar in Palestine[102], and the earliest metal using cultures north of the Alps (Mondsee, Pfyn, Cortaillod, etc.[103]). It also includes the so-called Circum-Pontic metallurgical province as definded by Černych[104] whose origins obviously reach back until the Proto Bronze Age.

Investigated archaeological sites and objects[105]

1. **Ai Bunar**, District of Stara Zagora, southern slopes of the Sredna Gora mountains, Thrace. – The mine 8 km northwest of Stara Zagora was discovered in 1934 by I. Azmanov and was investigated from 1971 to 1974 by E. N. Černych. A number of Chalcolithic pottery fragments from the vicinity of the mine yielded an unequivocal prehistoric date. Except for a few small traces of prospection no later exploitation has apparently occurred. – The hydrothermal mineralization consists of several almost vertical, subparallel 0.5 to 5 m wide veins in limestone, marl, and dolomite. The ore paragenesis is polymetallic with a well developed oxidation zone that varies in depth from 1 to 25 m. The outcrops were most likely conspicuous due to the occurrence of malachite and azurite. The mineralized zone extends within a broad crescent that stretches roughly NE–SW and crosses three hills that belong to the foot hills of the Sredna Gora mountains. – Altogether eleven prehistoric mines have been investigated. They consist of wedge-shaped trenches 10 to 100 m in length. In general, they were only 2–3 m deep but in richly mineralized areas their depth extended to 20–30 m, possibly all the way down to the groundwater table. The waste and the sterile country rock were backfilled so that nearly no dumps remained along the former veins. Former mining was indicated only by flat depressions that were not disturbed by later activities. In mine no. 4b the remains of an early Iron Age house covered part of the prehistoric exploitation area so that for this location a firm *terminus ante quem* is given. The pottery below the house floor is Chalcolithic

97 Unpublished; excavations under the direction of P. Stanev (Museum of Veliko Tărnovo).
98 Vajsov 1993, 137–142.
99 Roodenberg 1995.
100 Keighley 1986, 344–356.
101 Palmieri 1981.
102 Bar-Adon 1980, 15–134, 199, 239.
103 Ottaway 1982; Ottaway 1989.
104 Черных 1978, 265–267; Chernykh 1992, 54–167.
105 In the following catalogue the analyzed objects are desribed together with their find locations *(Fig. 4)*. Also given are their weights, their inventory numbers at the respective museums, if known, and the Heidelberg/Mainz laboratory number (HDM) that is generally used for reference in the text. Most of the heavy implements have already been published by Todorova 1981 a. These cases are marked as PBF IX/14, followed by the respective catalogue number. New finds and the majority of those published in the Bulgarian literature that may not be easily accessible have been put together in Plates II–VII. Objects of little or no

throughout so that the main mining activity can be dated into the second half of the fifth millennium B.C. The copper objects analyzed in this study probably derive from mine 1 where they are said to have been discovered by I. Azmanov in 1934[106]. They are now in the museum at Stara Zagora. – In the vicinity of the mines, within a distance of about 20 km and especially in the Thracian plain, there are a number of tell settlements that were also inhabitated in the Chalcolithic. A few of these have been totally (Tell Azmak) or partially (Stara Zagora-Hospital, Berekecka mogila, Starozagorski Mineralni bani, Kiril-Metodievo, Čatalka, and others) excavated. These excavations yielded numerous copper objects and copper ores. According to Černych the chemical composition of the ores largely agrees with the ores from Ai Bunar[107]. His chemical groups II, III and IV dominate, at Tells Azmak and Mădrec they comprise even all analyzed finds. New analyses of ore samples from Thracian tell sites yielded a somewhat different result[108].

Hammer axe, type Pločnik, var. B; Stara Zagora 1C3.786; 654 g;
PBF IX/14, 101; ASM 12687, MCG III. HDM 1940

Axe-adze, type Tîrgu Okna, stray find, Final Chalcolithic;
Stara Zagora 1C3.787; 995 g; PBF IX/14, 195; ASM 12686, MCG VI. HDM 1941

2. **Dăržanica**, Vidin district, western Bulgaria. – Hoard find consisting of two objects[109], now in the museum at Vidin. End of Middle Chalcolithic. Probably contemporaneous with Pločnik hoards.

Heavy axe, type Gumelniţa; VID M95; 640 g; PBF IX/14, 36;
ASM 10720, MCG I. HDM 2702

Heavy axe, type Sălcuţa; VID M96; 540 g;
PBF IX/14, 62; ASM 10721, MCG I. HDM 2703

3. **Debrene/Prilep**, District of Dobrič, Black Sea region.

Flat axe, type Goljamo Delčevo, stray find, Varna culture; DB-Ap0072;
143 g, *(Fig. 9,10)*. HDM 2140

4. **Devnja (Poveljanovo)**, District of Varna, northern Black Sea region. – Rescue excavations of cemetery I on the northern shore of the Varna lake in 1969 by H. Todorova unearthed 26 burials of the Late Chalcolithic Varna culture, phase III, including some with copper and gold objects[110]. Burial practice separated according to sex: Men are stretched out on their back, women are buried in crouched position to the right. The head points in all cases to the north. Cenotaphs occur also. The accompanying settlement is situated below the present level of Lake Varna. Copper objects are most often associated with burials of men and with cenotaphs. They are now in the Municipal Museum at Varna.

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typological value, like small borers, wires, or casting spills, have not been included in the Plates. Several objects of this study have already been analyzed by optical emission spectroscopy in the laboratory of the Archaeological Institute of the Academy of Sciences in Moscow and partly been published by Черных1978. The laboratory numbers of these analyses are given as ASM numbers. The data were used by Černych to sort the Chalcolithic objects from Bulgaria into nine groups designated by Roman numerals I to IX (Черных 1978). These are given as MCG numbers (for Moscow chemical groups). In a few cases analyses of the same objects were also made in the course of the analytical project at the Württembergisches Landesmuseum in Stuttgart and published in Junghans u. a. 1968. These laboratory numbers are listed as SAM (for Studien zu den Anfängen der Metallurgie).

[106] Черных 1978, 54–75.

[107] Черных 1978, 56–76.

[108] Gale u. a. 1991.

[109] Миков 1972, 91 Pl. 2. The axe-adze published by Mikov in the same article does not belong to the hoard.

[110] Тодорова-Симеонова 1971.

Grave 1 – destroyed burial.
Hammer axe, type Varna; V. I-1449; 253 g;
PBF IX/14, 141; ASM 9246, MCG II. HDM 1929

Heavy axe, type Gumelniţa; V. I-1451; 146 g;
PBF IX/14, 35. HDM 1933

Grave 4 – destroyed burial of a child.
Small hammer axe, type Čoka-Varna; V. I-1454; 66 g;
PBF IX/14, 132; ASM 9247, MCG V. HDM 1936

Grave 18 – adult male, rich grave goods including gold.
Hammer axe, type Čoka-Varna; V. I-1448 [111]; 473 g;
PBF IX/14, 134; ASM 9329, MCG IV. HDM 1928

Grave 23 – cenotaph.
Hammer axe, type Devnja with corroded remains of textiles;
V. I-1450; 458 g; PBF IX/14, 147; ASM 9330, MCG II. HDM 1930

Grave 24 – cenotaph.
Hammer axe, type Čoka-Varna; V. I-1452; 255 g;
PBF IX/14, 133; ASM 9328, MCG V. HDM 1934

Grave 26 – adult male.
Chisel; V. I-1453; 92 g; PBF IX/14, 5; ASM 9327, MCG II. HDM 1935

5. **Djakovo**, District of Kjustendil, western Bulgaria. – High altitude tell settlement in the upper Strymon valley, now partly below the water level of a reservoir. Rescue excavation 1980–1981 by St. Čochadžiev, settlement structure on terraces, phases I and IV of the Krivodol cultural complex. Object in the museum at Kjustendil [112].
Flat axe, type Kamenar, settlement phase II (Late Chalcolithic), from the
floor of a building; KU I-100(A74); 240 g, *(Fig. 9,1)*. HDM 1905

6. **Dragoman (Slivnica)**, District of Sofia, western Bulgaria. – Late Middle Chalcolithic hoard find consisting of three hammer axes, two of which are clearly miscasts [113]. Museum of the Archaeological Institute of the Academy of Sciences, Sofia.

Hammer axe, type Pločnik, var. A; AIM 638 b; 910 g, *(Fig. 12,2)*;
PBF IX/14, 94; ASM 20417, MCG II; SAM 3513. HDM 1910

Hammer axe, type Pločnik, var. A; AIM 638 a; 392 g, *(Fig. 12,3)*;
PBF IX/14, 93; ASM 20418, MCG I; SAM 3515. HDM 1911

7. **Durankulak**, District of Dobrič, northern Black Sea region. – Tell settlement on the larger one of two islands in the lagoon of Durankulak with an associated cemetery on the western shore of the lagoon. Excavations have been conducted under the direction of H. Todorova since 1975 and are still continuing *(Fig. 6)* [114]. – Presently, three settlement horizons of the Varna culture are known from the tell. In addition, there is one medieval layer (I) and a Late Bronze Age layer (II, with Coslogeni material). Layers III–VIII are Chalcolithic with a palace structure, cultic buildings, and megaron houses, mostly in stone architecture. The settlement horizon V has ended in a destructive

[111] Wrong inventory no. in Черных 1978.
[112] Čochadžiev 1984, 72 Fig. 11,11.
[113] Попов 1921, 25; Миков 1933, Fig. 52.
[114] Тодорова 1984 b, 30–31, 38, 44–45; Todorova 1989d.

fire. A fireplace in layer VI may have been used for melting of metal and smithing[115]. The presence of the Hamangia culture (Phases I and II) is documented by pit houses on the shore of the lagoon. Timber-frame and mud-wall houses as well as stone-built structures of Hamangia III and IV have been found on the island with the tell settlement (layers VII and VIII). – The excavation of the cemetery along the shore of the lagoon has been completed in 1991. It comprised more than 1200 burials of the Hamangia and Varna cultures[116] with a continuous development between the sixth and fifth millennium B.C. The burial practice is differentiated by sex – men are buried in stretched position while women are interred crouched towards the right. All burials are oriented with heads towards the north. The grave goods indicate a social and gender differentiation beginning as early as at the age of seven. The graves of the late Hamangia and the Varna cultures contain also gold ornaments and copper artifacts. – The earliest find, a long necklace consisting of malachite and possibly several copper beads from grave 626, belongs to the end of Hamangia II and, accordingly, is Late Neolithic. If it could be proven that metal beads were also used then this would represent one of the earliest well dated metal finds in Bulgaria[117]. The number of metal finds increases in the course of the Hamangia period as indicated by some burials with rich grave goods, some of them made of copper. After the termination of the Chalcolithic settlement there are only sporadic metal finds and Durankulak seems to have served only for very short periods as settlement and burial site, possibly for nomadic groups of the Usatovo and pre-Jamnaja cultures that infiltrated northeast Bulgaria from the north Pontic steppe areas. This is suggested by two interesting daggers of this period[118] one of which comes from a grave (no. 982) with a body crouched to the *left*.

The Early and Middle Bronze Age are not represented at Durankulak, possibly because of a rise at that time of the sea level. From the Late Bronze Age a fortified settlement of the Coslogeni and Babadag culture groups can be identified[119]. Later habitation remains date to the fourth and third centuries B.C. and to the fifth, tenth, and eleventh centuries A.D. In the intervening periods and from the Middle Ages on, the island was not inhabitated because of growing swamps. As of 1992 altogether 104 metal artifacts have been found at Durankulak. They are now stored in the museum at Dobrič. All of these have been analyzed, including objects that had been analyzed previously in the laboratories at the Academy of Sciences in Moscow.

Durankulak, cemetery

Grave 29 – adult female, Hamangia IV.
Bracelet, right forearm; K0080a; 29.6 g; ASM 45351, MCG IV. — HDM 1976
Bracelet, left forearm; K0080b; 23.2 g, *(Fig. 7,20)*;
ASM 45352, MCG V. — HDM 1977

Grave 171 – adult female, Varna III.
Bracelet; K0220; 41.3 g, *(Fig. 7,19)*; ASM 45353, MCG III. — HDM 1997

Grave 217 – child, Varna II.
Bracelet; K0256; 19.0 g, *(Fig. 7,9)*; ASM 36237, MCG I. — HDM 1979
Bracelet; K0253; 29.0 g, *(Fig. 7,7)*; ASM 36205, MCG VI. — HDM 1982

Grave 223 – child, female, with gold objects, Varna III.
Small bracelet; K0271; 3.6 g, *(Fig. 7,2)*; ASM 36220, MCG III. — HDM 2020

Grave 251 – adult male, rich grave goods, Varna III.
Bracelet; K1667a; 53.0 g, *(Fig. 7,18)*; ASM 45357, MCG V. — HDM 2141
Bracelet; K1667b; 26.0 g, *(Fig. 7,21)*. — HDM 2142

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115 Todorova 1997.
116 Todorova/Dimov 1989.
117 Вайсов 1987, 78–79 Fig. 2.
118 Vajsov 1992b, 61–65.
119 Тодорова 1984b, 68–69.

Grave 253 – cenotaph, Varna III.
Bracelet; K0259; 87 g; ASM 36214, MCG IV. HDM 1965
Bracelet; K0258; 61 g; ASM 36213, MCG VI. HDM 1978

Grave 262 – child, Varna II.
Bracelet; K0294; 5.5 g, *(Fig. 7,4)*; ASM 36239, MCG III. HDM 2017

Grave 272 – child, Varna II.
Bracelet; K0581; 4.5 g, *(Fig. 8,1)*; ASM 36240, MCG II. HDM 2016

Grave 273 – adult male, Varna II.
Bracelet; K0313; 19.2 g, *(Fig. 7,11)*; ASM 45358. HDM 2019

Grave 275 – adult male, early Varna I.
Bracelet; K0314; 38.0 g, *(Fig. 7,14)*; ASM 36212, MCG III. HDM 1968
Bracelet; K0315; 13.2 g, *(Fig. 7,16)*; ASM 36238, MCG II. HDM 1981

Grave 291 – adult female, Varna I.
Bracelet; K0308; 8.8 g, *(Fig. 7,6)*; ASM 36243, MCG III. HDM 2018

Grave 298 – adult male, Varna II.
Spiral ring; K0323a; 3.3 g; ASM 36210, MCG III. HDM 2035

Grave 309 – child, Varna I.
Bracelet; K0318; 39 g, *(Fig. 7,12)*; ASM 36207, MCG V. HDM 1972

Grave 317 – adult male, Varna III.
Bracelet; K0417a; 47 g, *(Fig. 7,13)*. HDM 1973

Grave 320 – adult male, Varna III.
Borer; K0324; 1.6 g; ASM 36203, MCG II. HDM 2010
Hammer axe, type Varna; K0321; 341 g; ASM 36199, MCG IV. HDM 2022

Grave 321 – adult male, Varna II.
Bracelet; K0319; 15.1 g, *(Fig. 7,10)*; ASM 36202, MCG III. HDM 1980

Grave 330 – adult male, Varna III.
Flat axe; K0320; 163 g; ASM 36200, MCG II. HDM 2021

Grave 389 – adult male, Varna III.
Bracelet; K0670; 91 g, *(Fig. 8,8)*; ASM 36236, MCG II. HDM 1946

Grave 390 – adult male, Varna II, *(Fig. 13,3)*.
Bracelet; K0676b; 68 g, *(Fig. 7,17)*; ASM 36235, MCG III. HDM 1944
Bracelet; K0676a; 67 g, *(Fig. 7,15)*; ASM 36234, MCG III. HDM 1945

Grave 395 – adult female, Varna II.
Bracelet; K0633; 37 g, *(Fig. 8,14)*; ASM 36257, MCG VI. HDM 1947

Grave 408 – adult female, Varna II.
Finger ring; K0517a; ASM 36233, MCG III. HDM 1942

Grave 433 – child, female, Varna I.
Broad finger ring; K0731. HDM 1943

Grave 447 – adult female, with gold objects, Varna II.
Spiral ring; K0749; 2.7 g; ASM 45360, MCG VI. HDM 2009
Bracelet; K0751a; 13.2 g, *(Fig. 7,1)*; ASM 45361, MCG III. HDM 2023
Bracelet; K0751b; 11.0 g, *(Fig. 7,8)*; ASM 45362, MCG III. HDM 2024
Finger ring; K0750; 3.8 g; ASM 45359, MCG V. HDM 2027

Grave 454 – adult female, Varna I.
Broad finger ring; K0786; 3.1 g; ASM 45363, MCG III. HDM 2028

Grave 455 – adult female, Varna I.	
Finger ring; K0796; 1.6 g; ASM 45364, MCG III.	HDM 2013
Broad finger ring; K0797; 5.4 g; ASM 45365, MCG V.	HDM 2029
Grave 460 – adult female, Varna I.	
Finger ring; K0818; ASM 45366, MCG VI.	HDM 2000
Grave 496 – adult female, Varna I.	
Finger ring; K1048; ASM 45367, MCG VI.	HDM 2002
Finger ring; K1049a.	HDM 2137
Finger ring; K1049b.	HDM 2138
Grave 501 – child, male, late Hamangia IV.	
Bracelet, right forearm; K0874; 4.7 g, *(Fig. 7,3)*; ASM 45369, MCG VI.	HDM 2025
Bracelet, left forearm; K0873; 5.0 g, *(Fig. 7,5)*; ASM 45368, MCG VI.	HDM 2026
Grave 502 – adult female, Varna I.	
Spiral ring; K2138; 2.0 g; ASM 45370, MCG IV.	HDM 2006
Grave 506 – adult female, Varna I.	
Finger ring; K2140; 1.4 g.	HDM 2031
Spiral ring; K0884; 2.3 g; ASM 45371, MCG III.	HDM 2036
Grave 512 – adult male, Varna I, *(Fig. 13,2)*.	
Bracelet; K0943; 32.8 g, *(Fig. 8,2)*; ASM 45372, MCG V.	HDM 2011
Grave 514 – adult female, Varna III.	
Finger ring, fragment; K0957; 7.1 g; ASM 45377, MCG III.	HDM 1962
Bracelet, right forearm; K0958b; 47 g, *(Fig. 8,15)*; ASM 45379, MCG V.	HDM 1963
Bracelet, left forearm; K0958a; 31 g, *(Fig. 8,10)*; ASM 45378, MCG V.	HDM 1964
Spiral ring; K0956a; 2.4 g; ASM 45373, MCG V.	HDM 2007
Finger ring; K0956b; 7.3 g; ASM 45374, MCG IV.	HDM 2008
Grave 530 – child, female, Varna I.	
Small bracelet; K1024; 3.5 g; ASM 45380, MCG III.	HDM 2012
Grave 533 – child, Varna I.	
Bracelet; K1029; 28.5 g, *(Fig. 8,7)*; ASM 45381, MCG IV.	HDM 2014
Bracelet; K1030; 24.6 g, *(Fig. 8,5)*; ASM 45382, MCG III.	HDM 2015
Grave 545 – adult female, Hamangia III.	
Finger ring; K1095; 1.2 g; ASM 45383, MCG III.	HDM 2005
Grave 554 – adult female, Varna I.	
Finger ring, fragment; K1113; 3.4 g; ASM 45384, MCG VI.	HDM 2003
Grave 559 – child, male, Varna II.	
Bracelet; K1123; 29.2 g; ASM 45387, MCG VI.	HDM 1985
Grave 611 – adult male, Hamangia II.	
Malachite bead; K1353.	HDM 2004
Grave 623 – adult male, Varna III.	
Flat axe, type Sălcuţa; K1301; 309 g, *(Fig. 9,6)*; ASM 45388, MCG II.	HDM 1971
Grave 626 – adult female, Hamangia IIb[120].	
Cu or malachite bead from necklace; K1443.	HDM 1969
Cu or malachite bead from necklace; K1442.	HDM 1970

[120] Вайсов 1987, Fig. 1–4; Vajsov 1992a, 97 Pl. 1,2.4.5.

Grave 643 – adult male, Hamangia III.
Three malachite beads; K1357. HDM 2210
HDM 2211
HDM 2212

Grave 648 – destroyed child's burial, Hamangia I.
Malachite bead; K1367. HDM 2209

Grave 660 – child, male, Varna II.
Bracelet; K1471; 16.0 g, *(Fig. 8,6)*; ASM 45389, MCG V. HDM 1984
Bracelet; K1472; 38.4 g, *(Fig. 8,4)*; ASM 45390, MCG II. HDM 1993

Grave 672 – adult male, Varna II.
Bracelet, left forearm; K1590; 8.8 g; ASM 45391, MCG III. HDM 1994
Bracelet, right forearm; K1591; 19.4 g, *(Fig. 8,13)*; ASM 45392, MCG VI. HDM 1995

Grave 679 – adult female, Varna II.
Finger ring; K1616; ASM 45393, MCG VI. HDM 1974

Grave 681 – adult female, Hamangia III.
Finger ring; K1611; 4.4 g; ASM 45394, MCG V. HDM 1975

Grave 684 – adult female, Varna II.
Finger ring; K1568; 1.2 g; ASM 45412, MCG V. HDM 2001

Grave 722 – child, male, Varna III.
Bracelet, right forearm; K1876; 19.8 g; ASM 45395, MCG V. HDM 1990

Grave 731 – adult female, Varna I.
Finger ring; K1834; 6.3 g; ASM 45396, MCG V. HDM 1967

Grave 732 – adult male, Varna I.
Bracelet; K1980; 9.2 g, *(Fig. 8,9)*; ASM 45397, MCG V. HDM 1986
Bracelet; K1981; 8.4 g, *(Fig. 8,3)*; ASM 45398, MCG V. HDM 1996

Grave 733 – destroyed child's burial, Varna I.
Bracelet; K1696a; 26.0 g; ASM 45399, MCG III. HDM 1987

Grave 739 – child, Varna III.
Bracelet; K1881; 7.9 g; ASM 45400, MCG III. HDM 1988

Grave 741 – adult female, Varna III.
Finger ring, fragment; K1847; 0.3 g; ASM 45401, MCG V. HDM 2030

Grave 756 – adult male, Varna I, *(Fig. 13,4)*.
Bracelet, right forearm; K1942; 44 g, *(Fig. 8,16)*; ASM 45402, MCG V. HDM 1989
Bracelet, left forearm; K1943; 38.6 g, *(Fig. 8,17)*; ASM 45403, MCG IV. HDM 1998

Grave 763 – adult male, Varna II, *(Fig. 13,1)*.
Bracelet, right forearm; K1859; 54.5 g, *(Fig. 8,11)*; ASM 45404, MCG IV. HDM 1991
Bracelet, left forearm; K1861; 60.0 g, *(Fig. 8,12)*; ASM 45405, MCG V. HDM 1992

Grave 867 – adult male, Varna III.
Flat axe; K2127; 274 g; ASM 45406, MCG V. HDM 1999

Grave 977 – adult male, Varna II/III.
Hammer axe, type Čoka-Varna; K2332; 309 g; ASM 45407, MCG V. HDM 1961

Grave 982 – barrow burial with stone ring, crouched to the left, with traces of red colouration, rectangular, flat burial pit, Usatovo (?), Celej group (?).
Dagger, type Nerušaj with handle plate of bone and rivet holes[121]; K2317; *(Fig. 12,7)*. HDM 1918

[121] Vajsov 1992b, 62 Fig. 1,6.

Grave 1183 – adult female, Varna II.	
Pin; K2856; 2.5 g.	HDM 2034
Grave 1202 – mature male, Varna II.	
Borer; K2909; 0.3 g.	HDM 2032
Burials of small children	
Bracelet, quadrant 23St/3; K0489; 20 g; ASM 36225, MCG III.	HDM 1948
Bracelet, quadrant 29B/2; K0496; 16 g; ASM 36251, MCG IV.	HDM 1949
Stray find	
Spiral-headed pin, Late Bronze Age; K1726; 1.7 g; ASM 45408.	HDM 2033

Durankulak, tell settlement on Goljamija ostrov (Great Island)

Wire, stray find; E 0213.	HDM 1950
Wire, stray find; E 0224; 4.1 g.	HDM 1951
Small dagger, stray find, Proto Bronze Age; E 0162; 3.6 g.	HDM 1952
Stylo, stray find, Hellenistic 4th to 3rd century B.C.; E 0225; 1.2 g, *(Fig. 11,19)*.	HDM 1953
Pin, Proto Bronze Age; Z 0279; 11.4 g.	HDM 1954
Metal band, settlement phase II, Late Bronze Age, Coslogeni culture group; Z 0211; 7.6 g.	HDM 1955
Small bracelet, settlement phase III, Late Chalcolithic, Varna III; E 0229; 3.4 g.	HDM 1956
Pin, settlement phase II, Late Bronze Age, Coslogeni culture group; E 0193; 4.5 g.	HDM 1957
Spiral-headed pin, settlement phase II, Late Bronze Age, Coslogeni culture group; E 0071; 4.0 g.	HDM 1958
Pin, settlement phase IV, Late Chalcolithic,Varna III; E 0215; 3.4 g.	HDM 1959
Bracelet, settlement phase III, quadrant E30/3, Late Chalcolithic; E 0228; 16.4 g.	HDM 1960
Dagger, variant of the Usatovo type[122], Proto Bronze Age; E 0004; ASM 36201, MCG VII.	HDM 1966
Bracelet, settlement phase IV, Varna III; E 0218; 45 g.	HDM 1983

8. **Galiče**, District of Vraca, western Bulgaria. – High altitude settlement near the village of Galiče, surface finds dating from the Transition Period from the Late Chalcolithic to the Early Bronze Age (Galatin group). The analyzed dagger has been published by B. Nikolov[123] and dated to the so-called *Scheibenhenkelhorizont* by I. Vajsov[124]. It is now in the museum at Vraca.

Dagger, type Cucuteni; VR-AI 661; 53 g.	HDM 2737

9. **Goljamo Delčevo**, District of Varna, Black Sea region. – A tell settlement and a cemetery along the Luda Kamčija river, 50 km west of the Black Sea coast have been excavated by H. Todorova from 1968 to 1970. Today the site is inundated by the lake of the Tsonevo dam. The tell, rising 4.9 m above ground, revealed 16 settlement horizons ranging from Early to Late Chalcolithic. It is situated at the border of the distribution areas of the Gumelniţa and the Varna cultural elements. According to the burial practices it should be attributed to the Gumelniţa culture. In contrast to

[122] Vajsov 1992b, 62, 65 Fig. 1,4.
[123] Николов 1962, 69.
[124] Vajsov 1993, 118 Fig. 15, 120.

Durankulak, burials are crouched to the left, looking east, regardless of sex. There are a few exceptions, however, which may be attributed to the Varna cultural group. – A large part of the metal finds has already been analyzed chemically and metallographically by E. N. Černych and N. N. Ryndina[125]. Some of the objects have been re-analyzed in this study. An interesting find is an installation that may have been used for the production of charcoal (horizons VII–IX) and was probably associated with a smithing workshop at the edge of the settlement[126].

Goljamo Delčevo, cemetery

Grave 25 – adult male, with gold bead, copper axe and other valuable grave goods.

Borer[127]; 3 g; ASM 12147, MCG III.	HDM 2129
Hooked wire[128]; G.D. 00354; 15.8 g; ASM 12146, MCG II.	HDM 2707

Grave 31 – adult female.

Borer[129]; 4.8 g; ASM 12148, MCG III.	HDM 2120
Double-spiral-headed pin[130]; 8.1 g, *(Fig. 12,4)*; ASM 12149, MCG VI.	HDM 2126

Goljamo Delčevo, tell settlement

Short borer, settlement phase X[131]; G.D. 00237; 2.2 g.	HDM 2119
Bracelet, settlement phase XIII[132]; G.D. 01919; 6.2 g; ASM 12703, MCG III.	HDM 2121
Spiral ring, settlement phase XV[133]; G.D. 00822; 1.2 g; ASM 12700, MCG II.	HDM 2122
Spiral ring, settlement phase XIV, 3 fragments[134]; G.D. 01394; 1.3 g; ASM 12701, MCG I.	HDM 2123
Borer, settlement phase III–IV[135]; G.D. 04720; 6.3 g; ASM 12699, MCG I.	HDM 2124
Borer, settlement phase XI[136]; G.D. 00572; 6.9 g.	HDM 2125
Borer, settlement phase XVII[137]; G.D. 02729; 2.6 g.	HDM 2127
Borer, settlement phase VIII[138]; G.D. 03510; 7.4 g.	HDM 2128
Borer, settlement phase XV[139]; G.D. 00164; 3.8 g.	HDM 2130

10. **Gradešnica**, District of Vraca, western Bulgaria. – On a large plateau, 20 km west of Vraca, and some 2 km west of the village, several settlements of the Early Neolithic, the Early Chalcolithic, and the Bronze Age were observed, mainly based on dislocated finds. Excavations have been conducted from 1961 to 1974 by B. Nikolov[140] and in 1985 by B. Nikolov and I. Vajsov at various locations[141]. Unfortunately, most of the metal finds are without inventory numbers and reliable information on their find contexts. They are stored in the museum at Vraca.

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125 Черных 1978, Pl. 8,2.3.6; 10,4; 11,11.13; 13,3; 15,36; 17,19; 19,24; Рындина/Орловская 1978.
126 Тодорова u. a. 1975, 44 Fig. 26; Todorova 1981 a, 1–8.
127 Тодорова u. a. 1975, 63; 239 Pl. 127,5.
128 In Черных 1978 erroneously cited as "Grave 27".
129 Тодорова u. a. 1975, 63–64; 241 Pl. 129,5.
130 Тодорова u. a. 1975, 63–64; 241 Pl. 129,6.
131 Тодорова u. a. 1975, 132 Pl. 59,22.
132 Тодорова u. a. 1975, 194 Pl. 80,28.
133 Тодорова u. a. 1975, 158 Pl. 44,4.
134 Тодорова u. a. 1975, 204 Pl. 91,6.
135 Тодорова u. a. 1975, 132 Pl. 18,10 Tab. 33,4.
136 Тодорова u. a. 1975, 167 Tab. 53,2.
137 Тодорова u. a. 1975, 203 Tab. 90,1.
138 Тодорова u. a. 1975, 167 Tab. 53,1.
139 Тодорова u. a. 1975, 179 Tab. 65,44.
140 Николов 1974; Николов/Вайсов 1985.
141 Тодорова/Вайсов 1993, 109 Fig. 97.

Borer[142], house 2; VR AI-000c; 6.5 g.	HDM 2729
Borer, stray find, Late Bronze Age; VR AI-000a; 15.6 g.	HDM 2731
Borer, Late Bronze Age; VR AI-961; 1.1 g.	HDM 2732
Loop-headed pin, Late Bronze Age; VR AI-000d; 1.5 g.	HDM 2734
Borer with broadened handle plate; VR AI-000e; 2.5 g.	HDM 2735

11. **Haramijska Dupka**, District of Smoljan, central Rhodope region. – A cave site near Trigrad was excavated by Ch. Vălčanova from 1982 to 1984. Three cultural layers have been observed. The two lower ones belong to the transition period (Jagodina group)[143] between the Chalcolithic and the Early Bronze Age. Two daggers have been found below house 2 of the uppermost horizon (Final Chalcolithic). Accordingly, they belong to the Proto Bronze Age. The objects are now in the museum at Smoljan.

Dagger with three rivet holes, type Jagodina[144]; SM I-198; 23.2 g; ASM 34405.	HDM 2740
Small dagger without rivet holes, type Sofievka[145]; SM I-142; 13.0 g; ASM 34406.	HDM 2741
Borer; SM I-109; 1.5 g.	HDM 2742

12. **Hotnica**, District of Veliko Tărnovo, central northern Bulgaria. – Tell settlement partially excavated by N. Angelov from 1956 to 1957. The uppermost horizon contains a burnt layer of the late third phase of the KGK VI complex (Late Chalcolithic). A small sondage excavation reached virgin soil so that the complete stratigraphic sequence is known. The settlement begins in the fourth phase of the Poljanica group (Boian IV). In burnt houses of the uppermost layer human skeletons with associated metal objects were found. In addition, a hoard find of gold objects was discovered[146]. All sampled objects are dated to phase IIIb of the KGK VI complex and are stored in the museum at Veliko Tărnovo. – A small settlement of the Proto Bronze Age on a terrace east of the water fall was excavated by V. Ilčeva from 1978 to 1992. The two lowest cultural layers are separated by sediment from an inundation and contain pit houses of the Pevec group, phase "Vodopada", which is contemporaneous with the Ulmeni phase of the Cernavoda I culture. Imports attest contacts with the Cucuteni-Tripolye culture[147]. The Proto Bronze Age settlement is partly overlain by a strongly eroded layer of the Late Bronze Age Tei culture. Both daggers derive from layer I of the settlement. Calibrated ^{14}C dates[148] range from 3625 to 3919 B.C. All objects are in the museum at Veliko Tărnovo.

Hotnica, tell settlement

Hammer axe, type Vidra, var. A, settlement phase I; VT-132n; 312 g; PBF IX/14, 105; ASM 10895, MCG II.	HDM 1937
Hammer axe, type Vidra, var. B, settlement phase I; VT-759n; 552 g; PBF IX/14, 125; ASM 10896, MCG II.	HDM 1938
Heavy axe, type Coţeana, settlement phase I; VT-104n; 418 g; PBF IX/14, 45; ASM 10897, MCG II.	HDM 1939
Borer, settlement phase I; VT-104r; ASM 10899, MCG II.	HDM 2143
Borer, settlement phase I; VT-0000; ASM 10900, MCG II.	HDM 2144

142 Drawings of this object and of HDM 2732 and 2735 have been published by Nikolov 1974, Fig. 72.
143 Аврамова 1981; Вълчанова 1984; Аврамова 1992; Vajsov 1993, with radiocarbon dates of sites.
144 Vajsov 1993, 118f. Fig. 12,6. The analyses are from the laboratory of the Archaeological Institute in Moscow and not in St. Petersburg, as stated in this article.
145 Vajsov 1993, 118f. Fig. 12,3.
146 Ангелов 1959; Ангелов 1961.
147 Todorova 1981a, 78, 194; Ilčeva 1986; Ilčeva 1996.
148 Vajsov 1993, 117.

Borer, settlement phase I; VT-0000; ASM 10901, MCG II. HDM 2145
Borer, settlement phase I; VT-0000. HDM 2146

Hotnica, Vodopada (waterfall)

Borer, settlement phase II; VT-1724; 1 g. HDM 2131
Dagger, type Mondsee[149]; VT-1432p; 8.1 g; ASM 36281. HDM 2134
Dagger, type Mondsee, settlement phase I[150]; VT-1509p; 2.7 g; ASM 36282. HDM 2135
Metal fragment, stray find, Late Bronze Age, late Tei culture; VT-223. HDM 2136

In the vicinity of Hotnica-Vodopada[151] also some stray finds of prehistoric metal objects were recovered.
Axe-adze, type Tîrgu Okna, early Pevec culture, Final Chalcolithic;
VT-529; 758 g, *(Fig. 10,2; 12,9)*; PBF IX/14, 194; ASM 20842, MCG II. HDM 1901
Flat axe, type Altheim, var. Ete, late Pevec culture, Proto Bronze Age;
VT-H.T. 310; 664 g, *(Fig. 9,13)*; PBF IX/14, 78; ASM 20844, MCG VI. HDM 1902
Dagger, Late Bronze Age; VT-513; 8.2 g. HDM 2132
Dagger, Late Bronze Age; VT-000; 13.4 g. HDM 2133

13. **Ivanovo**, District of Ruse, northeast Bulgaria.

Axe-adze, type Jászladány, with traces of hammering, stray find,
Final Chalcolithic; R 3515; 487 g, *(Fig. 12,8)*; PBF IX/14, 184. HDM 1919

14. **Jasen**, near Unia Alba, District of Vidin, western Bulgaria.

Flat axe, type Altheim, var. Ete, stray find, Proto Bronze Age;
VID-M94; 170 g, *(Fig. 12,1)*; PBF IX/14, 79; ASM 10719, MCG I. HDM 2701

15. **Kačica**, Suburb of Veliko Tărnovo, central northern Bulgaria. – The large plateau settlement is today part of a suburb of the town of Veliko Tărnovo. Rescue excavations by P. Stanev in 1973 and 1979 disclosed pit houses of the Late Neolithic Hotnica culture, timber-frame houses of the Middle Chalcolithic and pit houses of the Proto Bronze Age Pevec culture to which all three sampled objects are dated[152]. The objects are stored in the museum at Veliko Tărnovo.
Borer, quadrant IX-1, h 1.50; VT-I; *(Fig. 11,5)*. HDM 2147
Borer; VT-I; *(Fig. 11,2)*. HDM 2148
Borer, quadrant IX-7, h 1.80; VT-I; *(Fig. 11,1)*. HDM 2149

16. **Karanovo**, District of Sliven, Thrace. – The tell settlement, rising 12.5 m above ground, has been excavated with interruptions by N. Koičev, V. Mikov and G. Georgiev in the period from 1939 to 1956 and by S. Hiller from 1984 to 1996[153]. The Chalcolithic layers comprise 5.60 m and range from Karanovo V (Middle Chalcolithic) to Karanovo VI (Late Chalcolithic). Early Chalcolithic (Marica I–III) is missing at the tell[154]. Some additional metal finds have been analyzed by E. N. Černych. They are now in the Museum of the Archaeological Institute of the Academy of Sciences, Sofia.

Borer, Karanovo VI, phase III of the KGK VI complex; AIM 353;
ASM 12029, MCG II. HDM 2150

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149 Ilčeva 1986, Fig. 4,3; Vajsov 1992b, 62, 65 Fig. 1,2; Vajsov 1993, 117 Fig. 12,1.
150 Ilčeva 1986, Fig. 4,2, Vajsov 1992b, 65 Fig. 1,1; Vajsov 1993, 117 Fig. 12,2.
151 Илчева/Кокорков 1978.
152 Станев 1980.
153 Миков 1958; Georgiev 1961, 57–80; Hiller/Georgiev 1984–1987; Hiller/Nikolov 1988–1996.
154 Todorova 1981b, 208–216.

17. **Kočan**, District of Goce Delčevo, western Bulgaria. – High altitude settlement of phase III/IV of the KSB complex in the central Nestos valley. The implement was recovered by D. Serafimova, first publication here. Apparently the axe has been cast in an open mould, because its bottom surface shows a shallow depression. Its body, 29.5 cm long and 6 cm wide, is decorated on the top surface with grooves. The dating of the hammer axes type Mezökeresztes has been detailed by A. Vulpe[155] and P. Patay[156]. Both authors agree that they are contemporaneous with the much more common early axe-adzes type Jászladány and, accordingly, belong to the Bodrogkeresztúr culture (i.e. the beginning of the fourth millennium B.C.). This is corroborated by the overlapping distribution areas of both types from which our find is now the southernmost known example. This date agrees with the chronological position of the settlement at Kočan as well as that of the Jászladány-type axe from Teliš (see below). Because of the decoration the hammer axe from Kočan is closely related to a similar example from a hoard find from Petren, district of Bihor, Romania[157]. Another hammer axe, type Mezökeresztes (Final Chalcolithic), from present-day Bulgaria has been reported from the vicinity of Vidin[158].

Hammer axe, type Mezökeresztes, stray find; BL – 1000; 1630 g, *(Fig. 10,6)*. HDM 1916

18. **Kozlodujci**, District of Dobrič, Black Sea region.
Sickle, stray find from the surface of a Late Bronze Age settlement of the Coslogeni culture; now in the museum at Dobrič.
Ap. 0006; 12.8 g. HDM 2037

19. **Krivodol**, District of Vraca, western Bulgaria. – Multilayered tell settlement on the steep bank of the river Botunja, 2 km north of the town of Krivodol. The site has been excavated by V. Mikov over an area of 200 m². Further excavations were conducted by B. Nikolov in 1977 and 1978. The site is eponymous for the Late Chalcolithic culture in northwest Bulgaria, which is part of the large Krivodol-Sălcuţa-Bubanj complex[159]. Museum at Vraca.

Borer with rectangular cross section, h 2.10; VR-AI-2269; 18.2 g. HDM 2727
Knife, stray find from the vicinity of Oslen near Krivodol, Late Bronze Age, h 2.10; VR-AI-2868; 7.3 g. HDM 2730

20. **Lesura**, District of Vraca, western Bulgaria. – Prehistoric plateau settlement that is severely disturbed by an antique fortification. There are finds from the transition period between the Chalcolithic and the Early Bronze Age. The objects sampled were erroneously published as part of a Late Bronze Age hoard[160]. This was later corrected by Černych[161].

Dagger with four rivet holes and midrib; VR-AI-653; 27.6 g; ASM 10689, MCG VII. HDM 2738
Dagger with three rivet holes and engravings along the blade; VR-AI-654; 12.8 g; ASM 10690, MCG IX. HDM 2739

21. **Malorad**, District of Vraca, near Pčelina, western Bulgaria.

Dagger with three rivet holes, stray find, Proto Bronze Age; Museum at Vraca; VR-AI-960; 11.8 g. HDM 2736

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155 Vulpe 1970, 30–31.
156 Patay 1984, 58–59.
157 Vulpe, 1975, 59A.
158 Черных 1978, 9148; MCG I; Todorova 1981a, no. 151.
159 Миков 1948; Николов 1984; Тодорова 1986, 82.
160 Николов 1966, 49, Fig. 3 a, b.
161 Черных 1978, 234.

22. **Marica**, District of Haskovo, Thrace. – Tell settlement Devebargan. The site is eponymous for the Early Chalcolithic Marica culture[162]. Numerous finds from an unsystematic excavation were rescued by R. Popov in 1922[163]. The stratigraphy of the site has been discussed recently[164]. The lowest layer of the tell belongs to phase III of the Marica culture from which the sampled axe probably derives. Museum of the Archaeological Institute of the Academy of Sciences, Sofia.

Axe with oval cross section, typologically without parallel; AIM 930;
772 g, *(Fig. 9,12)*; PBF IX/14, 25; ASM 20420, MCG III; SAM 3521. HDM 1912

23. **Mezdra**, District of Vraca, western Bulgaria. – The Late Chalcolithic settlement is situated on a steep promontory on the bank of the river Iskar and is disturbed by a medieval castle. The prehistoric objects were found during an investigation of the latter and are now in the museum at Vraca.

Heavy axe, type Pločnik; VR-AI-2859; 620 g, *(Fig. 9,11)*. HDM 2726

24. **Onogur**, District of Dobrič, Black Sea region. – Late antique fortification in the southern Dobrugea with traces of a Late Chalcolithic settlement dating to Varna IIb. Museum at Dobrič.

Flat axe, type Sălcuţa(?), stray find; Ap. 0018; 57 g, *(Fig. 9,2)*. HDM 2139

25. **Plakuder**, District of Vidin, western Bulgaria. – Hoard of twelve axes, bound together[165]. Four pieces were lost later. Today eight axes are distributed among the Historic Museum at Vidin, the National Museum in Sofia and the Museum of the Archaeological Institute of the Academy of Sciences. Two axes belong to the Ariuşt and six to the Jászladány type, Final Chalcolithic[166].

Axe-adze, type Jászladány; AIM 3236A; 1050 g; PBF IX/14, 169;
ASM 9150, MCG I; SAM 3492. HDM 1906
Axe-adze, type Jászladány; AIM 3236D; 876 g; PBF IX/14, 166;
ASM 9152, MCG I. HDM 1907
Axe-adze, type Jászladány with notches; AIM 3236E; 762 g;
PBF IX/14, 165; ASM 9153, MCG I; SAM 3494. HDM 1908
Axe-adze, type Jászladány; AIM 3236B; 1028 g; PBF IX/14, 167;
ASM 9151, MCG I. HDM 1909
Axe-adze, type Jászladány; VID - M98; 1060 g; PBF IX/14, 170;
ASM 10723, MCG I. HDM 2705
Axe-adze, type Ariuşt; VID - M99; 880 g; PBF IX/14, 161;
ASM 10722, MCG I. HDM 2706

26. **Radlovci**, District of Kjustendil, western Bulgaria.
Hammer axe, type Pločnik (end of Middle Chalcolithic), rough surface from casting, no traces of use or wear, stray find; museum at Kjustendil,

KU - I-539; 972 g, *(Fig. 10,3; 12,10)*; ASM 39107, MCG I. HDM 1903

27. **Rebărkovo, Djugera**, District of Vraca, western Bulgaria. – Settlement on the bank of the Iskar river. Excavated by H. Todorova from 1982 to 1984. Single-phase site dating to phase IVa of the Krivodol culture, Final Chalcolithic[167]. Within the settlement a hoard of several axe-adzes was found that cannot be located at present. The object sampled is the only one remaining of this hoard. Museum at Vraca.

Axe-adze, type Ariuşt; VR-AI-2867; 750 g; PBF IX/14, 158. HDM 2724

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162 Gaul 1948, 72–74.
163 Попов 1925; Gaul 1948, 43–47 Pl. 34,11.
164 Todorova 1981b; Тодорова 1986, 57.
165 Миков 1961, Pl. 3.
166 Todorova 1981a, 160, 161, 165–170.
167 Georgieva 1988, 6–9 Pl. 6; Георгиева 1992, 341, 343.

28. **Ruse**, Northeastern Bulgaria. - The tell close to the Danube near the mouth of the river Rusenski Lom has been recognized since 1888[168]. Several excavations have been conducted: 1904 Škorpil, 1921-1922 Kostov[169], 1948-1953 N. Angelov and G. Georgiev[170], 1970-1980 G. Georgiev, 1980-1983 K. Kănčev, 1983-1990 - V. Popov[171]. The poor excavation technique applied by Kănčev has practically destroyed more than half of the tell. Moreover, although the cultural layers are inclined towards the Danube, Kănčev has continuously removed *horizontal* layers so that material from between two and four cultural horizons was mixed. Most probably Georgiev has used a similar technique also. Even burnt houses have been divided between two horizons so that any dating from the stratigraphical context is problematic. The last excavation by V. Popov using modern methods has established the relative stratigraphy of the lowest 2 m in the northern part of the tell. Thus the stratigraphy of the tell can be reconstructed within limits[172] so that the depth information of finds from the old excavations can be used with caution. By and large the tell has been occupied during the whole Chalcolithic as is indicated by the dominance of Boian-Vidra, Boian-Spanţov and Gumelniţa material. Hence, the majority of finds can be roughly dated into the fifth millennium B.C. In addition, the later so-called "Eneolithic cemetery" of Ruse, located on the tell, had disturbed parts of the settlement[173]. Late Bronze Age material has also been found on the surface of the tell as well as in pits that reach a depth of up to 2.50 m. - Analyses by Černych have indicated that several graves should be later than Chalcolithic, because of greatly different metal composition as indicated by samples ASM 10853 - h 2.80, 10863 - h 2.10, 10875 - h 2.50, 10882 - h 3.50. Their composition as well as their typological spectrum can be well accommodated within the known material from the Gumelniţa culture. We have sampled all available metal finds from Ruse that are now stored in the museum at Ruse. The respective depths from which the samples were recovered are given in meters in all cases.

Flat axe, type Kamenar, settlement phase I, h 0.10; R 5018/1986; 263 g, *(Fig. 9,7)*.	HDM 1920
Chisel; R 5016/1986; 112 g, *(Fig. 9,5)*.	HDM 1921
Chisel, settlement phase I, h 0.20; R 5017/1986; 100 g, *(Fig. 9,3)*.	HDM 1922
Heavy axe, type Gumelnita; R 5019/1986; 746 g, *(Fig. 9,15)*.	HDM 1923
Borer; R 3268/1953; 5.3 g.	HDM 2039
Borer, h 2.25; R 3259/1953; 5.8 g.	HDM 2040
Borer, sondage, h 3.10, Middle Chalcolithic; R 1225/1949; 5.9 g; ASM 10883, MCG II.	HDM 2041
Hook, h 1.35, age uncertain; R 0670/1950; 4.1 g, *(Fig. 11,12)*; ASM 10877, MCG II.	HDM 2042
Borer, h 2.25; R 3259/1953; 18.0 g.	HDM 2043
Borer, h 0.90; R 1107/1948; 2.9 g; ASM 10885, MCG II.	HDM 2044
Borer, sondage, h 4.40; R 1332/1949; 4.4 g; ASM 10878, MCG V.	HDM 2045
Borer, h 1.35, possibly Late Bronze Age; R 0673/1950; 2.3 g; ASM 10875, MCG X.	HDM 2046
Double-spiral-headed pin; R 5000/1952; 10 g.	HDM 2047
Bracelet, age uncertain; R 5635/1986; 6.4 g.	HDM 2048
Borer, sondage, h 4.65, Middle Chalcolithic; R 3444/1959; 4.8 g; ASM 10830, MCG I.	HDM 2049

168 Шкорпил/Шкорпил 1888, 34-36; Шкорпил 1914, 59-60 Pl. 8-11 Fig. 65, 71.
169 Костов 1925.
170 Георгиев/Ангелов 1952; Георгиев/Ангелов 1957.
171 V. Popov 1997.
172 Тодорова 1986, 88; V. Popov 1997.
173 Тодорова 1986, 74.

Borer, h 1.60; R 3253/1952; 6.6 g; ASM 10852, MCG IV.	HDM 2050
Borer, h 1.80; R–/1952; 2.5 g.	HDM 2051
Borer, h 1.80; R–/1952; 0.4 g.	HDM 2052
Chisel, settlement phase I, h 0.10; R 0119/1986; 67.5 g.	HDM 2053
Borer, h 2.70; R 1192/1949; 8.2 g; ASM 10823, MCG III.	HDM 2054
Borer from a hoard, h 1.20; R 3266/1952; 8.6 g; ASM 10850, MCG II.	HDM 2055
Borer, h 2.20; R 3.10 (unreadable inventory number); 5.3 g.	HDM 2056
Borer; R 5014; 24.2 g.	HDM 2057
Borer, h 2.00; R 5015/1986; 8.8 g.	HDM 2058
Borer, h 2.00; R 3250/1952; 5.8 g, *(Fig. 11,6)*; ASM 10861, MCG II.	HDM 2059
Borer, h 2.70; R 1191/1949; 8.6 g; ASM 10888, MCG III.	HDM 2060
Borer, h 2.10; R 3265/1952; 5.3 g; ASM 10847, MCG II.	HDM 2061
Borer, settlement phase I, h 0.70, age uncertain; R 0671/1950; 3.0 g; ASM 10876, MCG I.	HDM 2062
Borer, h 1.40; R 1761/1951; 3.7 g; ASM 10860, MCG III.	HDM 2063
Borer, sondage, h 5.70, Middle Chalcolithic; R 3434/1959; 6.2 g; ASM 10832, MCG III.	HDM 2064
Borer; R 5012/1986; 9.5 g.	HDM 2065
Borer, h 0.70, age uncertain; R 0772/1950; 8.3 g.	HDM 2066
Borer, h 2.10; R 3252/1952; 5.5 g; ASM 10808, MCG III.	HDM 2067
Borer; R 3260/1952; 4.8 g; ASM 10851, MCG III.	HDM 2068
Chisel, no context, age uncertain; R 0000 (no inventory number); 10.6 g.	HDM 2069
Borer, sondage, h 3.00; R 3460/1953; 9.1 g; ASM 10822, MCG IV.	HDM 2070
Borer, h 2.25; R 3264a/1952; 5.5 g, *(Fig. 11,13)*; ASM 10849, MCG IV.	HDM 2071
Borer, h 2.10; R 3251/1952; 5.4 g; ASM 10846, MCG IV.	HDM 2072
Fragment of a double-spiral-headed pin, h 1.25; R 5005/1952; 11.4 g.	HDM 2073
Borer, h 2.50; R 1179/1949; 6.5 g; ASM 10833, MCG II.	HDM 2074
Loop-headed pin, h 2.25; R 3264b/1952; 5.5 g.	HDM 2075
Borer, h 2.25; R 3263/1953; 6.6 g; ASM 10859, MCG IV.	HDM 2076
Borer, h 1.25; R 5003/1952; 5.7 g.	HDM 2077
Borer, h 1.25; R 5009a/1952; 5.8 g.	HDM 2078
Borer, h 1.20; R 5009b/1952; 9.6 g.	HDM 2079
Borer, sondage, h 4.65, Middle Chalcolithic; R 3444/1953; 5.3 g.	HDM 2080
Borer, h 2.70; R 1193/1949; 5.3 g, *(Fig. 11,11)*; ASM 10884, MCG II.	HDM 2081
Borer; R 1178/1949; 1.2 g.	HDM 2082
Borer, h 1.20; R 5007/1952; 3.8 g, *(Fig. 11,17)*.	HDM 2083
Borer, h 1.35; R 5001/1952; 4.9 g.	HDM 2084
Borer, h 0.75; R 5010/1952; 8.1 g.	HDM 2085
Borer, no context; R 3. .../1952 (unreadable inventory number); 7.3 g.	HDM 2086
Borer, no context; (no inventory number).	HDM 2087
Borer; R 5006/1952; 5.4 g.	HDM 2088
Fragment of a loop-headed pin, h 1.10; R 1783/1951; 2.3 g; ASM 10866, MCG II.	HDM 2089
Borer, h 2.70; R 1194/1949; 1.6 g; ASM 10879, MCG III.	HDM 2090
Borer, h 3.50; R 1244/1949; 5.7 g; ASM 10887, MCG II.	HDM 2091
Fragment of a loop-headed pin; R 5013/1986; 1.2 g.	HDM 2092
Fragment of a borer, h 1.30; R 1945/1951; 0.7 g; ASM 10865, MCG II.	HDM 2093
Pin, no context; no inventory number; 6.2 g.	HDM 2094
Pin, no context; no inventory number; 5.6 g.	HDM 2095
Borer, h 2.45; R 3267/1953; 4.2 g.	HDM 2096
Borer, h 1.00; R 1108/1948; 3.5 g; ASM 10827, MCG V.	HDM 2097
Pin, h 2.00; R 5002/1952; 7.5 g, *(Fig. 11,20)*.	HDM 2098

Fragments of two double-spiral-headed pins from a hoard that are
inseparable due to corrosion,h 1.20; R 266; together 15.8 g;
(*Fig. 11,8* left); ASM 10820, MCG II. HDM 2099
(*Fig. 11,8* right); ASM 10821, MCG II; SAM 3546. HDM 2100
Borer, h 1.75; R 1853/1951; 5.4 g; ASM 10858, MCG I. HDM 2101
Fragments of two borers, h 3.50; R 1243 a,b/1949; one fragment ASM 10886, MCG IV.
a) 3.5 g. HDM 2102
b) 2.8 g. HDM 2103
Loop-headed pin, settlement phase I, h 0.10; R 5011/1986; 8.2 g. HDM 2104

29. **Sadovec**, District of Pleven, central northern Bulgaria. – A large settlement of the Late Chalcolithic that is situated below the late antique fortification "Golemanovo Kale" near Sadovec, discovered and excavated by G. Bersu from 1935 to 1937[174]. Further excavations in 1979 by H. Todorova. Numerous stray finds are known from all over the plateau, including the implement sampled, now in the museum at Pleven.

Hammer axe, type Vidra, var. A, with traces of hammering[175];
PL 3057; 283 g, *(Fig. 10,5)*. HDM 1915

30. **Sava**, District of Varna, Black Sea region. – The tell settlement at Sava, on the right bank of the Kamčija river at a distance of around 50 km from the Black Sea, was explored from 1950 to 1953[176]. In 1959 a control excavation was conducted by D. Zlatarski and H. Todorova to clarify the stratigraphy of the site. Ten burials of the Late Chalcolithic cemetery on the river bank were investigated by G. Tončeva in 1977 and 1978. The tell is eponymous for the Early Chalcolithic Sava culture of the Black Sea region[177].

Grave 6 – adult male.
Heavy axe, type Coteana, late KGK VI complex; NIM 000;
298 g, *(Fig. 9,14)*; PBF IX/14, 40. HDM 1924

31. **Slatino**, District of Kjustendil, western Bulgaria. – The plateau settlement near Čardako was discovered by J. Gaul in 1938[178] and excavated by S. Čochadžiev between 1981 and 1989. Three settlement horizons belong to the Early Chalcolithic; the site is the eponym of the Early Chalcolithic Dikilatash-Slatino cultural complex that covers the area of the middle and lower Strymon valley[179]. The copper object analyzed has so far not been published and derives from a closed context. It is thus the earliest well dated heavy metal implement from Bulgaria.

Chisel with rectangular cross section; KU I-1100 (827); 290 g, *(Fig. 9,9)*. HDM 1904

32. **Smjadovo**, Čolak kladenec, District of Šumen, northeastern Bulgaria. – Chalcolithic tell settlement on the western edge of the village. Excavations have been performed by N. Popov until 1986. Several horizons of the Middle and Late Chalcolithic were identified[180].

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174 Bersu 1938.
175 Todorova 1992 b, 371 Fig. 5.
176 Мирчев/Златарски 1960.
177 Vajsova 1966, 20; Тодорова 1986, 75–76.
178 Gaul 1948, 61–63.
179 Čochadžiev 1986 a.
180 N. Popov 1978, Pl. 3.

Axe-adze, type Ariuşt, stray find from the vicinity of the tell, Final Chalcolithic; SUM-4574; 200 g.	HDM 2708
Strip of copper metal, settlement phase III, h 0.90; SUM-5388; 3.2 g.	HDM 2709
Chisel, settlement phase III, h 1.20; SUM-6614; 39.2 g, *(Fig. 9,4)*.	HDM 2710
Borer, settlement phase IV, h 2.50; SUM-6631; 1.7 g.	HDM 2711
Casting spill, settlement phase IV, h 2.50; SUM-6493; 18.2 g.	HDM 2712
Casting spill, settlement phase IV, h 2.50; SUM-6493; 5.8 g.	HDM 2713
Borer, settlement phase IV, h 2.50; SUM-6492.	HDM 2714
Ear ring, settlement phase IV, h 3.50; SUM-6430; 0.8 g.	HDM 2715
Borer, settlement phase III, h 0.90; SUM-3404.	HDM 2716
Borer, settlement phase II, h 0.75; SUM-2943; 2.4 g.	HDM 2717
Borer, surface find; SUM-0000; 2.2 g.	HDM 2718
Borer, settlement phase IV, h 2.20; SUM-5968; 3.9 g.	HDM 2719
Borer, settlement phase III, h 0.90; SUM-2572; 0.7 g.	HDM 2720
Borer, settlement phase III, h 0.90; SUM-5401; 1.4 g.	HDM 2721
Borer, settlement phase IV, h 2.20; SUM-5966; 3.1 g.	HDM 2722

33. **Sozopol**, District of Burgas, southern Black Sea region. – Submerged Chalcolithic settlement. The objects derive from a depth of about 4 m below the present sea level. They were recovered during construction works to increase the size of the harbour at Sozopol. Shore settlements are known from this depth and were partly investigated. They date from the Early Chalcolithic to the Proto Bronze Age with interruptions. Wooden logs from the former sea shore settlements have been dendrochronologically dated to the Early Bronze Age around 2500 B.C. by P. Kuniholm (private communication). They rest on remains of a Late Chalcolithic settlement that was founded on dry soil and did not extend into the sea as hitherto assumed. This stratigraphic context corroborates the ingression of the Black Sea in the fourth millennium B.C. The two heavy axes are completely mineralized but retained their shape. Typologically they date to the Late Chalcolithic[181].

Heavy axe, completely corroded with dark blue surface; 280 g.	HDM 2743
Heavy axe, completely corroded with yellowish brown surface; 210 g, *(Fig. 9,8)*.	HDM 2744

34. **Stolnik**, District of Sofia, near Elin Pelin, western Bulgaria. – The small axe-adze, type Jászladány (Final Chalcolithic), is apparently a miscast. Length 13.7 cm, width 5 cm. Its surface has a foamy appearance, the shafthole is circular and somewhat asymmetrically positioned. A shafthole ring is clearly visible. Both cutting edges are blunt and deformed. There are no traces of use or wear.

Small axe-adze, type Jászladány, private collection, stray find; 409 g, *(Fig. 10,1; 12,5)*.	HDM 1917

35. **Teliš**, District of Pleven, central northern Bulgaria. – The plateau settlement Teliš-Redutite was discovered in 1978 during construction work that destroyed almost all of the uppermost horizon of the Proto Bronze Age (Scheibenhenkelhorizont). The two lower horizons (II and III) date to the Final Chalcolithic (phase IV of the KSB complex). These were completely excavated over the entire exposed area by V. Gergov from 1977 to 1989. Several metal objects were recovered[182] that are now in the museum at Pleven. All 000-numbers designate objects from a hoard.

181 Draganov 1997, 201–203.
182 Гергов 1987.

Fragment of a heavy implement, presumably an axe-adze, settlement phase II[183]; PL-1979; 54 g. HDM 1913
Axe-adze, type Jászladány, from a sealed context below the ruins of a burnt house of settlement phase II, phase IV of the KSB complex[184]; PL-1849; 980 g, *(Fig. 12,11)*; ASM 28350, MCG II. HDM 1914
Double-spiral-headed pin, settlement phase III[185]; PL-2366; 17.4 g, *(Fig. 11,3)*. HDM 2105
Tip of a knife[186], settlement phase II[187]; PL-1846; 11.2 g, *(Fig. 11,16)*; PBF IX/14, 226. HDM 2106
Tip of a knife, settlement phase II; PL 2978; 10.0 g, *(Fig. 11,18)*; PBF IX/14, 227. HDM 2107
Borer, settlement phase III; PL-1848; 8.7 g, *(Fig. 11,10)*. HDM 2108
Pin with flat head[188]; PL-E 000; 1.9 g, *(Fig. 11,9)*. HDM 2109
Borer, settlement phase II; PL-F 000; 5.1 g. HDM 2110
Borer, settlement phase II; PL-2365; 6.4 g. HDM 2111
Borer, settlement phase II; PL-0317; 10.2 g. HDM 2112
Borer, settlement phase II; PL-C 000; 5.4 g, *(Fig. 11,7)*. HDM 2113
Borer, settlement phase II; PL-2719; 5.7 g. HDM 2114
Borer, settlement phase II; PL-2614; 6.6 g, *(Fig. 11,4)*. HDM 2115
Long borer, settlement phase II; PL-E 000; 22.8 g, *(Fig. 11,15)*. HDM 2116
Borer, settlement phase II; PL -A 000; 9.2 g, *(Fig. 11,14)*. HDM 2117
Borer, settlement phase II; PL-G 000; 0.8 g. HDM 2118

36. **Varna**, Black Sea region. – The cemetery of Varna I is situated at the western outskirts of the town of Varna, on a 10–12 m high promontory over the Varna lake. The associated settlement has not been found but it is presumed that it lies below the water table. There are numerous Late Chalcolithic settlements around the lake, such as Devnja (see above), but Varna seems to have been the central place of the region.
The cemetery was discovered in the fall of 1972 during construction work. The inventory of grave no. 1 was rescued by D. Zlatarski of the Dălgopol Museum. Regular excavations were conducted under the direction of G. Georgiev, M. Lazarov, and I. Ivanov in the period between 1972 and 1991. Until today 294 graves of the third phase of the Varna culture have been unearthed. Gender-specific burial practice is similar to Durankulak, i.e. men are buried in stretched and women in crouched position. There occur also burials of children, cenotaphs and a number of so-called "symbolic burials"[189] that are interpreted as burials of ceremonial idols[190]. Many metal objects have been found that are now stored in the museum at Varna. Černych has analyzed 44 of those; the results seem to suggest a greater variety of copper sources than at other sites. We have sampled only five objects.

Grave 1 – destroyed context, probably a life-size male idol. Among the grave goods are 76 objects made of gold and copper, third phase of the Varna culture group[191].

[183] Гергов 1987, 50 Fig. 11.
[184] Гергов 1987, 50 Fig. 9, 10. This is not Sălcuţa IV.
[185] Гергов 1987, 49 Fig. 7.
[186] This and the following object have been described as "tip of a harpoon" in Todorova 1981a.
[187] Гергов 1987, Fig. 6a.
[188] Гергов 1987, 49 Fig. 8a.
[189] I. Ivanov 1991, 127.
[190] Todorova 1993.
[191] Иванов 1975, 1–10.

Hammer axe, type Vidra, var. B[192]; V 1541; 285 g; PBF IX/14, 112. HDM 1926
Flat axe, type Sălcuţa[193]; V 1542; 84 g; PBF IX/14, 55. HDM 1927

Grave 4 – Symbolic burial of a life-size clay idol, probably male, made of low-temperature fired clay, rich grave goods of gold and copper, richest of all complexes of the cemetery, third phase of the Varna culture group.
Hammer axe, type Devnja[194]; V 0197; 174 g; PBF IX/14, 149;
ASM 13813, MCG III. HDM 1931
Chisel with hook[195]; V 0198; 126 g; PBF IX/14, 88; ASM 13815, MCG III. HDM 1932

Grave 36 – probably a depot of prestige goods[196], Varna IIIb, numerous gold objects, dentalium shells, ochre, etc.
Hammer axe, type Vidra, var. B[197]; V 0647; 394 g; PBF IX/14, 116;
ASM 13832, MCG III. HDM 1925

37. **Vidbol (Dunavci)**, District of Vidin, western Bulgaria.

Fragment of a flanged axe, stray find, Early Bronze Age;
Museum at Vidin; VID-M93; 270 g; ASM 10718, MCG VII. HDM 2704

38. **Vodnjanci**, District of Dobrič, southern Dobrugea, Black Sea region.

Double axe, Late Bronze Age, Coslogeni culture, stray find[198];
Museum at Dobrič; Ap. 0022. HDM 2038

39. **Zaminec**, District of Vraca, near Gorna Kremena, western Bulgaria. – High altitude settlement of the Late Chalcolithic Krivodol culture. Excavated by B. Nikolov from 1970 to 1973[199]. Bronze Age material has been found on the surface of the site. Museum at Vraca.

Fragment of a flat axe, level C; VR-AI-2272; 93 g. HDM 2725
Borer, level B; VR-AI-2270; 10 g. HDM 2728
Borer, level B; VR-AI-2271; 0.9 g. HDM 2733

40. **Živlovci**, District of Montana, western Bulgaria.

Axe-adze, type Jászladány, Final Chalcolithic, stray find; M – AI-1162;
(Fig. 10,4). HDM 2213

41. **Zlatarevo**, District of Šumen, northeastern Bulgaria. – Tell settlement of the Late Chalcolithic with some later prehistoric surface finds. Museum at Šumen.

Borer, possibly Late Bronze Age, stray find; SUM-6042; 2.2 g. HDM 2723

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192 I. Ivanov 1988, cat. no. 1/58.
193 I. Ivanov 1988, cat. no. 1/61.
194 I. Ivanov 1988, cat. no. 5/3.
195 I. Ivanov 1988, cat. no. 5/4.
196 Todorova 1993, 260–269.
197 I. Ivanov 1988, cat. no. 12/16.
198 Димов 1989.
199 Николов 1975; Тодорова 1986, 17 Pl. 22; 23.

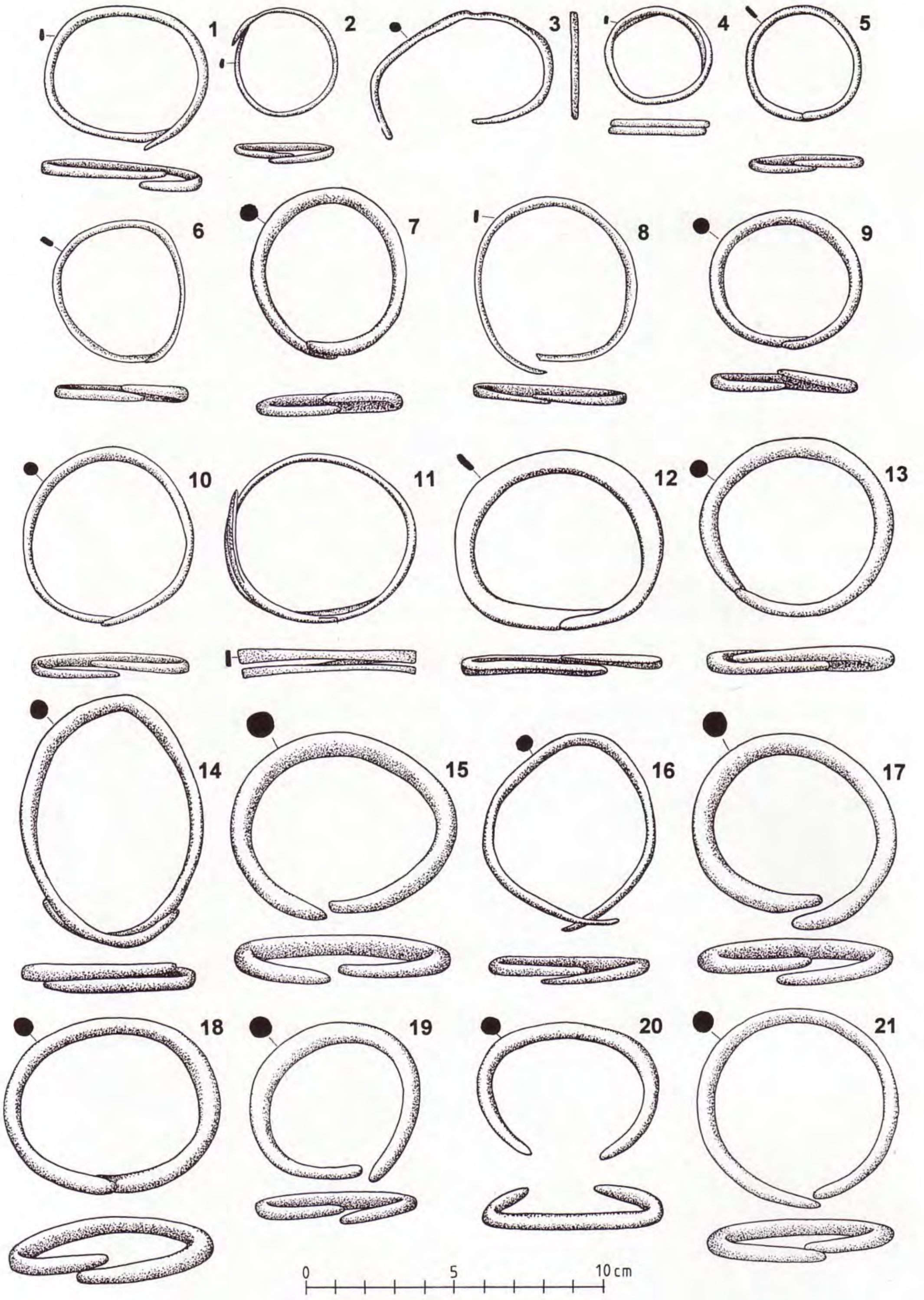

Fig. 7. Durankulak. Cemetery. Copper bracelets. 1.8 grave 447 (HDM 2023, 2024); 2 grave 223 (HDM 2020); 3.5 grave 501 (HDM 2025, 2026); 4 grave 262 (HDM 2017); 6 grave 291 (HDM 2018); 7.9 grave 217 (HDM 1982, 1979); 10 grave 321 (HDM 1980); 11 grave 273 (HDM 2019); 12 grave 309 (HDM 1972); 13 grave 317 (HDM 1973); 14.16 grave 275 (HDM 1968, 1981); 15.17 grave 390 (HDM 1945, 1944); 18.21 grave 251 (HDM 2141, 2142); 19 grave 171 (HDM 1997); 20 grave 29 (HDM 1977).

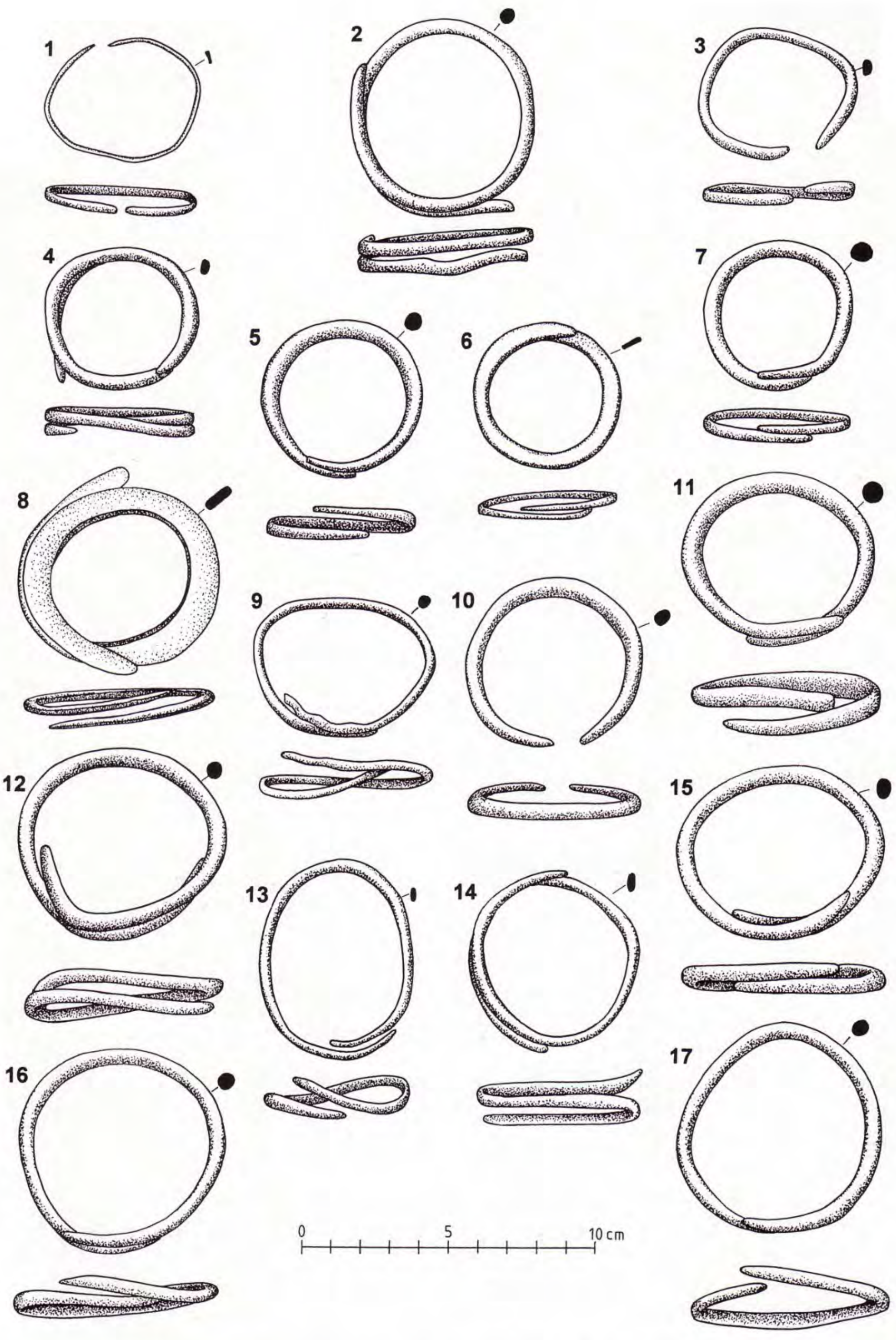

Fig. 8. Durankulak. Cemetery. Copper bracelets. 1 grave 272 (HDM 2016); 2 grave 512 (HDM 2011); 3.9 grave 732 (HDM 1996, 1986); 4.6 grave 660 (HDM 1993, 1984); 5.7 grave 533 (HDM 2015, 2014); 8 grave 389 (HDM 1946); 10.15 grave 514 (HDM 1964, 1963); 11.12 grave 763 (HDM 1991, 1992); 13 grave 672 (HDM 1995); 14 grave 395 (HDM 1947); 16.17 grave 756 (HDM 1989, 1998).

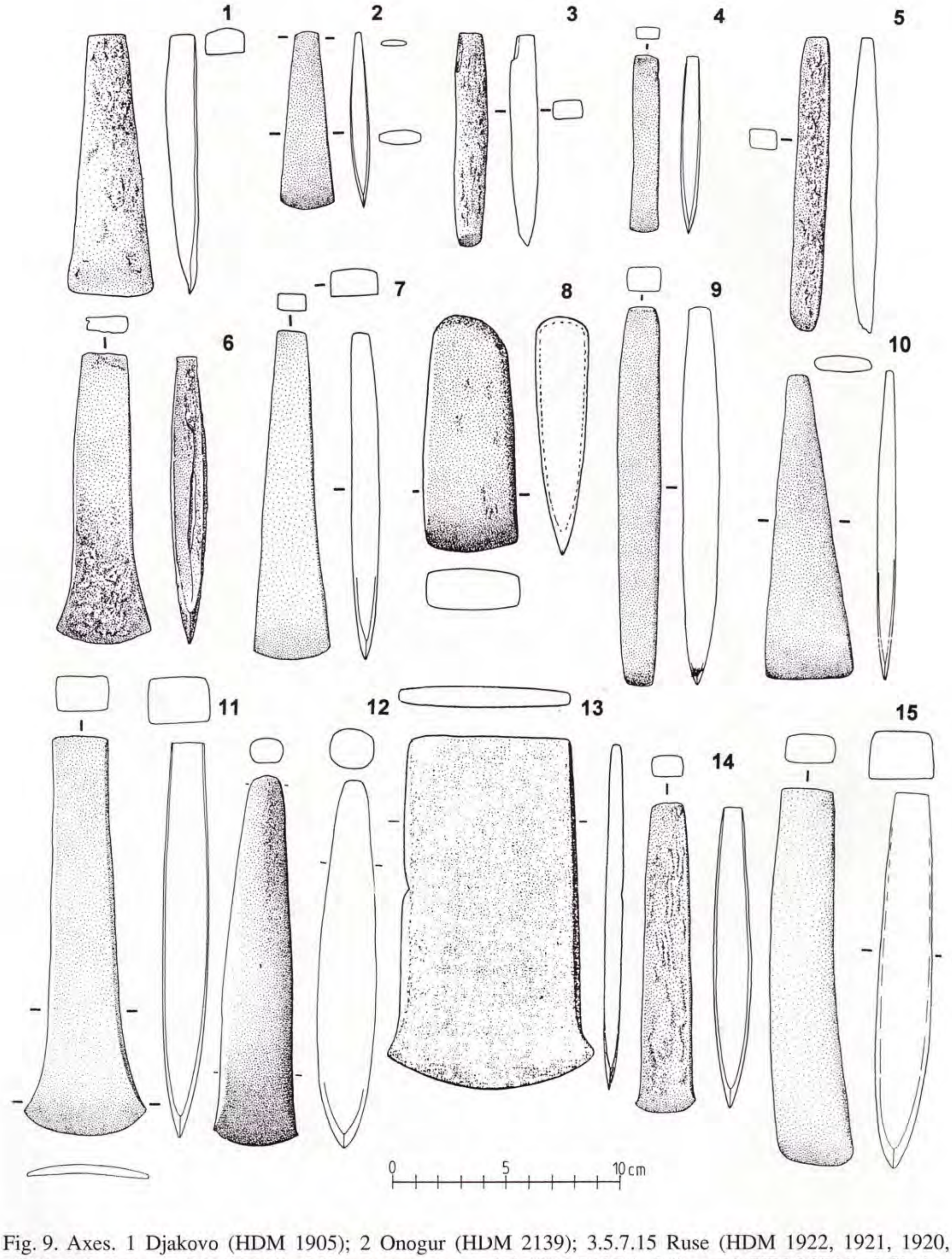

Fig. 9. Axes. 1 Djakovo (HDM 1905); 2 Onogur (HDM 2139); 3.5.7.15 Ruse (HDM 1922, 1921, 1920, 1923); 4 Smjadovo (HDM 2710); 6 Durankulak, grave 623 (HDM 1971); 8 Sozopol (HDM 2744); 9 Slatino (HDM 1904); 10 Debrene (HDM 2140); 11 Mezdra (HDM 2726); 12 Marica (HDM 1912); 13 Hotnica-Vodopada (HDM 1902); 14 Sava (HDM 1924).

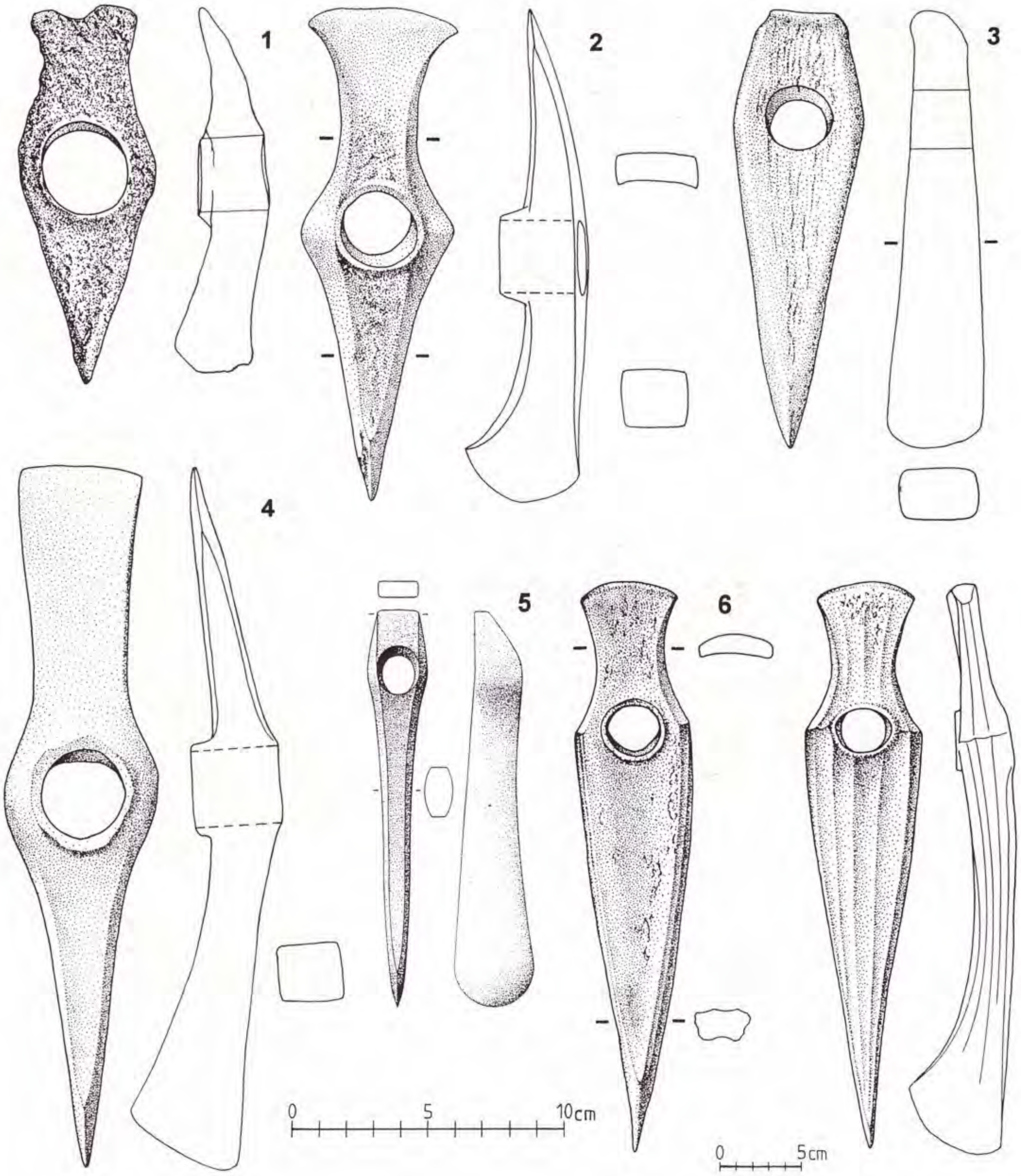

Fig. 10. Heavy implements. 1 Stolnik (HDM 1917); 2 Hotnica-Vodopada (HDM 1901); 3 Radlovci (HDM 1903); 4 Živlovci (HDM 2213); 5 Sadovec (HDM 1915); 6 Kočan (HDM 1916).

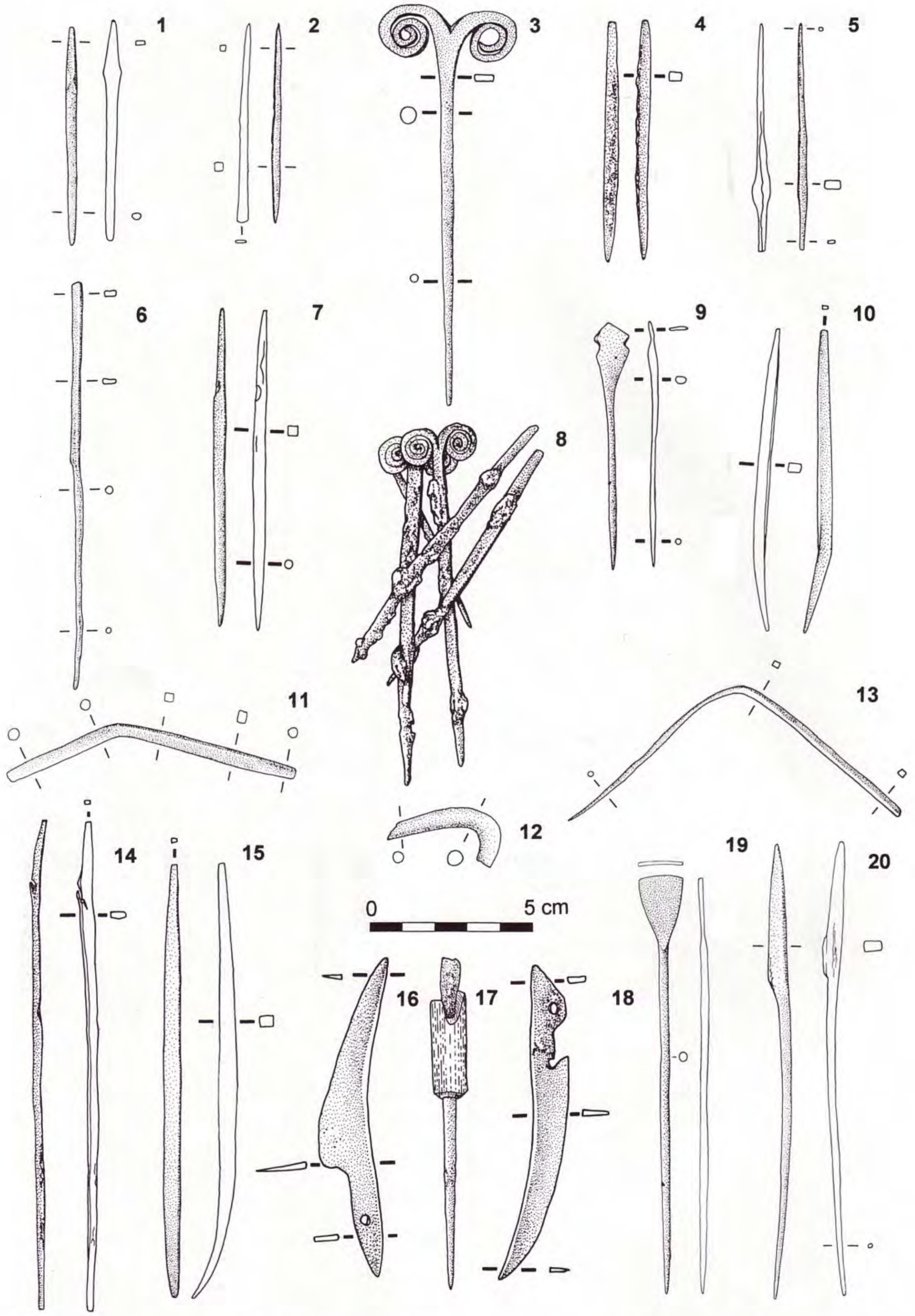

Fig. 11. Copper small finds. 1.2.5 Kačica (HDM 2149, 2148, 2147); 3.4.7.9.10.14-16.18 Teliš (HDM 2105, 2115, 2113, 2109, 2108, 2117, 2116, 2106, 2107); 6.8.11-13.17.20 Ruse (HDM 2059, 2099, 2100, 2081, 2042, 2071, 2083, 2098); Durankulak (HDM 1953).

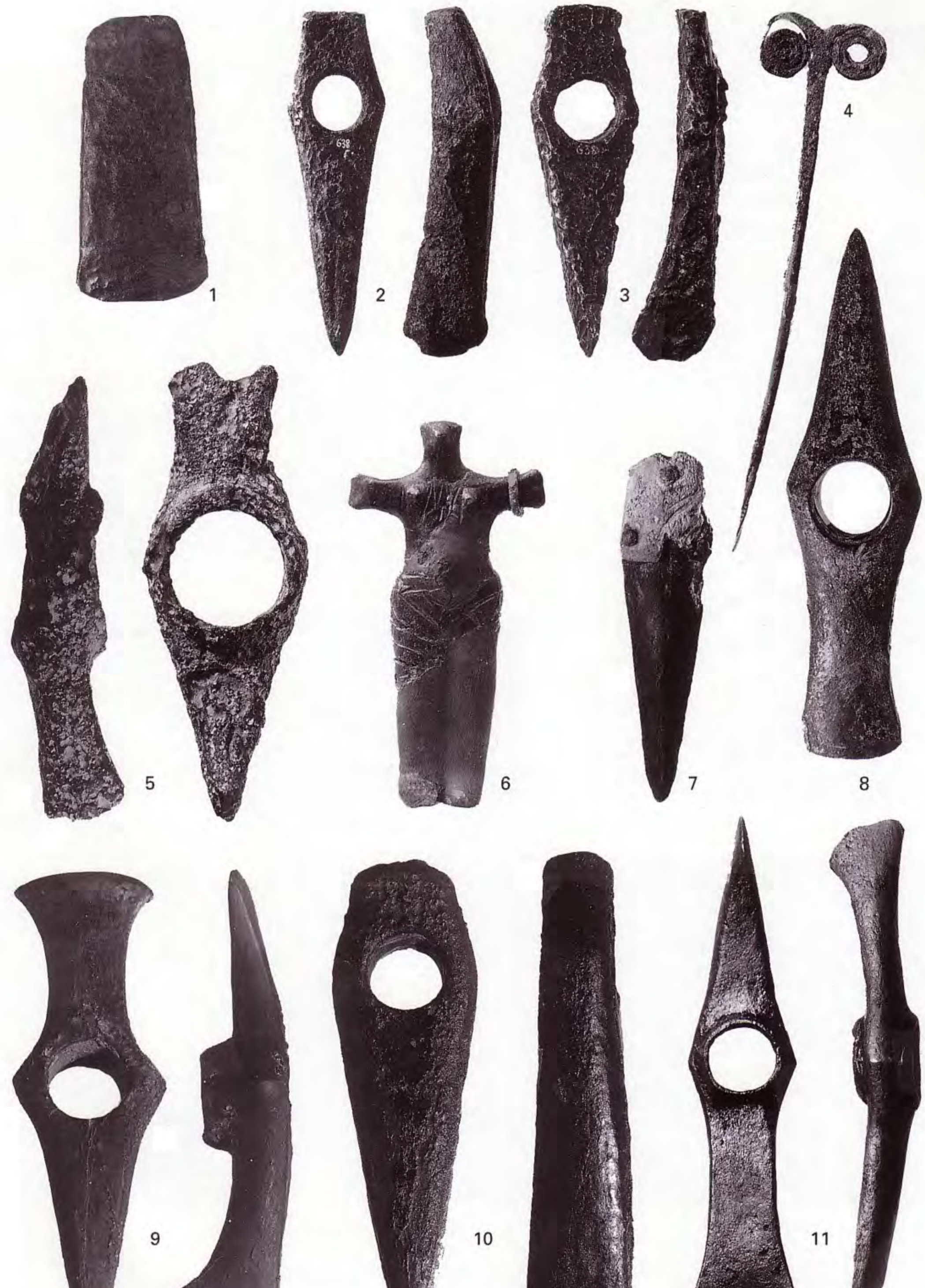

Fig. 12. 1 Flat axe, type Altheim, Jasen (HDM 2701); 2.3 hammer axes, type Pločnik, Dragoman (HDM 1910, 1911); 4 double-spiral-headed pin, Goljamo Delčevo (HDM 2126); 5 axe-adze, miscast, Stolnik (HDM 1917); 6 female idol with copper bracelet, h. 28 cm, Durankulak (not analysed); 7 dagger, type Nerušaj, Durankulak, grave 982 (HDM 1918); 8 axe-adze, type Jászladány, Ivanovo (HDM 1919); 9 axe-adze, type Tîrgu Okna, Hotnica-Vodopada (HDM 1901); 10 hammer axe, type Pločnik, Radlovci (HDM 1903); 11 axe-adze, type Jászladány, Teliš (HDM 1914).

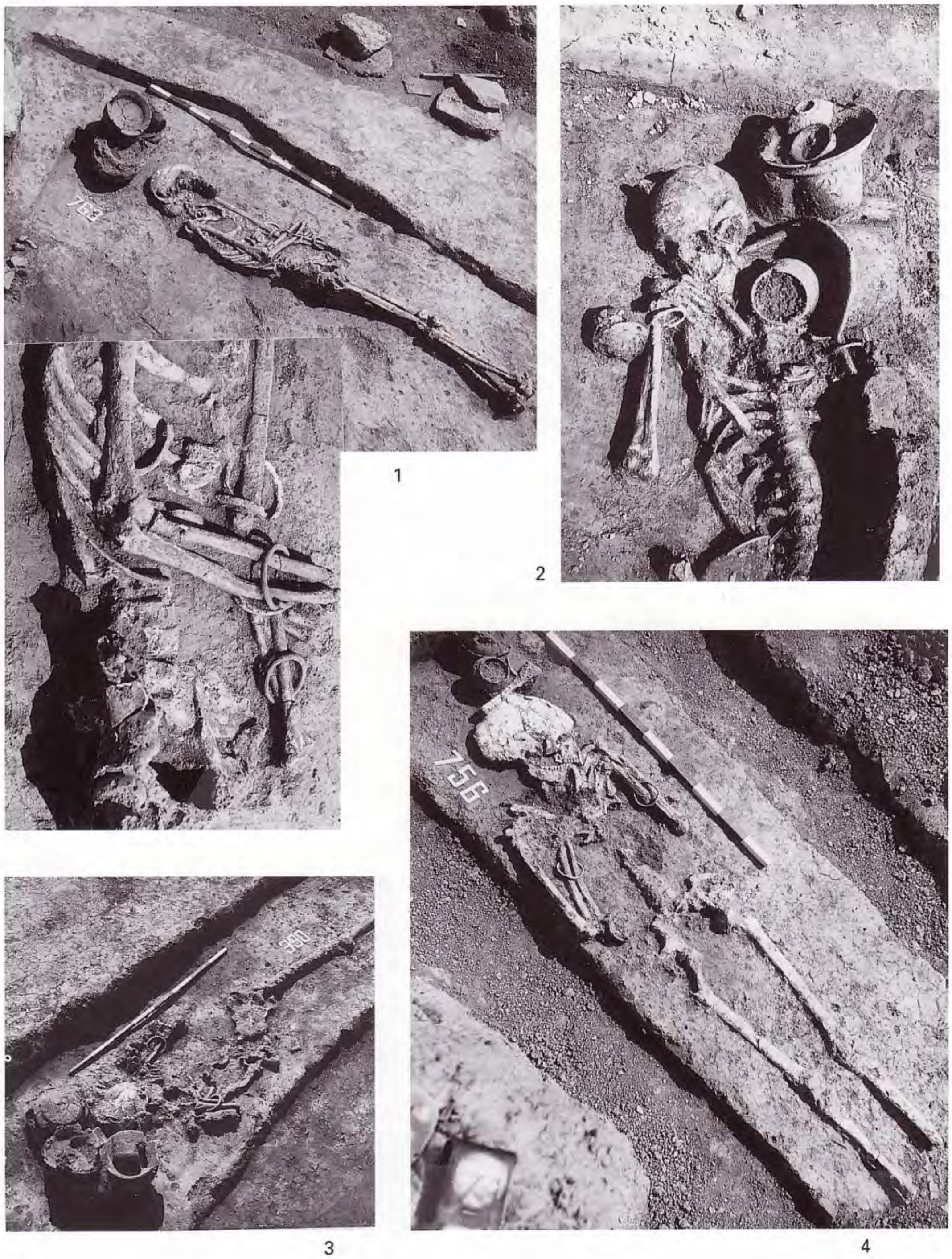

Fig. 13. Durankulak, cemetery. 1 grave 763 with enlargement showing the bracelets *in situ*; 2 grave 512; 3 grave 390; 4 grave 756.

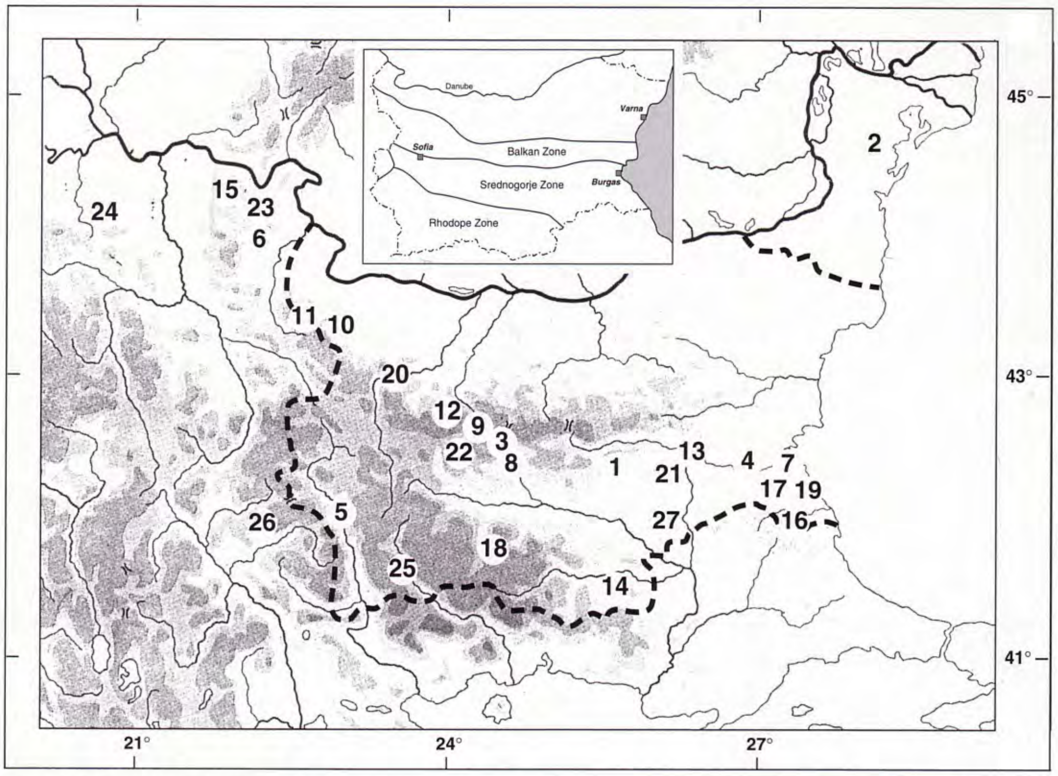

Fig. 14. Mineralized regions of Bulgaria and locations of analyzed copper ores. Numbers relate to the following sites.1 Ai Bunar; 2 Altin Tepe (Ro); 3 Assarel; 4 Bakadžik; 5 Boboševo; 6 Bor; 7 Burgas; 8 Car Assen; 9 Čelopeč; 10 Čiprovci; 11 Čuprene; 12 Elacite; 13 Gorno Alexandrovo; 14 Madžarovo, Popsko; 15 Majdanpek; 16 Malko Tărnovo, Strandža, Ikiztepe; 17 Medni Rid, Rosen, Zidarovo; 18 Mihalkovo; 19 Kiten; 20 Plakalnica; 21 Prochorovo; 22 Radka; 23 Rudna Glava; 24 Rudnik; 25 Skrebatno; 26 Sletovo; 27 Ustrem.

Bulgarian ore deposits and description of ore samples

Bulgaria is rich in base metal deposits. They are concentrated in eleven geographical districts[200] that form essentially three metallogenetic zones[201]: The Balkan, the Srednogorje and the Rhodope zone *(Fig. 14).*

West Balkan metallogenetic zone

In Bulgaria this zone is largely confined to the Stara Planina range. Although it is adjacent to the east Serbian copper belt, with Bor and Majdanpek as major deposits, the two are not genetically related. Three districts can be distinguished:

1) *Čuprene* is an iron skarn deposit. Magnetite[202] and siderite are the main minerals, with smaller amounts of sulfides including chalcopyrite. Genetically, the deposit resembles

[200] Ковачев 1994.

[201] Bogdanov 1982.

[202] Some of the mineral names used in this section may not be familiar to all archaeologists. Therefore, we summarize here their (idealized) chemical compositions. Copper minerals: azurite ($Cu_3[(OH)/(CO_3)_2]$), bornite (Cu_5FeS_4), chalcocite (Cu_2S), chalcopyrite ($CuFeS_2$), covelline (CuS), enargite (Cu_3AsS_4), malachite ($Cu_2[(OH)_2/CO_3]$), tennantite (Cu_3AsS_3). Iron minerals: hematite (Fe_2O_3), limonite (FeO(OH)), magnetite (Fe_3O_4), pyrite (FeS_2), siderite ($FeCO_3$). Lead mineral: galena (PbS). Zinc mineral: sphalerite (ZnS). Base metal ores are usually not monomineralic but mixtures of the above minerals and others as well.

Rudna Glava in eastern Serbia. Old workings have been observed on the steep flank of the river Goljama Reka but they have not been investigated in any detail.
2) The lead-silver deposit at *Čiprovci* occurs in the same marble beds of the diabase-phyllitoid formation as Čuprene. Small amounts of chalcopyrite have been reported but this certainly is not a copper deposit.
3) From the archaeological point of view a most interesting district in western Bulgaria is *Plakalnica* with several ore bodies containing mixed sulfides with bornite, chalcopyrite, sphalerite, galena, pyrite, and tennantite. We had available a sample of almost pure bornite (BG-4) from the present open-cast mine that is still in operation. Old workings have been reported but they seem to date to the medieval period or later. Close to the mine along the stream Leva Reka is an old pile of about 10,000 tons of copper slag. The age is unknown but judging from the appearance of the slags, with a high proportion of glassy phases, they are most probably medieval. Further mining sites in this district are Sedmočislenici, Kalamata, Gladna, Sokolec, and Lăkatnik.

Srednogorje metallogenetic zone

Having been formed during the Alpine folding this zone cuts Bulgaria from west to east and extends over 500 km from the Timok basin in Serbia to the Black Sea coast[203]. It encompasses copper and iron deposits of different genetic types. Within this zone at least seven districts, each comprising several deposits, can be distinguished:

1) *Sofia district*: There are no major copper deposits in this area but the low-temperature hydrothermal iron deposit at *Kremikovci* also contains some chalcopyrite. The deposit was probably not exposed at the surface in ancient times. Our sample (BG-5) consists of limonite and hematite, with inclusions of malachite and native copper.

2) *Panagjurište district*: This is the largest copper district in Bulgaria today. Its present production is based on porphyry copper deposits like the ones at Assarel (BG-8), Medet, and Elacite (BG-9) and on massive sulfide deposits like Car Assen (BG-10) and Radka (BG-11). The upper layers of these deposits have been almost completely removed by open-cast mining, comparable to the situation of the porphyry copper deposit of Majdanpek in Serbia.

We had a sample of native copper (BG-8a) and a piece of very rich ore with 57% Cu (BG-8b) from the mine at *Assarel*. The mineral association of the ore with chalcopyrite, chalcocite, and covelline is typical of a cementation zone. No old workings are known but intensive open-cast mining operations may have erased them. A further sample of native copper (BG-9a) derives from the mine at *Elacite* that is located at the contact of intrusive with metamorphic rocks.

From the massive sulfide deposits at *Car Assen* and *Radka* we had two samples each of native copper (BG-10a, BG-10b, BG-11a, BG-11b), a rather low-grade ore from Car Assen (BG-10c) and a typical sample of mixed sulfide primary ore from Radka (BG-11c) that contains chalcopyrite, pyrite, tennantite, bornite, galena, and sphalerite. No old workings have been reported. All samples from the Panagjurište district were taken from a collection in the museum of the Faculty of geology at the University of Sofia.

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[203] Note that this metallogenic zone is larger in extent than the geographical area called "Sredna Gora" that encompasses the mountain range between east of Sofia and Jambol, some 100 km west of Burgas.

At *Čelopeč* (BG-3) there is a relatively small mineralization, mined today, of mixed sulfides and gold well below the surface, with a predominance of copper minerals. Sample BG-3a derives from a depth of 300 m, it consists of bornite, pyrite, and chalcocite with minor chalcopyrite and covelline. Sample BG-3b, from the southern part of the deposit, consists mainly of enargite with minor chalcopyrite, pyrite, and bornite. No old workings are reported from the site.

3) *Northern Thrace*: Located in the eastern part of the Srednogorje metallogenetic zone the district is rather poor in copper. More common are iron skarn deposits and a polymetallic mineralization with gold, like at Bakadžik. However, some iron deposits apparently experienced a late-stage hydrothermal phase that also deposited some pyrite and chalcopyrite together with hematite and magnetite, as at Krumovo southeast of Stara Zagora.

The deposit at *Ai Bunar*, some 10 km north-west of Stara Zagora, where extensive Chalcolithic copper mining was attested by the excavations of Černych[204] and his colleagues from 1971 until 1974 seems to belong to this type. Based on pottery finds, Černych dated the workings to the Chalcolithic, the earliest pottery belonging to the Marica III phase of the Early Chalcolithic. The most important mineralizations are at Chrišteni, Ai Bunar, Tămjanka, N. Selo, Rakitnica, and Zmejovo. Our nine samples (BG-17a–i) consist mainly of limonite, hematite and some malachite. Only four of those contain more than 10% copper and can thus be considered to be the kind of high-grade ores that will have been smelted in prehistoric times. Samples BG-17e–i derive from a pile of waste ore at the rim of the Malka Tumba site (probably Černych's mining site no. 4) while the exact find location of samples BG-17a–d is not known.

Another mineralized area is situated around *Prochorovo*, some 50 km to the east of Ai Bunar. Numerous traces of ancient mining are known that resemble those at Ai Bunar. Surface finds of Chalcolithic pottery suggest that this area also may have been exploited in the fifth millennium B.C. but no excavations have been performed so far. Analyses by Černych[205] indicate that the copper produced from these ores should have relatively low concentrations of impurities.

Still further east is the ore field of *Gorno Alexandrovo*, with easily visible mineralized veins on the southern slopes of the Sărnena Gora mountains that form the eastern part of the Sredna Gora. Surface finds of native copper have been reported[206]. Other possible ancient mining sites are known at Tropoklovo, Glasnik, and Hadžíte.

4) *Burgas district*: South of the town of Burgas close to the Black Sea coast are three ore fields: Vărli Brjag, often also referred to as Medni Rid (BG-1), Zidarovo (BG-6), and Rosen (BG-7) of which the latter one is the most important. They can be classified as vein-type hydrothermal copper deposits. The *Vărli Brjag* ore field is confined to the centre of the Upper Cretaceous magmatic activity along the Dară-dere river west of Burgas. The ore veins are similar in size and composition to those of the Rosen field but they do not contain molybdenum and cobalt. Mining sites are Vărli Brjag (samples BG-1a, b), Kara Baïr, and Černy Vrah. Amov and Văkova[207] list Vărli Brjag together with Medni Rid. Černych[208] has chemically analyzed 15 ore samples from Kara Baïr and found them to be rather low-grade ores. The ore field is surrounded by several Chalcolithic tell settlements.

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204 Черных 1978, 56–78.
205 Черных 1978, 40 Fig. 21.
206 Черных 1978, 40–41.
207 Амов/Въкова 1994.
208 Черных 1978.

The *Zidarovo* ore field is located south-west of Lake Burgas on the western slopes of the Medni Rid ridge, with Zidarovo-Jurta (samples BG-6a, native copper; BG-6b, malachite; BG-6c, bornite from a depth of 50–60 m) and Kanarata as major mines. Smaller deposits are known at Kruševec and Izvor. Černych and Kovačev[209] considered these deposits to be different from Rosen but neither lead isotope data[210] nor structural observations allow to distinguish between them. Numerous old workings of unknown age, wooden mining tools, and slag piles are known from this district, although many have been destroyed by modern mining operations.

The *Rosen* ore field extends over 40 km along a narrow strip on the eastern slopes of the Medni Rid ridge. The ore veins contain primarily chalcopyrite and pyrite, with some cobalt and molybdenum minerals. In the oxidation zone occasional finds of native copper have been reported. Numerous old workings as well as slag piles are still present. No systematic survey or any excavation has been performed. Our samples from Rosen are one piece of native copper (BG-7a), two ore samples consisting mainly of malachite with some bornite and chalcopyrite (BG-7b, BG-7c), some sulfide ores of rather pure chalcopyrite (BG-7d), of chalcopyrite and pyrite (BG-1b), and bornite with pyrite and tetrahedrite (BG-1c), all from shallow depths (10–20 m) of the modern mines. The principal mining sites are Rosen-Central (samples BG-7a,b), Rosen-sărneško kladenče (samples BG-7c,d), Propadnala voda, and Čaplaka. Further mineralizations are known at Metocha, Djado Toma, Minna rota, Lobodovo kale, Kaleto, and at Burchamata.

Circumstantial evidence for a Chalcolithic exploitation is perhaps the existence nearby of submerged Chalcolithic settlements in the Black Sea, such as Sozopol. Copper mining and copper trade might explain the presence of these settlements in rather unfertile and swampy areas.

5) *Strandža district*: In this area in the southeastern corner of Bulgaria several copper and copper-iron skarn deposits occur, the largest of which is located near Malko Tărnovo. It is one of the largest copper deposits of Bulgaria.

Four samples of native copper were available (BG-12a–c,e) from *Brădceto* near Malko Tărnovo and a piece of low-grade malachite ore (BG-12d). Similar, and genetically related, deposits are known at Ikiztepe and Dereköy in Turkish Thrace. Both have been worked in antiquity[211] and it is possible that the ancient mining activities extended also into the Malko Tărnovo area. Major mining sites besides Brădceto are Gradište, Propada, and Mladenovo that have all been chemically analyzed by Černych[212]. Somewhat further north is the polymetallic deposit of Gramatikovo with chalcopyrite the predominant copper mineral. No samples have been analyzed from this deposit.

The *Rezovo* deposit is located near the Black Sea shore along the river Rezovska Reka. The mineralization is of the vein type, with iron and manganese minerals as main constituents and accessory copper minerals containing arsenic[213]. Numerous slag piles and old open casts are known. Deposits are known at Silistar (Zelenata Kanara), some 10 km south of Ahtopol near the Turkish border, at Kiten, Izvor, and Staro Rezovo. Today the occurrence of Zelenata Kanara rises just above sea level; the exposed veins of

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209 Черных 1978; Ковачев 1994.
210 Амов/Въкова 1994.
211 Wagner u. a. 1986, TG 188 and TG 189.
212 Черных 1978.
213 Ковачев 1994.

malachite and azurite easily could have been exploited in prehistoric times. A surface find of copper ore consisting of partly oxidized chalcopyrite with pyrite and chalcocite comes from Silistar (Zelenata Kanara, BG-14a), and a small piece of ore consisting of malachite and azurite from the prehistoric pile-dwelling site of Kiten (BG-15a) further south on the Black Sea coast.

Rhodope metallogenetic zone

This zone covers the Rhodope massif and part of the Serbo-Macedonian massif.

1) *East Rhodope district*: To the northeast the district is bound by the Marica valley and to the west by the Central Rhodope deep fault that cuts the Rhodope massif roughly north-south from Haskovo to the Aegean Sea. It contains, among others, numerous lead-zinc deposits where copper becomes occasionally dominant such as at Madžarovo.

The ore deposit of *Madžarovo* has been exploited at least since Roman times and was mined during the sixteenth and seventeenth centuries. It is polymetallic with galena, sphalerite, chalcopyrite, and pyrite as the main minerals. Over forty accessory minerals have been identified, with native copper and gold among them. We had two samples of native copper available for analysis (BG-13a,b).

Genetically quite different is the lead-zinc deposit of *Ustrem* that is actually situated north of the Marica valley and thus does not belong to the East Rhodope district *sensu strictu*. The mineralization seems to be due to the remobilization of lead and zinc from the country rocks. Today intensive mining operations are performed at Lesovo, Ustrem, and Barita that may have destroyed any earlier workings. On the other hand, no archaeological finds earlier than the Late Bronze Age have been found in this region suggesting that it may not have been important in the Chalcolithic.

2) *West Rhodope district*: It comprises the entire uplifted part of the Rhodope massif with some major lead-zinc deposits around Madan (Madan, Lăki, Nedelino, Davidkovo, Ardino and others). The main ore minerals are galena and sphalerite but these are almost always accompanied by other minerals, including copper-bearing ones such as chalcopyrite and various fahlore types. Nevertheless, Madan certainly was never primarily a copper mine but one for lead and silver. This district has been known and exploited since ancient times. In the Middle Ages the mines were operated by the Turks. From this time the name 'Madan' derives, which is the Turkish word for 'ore' as well as 'mine'.

3) *Osogovo district*: This area in the extreme west of present-day Bulgaria belongs to the least investigated ones, geologically and archaeologically speaking. Some finds of malachite and native copper from archaeological contexts derive from there so that it is at least feasible that mining was practiced in antiquity. There are several lead-zinc mineralizations known of which the one at Ruen is the most important. The main minerals are sphalerite, galena, and pyrite, with minor amounts of chalcopyrite, fahlore and other, silver, minerals. From the museum at Kjustendil we had available a relatively large sample of native copper (BG-2a) and a piece of oxidized copper ore with malachite and azurite (BG-2b), allegedly from Boboševo, which is located at the western fringe of the Rhodope zone. In the mineralogical literature there is no mineralization mentioned at Boboševo, however, and both samples may actually derive from Ruen in the Osogovo district further west and close to the Serbian border.

To summarize, we had available 22 copper ores from 14 locations. Clearly this number is too small for a reliable chemical and mineralogical characterization of the Bulgarian ore occurrences. Nevertheless, together with the published literature on the economic geology of Bulgaria the present data provide a rough overview of the principal ore types accessible to prehistoric metallurgists. Moreover, as far as the lead isotopy is concerned the situation is much more favorable because in addition to these results from a few scattered ore deposits there are data from the comprehensive compilation of Amov and Văkova[214] that contains more than 500 ore specimens from 70 deposits from all metallogenetic zones in Bulgaria, including a few deposits in adjacent Serbia and Turkey.

We had also available 14 samples of native copper from nine different locations. Although the number of samples is again small it nevertheless is obvious that native Cu will have been available to the ancients. The statement should therefore be viewed with some caution that "native copper seems not in fact to occur to any extent in Bulgarian copper deposits"[215], a view that, incidentally, would also appear to be at variance with oral reports that at Elisejna/Plakalnica as late as at the beginning of this century many donkey loads of native Cu to have been delivered.

Results and interlaboratory comparison

Our procedures for the chemical and isotope analysis of copper-based alloys were as already published[216] with later amendments[217]. Ore samples were first investigated by optical microscopy of polished sections to determine the major mineral components. Bulk concentrations of copper and bismuth were determined by atomic absorption spectroscopy with a flame atomizer, all other elements except lead in artifacts and ores by instrumental neutron activation (INAA)[218]. The results for artifacts, ores, and native copper are given in *Tab. A1–A3* in the Appendix. Lead contents were measured by isotope dilution mass spectrometry in the course of the determination of the lead isotope abundance ratios. This was accomplished by thermal ionization mass spectrometry after dissolution of the samples in ultra-high-purity acids and chemical separation of lead.

Since a large number of Chalcolithic objects from Bulgaria had already been analyzed by optical emission spectroscopy in the laboratory of the Archaeological Institute of the Academy of Sciences in Moscow it is perhaps of some interest to compare the results of the two methods in some detail. Unfortunately, this is not possible in as large a number of instances as one might expect because there are a number of cases where the documentation is contradictory in itself or simply not good enough to unambiguously decide whether the respective samples indeed come from the same objects. Such cases have been left out of the comparison; we use only such where we feel reasonably sure that the analytical data pertain to the same artifacts.

The result is shown in *Fig. 15*. In each panel the element concentrations reported by Černych are plotted on the ordinate, the ones from the present study on the abscissa; in the case of a perfect agreement all data points should fall on the diagonal. Clearly this is

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[214] Амов/Въкова 1994.
[215] Gale u. a. 1991.
[216] Pernicka 1984; Pernicka u. a. 1984.
[217] Pernicka u. a. 1993; Kuleff/Pernicka 1995.
[218] For experimental details see Kuleff/Pernicka 1995.

not the case. Nevertheless, for arsenic, antimony, silver, nickel, and gold most points fall into the shaded bands that indicate agreement of the respective concentrations within a factor of three. Except for nickel there is perhaps a tendency for the Moscow values to be higher than ours; for lead *(Fig. 15)* this is quite clearly so. The important point, however, is that Černych's classification into chemical groups of the artifacts is hardly affected by these discrepancies. Using our analytical data but retaining the chemical groups as defined by Černych results in a distribution among the groups that is not very different from the original one as it was obtained with the Moscow data. This is also true if we use for this purpose our data for all Chalcolithic artifacts.

In the subsequent discussion we shall not resort to the chemical categories as they were defined by Černych, however, but search for structure in the data by average-link cluster analysis. This method combines into groups objects with similar trace element abundances where, in the present instance, the similarity is based on the logarithms of the concentrations of the elements As, Sb, Co, Ni, Ag, Au, and Se. The justification for this particular selection and some details of the method have been outlined before[219] except that now we have added discriminant analysis as a procedure to calculate for each object the posterior probability that it belongs to the cluster it was assigned to originally. In a few cases this posterior probability led to a regrouping of an object or it turned out that some objects could not reasonably be assigned to a specific cluster at all because the posterior probabilities were about the same for two or three different clusters. Actually, this is to be expected because there is no clearly differentiated multivariate structure in the data and, accordingly, clusters overlap and about 25% of the samples have a significant probability to belong to more than one cluster. This has to be kept in mind when discussing the chemical groups in detail. Nevertheless, most groups are definitely discernible from one another and are stable in the majority of their members.

For the clustering procedure we have used the present data sets for all artifacts from periods up to, and including, the Proto Bronze Age. Because of their geographical and cultural relationship we have added to this 77 contemporaneous Serbian metal objects for which the analyses have been published before[220]. As a result our suite of samples then comprises 335 objects *(Tab. 1)*. According to the clustering procedure more than 95% of these can be sorted into nine clusters, with between 7 and 102 artifacts per cluster.

The chemical characteristics of the nine major clusters are summarized in *Tab. 2* and *Fig. 16*. It is evident that cluster #8 is set apart from all others by exceedingly low concentrations of Sb and Au. Clusters #1, #3, and #4 are quite similar in their Ag, Au, and Se concentrations, but in cluster #3 Sb is high, and clusters #1 and #4 can be told apart by their contents of As and Co. Although there is some overlap between different clusters if only one element is considered, almost complete discrimination is possible even though Co and Ni are rather similar in all groups and do not contribute much to the discrimination in our sample suite. We have nevertheless included them in our clustering procedure to be compatible with the earlier work on Serbia[221].

The cluster assignment of the individual artifacts is listed in *Tab. 3*. In some instances objects from Serbia now fall into the same cluster that, when clustering only Serbian artifacts, belonged to different ones. This is a distinct feature of the clustering algo-

[219] Pernicka u. a. 1990; Pernicka u. a. 1993.
[220] Pernicka u. a. 1993.
[221] Pernicka u. a. 1993.

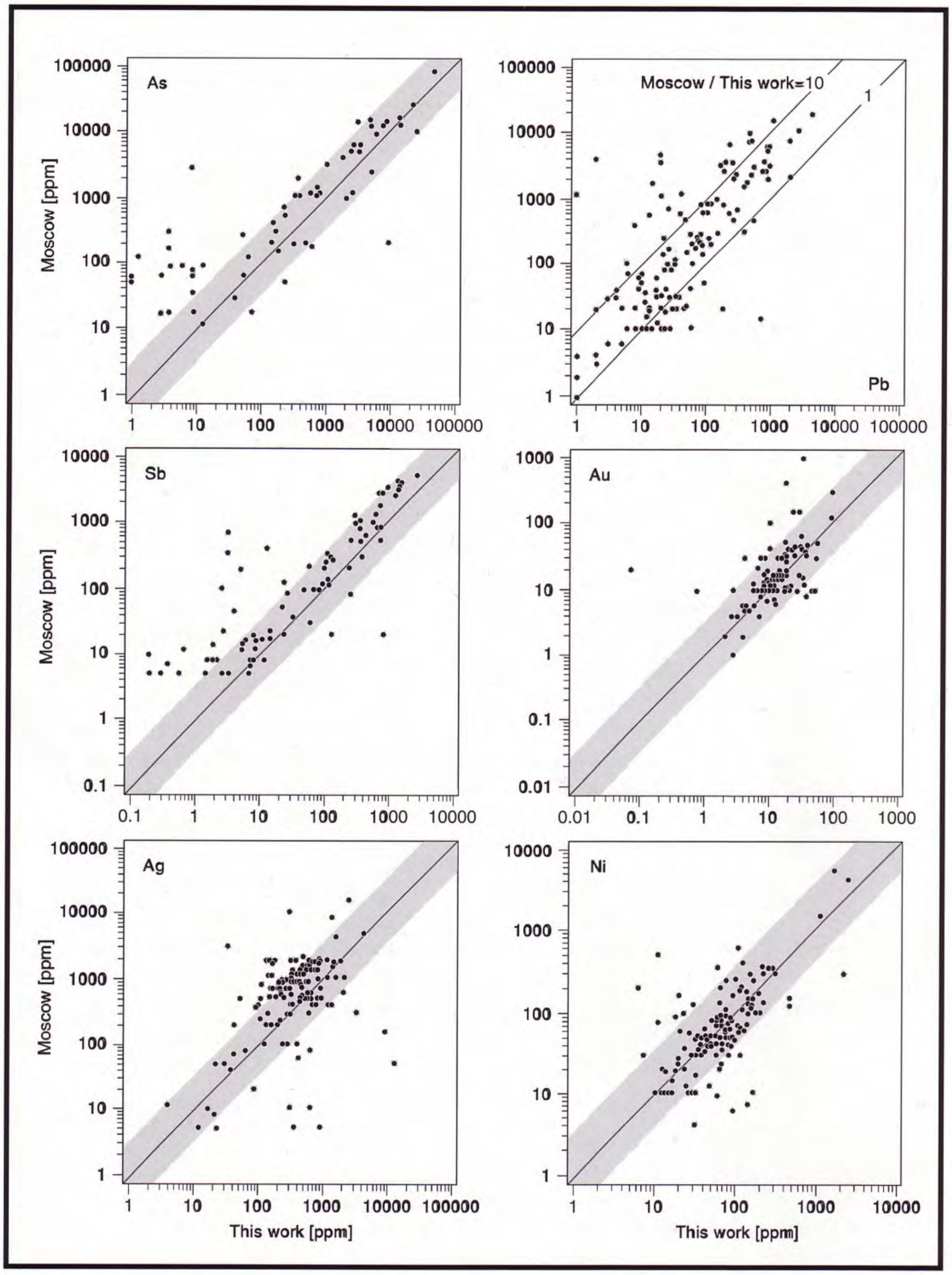

Fig. 15. Comparison of element concentrations determined on aliquants of the same objects via neutron activation analysis and isotope dilution mass spectrometry (for lead) with optical emission spectroscopy results (Moscow). For data points within the shaded bands the respective concentrations agree to within a factor of three.

	Serbia	Bulgaria						
		W	NC	NE	BSC	THR	RHO	Total
Neolithic*	13	–	–	–	7	–	–	20
Chalcolithic								
Early	2	1	–	–	2	1	–	6
Middle	19	6	–	4	6	1	–	36
Late	2	5	8	70	106	1	–	192
Final	52**	10**	17	2	–	1	–	82***
Bronze Age								
Proto	2	4	6	–	4	–	3	19
Early	–	2	–	–	-	–	–	2
Late	–	4	4	–	6	–	–	14
Classical	–	–	–	–	1	–	–	1
Uncertain	–	2	–	11	2	–	–	15
Total	90**	34**	35	87	134	4	3	387

* All Neolithic objects are malachites
** One sample not analyzed for its lead isotopy
*** Two samples not analyzed for their lead isotopy

Table 1. Summary of archaeological ages and find regions of artifacts investigated.

	Cluster #1 (102 samples)			Cluster #2 (58 samples)			Cluster #3 (42 samples)			Cluster #4 (42 samples			Cluster #5 (23 samples)		
As	1.0	**3**	17	4	**15**	99	84	**770**	5100	8	**61**	350	1.3	**4**	44
Sb	0.9	**6**	100	2.0	**5**	14	120	**450**	1500	1.2	**6**	26	0.6	**4**	14
Co	0.4	**2**	6	0.4	**1.2**	4	0.7	**2**	4	5	**15**	85	1.2	**4**	8
Ni	16	**41**	140	23	**53**	94	14	**39**	120	60	**99**	280	41	**120**	200
Ag	200	**450**	1200	9	**19**	59	330	**630**	890	63	**170**	450	20	**40**	130
Au	5	**11**	33	0.2	**0.4**	0.8	6	**14**	38	0.9	**8**	37	0.1	**0.8**	1.8
Se	28	**72**	220	0.5	**1.4**	5	33	**100**	300	13	**42**	100	14	**38**	150

	Cluster #6 (16 samples)			Cluster #7 (17 samples)			Cluster #8 (15 samples)			Cluster #9 (7 samples)		
As	11	**36**	550	560	**8700**	49000	1.0	**1.5**	2.9	620	**3800**	26000
Sb	9	**37**	160	38	**150**	390	0.01	**0.2**	0.6	140	**270**	3400
Co	0.6	**2**	3	0.3	**2**	7	1.3	**13**	56	9	**25**	109
Ni	71	**160**	510	25	**73**	240	7	**32**	47	53	**190**	1200
Ag	3000	**1300**	13000	68	**150**	450	3	**6**	21	48	**400**	990
Au	4	**10**	53	0.25	**3**	7	0.02	**0.07**	0.2	4.8	**13**	59
Se	3	**21**	200	9	**23**	56	0.7	**1.7**	5.5	1.1	**8**	25

Table 2. Composition of nine major chemical clusters among 335 copper-based early metal artifacts from Bulgaria and Serbia. For each element the following parameters are given in µg/g in ascending order: The 10% percentile, the median, and the 90% percentile of the distribution of elemental concentrations. Results were rounded to one or two significant digits. Clusters were obtained by average-link cluster analysis excluding Neolithic malachites and samples younger than Proto Bronze Age.

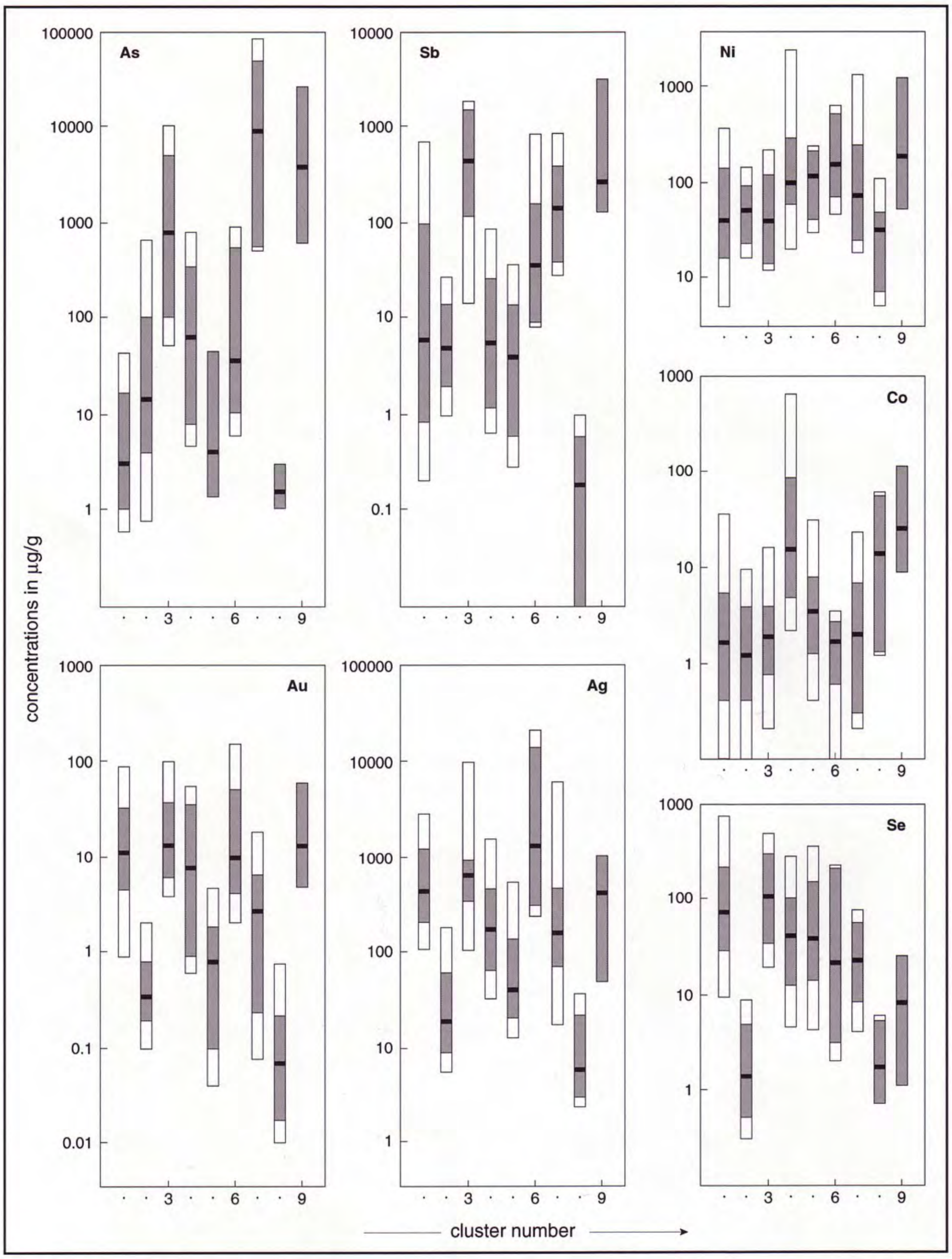

Fig. 16. Chemical characteristics of the nine major clusters found for 335 metal samples from Bulgaria (258) and Serbia (77) covering the Chalcolithic and Proto Bronze Age. Plotted for each element are the total range of concentrations (light), the range comprising 80% of the members of each cluster (grey), and the median concentration (black).

Table 3

HDM number	site	region	context	object	grouplet	cluster	ASM anal. nr.	ASM group
Late Neolithic and Early Chalcolithic malachite samples								
1481	Selevac	SRB	b.h. 76: II	malachite	rPb	–		
1482	Selevac	SRB	b.h. 76: III	malachite	–	–		
1484	Selevac	SRB	b.h. 76: V	malachite	1	–		
1485	Selevac	SRB	b.h. 76: III	malachite	1	–		
1486	Selevac	SRB	b.h. 76: I–V	malachite	–	–		
1487	Selevac	SRB	b.h. 76: III	malachite	1	–		
1488	Selevac	SRB	b.h. 76: V	malachite	–	–		
1489	Selevac	SRB	b.h. 76: III	malachite	1	–		
1490	Selevac	SRB	b.h. 76: V	malachite	–	–		
1491	Selevac	SRB	b.h. 76: V	malachite	5	–		
1492	Selevac	SRB	b.h. 76: IV	malachite	6, 10	–		
1493	Selevac	SRB	b.h. 76: V	malachite	9, 10	–		
1495	Selevac	SRB	b.h. 76: V	malachite	8	–		
1969	Durankulak	BSC	G626	malachite bead	6, 9	–		
1970	Durankulak	BSC	G626	malachite bead	9	8		
2004	Durankulak	BSC	G611	malachite bead	7	–		
2209	Durankulak	BSC	G648	malachite bead	3	8		
2210	Durankulak	BSC	G643	malachite bead	3	–		
2211	Durankulak	BSC	G643	malachite bead	6, 9	–		
2212	Durankulak	BSC	G643	malachite bead	3	–		
Early Chalcolithic								
1483	Selevac	SRB	b.h. 76: III	copper prill	rPb	8		
1494	Selevac	SRB	b.h. 76: V	copper prill	rPb	8		
1904	Slatino	W	tell	chisel	2	1		
1912	Marica (Devebargan)	THR	tell	heavy axe, oval cross section	6	1	20420	III
1975	Durankulak	BSC	G 681	finger ring	2	3	45394	V
2005	Durankulak	BSC	G 545	finger ring	rPb	1	45383	III
Middle Chalcolithic								
1308	Šumrakovac	SRB	stray find	hammer axe (Pločnik)	10	7		
1496	Gomolava	SRB	G 12/76	bracelet	5	8		
1555	Pločnik	SRB	railtrack	chisel	2	2		
1556	Pločnik	SRB	hoard	flat axe	2	3		
1557	Pločnik	SRB	hoard	flat axe	2	3		
1558	Pločnik	SRB	railtrack	hammer axe (Pločnik)	–	8		
1559	Pločnik	SRB	hoard	hammer axe (Pločnik)	7	8		
1560	Pločnik	SRB	hoard	chisel	–	8		
1561	Pločnik	SRB	hoard	chisel	10	8		
1562	Pločnik	SRB	hoard	chisel	4	1		
1563	Pločnik	SRB	hoard	chisel	3	1		
1564	Pločnik	SRB	hoard	chisel	2	1		
1565	Pločnik	SRB	hoard	chisel	2	1		
1566	Pločnik	SRB	stray find	chisel	8	1		
1567	Pločnik	SRB	hoard	chisel	2	3		
1568	Pločnik	SRB	hoard	chisel	7	8		
1569	Pločnik	SRB	hoard	chisel	2	1		
1570	Pločnik	SRB	hoard	chisel	10	1		

Table 3 (continued)

HDM number	site	region	context	object	grouplet	cluster	ASM nr.	ASM group
1571	Pločnik	SRB	railtrack	hammer axe (Pločnik)	2	3		
1903	Radlovci	W	stray find	hammer axe (Pločnik)	1	2	39107	I
1910	Dragoman	W	hoard	hammer axe (Pločnik A)	1	2, 5	20417	II
1911	Dragoman	W	hoard	hammer axe (Pločnik A)	1	2	20418	I
1940	Ai Bunar	THR	hoard	hammer axe (Pločnik B)	3	1	12687	III
1976	Durankulak	BSC	G 29	bracelet	rPb	1	45351	IV
1977	Durankulak	BSC	G 29	bracelet	2	3	45352	V
2011	Durankulak	BSC	G 512	bracelet	9	7	45372	V
2025	Durankulak	BSC	G 501	bracelet	9	4	45369	VI
2026	Durankulak	BSC	G 501	bracelet	9	4	45368	VI
2041	Tell Ruse	NE	3.10	borer	4	6	10883	II
2049	Tell Ruse	NE	4.65	borer	4	1	10830	I
2064	Tell Ruse	NE	5.70	borer	1	5	10832	III
2080	Tell Ruse	NE	4.65	borer	1	–		
2124	Goljamo Delčevo	BSC	b.h. 3–4	borer	–	4	12699	I
2702	Dăržanica	W	hoard	heavy axe (Gumelniţa)	rPb	8	10720	I
2703	Dăržanica	W	hoard	heavy axe (Sălcuţa)	1	2	10721	I
2726	Mezdra	W	tell	heavy axe (Pločnik)	1	2		
Late Chalcolithic								
1422	Bubanj, Novo Selo	SRB	b.h. 1	chisel	1	5		
1431	Stari Kostolac	SRB	hoard	chisel	1	2		
1905	Djakovo	W	b.h. 2	flat axe	3	1		
1915	Sadovec	NC	tell	hammer axe (Vidra A)	3	1		
1920	Tell Ruse	NE	0.10	flat axe (Kamenar)	7	4		
1921	Tell Ruse	NE	unknown	chisel	2	3		
1922	Tell Ruse	NE	0.20	chisel	rPb	4		
1923	Tell Ruse	NE	unknown	heavy axe (Gumelniţa)	2	1		
1924	Sava, cemetery	BSC	G 6	heavy axe (Coţeana)	2	1		
1925	Varna I	BSC	G 36	hammer axe (Vidra B)	6	1	13832	III
1926	Varna I	BSC	G 1	hammer axe (Vidra B)	8	1		
1927	Varna I	BSC	G 1	flat axe (Sălcuţa)	4	1		
1928	Devnja	BSC	G 18	hammer axe (Čoka-Varna)	–	3	9329	IV
1929	Devnja	BSC	G 1	hammer axe (Varna)	3	1	9246	II
1930	Devnja	BSC	G 23	hammer axe (Devnja)	rPb	1	9330	II
1931	Varna I	BSC	G 4	hammer axe (Devnja)	4	1	13813	III

Table 3 (continued)

HDM number	site	region	context	object	grouplet	cluster	ASM nr.	ASM group
1932	Varna I	BSC	G 4	chisel	6	1	13815	III
1933	Devnja	BSC	G 1	heavy axe (Gumelniţa)	2	1		
1934	Devnja	BSC	G 24	hammer axe (Čoka-Varna)	2	3	9328	V
1935	Devnja	BSC	G 26	chisel	6	1	9327	II
1936	Devnja	BSC	G 4	hammer axe (Čoka-Varna)	2	3	9247	V
1937	Hotnica Tell	NC	b.h. 1	hammer axe (Vidra A)	1	1	10895	II
1938	Hotnica Tell	NC	b.h. 1	hammer axe (Vidra B)	6	4	10896	II
1939	Hotnica Tell	NC	b.h. 1	heavy axe (Coţeana)	8	8	10897	II
1942	Durankulak	BSC	G 408	finger ring	4	1	36233	III
1943	Durankulak	BSC	G 433	finger ring	5	4		
1944	Durankulak	BSC	G 390	bracelet	5	1	36235	III
1945	Durankulak	BSC	G 390	bracelet	5	1	36234	III
1946	Durankulak	BSC	G 389	bracelet	rPb	1	36236	II
1947	Durankulak	BSC	G 395	bracelet	rPb	4	36257	VI
1948	Durankulak	BSC	child burial	bracelet	5	4	36225	III
1949	Durankulak	BSC	child burial	bracelet	rPb	4	36251	IV
1956	Durankulak	BSC	b.h. 3	bracelet	9	7		
1959	Durankulak	BSC	b.h. 4	pin	4	–		
1960	Durankulak	BSC	b.h. 3	bracelet	9	7		
1961	Durankulak	BSC	G 977	hammer axe (Čoka-Varna)	2	1	45407	V
1962	Durankulak	BSC	G 514	finger ring	rPb	4	45377	III
1963	Durankulak	BSC	G 514	bracelet	2	3	45379	V
1964	Durankulak	BSC	G 514	bracelet	2	3	45378	V
1965	Durankulak	BSC	G 253	bracelet	rPb	4	36214	IV
1967	Durankulak	BSC	G 731	finger ring	2	3	45396	V
1968	Durankulak	BSC	G 275	bracelet	rPb	4	36212	III
1971	Durankulak	BSC	G 623	flat axe (Sălcuţa)	10	1	45388	II
1972	Durankulak	BSC	G 309	bracelet	9	7	36207	V
1973	Durankulak	BSC	G 317	bracelet	rPb	4		
1974	Durankulak	BSC	G 679	finger ring	rPb	4	45393	VI
1978	Durankulak	BSC	G 253	bracelet	2	3	36213	VI
1979	Durankulak	BSC	G 217	bracelet	3	8	36237	I
1980	Durankulak	BSC	G 321	bracelet	rPb	1, 4	36202	III
1981	Durankulak	BSC	G 275	bracelet	10	1	36238	II
1982	Durankulak	BSC	G 217	bracelet	7	1	36205	VI
1983	Durankulak	BSC	b.h. 4	bracelet	5	1		
1984	Durankulak	BSC	G 660	bracelet	2	3	45389	V
1985	Durankulak	BSC	G 559	bracelet	3	1	45387	VI
1986	Durankulak	BSC	G 732	bracelet	1, 4, 8	3	45397	V
1987	Durankulak	BSC	G 733	bracelet	7	1	45399	III
1988	Durankulak	BSC	G 739	bracelet	rPb	1	45400	III
1989	Durankulak	BSC	G 756	bracelet	2	1	45402	V
1990	Durankulak	BSC	G 722	bracelet	2	3	45395	V
1991	Durankulak	BSC	G 763	bracelet	2	3	45404	IV
1992	Durankulak	BSC	G 763	bracelet	2	3	45405	V
1993	Durankulak	BSC	G 660	bracelet	rPb	4	45390	II

Table 3 (continued)

HDM number	site	region	context	object	grouplet	cluster	ASM nr.	ASM group
1994	Durankulak	BSC	G 672	bracelet	rPb	1	45391	III
1995	Durankulak	BSC	G 672	bracelet	rPb	4	45392	VI
1996	Durankulak	BSC	G 732	bracelet	2	3	45398	V
1997	Durankulak	BSC	G 171	bracelet	5	4	45353	III
1998	Durankulak	BSC	G 756	bracelet	2	1	45403	IV
1999	Durankulak	BSC	G 867	flat axe	8	1	45406	V
2000	Durankulak	BSC	G 460	finger ring	rPb	4	45366	VI
2001	Durankulak	BSC	G 684	finger ring	1	9	45412	V
2002	Durankulak	BSC	G 496	finger ring	rPb	4	45367	VI
2003	Durankulak	BSC	G 554	finger ring	–	1	45384	VI
2006	Durankulak	BSC	G 502	spiral ring	9	1, 6	45370	IV
2007	Durankulak	BSC	G 514	spiral ring	2	3	45373	V
2008	Durankulak	BSC	G 514	finger ring	rPb	4	45374	IV
2009	Durankulak	BSC	G 447	spiral ring	–	1	45360	VI
2010	Durankulak	BSC	G 320	borer	10	4	36203	II
2012	Durankulak	BSC	G 530	bracelet	5	8	45380	III
2013	Durankulak	BSC	G 455	finger ring	4	1	45364	III
2014	Durankulak	BSC	G 533	bracelet	rPb	4	45381	IV
2015	Durankulak	BSC	G 533	bracelet	9	1	45382	III
2016	Durankulak	BSC	G 272	bracelet	7	1	36240	II
2017	Durankulak	BSC	G 262	bracelet	rPb	1	36239	III
2018	Durankulak	BSC	G 291	bracelet	rPb	4	36243	III
2019	Durankulak	BSC	G 273	bracelet	–	1	45358	
2020	Durankulak	BSC	G 223	bracelet	–	1	36220	III
2021	Durankulak	BSC	G 330	flat axe	10	1	36200	II
2022	Durankulak	BSC	G 320	hammer axe (Varna)	4	1	36199	IV
2023	Durankulak	BSC	G 447	bracelet	rPb	1	45361	III
2024	Durankulak	BSC	G 447	bracelet	rPb	1	45362	III
2027	Durankulak	BSC	G 447	finger ring	2	3	45359	V
2028	Durankulak	BSC	G 454	finger ring	rPb	4	45363	III
2029	Durankulak	BSC	G 455	finger ring	2	3	45365	V
2030	Durankulak	BSC	G 741	finger ring	2	3	45401	V
2031	Durankulak	BSC	G 506	finger ring	rPb	4		
2032	Durankulak	BSC	G 1202	borer	2	1		
2034	Durankulak	BSC	G 1183	pin	3	1		
2035	Durankulak	BSC	G 298	spiral ring	2	1	36210	III
2036	Durankulak	BSC	G 506	spiral ring	rPb	–	45371	III
2039	Tell Ruse	NE	unknown	borer	5	7, 3		
2040	Tell Ruse	NE	2.25	borer	4	1		
2043	Tell Ruse	NE	2.25	borer	7	5		
2044	Tell Ruse	NE	0.90	borer	2	1	10885	II
2045	Tell Ruse	NE	4.40	borer	2	3	10878	V
2047	Tell Ruse	NE	unknown	double-spiral-headed pin	5	1		
2050	Tell Ruse	NE	1.60	borer	1	4	10852	IV
2051	Tell Ruse	NE	1.80	borer	5	3		
2052	Tell Ruse	NE	1.80	borer	5	–		
2053	Tell Ruse	NE	0.10	chisel	2	3		
2054	Tell Ruse	NE	2.70	borer	3	1	10823	III
2055	Tell Ruse	NE	1.20	borer	5	6	10850	II
2056	Tell Ruse	NE	2.20	borer	8	5		
2057	Tell Ruse	NE	unknown	borer	2	3		

Table 3 (continued)

HDM number	site	region	context	object	grouplet	cluster	ASM nr.	ASM group
2058	Tell Ruse	NE	2.00	borer	1	2		
2059	Tell Ruse	NE	2.00	borer	7	5	10861	II
2060	Tell Ruse	NE	2.70	borer	6	1	10888	III
2061	Tell Ruse	NE	2.10	borer	10	5	10847	II
2063	Tell Ruse	NE	1.40	borer	rPb	5	10860	III
2065	Tell Ruse	NE	unknown	borer	1	4		
2067	Tell Ruse	NE	2.10	borer	4	1	10808	III
2068	Tell Ruse	NE	unknown	borer	7	1	10851	III
2070	Tell Ruse	NE	3.00	borer	2	1	10822	IV
2071	Tell Ruse	NE	2.25	borer	2	1	10849	IV
2072	Tell Ruse	NE	2.10	borer	3	4	10846	IV
2073	Tell Ruse	NE	1.25	double-spiral-headed pin	3	1		
2074	Tell Ruse	NE	2.50	borer	9	1	10833	II
2075	Tell Ruse	NE	2.25	loop-headed pin	8	5		
2076	Tell Ruse	NE	2.25	borer	2	3	10859	IV
2077	Tell Ruse	NE	1.25	borer	4	1		
2078	Tell Ruse	NE	1.25	borer	4	1		
2079	Tell Ruse	NE	1.20	borer	3	1		
2081	Tell Ruse	NE	2.70	borer	4	1	10884	II
2082	Tell Ruse	NE	unknown	borer	3	1		
2083	Tell Ruse	NE	1.20	borer	3	1		
2084	Tell Ruse	NE	1.35	borer	3	4		
2085	Tell Ruse	NE	0.75	borer	rPb	5		
2088	Tell Ruse	NE	unknown	borer	6	1		
2089	Tell Ruse	NE	1.10	loop-headed pin	7	4	10866	II
2090	Tell Ruse	NE	2.70	borer	8	–	10879	III
2091	Tell Ruse	NE	3.50	borer	rPb	1	10887	II
2092	Tell Ruse	NE	unknown	loop-headed pin	1	6		
2093	Tell Ruse	NE	1.30	borer	3	6, 1	10865	II
2096	Tell Ruse	NE	2.45	borer	3	6, 1		
2097	Tell Ruse	NE	1.00	borer	4	–	10827	V
2098	Tell Ruse	NE	2.00	pin	2	3		
2099	Tell Ruse	NE	1.20	double-spiral-headed pin	4	1	10820	II
2100	Tell Ruse	NE	1.20	double-spiral-headed pin	5	6	10821	II
2101	Tell Ruse	NE	1.75	borer	7	5	10858	I
2102	Tell Ruse	NE	3.50	borer	7	1		
2103	Tell Ruse	NE	3.50	borer	7	1		
2104	Tell Ruse	NE	0.10	loop-headed pin	1	5		
2119	Goljamo Delčevo	BSC	b.h. 10	borer	6	1		
2120	Goljamo Delčevo	BSC	G 31	borer	6	1	12148	III
2121	Goljamo Delčevo	BSC	b.h. 13	bracelet	2	3	12703	III
2122	Goljamo Delčevo	BSC	b.h. 15	spiral ring	10	1	12700	II
2123	Goljamo Delčevo	BSC	b.h. 14	spiral ring	2	3	12701	I
2125	Goljamo Delčevo	BSC	b.h. 11	borer	rPb	3		
2126	Goljamo Delčevo	BSC	G 31	double-spiral-headed pin	4	3	12149	VI
2127	Goljamo Delčevo	BSC	b.h. 17	borer	rPb	1		
2128	Goljamo Delčevo	BSC	b.h. 8	borer	2	3		
2129	Goljamo Delčevo	BSC	G 25	borer	5	1	12147	III
2130	Goljamo Delčevo	BSC	b.h. 15	borer	6	1		

Table 3 (continued)

HDM number	site	region	context	object	grouplet	cluster	ASM nr.	ASM group
2137	Durankulak	BSC	G 496	finger ring	rPb	4		
2138	Durankulak	BSC	G 496	finger ring	5	4		
2139	Onogur castle	BSC	stray find	flat axe (Sălcuţa?)	3	3		
2140	Debrene/Prilep	BSC	stray find	flat axe (Goljamo Delčevo)	1	7		
2141	Durankulak	BSC	G 251	bracelet	2	3	45357	V
2142	Durankulak	BSC	G 251	bracelet	2	-		
2143	Hotnica Tell	NC	b.h. 1	borer	1	3	10899	II
2144	Hotnica Tell	NC	b.h. 1	borer	3	1	10900	II
2145	Hotnica Tell	NC	b.h. 1	borer	3	1	10901	II
2146	Hotnica Tell	NC	b.h. 1	borer	3	5		
2150	Karanovo Tell	THR	tell	borer	3	6	12029	II
2707	Goljamo Delčevo	BSC	G 25	hooked wire	3	6	12146	II
2709	Smjadovo	NE	b.h. 3	copper strip	4	5		
2710	Smjadovo	NE	b.h. 3	chisel	4	1		
2711	Smjadovo	NE	b.h. 4	borer	4	1		
2712	Smjadovo	NE	b.h. 4	casting spill	3	1		
2713	Smjadovo	NE	b.h. 4	casting spill	3	1		
2714	Smjadovo	NE	b.h. 4	borer	4	1		
2715	Smjadovo	NE	b.h. 4	ear ring	2	3		
2716	Smjadovo	NE	b.h. 3	borer	rPb	1		
2717	Smjadovo	NE	b.h. 2	borer	5	1		
2718	Smjadovo	NE	stray find	borer	3	1		
2719	Smjadovo	NE	b.h. 4	borer	4	1		
2720	Smjadovo	NE	b.h. 3	borer	9	9		
2721	Smjadovo	NE	b.h. 3	borer	9	1		
2722	Smjadovo	NE	b.h. 4	borer	2	3		
2725	Zaminec	W	b.h. C	flat axe	4	6		
2727	Krivodol	W	2.10	borer	1	4		
2728	Zaminec	W	b.h. B	borer	1	2		
2733	Zaminec	W	b.h. B	borer	2	9		
2743	Sozopol	BSC	under water	heavy axe (Gumelniţa?)	n.m.	-		
2744	Sozopol	BSC	under water	heavy axe (Gumelniţa?)	n.m.	-		
Final Chalcolithic								
1301	Dušanovac	SRB	stray find	axe-adze (Mezőkeresztes)	1	2		
1302	eastern Serbia	SRB	stray find	axe-adze (Jászladány)	1	2		
1303	Urovica	SRB	stray find	axe-adze (Jászladány)	7	5		
1304	Šarkamen, Ostava	SRB	stray find	axe-adze (Jászladány)	1	2		
1306	Jabukovac	SRB	stray find	axe-adze (Jászladány)	1	2		
1307	Urovica	SRB	stray find	shafthole axe	3	9		
1309	Šarkamen, Ostava	SRB	stray find	axe-adze (Jászladány)	1	2		
1310	Šarkamen, Ostava	SRB	stray find	axe-adze (Jászladány)	1	2		

Table 3 (continued)

HDM number	site	region	context	object	grouplet	cluster	ASM nr.	ASM group
1311	Vrazogrnac	SRB	stray find	axe-adze (Jászladány)	3	5		
1312	Šarkamen, Ostava	SRB	stray find	axe-adze (Jászladány)	1	2		
1313	eastern Serbia	SRB	stray find	axe-adze (Jászladány)	1	2		
1314	Šarkamen, Ostava	SRB	stray find	axe-adze (Jászladány)	1	2		
1315	Šarkamen, Ostava	SRB	stray find	axe-adze (Jászladány)	1	2		
1317	Voluja	SRB	stray find	hammer axe	1	2		
1318	Glogovica	SRB	stray find	axe-adze (Jászladány)	1	4		
1319	Donja Bela Reka	SRB	stray find	axe-adze (Jászladány)	1	2		
1320	Gradac	SRB	stray find	hammer axe	1	5		
1321	Zlotska pečina	SRB	cave	flat axe	1	2		
1322	Donji Milanovac	SRB	stray find	axe-adze (Jászladány)	4	2		
1323	Serbia	SRB	stray find	axe-adze (Jászladány)	1	2		
1325	Osnić	SRB	stray find	axe-adze (Jászladány)	1	2		
1326	Krivelj	SRB	stray find	axe-adze (Jászladány)	1	2		
1327	Zlotska pečina	SRB	cave	chisel	1	2		
1328	Zlotska pečina	SRB	cave	chisel	8	8		
1329	Zlotska pečina	SRB	cave	borer	1	5		
1330	Zlotska pečina	SRB	cave	borer	–	–		
1331	Zlotska pečina	SRB	cave	borer	1	4, 5		
1332	Zlotska pečina	SRB	cave	borer	–	–		
1333	Zlotska pečina	SRB	cave	borer	1	2		
1334	eastern Serbia	SRB	stray find	pin	1	2		
1335	Zlotska pečina	SRB	cave	borer	1	2		
1336	Zlotska pečina	SRB	cave	awl	1	2		
1337	Krivelj	SRB	stray find	borer	1	6		
1338	Zlotska pečina	SRB	cave	dagger	n.m.	3		
1401	Stojačak	SRB	stray find	axe-adze (Jászladány)	10	4		
1402	Stojačak	SRB	stray find	axe-adze (Jászladány)	3,6	–		
1405	Svilajnc	SRB	stray find	axe-adze (Jászladány)	1	2		
1408	Selo Vrba	SRB	stray find	axe-adze (Jászladány)	–	2		
1409	Serbia	SRB	stray find	flat axe	1	2		
1413	Makrešane	SRB	stray find	axe-adze (Jászladány)	1	2		
1414	eastern Serbia	SRB	stray find	axe-adze (Jászladány)	1	2		
1415	Polna/Blagotin	SRB	stray find	chisel	1	5		
1416	vicinity of Niš	SRB	stray find	axe-adze (Jászladány)	1	2		

Table 3 (continued)

HDM number	site	region	context	object	grouplet	cluster	ASM nr.	ASM group
1417	Krivi Vir, Niš	SRB	stray find	axe-adze (Jászladány)	1	2		
1418	Lazinje, Mokra	SRB	stray find	axe-adze (Jászladány)	1	2		
1419	Pesak, Varoš	SRB	stray find	axe-adze (Jászladány)	1	2		
1420	Crvena stena	SRB	stray find	flat axe	1	2		
1421	Jelašnice	SRB	stray find	chisel	10	5		
1426	Prižren	SRB	stray find	axe-adze (Jászladány)	1	2		
1427	Veliko Laole	SRB	stray find	axe-adze (Jászladány)	8	2		
1428	Starčevo (M. Bubanj)	SRB	stray find	axe-adze (Jászladány)	1	2		
1430	Dvorište	SRB	stray find	axe-adze (Jászladány)	1	2		
1901	Hotnica	NC	stray find	axe-adze (Tîrgu Okna)	1	2	20842	II
1906	Plakuder	W	hoard	axe-adze (Jászladány)	1, 2	2	9150	I
1907	Plakuder	W	hoard	axe-adze (Jászladány)	1	2	9152	I
1908	Plakuder	W	hoard	axe-adze (Jászladány)	1	2	9153	I
1909	Plakuder	W	hoard	axe-adze (Jászladány)	1	2	9151	I
1913	Teliš	NC	b.h. 2	axe-adze (Jászladány?)		5, 1		
1914	Teliš	NC	b.h. 2	axe-adze (Jászladány)	1	7	28350	I
1916	Kočan	W	stray find	hammer axe (Mezökeresztes)	1	2		
1917	Stolnik/Elin Pelin	W	stray find	axe-adze (Jászladány)	1	2		
1919	Ivanovo	NE	stray find	axe-adze (Jászladány)	6	–		
1941	Ai Bunar	THR	hoard	axe-adze (Tîrgu Okna)	4	7	12686	VI
2105	Teliš	NC	b.h. 3	double-spiral-headed pin	8	3		
2106	Teliš	NC	b.h. 2	knife	8	4		
2107	Teliš	NC	b.h. 2	knife	6	6		
2108	Teliš	NC	b.h. 3	borer	1	4		
2109	Teliš	NC	unknown	pin	3	1		
2110	Teliš	NC	b.h. 2	borer	6	1		
2111	Teliš	NC	b.h. 2	borer	1	2		
2112	Teliš	NC	b.h. 2	borer	8	5		
2113	Teliš	NC	b.h. 2	borer	1	5		
2114	Teliš	NC	b.h. 2	borer	3	–		
2115	Teliš	NC	b.h. 2	borer	1	4		
2116	Teliš	NC	b.h. 2	borer	–	5		

Table 3 (continued)

HDM number	site	region	context	object	grouplet	cluster	ASM nr.	ASM group
2117	Teliš	NC	b.h. 2	borer	5	6		
2118	Teliš	NC	b.h. 2	borer	6	4		
2213	Živlovci/Montana	W	stray find	axe-adze (Jászladány)	n.m.	4		
2705	Plakuder	W	hoard	axe-adze (Jászladány)	1	2	10723	I
2706	Plakuder	W	hoard	axe-adze (Ariuşt)	1	2	10722	I
2708	Smjadovo	NE	stray find	axe-adze (Ariuşt)	9	6		
2724	Rebărkovo	W	hoard	axe-adze (Ariuşt)	1	2		
Proto Bronze Age								
1339	Zlotska pečina	SRB	cave	dagger	1	2		
1410	Serbia	SRB	stray find	flat axe (Altheim)	8	7		
1902	Hotnica, region	NC	stray find	flat axe (Altheim, var. Ete)	1	7	20844	VI
1918	Durankulak	BSC	G 982	dagger (Nerušaj)	1	7		
1952	Durankulak	BSC	tell	dagger	2	9		
1954	Durankulak	BSC	tell	pin	1, 2, 4	1		
1966	Durankulak	BSC	tell	dagger (Usatovo)	3	7	36201	VII
2134	Hotnica-Vodopada	NC	stray find	dagger (Mondsee)	1	7	36281	
2135	Hotnica-Vodopada	NC	b.h. 1	dagger (Mondsee)	8	7	36282	
2147	Kačica	NC	1.50	borer	3	–		
2148	Kačica	NC	unknown	borer	3	9		
2149	Kačica	NC	1.80	borer	3	6		
2701	Jasen	W	stray find	flat axe (Altheim, var. Ete)	1	2	10719	I
2736	Malorad	W	stray find	dagger	7	–		
2737	Galiče	W	stray find	dagger (Cucuteni)	8	7		
2738	Lesura	W	stray find	dagger	–	9	10689	VII
2740	Haramijska dupka	RHD	b.h. 2	dagger (Jagodina)	6	6	34405	
2741	Haramijska dupka	RHD	b.h. 2	dagger (Sofievka)	9	7	34406	
2742	Haramijska dupka	RHD	b.h. 2	borer	3	6		
Early Bronze Age								
2704	Vidbol	W	stray find	flanged axe	3, 8	7	10718	VII
2739	Lesura	W	stray find	dagger	4	7	10690	IX
Late Bronze Age								
1955	Durankulak	BSC	b.h. 2	metal band	h	7		
1957	Durankulak	BSC	b.h. 2	pin	h	7		
1958	Durankulak	BSC	b.h. 2	spiral-headed pin	h	7		
2033	Durankulak	BSC	stray find	spiral-headed pin	4	7	45408	
2037	Koslodujči	BSC	stray find	sickle	h	7		
2038	Vodnjanci	BSC	stray find	double axe	9	7		
2131	Hotnica-Vodopada	NC	b.h. 2	borer	9	7		
2132	Hotnica, region	NC	stray find	dagger	h	7, 6		
2133	Hotnica, region	NC	stray find	dagger	9	7		
2136	Hotnica-Vodopada	NC	stray find	metal fragment	9	–		
2730	Krivodol/Oslen	W	stray find	knife	h	7		
2731	Gradešnica	W	stray find	borer	4	7		
2732	Gradešnica	W	stray find	borer	–	1		
2734	Gradešnica	W	stray find	loop-headed pin	h	7		

Table 3 (continued)

HDM number	site	region	context	object	grouplet	cluster	ASM nr.	ASM group
Classical Period								
1953	Durankulak	BSC	tell	stylo	9	7		
Unstratified or uncertain								
1950	Durankulak	BSC	stray find	wire	–	3		
1951	Durankulak	BSC	stray find	wire	9	3		
2042	Tell Ruse	NE	1.35	hook	8	4	10877	II
2046	Tell Ruse	NE	1.35	borer	2	7	10875	X
2048	Tell Ruse	NE	unknown	bracelet	3	9, 7		
2062	Tell Ruse	NE	0.70	borer	rPb	5	10876	I
2066	Tell Ruse	NE	0.70	borer	4	7, 9		
2069	Tell Ruse	NE	unknown	chisel	h	3, 7		
2086	Tell Ruse	NE	unknown	borer	2	3		
2087	Tell Ruse	NE	unknown	borer	2	3		
2094	Tell Ruse	NE	unknown	pin	5	7		
2095	Tell Ruse	NE	unknown	pin	10	4		
2723	Zlatarevo	NE	stray find	borer	8	7		
2729	Gradešnica	W	house 2	borer	1	5		
2735	Gradešnica	W	unclear	borer	–	7		

Table 3. Summary of relevant archaeological information with chemical clusters and isotope grouplets of all samples studied. Where appropriate, the specific type of an object has been included in brackets. Regions are abbreviated as follows: BSC = Black Sea coast, NC = northern central Bulgaria, NE = north-east Bulgaria, RHD = Rhodope mountains, SRB = Serbia, THR = Bulgarian Thrace, W = western Bulgaria. In the "context" column: b.h. = building horizon, G = grave. For finds from Ruse the depth below surface is given in meters. Malachite finds from Selevac have been included[222]. Ambiguous cluster and grouplet assignments are indicated by listing the various possibilities; n.m. = not measured. Samples that can be assigned to more than two clusters or belong to a cluster with less than four members are designated "outliers" ("–"). ASM = analysis number and group assignment after Черных 1978. Samples younger than Proto Bronze Age have been assigned to the clusters as established by average-link cluster analysis of all metal objects older than, and including, Proto Bronze Age.

rithm; it does not simply sort objects into procrustean bins with predetermined upper and lower concentration limits of certain elements but searches anew for similarities among the objects once the set of input data is changed, be it by replacing samples or by changing the number of samples in the set.

A comparison of the structure within the data as it shows up when they are subjected to average-link cluster analysis, on the one hand, and when grouped as suggested by Černych, reveals that cluster #2 corresponds well with Černych's group I, cluster #3 with group V, and cluster #1 with groups II and III[223]. In all other cases each cluster contains members from several of Černych's groups, and vice versa.

For a detailed analysis of the lead isotope data *(Tab. A4)* we have subdivided the artifacts into 11 grouplets[224] with between 82 and 11 members per grouplet *(Tab. 4)*. This

[222] Pernicka u. a. 1993

[223] In this particular case it is solely the bismuth content, not measured by us, that splits one cluster into two Černych groups.

[224] In the subsequent discussion we shall refer to the isotope clusters as "grouplets" and reserve "cluster" for the chemically defined subsets.

leaves some 13 objects ungrouped. This number is somewhat arbitrary, however, because, to some extent, it is a question of definition how narrowly one defines a grouplet and whether one ascribes a set of abundance ratios to one of the grouplets or calls it an outlier. This ambiguity is evident from the fact that some of the grouplets are very tight and well separated from their neighbors while others are much looser and in at least one of the panels of *Figs. 17* and *18* show some overlap with one or more of the adjacent ones. In a few instances samples fit equally well into more than one grouplet. Such cases are marked in *Tab. 3* by listing all possibilities. A special case is the grouplet designated rPb. We have assigned to it all objects with a $^{208}Pb/^{206}Pb$ ratio below 2.050 (a somewhat arbitrary value). For such "radiogenic" lead the laws of physics and of geochemistry cause the three ^{206}Pb-normalized abundance ratios to be strongly correlated. They also may cause a wide variation in lead isotope abundance ratios in ore samples that otherwise are quite similar. This, then, makes it reasonable to combine such objects in a single grouplet.

The position of the grouplets in three-isotope plots is depicted in *Figs. 17* and *18*. Also shown are the contour lines of Bulgarian ore isotope fields as defined by Amov and Văkova[225], henceforth referred to as A&V. Although one may argue the reality as independent entities of some of these isotope ore fields we shall, for the sake of simplicity, retain them as defined by A&V. We also retain, for the present purpose, a partial presentation of the results in a $^{207}Pb/^{204}Pb$ vs. $^{208}Pb/^{204}Pb$ plot as it was chosen by A&V *(Fig. 18)*. There are some ore fields in A&V, however, that are entirely made up of samples other than copper ores; they have been left out in order not to clutter the figures. Our own results for ores and native copper extend field G but otherwise fall into regions of the diagram that are populated by other Bulgarian ores already. The only notable exception is BG-6b, a malachite sample from Zidarovo, near Burgas. Since it will be of importance in the discussion on the provenance of the artifacts we mention already here that, in the left panel of *Fig. 17*, this sample falls into the empty space between fields F and G. Actually, a closer scrutiny of the A&V compilation reveals that this void is not quite as empty as it appears. There are two ore occurrences in their tables that fall into this region but, for reasons unknown to us, have not been considered in defining the isotope fields. One is a set of seven samples from Zidarovo, Jurta (#42)[226] with mean isotope abundance ratios of $^{208}Pb/^{206}Pb = 2.0757$, $^{207}Pb/^{206}Pb = 0.8411$, $^{204}Pb/^{206}Pb = 0.05390$ that agree well with our ore from Zidarovo. The other instance where samples from the compilation of A&V plot in this region of the diagram is three specimens of Cu ore from Lăkatnik (#23) in the Isremec zone north of Sofia with abundance ratios $^{208}Pb/^{206}Pb = 2.0771$, $^{207}Pb/^{206}Pb = 0.8423$, and $^{204}Pb/^{206}Pb = 0.05380$.

As is evident from *Figs. 17* and *18*, some of the artifact grouplets are clearly associated with certain ore fields like, e.g., grouplet #2 with field F, #3 with field L, #5 with C, and #6 with field G. In other cases (grouplets #1, 4, and 8) a correspondence is less clear, and finally for grouplet #7 in the lower left of the $^{208}Pb/^{206}Pb$ vs. $^{207}Pb/^{206}Pb$ plot there are (as yet) no Bulgarian or Serbian Cu ores known with a fitting lead isotope signature. Although the general problem of provenance of the Cu in the Bulgarian artifacts will be discussed below, we mention already here that grouplet #1 (with 82 members) is isotopically matched by the ores from Majdanpek[227] and grouplet #2 (n = 54) by those from Ai Bunar.

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225 Amov/Văkova 1994.

226 Sample numbers of Amov/Văkova 1994 refer to their compilation, not to our *Fig. 14*.

227 Pernicka u. a. 1993.

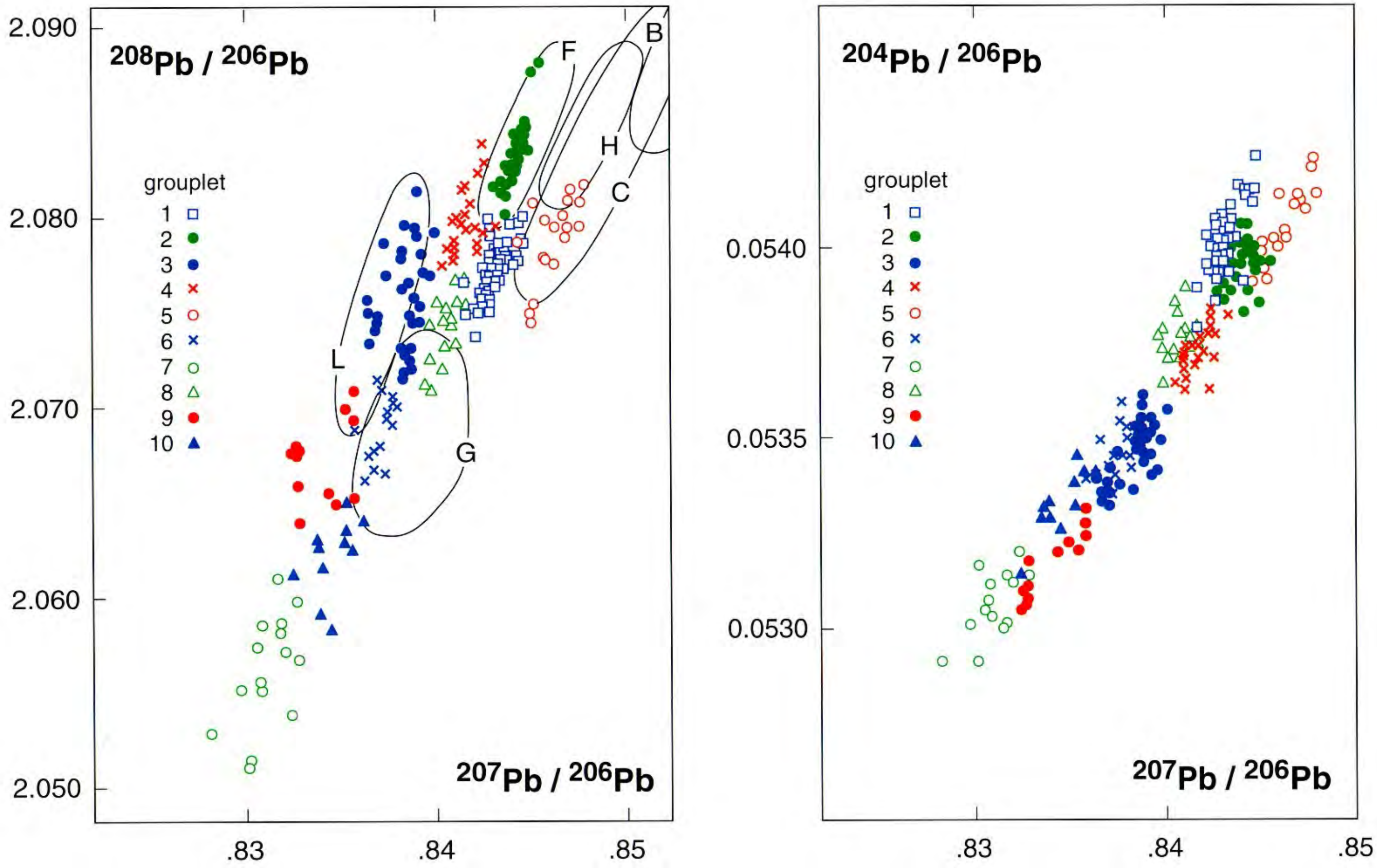

Fig. 17. ^{206}Pb-normalized three-isotope plots for lead isotope abundance grouplets of lead in copper artifacts. Also shown, as visual aid, are Bulgarian copper ore fields as suggested by Amov/Vǎkova 1994. Note that the artifact isotope grouplets are defined completely independent of the "ore fields".

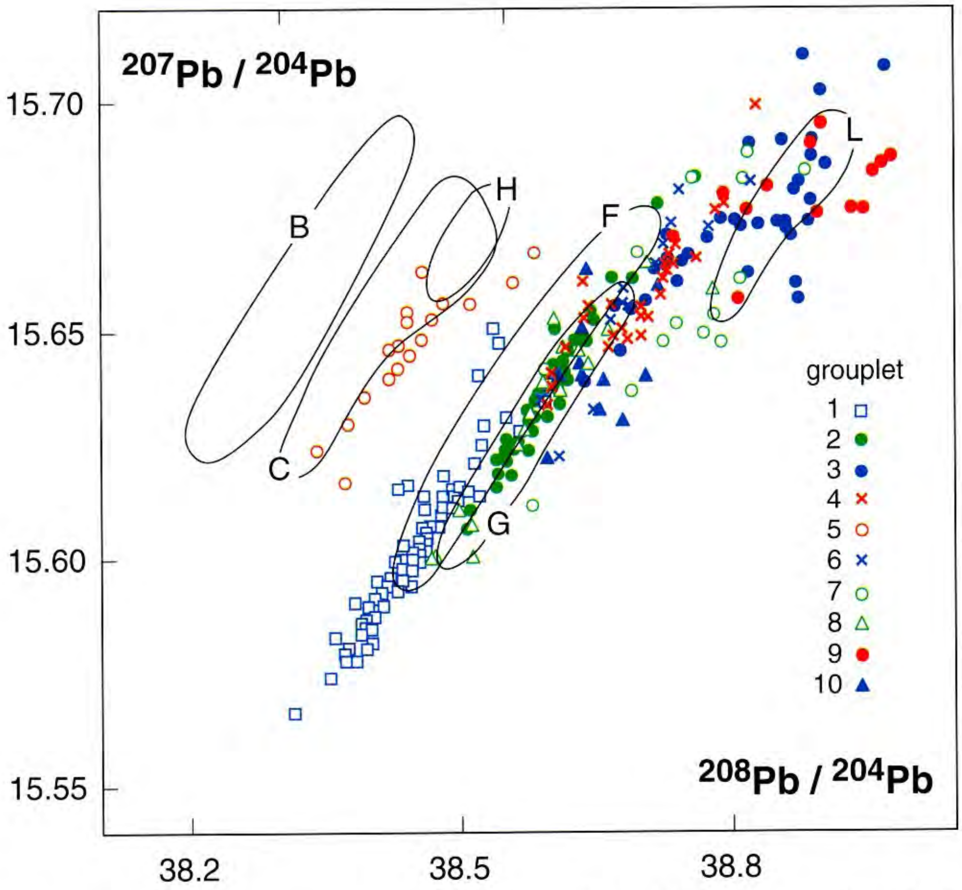

Fig. 18. Presentation of the lead isotope abundance ratios, normalized to ^{204}Pb, and contour lines of Bulgarian copper ore isotope fields as proposed by Amov/Văkova 1994.

Unfortunately, these assignments are not unique, however, because generally there is more than one occurrence whose ores match the lead isotope signatures of the grouplets as they are defined here *(Fig. 19)*:

Grouplet #1: Aside from Majdanpek fitting ores are also found at Čelopeč east of Sofia (BG-3a) and in the Burgas-Strandža districts near the Black Sea coast (BG-6b, BG-15, TG 188B-4, A&V 42.7).

Grouplet #2: The lead isotopy of Ai Bunar is matched by ores some 100 km to the west (BG-3a,b and BG-11c, A&V 36, 39, 40 from the Panagjurište district), by ores from east of Ai Bunar at Prochorovo (A&V 41), and by a sample consisting of malachite-chalcopyrite (TG 188A-1) from Ikiztepe on the Turkish side in the Strandža mountains.[228]

Grouplet #3: Sletovo and Trepča (A&V 50 and 51) in eastern Macedonia; four occurrences near Popsko in the eastern Rhodope mountains (A&V 66–69), two near Burgas (A&V 42.4 and 43) and one from Malko Tărnovo(A&V 47) in the Strandža district.

Grouplet #4: Čelopeč (BG-3b and A&V 40) and Elacite (A&V 39) from the western part of the Srednogorje metallogenetic zone east of Sofia, from Prochorovo (A&V 41) in its eastern part, and from Popsko (A&V 67) in the eastern Rhodope mountains.

[228] Wagner u. a. 1986.

Grouplet #5: In the Balkan metallogenetic zone fitting ores are found at Čuprene (A&V 7) in northwestern Bulgaria and in the vicinity of Vraca north of Sofia (BG-4, A&V 10, 11, 12, 16), and also in the Burgas/Rosen region at Medni Rid (BG-1c).

Grouplet #6: Skrebatno (A&V 53) and Boboševo (BG-2b) in western Bulgaria, Mihalkovo (A&V 58) in the central Rhodope range and Madžarovo (A&V 70) in its eastern part. And again there are several fitting ore deposits in the Burgas-Strandža region (BG-1b, 6c, 12d, A&V 42.4, 43, 44, 47, 48).

Grouplet #7: No ores with a matching lead isotopic composition are known in Bulgaria and Serbia. Of the "archaeological" ores, however, a malachite bead from Durankulak (HDM 2001) and one from Selevac (HDM 1482) are very close.

Grouplet #8: In the west of the Balkan metallogenetic zone north-east of Sofia at Delibirat (A&V 16) and in south-east Bulgaria at Zidarovo (A&V 42.7), at Bakadžik (A&V 43) and again in the Turkish part of the Strandža mountains at Ikiztepe (TG 188B-4).

Grouplet #9: In the western Rhodope mountains at Skrebatno (A&V 53), in the central parts at Mihalkovo (A&V 58) and more to the east at Madžarovo (A&V 70), and in the Strandža mountains at Malko Tărnovo(BG-12d) on the Bulgarian-Turkish border.

Grouplet #10: Two occurrences near Burgas (Medni Rid, BG-1b and A&V 44; Rosen, BG-7c) and again BG-12d from Malko Tărnovo that fits grouplets #9 and #10 about equally well.

Regrettably, most of these deposits are chemically not sufficiently well characterized to allow any reasonably meaningful predictions into which chemical cluster the metal smelted from them might fall. Most likely, however, different ores belonging to the same lead isotope grouplet will not always yield metal with the same, or very similar, trace element concentrations. This then, is the explanation that there is not a one-to-one correspondence between chemical cluster and isotope grouplets (see below). But it means also that, for the time being, the metal provenancing problem can only be attacked via the lead isotopic composition - aside from plausibility arguments and circumstantial evidence. Until the ore occurrences have been fully characterized chemically, some of the conclusions should therefore be considered as tentative.

For the 38 artifacts with $^{208}Pb/^{206}Pb$ abundance ratios below 2.050 a regression analysis reveals that all data points lie on well defined straight lines; the correlation coefficient between the $^{208}Pb/^{206}Pb$ and $^{207}Pb/^{206}Pb$ ratios is 0.99932, that between the $^{204}Pb/^{206}Pb$ and $^{207}Pb/^{206}Pb$ ratios is 0.99962[229]. A possible explanation of the wide spread in abundance ratios among these samples is that we are dealing with mixtures, in varying proportions, between a very radiogenic lead and one, or several, "normal" kind(s) of lead. The radiogenic variety would have to have $^{208}Pb/^{206}Pb$, $^{207}Pb/^{206}Pb$, and $^{204}Pb/^{206}Pb$ abundance ratios equal to, or lower than, the lowest ones encountered in any of the artifacts; the "normal" lead would have to fall on the extension to higher ratios of the regression lines[230]. It is impossible to determine precisely where such a mixing of two types of lead may have occurred. Often this already happens in the ores themselves; but it may also come about in the case of smelting of the ores, in particular if flux materials were added, or during the final processing of the artifacts. The important point is that in this mixing model a single kind of radiogenic lead suffices to account for the whole sequence of low ratios measured.

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[229] The $^{207}Pb/^{204}Pb$ ratios vary by less than 1.3% which results in a lower correlation coefficient between the $^{207}Pb/^{204}Pb$ and $^{206}Pb/^{204}Pb$ ratios of only 0.9140.

[230] Mixtures of two such types of lead form linear arrays only if the pairs of abundance ratios utilized have the same denominator.

Grouplet 1

HDM	site	region	object	per.	cl.	HDM	site	region	object	per.	cl.
1937	Hotnica Tell	NC	hammer axe	LC	1	1405	Svilajnc	SRB	axe-adze	FC	2
1913	Teliš	NC	axe-adze	FC	1	1409	Serbia	SRB	flat axe	FC	2
1954	Durankulak	BSC	pin	PB	1	1413	Makrešane	SRB	axe-adze	FC	2
						1414	eastern Serbia	SRB	axe-adze	FC	2
1903	Radlovci	W	hammer axe	MC	2	1416	vicinity of Niš	SRB	axe-adze	FC	2
1910	Dragoman	W	hammer axe	MC	2	1417	Krivi Vir, Niš	SRB	axe-adze	FC	2
1911	Dragoman	W	hammer axe	MC	2	1418	Lazinje, Mokra	SRB	axe-adze	FC	2
2703	Dăržanica	W	hammer axe	MC	2	1419	Pesak, Varoš	SRB	axe-adze	FC	2
2726	Mezdra	W	heavy axe	MC	2	1420	Crvena stena	SRB	flat axe	FC	2
1431	Stari Kostolac	SRB	chisel	LC	2	1426	Prižren	SRB	axe-adze	FC	2
2058	Tell Ruse	NE	borer	LC	2	1428	Starčevo (M. B.)	SRB	axe-adze	FC	2
2728	Zaminec	W	borer	LC	2	1430	Dvorište	SRB	axe-adze	FC	2
1301	Dušanovac	SRB	axe-adze	FC	2	1901	Hotnica	NC	axe-adze	FC	2
1302	eastern Serbia	SRB	axe-adze	FC	2	1906	Plakuder	W	axe-adze	FC	2
1304	Šarkamen, Ost.	SRB	axe-adze	FC	2	1907	Plakuder	W	axe-adze	FC	2
1306	Jabukovac	SRB	axe-adze	FC	2	1908	Plakuder	W	axe-adze	FC	2
1309	Šarkamen, Ost.	SRB	axe-adze	FC	2	1909	Plakuder	W	axe-adze	FC	2
1310	Šarkamen, Ost.	SRB	axe-adze	FC	2	1916	Kocan	W	hammer axe	FC	2
1312	Šarkamen, Ost.	SRB	axe-adze	FC	2	1917	Stolnik/E. P.	W	axe-adze	FC	2
1313	eastern Serbia	SRB	axe-adze	FC	2	2111	Teliš	NC	borer	FC	2
1314	Šarkamen, Ost.	SRB	axe-adze	FC	2	2705	Plakuder	W	axe-adze	FC	2
1315	Šarkamen, Ost.	SRB	axe-adze	FC	2	2706	Plakuder	W	axe-adze	FC	2
1317	Voluja	SRB	hammer axe	FC	2	2724	Rebarkovo	W	axe-adze	FC	2
1319	Donja Bela R.	SRB	axe-adze	FC	2	1339	Zlotska pečina	SRB	dagger	PB	2
1321	Zlotska pečina	SRB	flat axe	FC	2	2701	Jasen	W	flat axe	PB	2
1323	Serbia	SRB	axe-adze	FC	2						
1325	Osnič	SRB	axe-adze	FC	2	1986	Durankulak	BSC	bracelet	LC	3
1326	Krivelj	SRB	axe-adze	FC	2	2143	Hotnica Tell	NC	borer	LC	3
1327	Zlotska pečina	SRB	chisel	FC	2						
1333	Zlotska pečina	SRB	borer	FC	2	2050	Tell Ruse	NE	borer	LC	4
1334	eastern Serbia	SRB	pin	FC	2	2065	Tell Ruse	NE	borer	LC	4
1335	Zlotska pečina	SRB	borer	FC	2	2727	Krivodol	W	borer	LC	4
1336	Zlotska pečina	SRB	awl	FC	2	1318	Glogovica	SRB	axe-adze	FC	4

Grouplet 1 (continued)

HDM	site	region	object	per.	cl.
1331	Zlotska pečina	SRB	borer	FC	4
2108	Teliš	NC	borer	FC	4
2115	Teliš	NC	borer	FC	4
1910	Dragoman	W	hammer axe	MC	5
2064	Tell Ruse	NE	borer	MC	5
1422	Bubanj, N. S.	SRB	chisel	LC	5
2104	Tell Ruse	NE	pin	LC	5
1320	Gradac	SRB	hammer axe	FC	5
1329	Zlotska pečina	SRB	borer	FC	5
1331	Zlotska pečina	SRB	borer	FC	5
1415	Polna/Blagotin	SRB	chisel	FC	5
1913	Teliš	NC	axe-adze	FC	5
2113	Teliš	NC	borer	FC	5
2729	Gradešnica	W	borer	.	5
2092	Tell Ruse	NE	pin	LC	6
1337	Krivelj	SRB	borer	FC	6
2140	Debrene/Prilep	BSC	flat axe	LC	7
1914	Teliš	NC	axe-adze	FC	7
1902	Hotnica, region	NC	flat axe	PB	7
1918	Durankulak	BSC	dagger	PB	7
2134	Hotnica-Vodop.	NC	dagger	PB	7
2001	Durankulak	BSC	finger ring	LC	9
1484	Selevac	SRB	malachite	EC	-
1485	Selevac	SRB	malachite	EC	-
1487	Selevac	SRB	malachite	EC	-
1489	Selevac	SRB	malachite	EC	-
2080	Tell Ruse	NE	borer	MC	-

Grouplet 2

HDM	site	region	object	per.	cl.
1904	Slatino	W	chisel	EC	1
1564	Pločnik	SRB	chisel	MC	1
1565	Pločnik	SRB	chisel	MC	1
1569	Pločnik	SRB	chisel	MC	1
1923	Tell Ruse	NE	heavy axe	LC	1
1924	Sava, cemetery	BSC	heavy axe	LC	1
1933	Devnja	BSC	heavy axe	LC	1
1961	Durankulak	BSC	hammer axe	LC	1
1989	Durankulak	BSC	bracelet	LC	1
1998	Durankulak	BSC	bracelet	LC	1
2032	Durankulak	BSC	borer	LC	1
2035	Durankulak	BSC	spiral ring	LC	1
2044	Tell Ruse	NE	borer	LC	1
2070	Tell Ruse	NE	borer	LC	1
2071	Tell Ruse	NE	borer	LC	1
1954	Durankulak	BSC	pin	PB	1
1555	Pločnik	SRB	chisel	MC	2
1906	Plakuder	W	axe-adze	FC	2
1975	Durankulak	BSC	finger ring	EC	3
1556	Pločnik	SRB	flat axe	MC	3
1557	Pločnik	SRB	flat axe	MC	3
1567	Pločnik	SRB	chisel	MC	3
1571	Pločnik	SRB	hammer axe	MC	3
1977	Durankulak	BSC	bracelet	MC	3
1921	Tell Ruse	NE	chisel	LC	3
1934	Devnja	BSC	hammer axe	LC	3
1936	Devnja	BSC	hammer axe	LC	3
1963	Durankulak	BSC	bracelet	LC	3
1964	Durankulak	BSC	bracelet	LC	3

Grouplet 2 (continued)

HDM	site	region	object	per.	cl.
1967	Durankulak	BSC	finger ring	LC	3
1978	Durankulak	BSC	bracelet	LC	3
1984	Durankulak	BSC	bracelet	LC	3
1990	Durankulak	BSC	bracelet	LC	3
1991	Durankulak	BSC	bracelet	LC	3
1992	Durankulak	BSC	bracelet	LC	3
1996	Durankulak	BSC	bracelet	LC	3
2007	Durankulak	BSC	spiral ring	LC	3
2027	Durankulak	BSC	finger ring	LC	3
2029	Durankulak	BSC	finger ring	LC	3
2030	Durankulak	BSC	finger ring	LC	3
2045	Tell Ruse	NE	borer	LC	3
2053	Tell Ruse	NE	chisel	LC	3
2057	Tell Ruse	NE	borer	LC	3
2076	Tell Ruse	NE	borer	LC	3
2098	Tell Ruse	NE	pin	LC	3
2121	Goljamo Delčevo	BSC	bracelet	LC	3
2123	Goljamo Delčevo	BSC	spiral ring	LC	3
2128	Goljamo Delčevo	BSC	borer	LC	3
2141	Durankulak	BSC	bracelet	LC	3
2715	Smjadovo	NE	ear ring	LC	3
2722	Smjadovo	NE	borer	LC	3
2086	Tell Ruse	NE	borer	.	3
2087	Tell Ruse	NE	borer	.	3
2046	Tell Ruse	NE	borer	.	7
2733	Zaminec	W	borer	LC	9
1952	Durankulak	BSC	dagger	PB	9
2142	Durankulak	BSC	bracelet	LC	–

Grouplet 3

HDM	site	region	object	per.	cl.
1563	Pločnik	SRB	chisel	MC	1
1940	Ai Bunar	THR	hammer axe	MC	1
1905	Djakovo	W	flat axe	LC	1
1915	Sadovec	NC	hammer axe	LC	1
1929	Devnja	BSC	hammer axe	LC	1
1985	Durankulak	BSC	bracelet	LC	1
2034	Durankulak	BSC	pin	LC	1
2054	Tell Ruse	NE	borer	LC	1
2073	Tell Ruse	NE	pin	LC	1
2079	Tell Ruse	NE	borer	LC	1
2082	Tell Ruse	NE	borer	LC	1
2083	Tell Ruse	NE	borer	LC	1
2093	Tell Ruse	NE	borer	LC	1
2096	Tell Ruse	NE	borer	LC	1
2144	Hotnica Tell	NC	borer	LC	1
2145	Hotnica Tell	NC	borer	LC	1
2712	Smjadovo	NE	casting spill	LC	1
2713	Smjadovo	NE	casting spill	LC	1
2718	Smjadovo	NE	borer	LC	1
2109	Teliš	NC	pin	LC	1
2139	Onogur castle	BSC	flat axe	LC	3
2072	Tell Ruse	NE	borer	LC	4
2084	Tell Ruse	NE	borer	LC	4
2146	Hotnica Tell	NC	borer	LC	5
1311	Vrazogrnac	SRB	axe-adze	FC	5
2093	Tell Ruse	NE	borer	LC	6
2096	Tell Ruse	NE	borer	LC	6
2150	Karanovo Tell	THR	borer	LC	6

Grouplet 3 (continued)

HDM	site	region	object	per.	cl.	HDM	site	region	object	per.	cl.
2707	Goljamo Delčevo	BSC	hooked wire	LC	6	1307	Urovica	SRB	shafthole axe	FC	9
2149	Kačica	NC	borer	PB	6	2148	Kačica	NC	borer	PB	9
2742	Haramijska d.	RHD	borer	PB	6	2048	Tell Ruse	NE	bracelet	.	9
1966	Durankulak	BSC	dagger	PB	7	2210	Durankulak	BSC	malachite bead	LN	-
2704	Vidbol	W	flanged axe	EB	7	2212	Durankulak	BSC	malachite bead	LN	-
2048	Tell Ruse	NE	bracelet	.	7	1402	Stojačak	SRB	axe-adze	FC	-
2209	Durankulak	BSC	malachite bead	LN	8	2114	Teliš	NC	borer	FC	-
1979	Durankulak	BSC	bracelet	LC	8	2147	Kačica	NC	borer	PB	-

Grouplet 4

HDM	site	region	object	per.	cl.	HDM	site	region	object	per.	cl.
1562	Pločnik	SRB	chisel	MC	1	1986	Durankulak	BSC	bracelet	LC	3
2049	Tell Ruse	NE	borer	MC	1	2126	Goljamo Delčevo	BSC	pin	LC	3
1927	Varna I	BSC	flat axe	LC	1						
1931	Varna I	BSC	hammer axe	LC	1	2709	Smjadovo	NE	copper strip	LC	5
1942	Durankulak	BSC	finger ring	LC	1						
2013	Durankulak	BSC	finger ring	LC	1	2041	Tell Ruse	NE	borer	MC	6
2022	Durankulak	BSC	hammer axe	LC	1	2725	Zaminec	W	flat axe	LC	6
2040	Tell Ruse	NE	borer	LC	1						
2067	Tell Ruse	NE	borer	LC	1	1941	Ai Bunar	THR	axe-adze	FC	7
2077	Tell Ruse	NE	borer	LC	1	2739	Lesura	W	dagger	EB	7
2078	Tell Ruse	NE	borer	LC	1	2033	Durankulak	BSC	spiral-h. pin	LB	7
2081	Tell Ruse	NE	borer	LC	1	2731	Gradešnica	W	borer	LB	7
2099	Tell Ruse	NE	pin	LC	1	2066	Tell Ruse	NE	borer	.	7
2710	Smjadovo	NE	chisel	LC	1						
2711	Smjadovo	NE	borer	LC	1	2066	Tell Ruse	NE	borer	.	9
2714	Smjadovo	NE	borer	LC	1						
2719	Smjadovo	NE	borer	LC	1	1959	Durankulak	BSC	pin	LC	-
1954	Durankulak	BSC	pin	PB	1	2097	Tell Ruse	NE	borer	LC	-
1322	Donji Milanovac	SRB	axe-adze	FC	2						

<u>Grouplet 5</u>

HDM	site	region	object	per.	cl.	HDM	site	region	object	per.	cl.
1944	Durankulak	BSC	bracelet	LC	1	2055	Tell Ruse	NE	borer	LC	6
1945	Durankulak	BSC	bracelet	LC	1	2100	Tell Ruse	NE	pin	LC	6
1983	Durankulak	BSC	bracelet	LC	1	2117	Teliš	NC	borer	FC	6
2047	Tell Ruse	NE	pin	LC	1						
2129	Goljamo Delčevo	BSC	borer	LC	1	2039	Tell Ruse	NE	borer	LC	7
2717	Smjadovo	NE	borer	LC	1	2094	Tell Ruse	NE	pin	.	7
2039	Tell Ruse	NE	borer	LC	3	1496	Gomolava	SRB	bracelet	MC	8
2051	Tell Ruse	NE	borer	LC	3	2012	Durankulak	BSC	bracelet	LC	8
1943	Durankulak	BSC	finger ring	LC	4	1491	Selevac	SRB	malachite	EC	–
1948	Durankulak	BSC	bracelet	LC	4	2052	Tell Ruse	NC	borer	LC	–
1997	Durankulak	BSC	bracelet	LC	4						
2138	Durankulak	BSC	finger ring	LC	4						

<u>Grouplet 6</u>

HDM	site	region	object	per.	cl.	HDM	site	region	object	per.	cl.
1912	Marica (Deveb.)	THR	heavy axe	EC	1	2118	Teliš	NC	borer	FC	4
1925	Varna I	BSC	hammer axe	LC	1						
1935	Devnja	BSC	chisel	LC	1	2107	Teliš	NC	knife	FC	6
2060	Tell Ruse	NE	borer	LC	1	2740	Haramijska d.	RHD	dagger	PB	6
2088	Tell Ruse	NE	borer	LC	1						
2119	Goljamo Delčevo	BSC	borer	LC	1	1969	Durankulak	BSC	malachite bead	LN	–
2120	Goljamo Delčevo	BSC	borer	LC	1	2211	Durankulak	BSC	malachite bead	LN	–
2130	Goljamo Delčevo	BSC	borer	LC	1	1492	Selevac	SRB	malachite	EC	–
2110	Teliš	NC	borer	FC	1	1932	Varna I	BSC	chisel	LC	–
						1402	Stojačak	SRB	axe-adze	FC	–
1938	Hotnica Tell	NC	hammer axe	LC	4	1919	Ivanovo	NE	axe-adze	FC	–

Grouplet 7

HDM	site	region	object	per.	cl.	HDM	site	region	object	per.	cl.
1982	Durankulak	BSC	bracelet	LC	1	2043	Tell Ruse	NE	borer	LC	5
1987	Durankulak	BSC	bracelet	LC	1	2059	Tell Ruse	NE	borer	LC	5
2016	Durankulak	BSC	bracelet	LC	1	2101	Tell Ruse	NE	borer	LC	5
2068	Tell Ruse	NE	borer	LC	1	1303	Urovica	SRB	axe-adze	FC	5
2102	Tell Ruse	NE	borer	LC	1						
2103	Tell Ruse	NE	borer	LC	1	1559	Pločnik	SRB	hammer axe	MC	8
						1568	Pločnik	SRB	chisel	MC	8
1920	Tell Ruse	NE	flat axe	LC	4						
2089	Tell Ruse	NE	pin	LC	4	2004	Durankulak	BSC	malachite bead	LN	–
						2736	Malorad	W	dagger	PB	–

Grouplet 8

HDM	site	region	object	per.	cl.	HDM	site	region	object	per.	cl.
1566	Pločnik	SRB	chisel	MC	1	2112	Teliš	NC	borer	FC	5
1926	Varna I	BSC	hammer axe	LC	1						
1999	Durankulak	BSC	flat axe	LC	1	1410	Serbia	SRB	flat axe	PB	7
						2135	Hotnica-Vodop.	NC	dagger	PB	7
1427	Veliko Laole	SRB	axe-adze	FC	2	2737	Galiče	W	dagger	PB	7
						2704	Vidbol	W	flanged axe	EB	7
1986	Durankulak	BSC	bracelet	LC	3	2723	Zlatarevo	NE	borer	.	7
2105	Teliš	NC	double-sp. pin	FC	3						
						1939	Hotnica Tell	NC	heavy axe	LC	8
2106	Teliš	NC	knife	FC	4	1328	Zlotska pečina	SRB	chisel	FC	8
2042	Tell Ruse	NE	hook	.	4						
						1495	Selevac	SRB	malachite	EC	–
2056	Tell Ruse	NE	borer	LC	5	2090	Tell Ruse	NE	borer	LC	–
2075	Tell Ruse	NE	pin	LC	5						

Grouplet 9

HDM	site	region	object	per.	cl.	HDM	site	region	object	per.	cl.
2006	Durankulak	BSC	spiral ring	LC	1	1972	Durankulak	BSC	bracelet	LC	7
2015	Durankulak	BSC	bracelet	LC	1	2741	Haramijska d.	RHD	dagger	PB	7
2074	Tell Ruse	NE	borer	LC	1	2038	Vodnjanci	BSC	double axe	LB	7
2721	Smjadovo	NE	borer	LC	1	2131	Hotnica-Vodop.	NC	borer	LB	7
						2133	Hotnica, region	NC	dagger	LB	7
1951	Durankulak	BSC	wire		3	1953	Durankulak	BSC	stylo	C	7
2025	Durankulak	BSC	bracelet	MC	4	1970	Durankulak	BSC	malachite bead	LN	8
2026	Durankulak	BSC	bracelet	MC	4						
						2720	Smjadovo	NE	borer	LC	9
2006	Durankulak	BSC	spiral ring	LC	6						
2708	Smjadovo	NE	axe-adze	FC	6	1969	Durankulak	BSC	malachite bead	LN	–
						2211	Durankulak	BSC	malachite bead	LN	–
2011	Durankulak	BSC	bracelet	MC	7	1493	Selevac	SRB	malachite	EC	–
1956	Durankulak	BSC	bracelet	LC	7	2136	Hotnica-Vodop.	NC	metal fragment	LB	–
1960	Durankulak	BSC	bracelet	LC	7						

Grouplet 10

HDM	site	region	object	per.	cl.	HDM	site	region	object	per.	cl.
1570	Pločnik	SRB	chisel	MC	1	2061	Tell Ruse	NE	borer	LC	5
1971	Durankulak	BSC	flat axe	LC	1	1421	Jelašnice	SRB	chisel	FC	5
1981	Durankulak	BSC	bracelet	LC	1						
2021	Durankulak	BSC	flat axe	LC	1	1308	Šumrakovac	SRB	hammer axe	MC	7
2122	Goljamo Delčevo	BSC	spiral ring	LC	1						
						1561	Pločnik	SRB	chisel	MC	8
2010	Durankulak	BSC	borer	LC	4						
1401	Stojačak	SRB	axe-adze	FC	4	1492	Selevac	SRB	malachite	EC	–
2095	Tell Ruse	NE	pin		4	1493	Selevac	SRB	malachite	EC	–

Grouplet "high"

HDM	site	region	object	per.	cl.	HDM	site	region	object	per.	cl.
2069	Tell Ruse	NE	chisel	.	3	1958	Durankulak	BSC	spiral-h. pin	LB	7
						2037	Kozlodujci	BSC	sickle	LB	7
2132	Hotnica, region	NC	dagger	LB	6	2132	Hotnica, region	NC	dagger	LB	7
						2730	Krivodol/Oslen	W	knife	LB	7
1955	Durankulak	BSC	metal band	LB	7	2734	Gradešnica	W	loop-h. pin	LB	7
1957	Durankulak	BSC	pin	LB	7	2069	Tell Ruse	NE	chisel	.	7

Grouplet rPb (radiogenic lead)

HDM	site	region	object	per.	cl.	HDM	site	region	object	per.	cl.
2005	Durankulak	BSC	finger ring	EC	1	1974	Durankulak	BSC	finger ring	LC	4
1976	Durankulak	BSC	bracelet	MC	1	1980	Durankulak	BSC	bracelet	LC	4
1930	Devnja	BSC	hammer axe	LC	1	1993	Durankulak	BSC	bracelet	LC	4
1946	Durankulak	BSC	bracelet	LC	1	1995	Durankulak	BSC	bracelet	LC	4
1980	Durankulak	BSC	bracelet	LC	1	2000	Durankulak	BSC	finger ring	LC	4
1988	Durankulak	BSC	bracelet	LC	1	2002	Durankulak	BSC	finger ring	LC	4
1994	Durankulak	BSC	bracelet	LC	1	2008	Durankulak	BSC	finger ring	LC	4
2017	Durankulak	BSC	bracelet	LC	1	2014	Durankulak	BSC	bracelet	LC	4
2023	Durankulak	BSC	bracelet	LC	1	2018	Durankulak	BSC	bracelet	LC	4
2024	Durankulak	BSC	bracelet	LC	1	2028	Durankulak	BSC	finger ring	LC	4
2091	Tell Ruse	NE	borer	LC	1	2031	Durankulak	BSC	finger ring	LC	4
2127	Goljamo Delčevo	BSC	borer	LC	1	2137	Durankulak	BSC	finger ring	LC	4
2716	Smjadovo	NE	borer	LC	1						
						2063	Tell Ruse	NE	borer	LC	5
2125	Goljamo Delčevo	BSC	borer	LC	3	2085	Tell Ruse	NE	borer	LC	5
						2062	Tell Ruse	NE	borer	.	5
1922	Tell Ruse	NE	chisel	LC	4						
1947	Durankulak	BSC	bracelet	LC	4	1483	Selevac	SRB	copper prill	EC	8
1949	Durankulak	BSC	bracelet	LC	4	1494	Selevac	SRB	copper prill	EC	8
1962	Durankulak	BSC	finger ring	LC	4	2702	Dăržanica	W	heavy axe	MC	8
1965	Durankulak	BSC	bracelet	LC	4						
1968	Durankulak	BSC	bracelet	LC	4	1481	Selevac	SRB	malachite	EC	–
1973	Durankulak	BSC	bracelet	LC	4	2036	Durankulak	BSC	spiral ring	LC	–

no specific grouplet assignment

HDM	site	region	object	per.	cl.	HDM	site	region	object	per.	cl.
2003	Durankulak	BSC	finger ring	LC	1	2735	Gradešnica	W	borer	.	7
2009	Durankulak	BSC	spiral ring	LC	1						
2019	Durankulak	BSC	bracelet	LC	1	1558	Pločnik	SRB	hammer axe	MC	8
2020	Durankulak	BSC	bracelet	LC	1	1560	Pločnik	SRB	chisel	MC	8
2732	Gradešnica	W	borer	LB	1						
						2738	Lesura	W	dagger	PB	9
1408	Selo Vrba	SRB	axe-adze	FC	2						
						1482	Selevac	SRB	malachite	EC	–
1928	Devnja	BSC	hammer axe	LC	3	1486	Selevac	SRB	malachite	EC	–
1950	Durankulak	BSC	wire	.	3	1488	Selevac	SRB	malachite	EC	–
						1490	Selevac	SRB	malachite	EC	–
2124	Goljamo Delčevo	BSC	borer	MC	4	1330	Zlotska pečina	SRB	borer	FC	–
						1332	Zlotska pečina	SRB	borer	FC	–
2116	Teliš	NC	borer	FC	5						

Table 4. Assignment of samples to isotope grouplets and chemical clusters (cl.). In cases where more than one grouplet or cluster assignment is possible, the sample is listed more than once. Regions and periods (per.) are abbreviated as follows: BSC = Black Sea coast, NC = north-central Bulgaria, NE = north-east Bulgaria, RHD = Rhodope mountains, SRB = Serbia, THR = Bulgarian Thrace, W = western Bulgaria; LN = Late Neolithic, EC = Early Chalcolithic, MC = Middle Chalcolithic, LC = Late Chalcolithic, FC = Final Chalcolithic, PB = Proto Bronze Age, EB = Early Bronze Age, LB = Late Bronze Age, C = Classical period, blank = unstratified or uncertain.

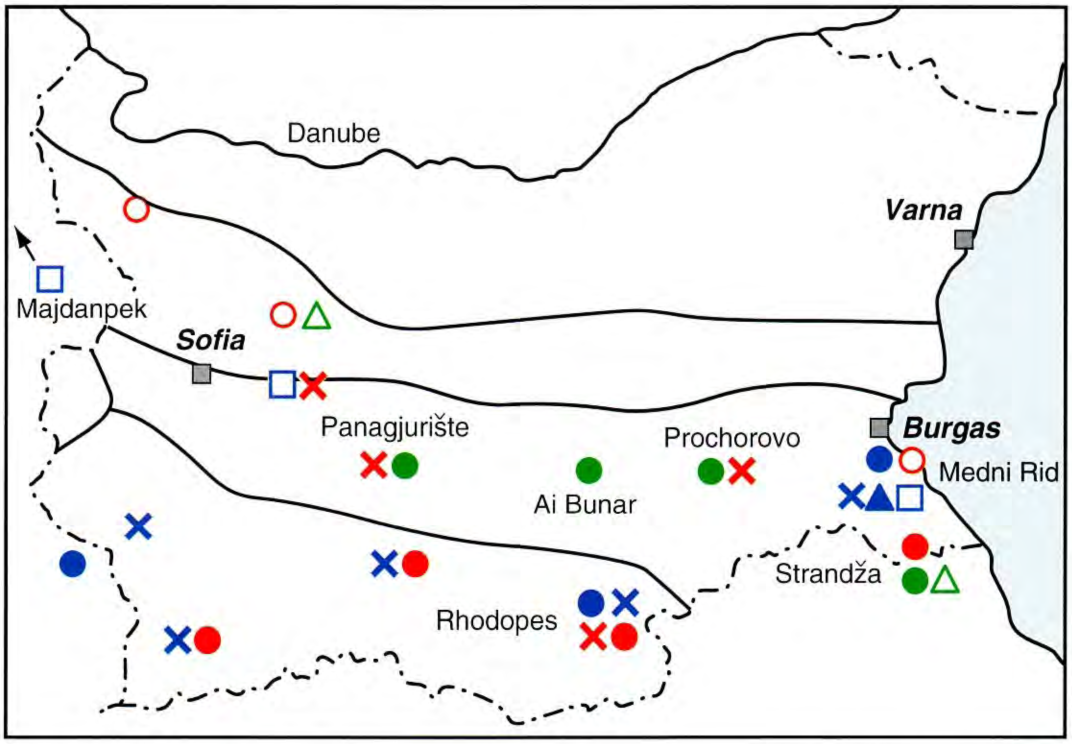

Fig. 19. Localities in Bulgaria and adjacent regions where copper ores occur with lead isotope signatures matching those of the different artifact grouplets (grouplet signatures are the same as in *Figs. 17* and *18*). As a rule there is more than one ore occurrence per grouplet but note also that for grouplet #7 (green circles) there is as yet no fitting ore known at all.

Chemically, of the objects with $^{208}Pb/^{206}Pb$ ratios below 2.05, nineteen belong to cluster #4, and twelve to cluster #1 *(Tab. 4*; HDM 1980 could be attributed to cluster #1 or #4 and is not counted here*)*. The lead content of all these samples is low, below one hundredth of one percent. Note, however, that the opposite is not true. There are many more samples with equally low, or even lower, Pb content that are perfectly "normal" in their isotopy. In fact, for artifacts with lead concentrations below 100 ppm the number frequency distribution of the Pb contents in objects with $^{208}Pb/^{206}Pb < 2.05$ very much mimics the distribution for *all* samples.

Concerning the provenance of copper with such radiogenic lead we note that, judged from our (limited) number of analyses, *native* copper from the Balkans does not qualify *(Tab. 5)*. Also only seldomly encountered is such lead among ores of copper *(Tab. A5)*; the A&V compilation does not contain a single sample with a $^{208}Pb/^{206}Pb$ ratio below 2.060. Still, there are a few instances where we have come across such ores: a malachite from Rosen/Burgas (BG-7b), a mixed-ore sample from a slag pile at Dereköy-Catak Tepe on the Turkish side in the Strandža Mountains[231], and at least thirteen out of our suite of seventeen ore specimens from Rudna Glava and five copper slags from Crnajka, both in Serbia[232]. All of these fall more or less exactly on the regression lines as they are defined by the artifacts.

[231] Wagner u. a. 1986, TG 189A-3.
[232] Pernicka u. a. 1993.

Sample no.	Location	Region	lead concentration [µg/g]	$^{208}Pb/^{206}Pb$	$^{207}Pb/^{206}Pb$	$^{204}Pb/^{206}Pb$
BG-2a	Boboševo	W	<0.2	2.0824	0.8453	0.05394
BG-6a	Zidarovo	BSC	<0.8	2.0664	0.8366	0.05332
BG-7a	Rosen	BSC	0.8	2.0721	0.8401	0.05368
BG-8a	Assarel	W	100	2.0663	0.8339	0.05323
BG-9a	Elacite	W	<0.6	2.0841	0.8404	0.05360
BG-10a	Car Assen	W	0.7	2.0609	0.8340	0.05337
BG-11a	Radka	W	<0.4	2.0703	0.8366	0.05339*
BG-11b	Radka	W	0.5	2.0819	0.8420	0.05376
BG-12a	M. Tărnovo-Brădceto	STR	<0.04	2.0733	0.8369	0.05341**
BG-12b	M. Tărnovo-Brădceto	STR	0.2	2.0689	0.8360	0.05354
BG-12c	M. Tărnovo-Brădceto	STR	40	2.0671	0.8341	0.05316
BG-12e	M. Tărnovo-Brădceto	STR	0.8	2.0731	0.8351	0.05333
BG-13a	Madžarovo (coll. W. Atanasov)	RHD	210	2.0667	0.8340	0.05318
BG-13b	Madžarovo (coll. V. Kovačev)	RHD	4000	2.0666	0.8340	0.05317

* and **: Uncertainty in $^{204}Pb/^{206}Pb$ ratio is 0.22% and 0.68%, respectively. In all other cases it is $2\sigma < 0.1\%$.

Table 5. Lead isotope abundance ratios of native copper samples from Bulgaria. Regions are abbreviated as follows: BSC = Black Sea coast, RHD = Rhodope mountains, STR = Strandža mountains, W = western Bulgaria.

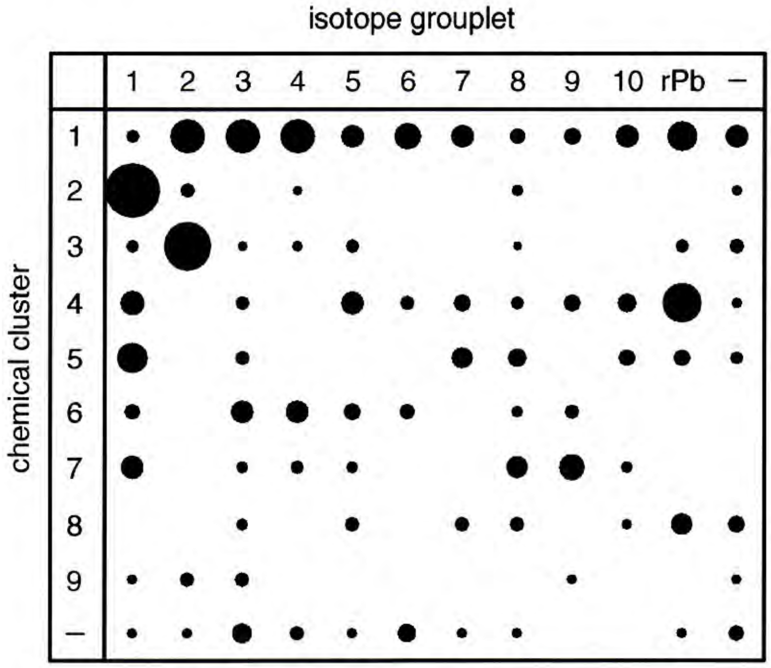

Fig. 20. The classification matrix of Chalcolithic and PBA copper artifacts from Bulgaria and Serbia shows that trace element abundances, characterized by cluster number, and lead isotope abundance ratios, characterized by grouplet number, are not uniquely correlated. The information to be gained from the two sets of data is complementary, not redundant; only when both sets are available is an assignment of an artifact to a specific ore occurrence possible with a reasonable degree of probability.

Hence, as far as the lead isotopy is concerned these are candidate ores from which the copper may ultimatively have been derived. We shall return to this problem below.

The two sets of material science information presented here, chemical clusters and isotope grouplets, are not uniquely correlated. According to *Fig. 20* there is a close correspondence only between cluster #2 and grouplet #1, cluster #3 with grouplet #2, and cluster #4 with rPb; in all other instances are artifacts from the same cluster distributed among several grouplets, and vice versa. This is fortunate because it gives us two independent parameters to characterize the material properties of an artifact and to search for potential source ores. Had it been otherwise, the information to be gained from the two sets of data would be redundant and, as far as provenance studies are concerned, one might well wonder whether an analysis for both features would be worth while.

Of course, the implication of this result is also that all previous assignments of artifacts to specific ore occurrences based on trace element patterns alone must be considered with due caution and to be tentative at best. In the present case this is true in particular for all those artifacts that belong to cluster #1, corresponding to Černych's groups II and III because they are rather evenly distributed among many isotope grouplets and quite clearly do not derive from one but from a number of different ore deposits.

Discussion

Native copper and copper ores

The native copper specimens available to us were millimeter-sized framboidal aggregates, which makes them prone to be easily contaminated. Their Cu contents were nevertheless around 90% or higher indicating that the samples were quite pure, without adhering or intergrown minerals that might have affected the trace element concentrations. The lead content puts the samples into two groups, one with concentrations below 1 μg/g and the other with values between 40 and 4000 μg/g. Since the isotopic compositions of the four samples with high lead content (BG-8a, BG-12c, BG-13a,b) are indistinguishable from one another *(Tab. 5)* we suspect these samples to be heavily contaminated, all with the same type of lead. Since they all come from the same collection this does appear to be entirely possible. We shall not consider them in the subsequent discussion of the lead isotope results.

Most samples contain Hg in low but measurable amounts. Although perhaps intuitively considered to be a very volatile element, mercury is largely retained even upon extensive heat treatment of copper[233]. On the other hand, smelting of copper ores removes mercury very effectively, so that this element might possibly serve as a useful indicator to distinguish between artifacts made of native copper and such made from smelted copper[234]. The other elements listed in *Tab. A3a* are in the low concentration ranges as they are typical of native copper. Note, that for each element there are occasional outliers where

[233] Unpublished experiments have shown that even heating at 800 °C for 15 minutes reduces the original Hg content of copper only to around 30% of the original concentration. At 1000 °C a few percent of the original amount remain and only upon melting is more than 99% of the Hg removed.

[234] Some lithophile elements, like Ba, Ce, Cr, Cs, Sc, Th, and U, that can be measured with very high sensitivity are included in *Table A3b* although their concentrations are expected to be drastically altered on melting. They may, however, be useful for future provenance studies of unmelted native copper.

the concentrations are very much higher than normal. Except for gold, this has previously been observed in native copper from Anatolia and Serbia and by Rapp in his survey of samples from Cornwall, U.K. (104 samples), the former USSR (10), Norway (2), Germany (1) and Spain (1)[235]. But gold contents of native copper in the ppm-range, as found in the specimens from the Panagjurište and Madžarovo districts, so far are unique. They are also unaccountably high if the native copper were formed by reduction from solution in the cementation zone of an ore body. It is possible that the specimens with higher gold contents formed by another process which may also be hinted at by the slightly higher concentrations in the same samples of Sb and Se.

As to the chemical composition of the ores, given in *Tab. A2*, there is not much to comment because, except for Ai Bunar, their number is too low for any generalizing statements. For Ai Bunar, however, a typical impurity pattern can be predicted that will result upon smelting Cu from the kind of ores as they have been studied: The As content is expected to be in the low percent range, Sb should vary between 0.1 and 1%, Co between 10 and 100 μg/g, Ni should be lower than 200 μg/g, Ag should range between 0.01 and 0.1%, Au between 1 and 100 μg/g, and Se between 0.01 and 0.1%. According to *Tab. 2* these are the specifications of cluster #3. For complex sulfide ores as starting material the predictions would be more uncertain because such ores require several steps to produce copper, including an oxidizing one. It is unlikely, however, that sulfide ores were used to any significant extent in the Chalcolithic and there is also no indication in the present data that this may have occurred[236]. Thus we feel quite confident that the impurity pattern predicted for metal produced from Ai Bunar ores can be used, in addition to the lead isotope ratios, for tracing copper from this site to Chalcolithic copper artifacts. The major source of uncertainty at present are the relatively low copper contents in all samples except for one (BG-17b). This sample would produce copper with significantly lower As and Sb contents but otherwise resembles the lower-grade ores that derive from a pile of waste ore. Taking this into account it is therefore possible that Ai Bunar produced copper with somewhat lower As and Sb contents than listed above as our prediction.

Native vs. smelted copper in artifacts

The question whether an artifact is made of native copper is decided best by investigating its metallographic structure[237]. This works only, however, if the object has not been cast but been formed simply by hammering, and possibly tempering, of native copper. But by such a technique only small objects can be manufactured and this, in turn, makes it usually difficult to procure a sample for such a metallographic study because it is, by necessity, destructive to some extent.

An alternative suggestion to possibly distinguish between native and smelted copper goes back to the last century already when it was proposed that the high purity of Neolithic and Chalcolithic copper artifacts from southeastern Europe should be evidence,

[235] Unpublished results by G. Rapp jun.

[236] Sulfur contents of up to 17% in copper ore samples from Chalcolithic sites in the vicinity of Ai Bunar (Gale u. a. 1991, 60 Table 7) do not contradict this view. Practically all oxidic copper ores ultimately derive from sulfide minerals and thus usually contain varying amounts of sulfur depending on the degree of oxidation. It is nevertheless unlikely that this was recognized by Chalcolithic smelters, because all of these ores appear green or blue due to the intense colours of malachite and azurite.

[237] cf. e.g. Maddin u. a. 1980.

if not proof, that they were manufactured from native copper. This suggestion was taken up by Otto and Witter[238] who substantiated the purity of the metal through analyses by optical emission spectroscopy. Junghans and coworkers[239], in their very comprehensive study of prehistoric metal objects from all over Europe, observed a comparatively high proportion of spectroscopically pure copper – their metal types N and E00 – in southeast Europe. They followed the suggestion of Otto and Witter that this was most probably native copper, and so did Charles[240], also on the basis of the chemical purity of two Chalcolithic axe-adzes from Hungary and former Yugoslavia. Černych[241] simply states that his chemical groups I and II (that are more or less comparable to the Stuttgart groups N and E00) must derive from monometallic copper ores whereby he left it open if he meant native copper or oxidic copper ores. Relating to the same analyses by Černych, Gale and coworkers, on the other hand, completely dismiss the use of native copper in the Bulgarian Chalcolithic[242]. Their argument is that "even the purest of the Bulgarian Eneolithic copper objects contain an order of magnitude more of certain elements, such as nickel, than do the large number of native coppers analyzed by Patterson (1971)". Although we concur with their conclusion we nevertheless take exception to the way it was arrived at. First, the "large number of native coppers analyzed by Patterson" are actually just eleven samples, with eight of them from the Americas and one each from Australia, from Cornwall and from Cyprus[243]. Secondly, the selective reference to Patterson, completely ignoring the more recent work on native copper[244] would appear to be unnecessarily lopsided. Moreover, although it is true that Patterson reported only 4 µg/g Ni or less in all his samples, there are 156 objects among Černych's 1,233 analyses that contain also less than 10 µg/g Ni. Seventy six of those contain, in addition, less than 10 µg/g of Co, Sb, and Au while their Ag concentrations range up to 0.1%. At least these would qualify to be potentially made of native copper, even according to Patterson's trace element data. Finally, Patterson's compilation of "typical impurity levels" in native copper is nowadays outdated although the general tendency is still correct: Native copper is usually a high-purity metal, with Ag (and occasionally As) as major impurities. This is apparently also true for native copper from Anatolia and the Balkans *(Tab. A3)*.

But, to make the argument compelling that low trace element contents of artifacts are indicative of their having been manufactured from native copper requires also that in copper smelted from ores the trace element concentrations are invariably high. Experimental data on this question are rare, most of them pertain to copper as it has been smelted in the laboratory under conditions that are presumed to be identical to those in antiquity. Recently, however, results have been reported that should be more relevant because they were obtained on millimeter-sized copper prills as they were found in Chalcolithic and Early Bronze Age slags from Feinan, Jordan[245]. They demonstrate that silver, gold, and antimony in smelted copper can, for all practical purposes, be as low as they are typically found in native copper; that arsenic shows a wide distribution in concentra-

[238] Otto/Witter 1952.
[239] Junghans u. a. 1968.
[240] Charles 1969.
[241] Черных 1978.
[242] Gale u. a. 1991.
[243] Patterson 1971.
[244] Friedman u. a. 1966; Hancock u. a. 1991; in particular, the hundreds of results by Rapp 1982.
[245] Hauptmann u. a. 1992.

tions overlapping that observed in native copper, but that nickel and cobalt are almost invariably higher than in native copper *(Fig. 21)*. It would appear at present that these two elements, in particular when used in combination, are indeed useful markers to distinguish between native and smelted copper[246] although, in order to be applicable to artifacts from the Balkans, the results clearly need to be confirmed for Chalcolithic copper smelted from ore occurrences other than those at Feinan. Ores of the kind represented by several of the Neolithic malachites from Durankulak and from Selevac would be especially interesting in this respect because they are very low in all trace elements, including cobalt and nickel. But then perhaps some of these beads originally were native copper that, in the course of time, was converted into malachite.

For artifacts from periods earlier than the Proto Bronze Age the results of the present study are presented by way of the histograms of *Fig. 22*. The distributions are fairly wide for all elements but even for the most abundant contaminants iron[247], arsenic, silver and lead do the concentrations only rarely reach the percent level. Thus, generally speaking, all objects are made of rather pure copper, a result in accord with earlier studies[248]. Interesting in the present context, whether the artifacts are made from native copper or from smelted copper, are the distributions for the marker elements cobalt and nickel that might allow to decide between the two possibilities *(Fig. 23)*. Clearly, for nickel in particular, the two distributions are quite distinct.

Chalcolithic tin bronzes?

An interesting feature of the Chalcolithic in southeastern Europe is the seemingly increasing evidence for an early use of tin bronze. Recently, at least fourteen such tin bronzes from former Yugoslavia and Bulgaria have been claimed to belong to the fifth millennium B.C. and their appearance has been related to cassiterite occurrences in southeastern Europe[249]. Moreover, based on a crucible with adhering copper-tin slag of the Baden culture from Okukalj in Croatia[250] and at least two samples from Sitagroi IV in

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[246] This is largely in agreement with similar studies to distinguish between native and modern smelted copper used by North American Inuit. See e.g. Wayman u. a. 1985; Hancock u. a. 1993. Generally speaking it would appear that the possibility to differentiate native and smelted copper based on their trace element pattern has been underestimated in the past and it may equally be the case that the possibility to identify native copper by metallographic investigation has been overestimated. Moesta 1991 reported that a chisel from Drama, dated to Karanovo V (Middle Chalcolithic in our terms), was clearly cast and hammered, because of the presence of stringers of Cu/Cu_2O eutectic in the metal. Yet he also identified small (2–5 µm) particles of silver that are usually interpreted to indicate native copper. Indeed, he concluded that native copper was melted for a very short time and that oxide films protected the silver particles that supposedly must have existed already within the native copper. However, the particles were detected within a small void from casting that was partly filled with Cu_2O. It is thus conceivable that the silver particles formed on oxidation of argentiferous copper, a process that is well-known among mineralogists and that is responsible for the occurrence of metallic silver in native copper. Even within and on the surface of archaeological artifacts newly formed native silver has been observed (e.g. Martinek 1995). In this case only chemical analysis could possibly help to decide, if the chisel was made of native copper or not.

[247] Iron is not plotted in *Fig. 22*, because in most instances only upper limits could be determined that, moreover, are not the same for all samples.

[248] Junghans u. a. 1968; Черных 1978.

[249] Glumac/Todd 1991 b, 14 f.

[250] Poster presented by J. Lozuk in 1990 at the International Symposium on Ancient Mining and Metallurgy in Southeastern Europe at Donji Milanovac, Yugoslavia.

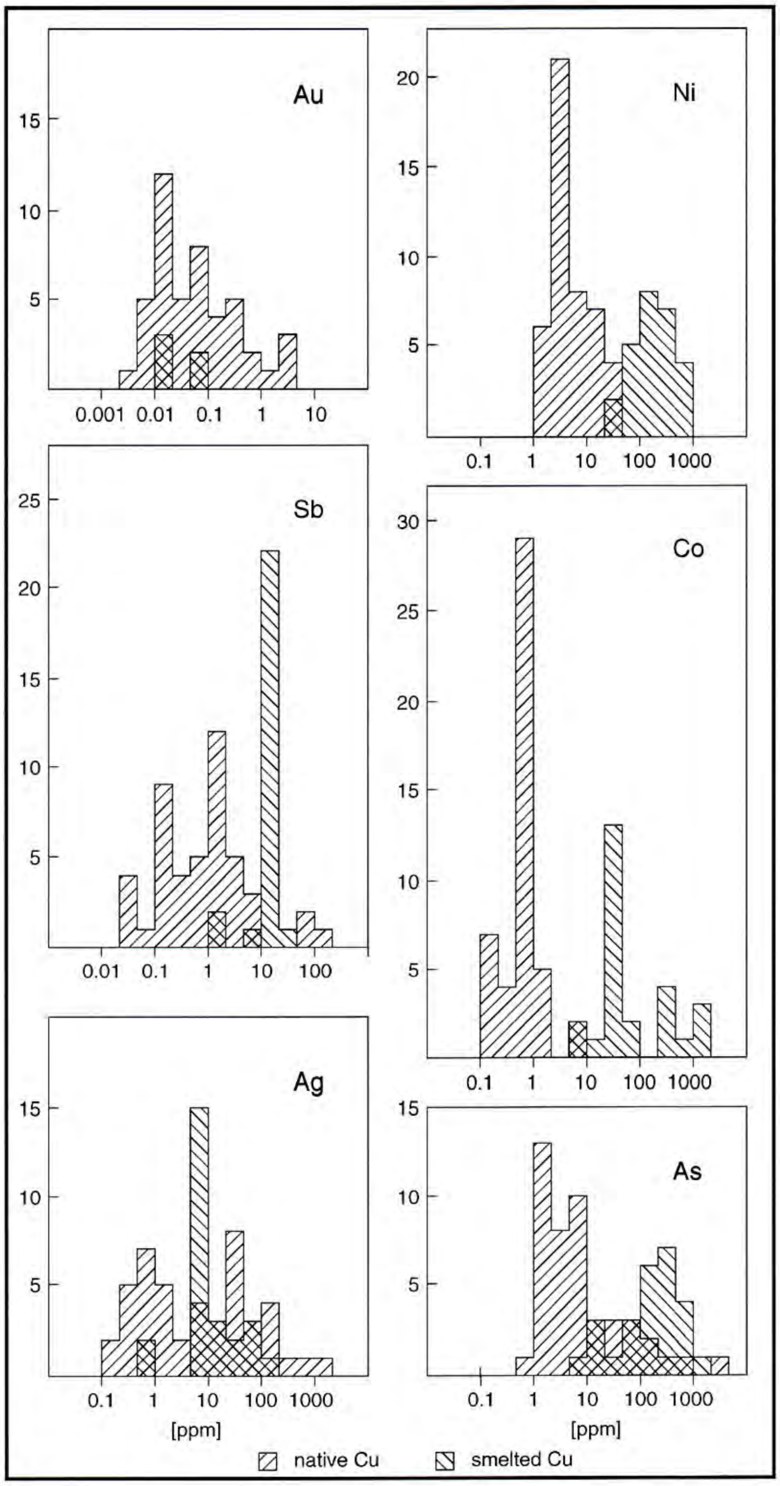

Fig. 21. Trace element contents in native copper from the Balkans and Anatolia as compared to copper prills from Chalcolithic and Early Bronze Age slags from Feinan, Jordan. Arsenic, silver, antimony, and gold in smelted copper are as low as in native copper; low concentrations of these elements in an artifact are no evidence that it should be made from native copper. Only cobalt and nickel are notably more abundant in smelted than in native copper and may thus be useful marker elements to distinguish between the two possibilities (Feinan data from Hauptmann u. a. 1992).

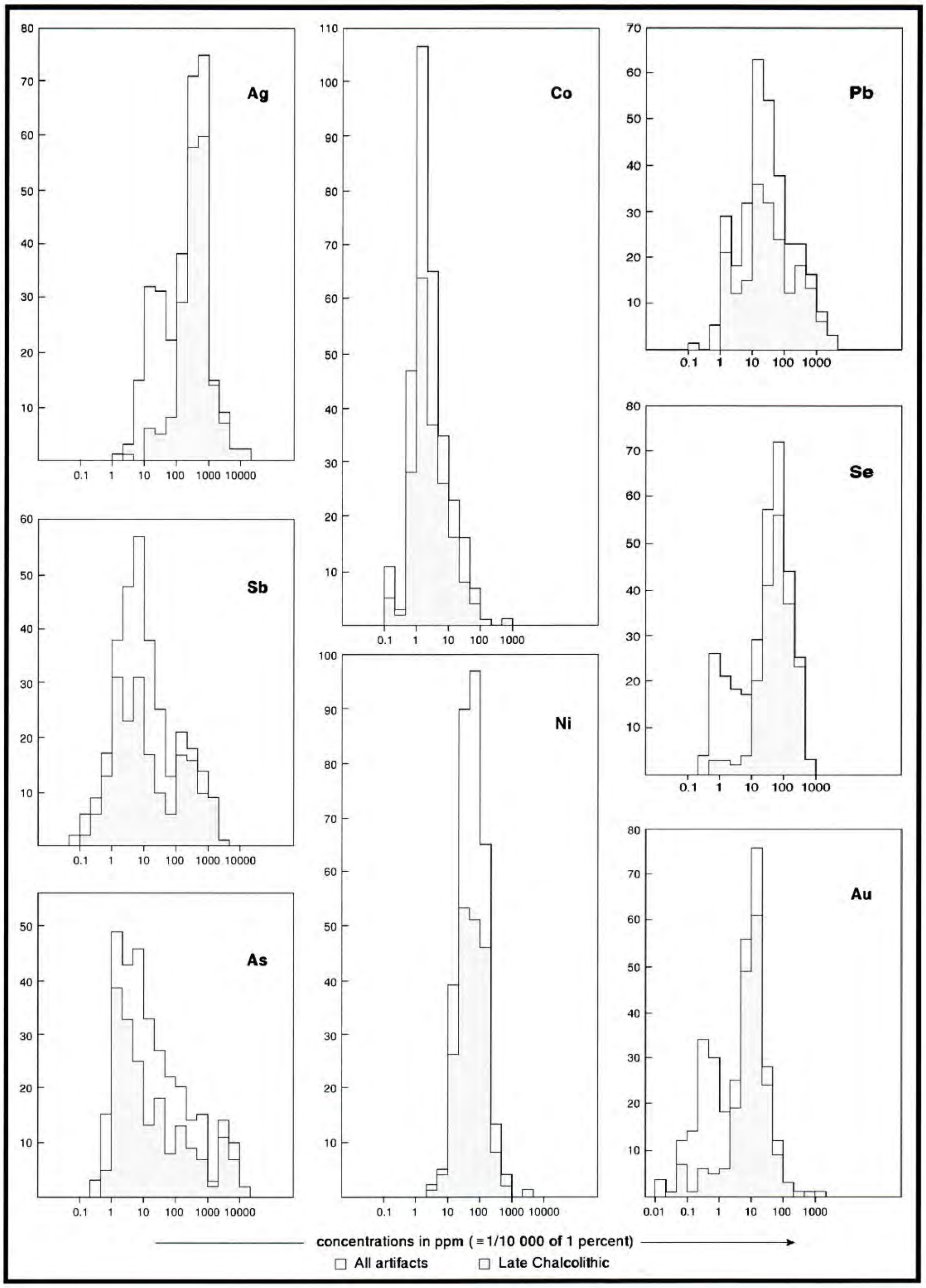

Fig. 22. Number frequency distribution of trace element concentrations in Chalcolithic and PBA copper artifacts from Bulgaria and Serbia. The grey areas for the subset of Late Chalcolithic objects very much mimic the distribution for the full set, except for silver, gold, and selenium. These elements have pronounced secondary low-concentration peaks due to the Final Chalcolithic objects from Serbia.

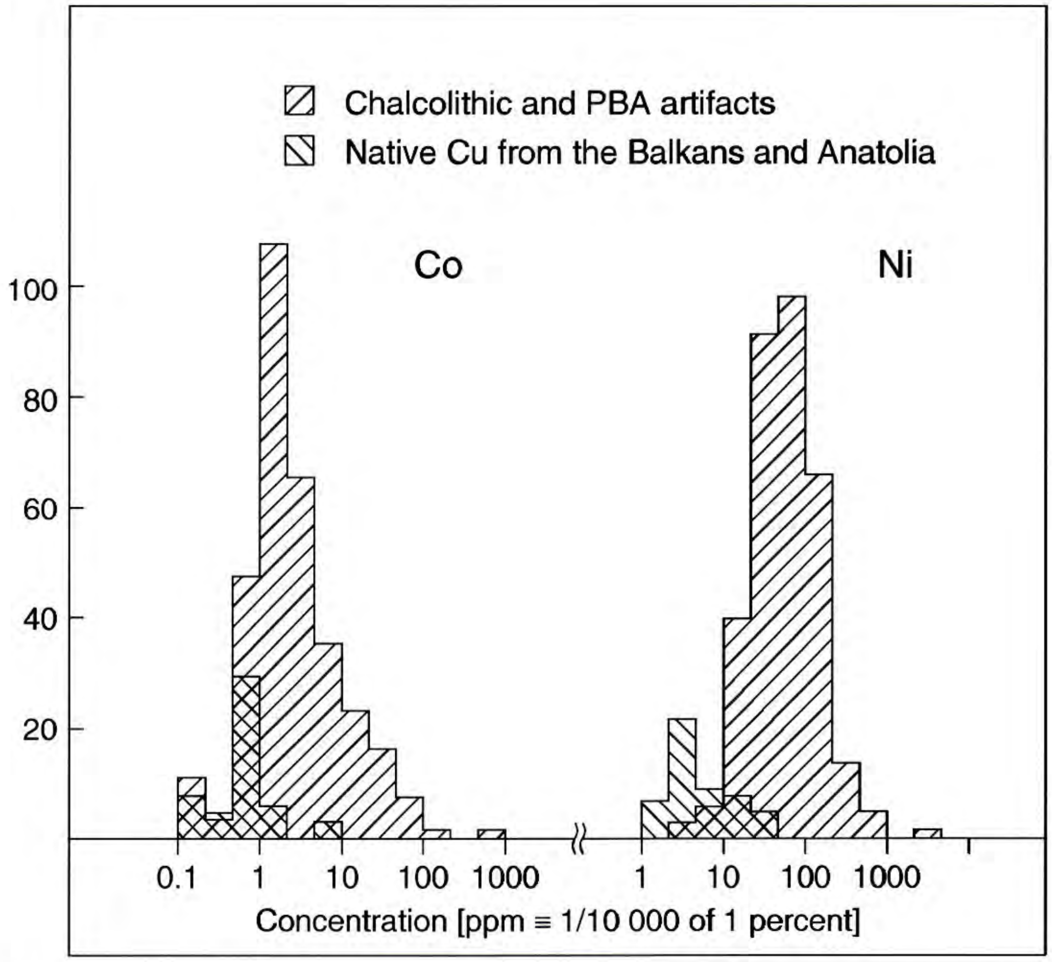

Fig. 23. Concentrations of Ni and Co in native copper from the Balkans and Anatolia tend to be lower than in Bulgarian artifacts. Nickel in particular appears to be a useful marker element to distinguish between artifacts made from smelted copper and such made from native copper.

northern Greece[251], it has been suggested that this alloying practice was continued in the fourth millennium B.C. Now we have with three tin-containing borers (two from the Late Chalcolithic, HDM 2720 and 2733, and one from the Proto Bronze Age, HDM 2148) further finds that would seem to corroborate such a view.

The problem with these finds is that their shape generally yields little chronological information so that in the absence of a reliable stratigraphic context there remains some doubt as to their age[252]. In our case the two samples HDM 2733 and 2148 from Zaminec and Kačica , respectively, are not beyond doubt regarding their stratigraphic position. Kačica was a rescue excavation at a site with a complex stratigraphy containing many pits and at Zaminec Bronze Age material was also found. HDM 2720 is one of fourteen

[251] McGeehan-Liritzis/Gale 1988.

[252] Deliberate tampering with the evidence occasionally may add to the confusion. An axe-adze of the Jászladány type in the Municipal Museum of Geneva, e.g., reportedly found at Szuskó in Hungary, contains 1.1% tin but is now suspected to be a modern fake rather than to indicate early tin alloying in the eastern part of Central Europe. The axe-adze was analyzed in the course of the analytical program at the Württembergisches Landesmuseum (Junghans u. a. 1968) and has the laboratory number SAM 7037.

Late Chalcolithic metal objects from the tell settlement at Smjadovo. There seems to be nothing wrong with the stratigraphy but we note that this object is chemically distinctly different from the remaining thirteen not only because of its tin content. This is reflected in the observation that most samples from Smjadovo are grouped into cluster #1 while HDM 2720 belongs to cluster #9 that contains predominantly late material. Not quite unexpectedly several early tin bronzes were also discovered from the transitional phase to the Early Bronze Age that we call Proto Bronze Age. Such finds from contexts of the Corded Ware and Bell Beaker cultures, however, seem to be concentrated in central and not in southeastern Europe[253].

If we take a closer look at the proposed early finds of tin bronze in southeastern Europe not a single one stands up to close scrutiny. A small (0.2 by 2 cm) piece of slag that supposedly derives from grave 230 of the cemetery at the Lengyel-culture site of Zengövarkony in Hungary has been presented as major evidence of intentional alloying of copper with tin[254]. The slag contained minerals of copper and tin, among others, and may represent a dross from melting or casting. However, in a review of the early metallurgy in southern Transdanubia where all finds from Zengövarkony related with metallurgy are listed and presented as drawings there is no mention of such a slag[255]. Although the excavation is comparatively well documented[256] the slag is also not recorded in the excavation report[257].

Three metal objects from doubtful or poorly documented contexts from the area of former Yugoslavia[258] form rather slim evidence for the use of tin bronze in the fifth millennium in southeastern Europe. Glumac and Todd also mention some ten contemporaneous pins or awls from Bulgaria[259], mainly from Karanovo and Ruse, that contain between 6 and 10% tin. Actually, Černych's analyses include nine Late Chalcolithic artifacts, two from Gradešnica without any reliable information on their context, four from Ruse including one that we have classified as uncertain (HDM 2046)[260], two from Karanovo, and one from the Berekecka Mogila in eastern Thrace. The finds from Karanovo are from the old excavation by M. Mikov and G. Georgiev[261]. Although this does not necessarily mean

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[253] Schickler 1981, 436ff. has listed some hundred such cases of which only about a dozen derive from Slovakia, Hungary, or Croatia. A Proto Bronze Age knife from Velika Humska Čuka in Serbia (SAM 2101) with more than 10% tin has been interpreted on typological grounds as imported from the Aegean (Spindler 1971, 216) where tin bronze was already in use in the third millennium B.C. This typological association was based on the "short chronology" of southeastern Europe and cannot be seriously considered today, because if the context were correct then the knife would be about a millennium earlier than its Aegean parallels.

[254] Glumac/Todd 1991a, 160-161; Glumac/Todd 1991b, 14.

[255] Ecsedy 1990.

[256] Dombay 1939; Dombay 1960.

[257] The metal finds from Zengövarkony in the Museum of Pecs have been kept on shelves in open boxes in the museum magazine over more than some fifty years before the slag was discovered. According to Dr. Ecsedy it is not inconceivable that during this long time some material from different contexts and even different sites may have been mixed.

[258] The objects are a ring from Gomolava (Ottaway 1979), a borer from the Zlotska pećina (Pernicka u. a. 1993), and a fragment of sheet metal from Ratina (Valovič 1985). The date of the ring from Gomolava was already questioned by Chapman 1981 and, in a personal communication, B. Ottaway has confirmed its doubtful context. In the Zlotska pečina three cultural layers were identified (Late Chalcolithic, Sălcuţa; Early Bronze Age Baden-Kostolac, Coţofeni; Early Iron Age, Basarabi). However, these three layers comprised only 20 cm of sediment in part of the cave and accidental admixture of younger material seems quite possible. The site of Ratina is reported to contain Vinča material but otherwise is poorly documented.

[259] Glumac/Todd 1991b, 15, with reference to Черных 1978.

[260] The other three have not been sampled by us.

[261] Миков 1958; Georgiev 1961, 57-80.

that their contexts are unreliable one has to take into account that in those days the stratigraphic positions of finds were generally recorded much less precisely than today. Berekecka Mogila has a complex stratigraphy with Early Bronze Age graves dug into Late Chalcolithic layers of the Karanovo VI period[262]. A metal disc from Strašimirovo that has been considered to be Late Chalcolithic can now be excluded. Strašimirovo is a lake-side dwelling on the shore of Lake Varna that was found during dredging works. Accordingly, there is no stratigraphic information at all available but the material recovered is mainly from the Early and the Late Bronze Age. All of these artifacts are typologically nondescript, i.e. they can only be roughly described as pins, awls, borers, wires, or sheet metal. Two further objects do not even have an exact find location. They are a fishing hook from "Bulgaria" and an axe-adze from the region of Sofia[263]. All this is not very compelling evidence to suggest that tin bronze was indeed used much earlier in the Balkans than in southwest Asia and the Aegean, from where the earliest well documented tin bronzes are presently known[264]. It seems that the significance of these finds from southeastern Europe has been as much exaggerated as the potential of small occurrences of cassiterite in the same area[265].

Less spectacular but nevertheless very interesting, however, is the occurrence in southeast Europe of tin bronze in the late fourth and early third millennium B.C. This is about the same time that this new material appears in northern Greece (Sitagroi)[266], Anatolia (Ikiztepe, Cudeyde)[267], and in northern Mesopotamia (Tepe Gawra)[268]. All these objects are low-tin bronzes, containing between 1 and 6% tin. At Ezerovo II 12 metal samples with such a composition[269] have been found, two borers, a flat axe, and the rest casting spills[270]. Ten more samples, mainly chisels, were detected at Emenska pest, although this site may also well range into the third millennium B.C. Of four samples (one bead, one pin and two nondescript pieces of copper) from Sitagroi IV dated to ca. 3500–3110 B.C.[271] all contained tin between 0.12 and 5.9%[272]. In this context our borer from Kačica with 2.35% tin fits well into this pattern. It seems that tin bronze came into use occasionally by the end of the fourth millennium B.C. but continued to be a rare commodity until the beginning of the European Early Bronze Age in the second half of the third millennium. The earlier extended use e.g. at Troia, Poliochni, Alaca Hüyük, and at Ur in Mesopotamia seems to have been restricted to a social elite.

[262] Parzinger 1993, 115. The excavation itself is as yet unpublished.
[263] Glumac/Todd 1987.
[264] Muhly 1985.
[265] McGeehan-Liritzis/Taylor 1987. In this article even minute occurrences of cassiterite identified only by ore microscopy in mixed sulfide ores from Romania (Radulescu/Dimitrescu 1966) are listed as potential sources of tin.
[266] McGeehan-Liritzis/Gale 1988.
[267] R. Braidwood/L. Braidwood 1960, 300ff.; Bilgi 1984, 53f.
[268] Muhly 1985.
[269] Черных 1978.
[270] Todorova/Tončeva 1975; Tončeva 1981. Altogether four lake side dwellings have been discovered at Ezerovo at Lake Varna. Ezerovo IV is considered Eneolithic but Ezerovo II and III are dated to the Early Bronze Age.
[271] Renfrew 1986a, 24.
[272] McGeehan-Liritzis/Gale 1988, 216 Tab. 4.

Variation of metal composition with time

Cultural periods often are named after material properties of artifacts that are typical of, or came into being, during these periods. Thus, it is almost by definition that, with the advent of tin, there should have been a change from the Chalcolithic to the Bronze Age of the chemical composition of many of the metal artifacts from these periods. Other important developments have occurred, however, without being recognized by naming a cultural period after them so that material changes will also be present in artifacts from within the same periods. A case in point would appear to be the use of arsenic. Although it improved some of the essential properties of pure copper to the extent that only after its introduction could many of the cutting tools be used to their full potential there is no cultural term to acknowledge this fact.

Presumably, the first use of arsenic as an alloying element was not intentional but rather came about because copper ores became to be utilized that happened to contain arsenic, or were associated with arsenical ores. An interesting question then is why there was such a change of source ores and when it took place. Did it happen within a given cultural period, without evidence for a change in cultural tradition to have occurred at the same time? Then it might just be that the old sources were exhausted or, because of increasing demand, had to be supplemented by others. Or did the changes coincide with such of typology, say at the transition from Middle Chalcolithic to Late Chalcolithic or from Late Chalcolithic to Final Chalcolithic? Then perhaps outside forces caused a change of trade pattern that was paralleled by one in style or taste.

For copper objects from Poliochni on the island of Lemnos and from Thermi on Lesbos it is well documented that material properties changed from one cultural level to the next[273]. Certainly, these two settlements are atypical in so far as on both islands there are no local ores available that could have been exploited. In such instances, if all raw metal or finished objects had to be imported, it stands to reason that a change of supplier due to changing trade connections will have occurred rather frequently, at least it will have occurred more frequently than at such sites that are located in the close vicinity of rich and abundant ores.

In present-day Bulgaria the situation will have been quite different. It is a region where copper ores are abundant and where Chalcolithic people knew how to utilize them. After all, at Ai Bunar in Thrace the earliest mining activities date back to the end of the fifth millennium B.C.[274] and at Durankulak, in context with a well-dated cultural level, crucible sherds have been discovered[275] that also date to ca. 4300 B.C. They testify at least to melting and casting but possibly also to small-scale smelting of ores. Hence, there can be no doubt that from the Late Chalcolithic on local ores were exploited and it is very likely that extractive metallurgy was mastered. Nevertheless, here we find also clear evidence in the data for a dependence on cultural level, in the chemical composition of the artifacts as well as in the isotope signature of their lead. *Fig. 24* displays the distribution among the chemical clusters of the artifacts from the different archaeological periods. Of the objects from the Early and Middle Chalcolithic 20% fall into cluster #8 that is almost completely absent from all later periods. During the Late Chalcolithic clusters #1, 3 and 4 dominate (80%), only to give way, during the Final Chalcolithic, to cluster #2 (61%). Finally, during the Proto Bronze Age, clusters #7, 6, and 9 become prominent and make up more than two thirds of the total.

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[273] Pernicka u. a. 1990; Begemann u. a. 1995.
[274] Черных 1978.
[275] Todorova 1995.

The lead isotope data show a similar picture; they also show changes with time that mirror those of the chemical composition. Taken together the lead in the artifacts is isotopically quite diverse in its composition, much more so than in the ores and the native copper. All three ^{206}Pb-normalized isotope abundance ratios vary by some 20%. A breakdown of the isotope data according to time periods shows that during the Early and Middle Chalcolithic as well as the Final Chalcolithic the lead isotope composition is rather narrowly constrained *(Fig. 25)*. It is objects from the period in between, the Late Chalcolithic (~ 4400–4100 B.C.), that are essentially responsible for the wide range in abundance ratios, in particular for the extension to the side of low ratios. Indeed, the correspondence between cultural phase and lead isotopy is possibly even more stringent than is apparent from the histogram. The object from the Middle Chalcolithic (HDM 1976) that, in the lower panel of *Fig. 25*, plots on the extreme left actually dates to the transition phase from Middle to Late Chalcolithic and might therefore, with equal justification, be assigned to the Late Chalcolithic. Objects from the Proto Bronze Age and younger then add to the total spread by extending the range on the high side (top panel). Without exception the objects with the highest ratios are all tin bronzes. It is not clear at present whether such high abundance ratios are a general feature of all Bulgarian copper-based artifacts from the end of the second millennium B.C. or whether the high ratios are restricted to, and are typical of, Bulgarian *bronzes* from this period.

Late Neolithic

The Bulgarian artifacts from this period are all small beads from Durankulak that appear to consist of malachite, possibly intergrown with higher-grade copper minerals such as cuprite (Cu_2O) or even with metallic copper. This suggestion is supported by the observation that under the binocular microscope one sample (HDM 1970) contained a layer of reddish material in the center of the bead that might be cuprite or copper[276]. Although such mineral associations are occasionally observed in nature, it is also possible that these beads actually are completely corroded copper metal. Trace elements unfortunately do not help to decide between the two possibilities. The low silver contents of 4 ppm or less are compatible with native copper, but the high cobalt concentrations of 280 ppm or more of three of the samples, and the nickel content of all samples, speak clearly against it. Actually, both the extremely low silver concentrations as well as the high cobalt contents are somewhat puzzling. As is evident from *Tab. A1*, with but one exception (HDM 1974, a Late Chalcolithic finger ring from Durankulak, grave 679) cobalt contents of 200 ppm or more are not encountered in any of the Chalcolithic artifacts; they only occur first in tin bronzes of the Late Bronze Age. Similarly, silver contents as low as 4 ppm or less are also only met in two of the 290 Bulgarian metal objects. Hence, if these beads from Durankulak originally should indeed have been made of malachite then such malachite cannot have been used for the production of the metal of any of the later objects. Interestingly, the same holds also true for the malachites excavated at Eneolithic Selevac in Serbia. There, the silver content of all 15 specimens was also in the low ppm range or even below 1 ppm[277]. The situation was different in so far, however, as 13 of the 75 Serbian metal artifacts had silver contents of less than 10 ppm also.

[276] Since this sample was rather small, no attempt was made to identify the mineralogical composition of the object further, e.g., by preparation of a polished section.

[277] Pernicka u. a. 1993.

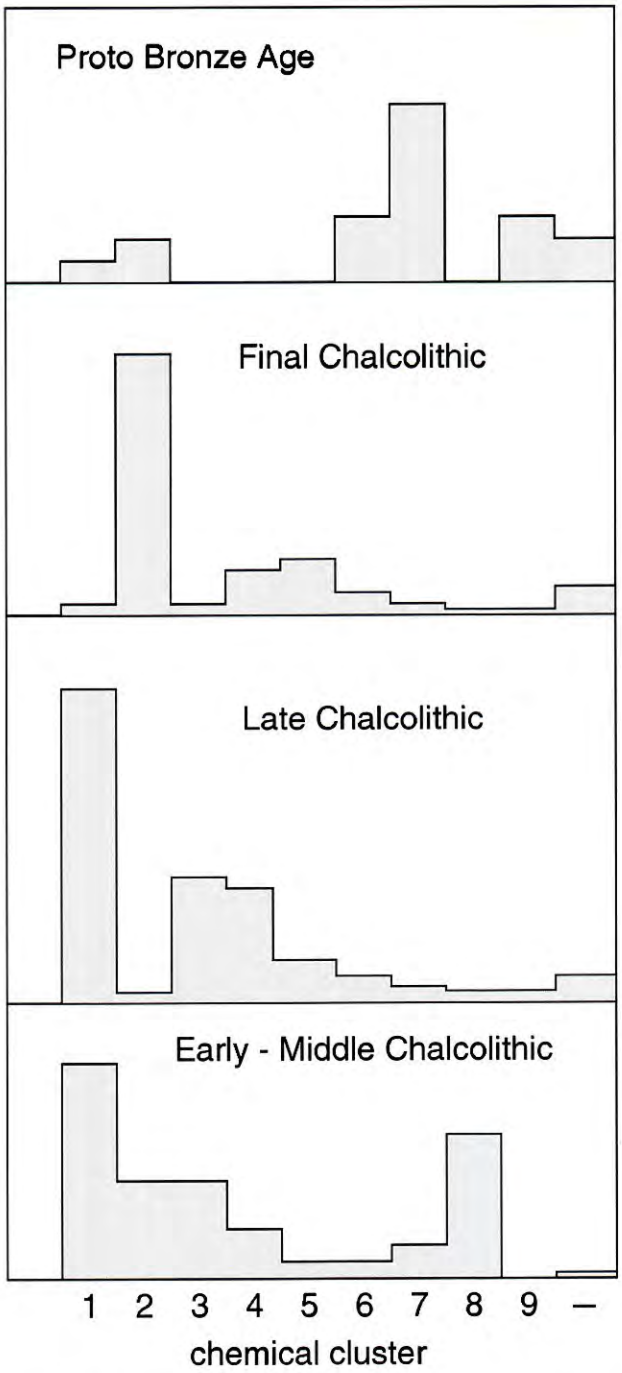

Fig. 24. Distribution among the chemical clusters of artifacts from different periods. The change with time of the trace element signature indicates a change with time of the copper supply.

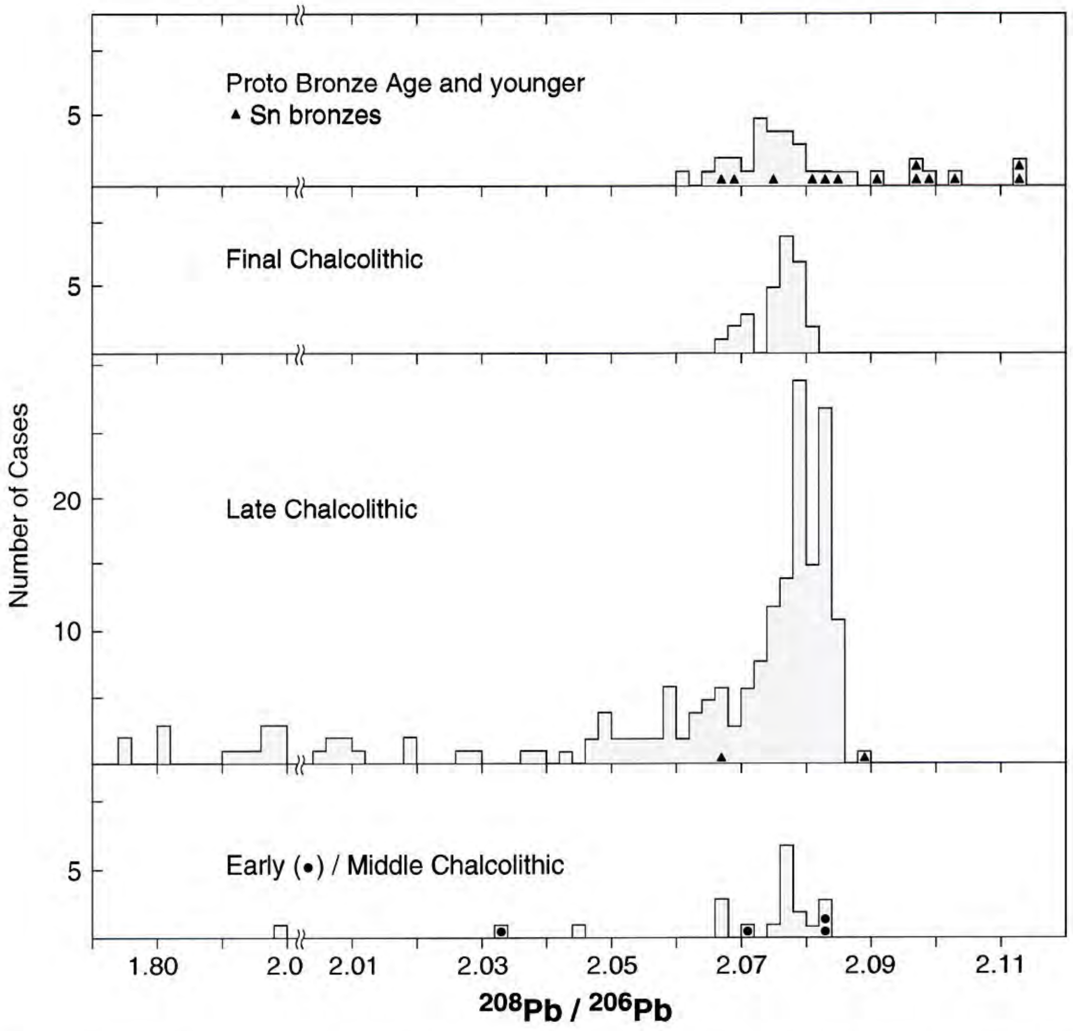

Fig. 25. Number frequency distribution of $^{208}Pb/^{206}Pb$ ratios in copper artifacts from Bulgaria, subdivided according to archaeological periods. Note the change in scale below $^{208}Pb/^{206}Pb = 2.00$.

Early Chalcolithic

At present the earliest reasonably well dated heavy metal implement from southeastern Europe is a chisel of 290 g from Slatino I/II in west Bulgaria (HDM 1904). We have also assigned to this period a slim axe, typologically unique with round to oval cross section from Devebargan in the Marica valley (HDM 1912) although its find context is not beyond doubt. And, finally, there are from this period two finger rings from Durankulak (HDM 1975 and 2005).

Chemically, three of these four objects consist of very pure copper, with silver as the major impurity *(Tab. A1)*. The fourth one, one of the finger rings from Durankulak (HDM 1975), has considerably higher contents of Pb, As and Sb but even in this case these combine to less than 0.5 percent of impurities. Using the criteria, outlined above, to distinguish between native copper and smelted copper it turns out that in all four artifacts the silver and nickel contents are at the very high side of what is typical of native copper from the Balkans. Based on these criteria we would argue that all four objects were manufactured from smelted copper. Isotopically, the lead in HDM 1904 and 1975 is indistinguishable from the lead in ores from Ai Bunar. However, only in the finger ring from Durankulak are the trace element abundances barely compatible with such an origin of the metal. In the case of the chisel from Slatino (HDM 1904) the very low concentrations of arsenic and antimony make a connection with Ai Bunar rather unlikely. This will be discussed in more detail below.

Middle Chalcolithic

Seventeen artifacts were available for analysis; seven axes - mostly of the Pločnik type; five bracelets from Durankulak, and five small items that we have called borers. The axes belong to the early types which were found in hoards that usually are notoriously difficult to date. The problems are exemplified already by the objects from Pločnik itself where it has been argued that even these eponymous objects derive from at least three distinct hoards and that it is impossible to know the stratigraphic layer to which they belong chronologically[278].

The chemical composition of the metal from this period is quite diverse *(Tab. A1)*[279]; the objects are rather evenly distributed between clusters #1-8. Similarly diverse is the isotopic composition of the lead *(Fig. 26)*. Nevertheless, there is some structure to be observed in the data that, moreover, is different in the Bulgarian and Serbian subsets. Five objects, all axes from western Bulgaria, belong to the combination gr/cl 1/2 that also characterizes the (few) ores from Majdanpek. Another five artifacts, this time four implements from Pločnik but only one from Bulgaria (HDM 1977), are gr/cl 2/3, a combination that so far is unique for ores from Ai Bunar.

Late Chalcolithic

This period, while poorly represented in our suite of metal artifacts from Serbia, makes up most of the objects from the present study. However, even in Bulgaria are the metal finds very unevenly distributed *(Tab. 1)*. Out of 190 samples only five derive from western Bulgaria (HDM 1905, 2725, 2727, 2728, 2733) and only one from Thrace (HDM 2150). All remaining objects were found along the Danube or in northeastern Bulgaria at or near the Black Sea coast. Moreover, some 70% of those come from only two sites, namely Durankulak (79) and Ruse (56).

As far as the trace element contents are concerned the composition is rather varied although it should be noted that with but six exceptions[280] we are still dealing with 99% pure copper. In the number frequency vs. concentration histogram *(Fig. 22)* silver, nickel, gold, and selenium show a rather narrow, nearly log-normal distribution with more than two thirds of all objects falling into the three most populated bins; among them the range of concentrations is about a factor of ten. In the case of arsenic and cobalt the

[278] Parzinger 1993.

[279] The chemical and isotope data for the Serbian objects are not repeated here. They have been published by Pernicka u. a. 1993.

[280] Two samples each contain more than one percent of arsenic (HDM 1978 and 2140), silver (HDM 2055 and 2092) and iron (HDM 1968 and 2138).

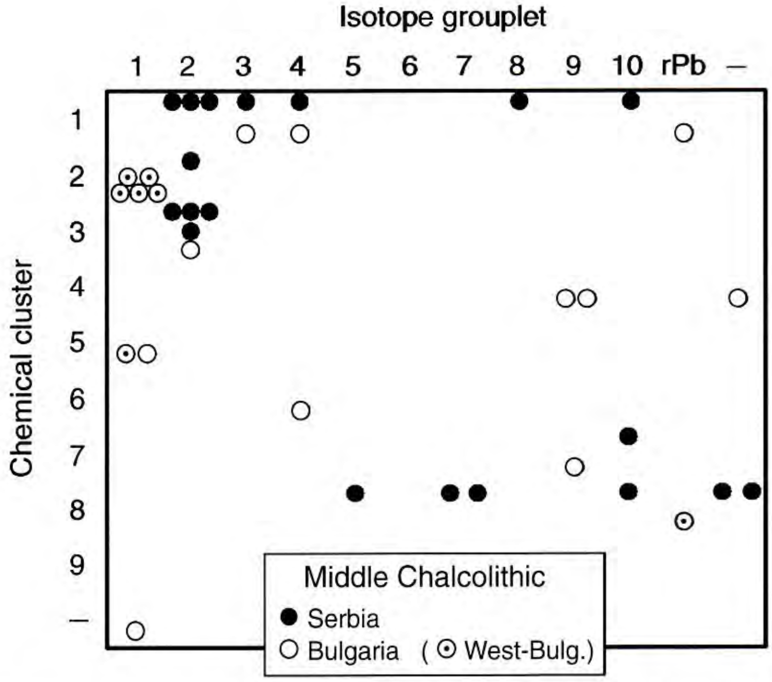

Fig. 26. Classification matrix of Middle Chalcolithic copper objects from the Balkans. Note the differences between Serbia and Bulgaria and also between west Bulgaria, on the one hand, and the remainder of the Bulgarian samples from the Black Sea coast and northeast Bulgaria, on the other.

distributions show a pronounced tail towards high concentrations as it is to be expected for elements in accessory minerals that are present in widely varying amounts. Antimony and lead, finally, are bimodally distributed. Moreover, the objects in the high-antimony peak also tend to be high in lead although the correlation is far from perfect[281].

Isotopically the metal is also quite diverse; peaks in the histogram on the left of the matrix in *Fig. 27* are seen to occur in grouplet #2 and for "radiogenic lead" (rPb). According to this matrix much of the variation in the isotopic composition of lead is due to artifacts that belong to (chemical) cluster #1. The matrix also shows that the box with the largest number of objects[282] is the combination gr/cl 2/3. As mentioned already, among all Bulgarian and Serbian ores analysed so far this combination is unique to Ai Bunar. Fully compatible with a derivation from Ai Bunar of the copper in these objects is the observation that the lead content in 23 of the 27 artifacts puts them into the high-lead peak of the number frequency distribution *(Fig. 22)*.

The second-most frequent combination among the Late Chalcolithic objects is gr/cl rPb/4 (19 cases). As far as the lead isotopy is concerned suitable ores to yield this kind of radiogenic lead are those from Rudna Glava in Serbia, from Rosen/Burgas and from Devenköy-Çatak Tepe on the Turkish side in the Strandža Mountains. Of these Rudna Glava can be eliminated because its ores do not meet the trace element requirements. Tylecote[283] reports copper smelted from Rudna Glava ores to contain trace elements in con-

281 Pb/Sb abundance ratios in these objects range between less than 0.1 and about 6.
282 For the exact number of samples in each box of the matrix cf. Table 4.
283 Tylecote 1982.

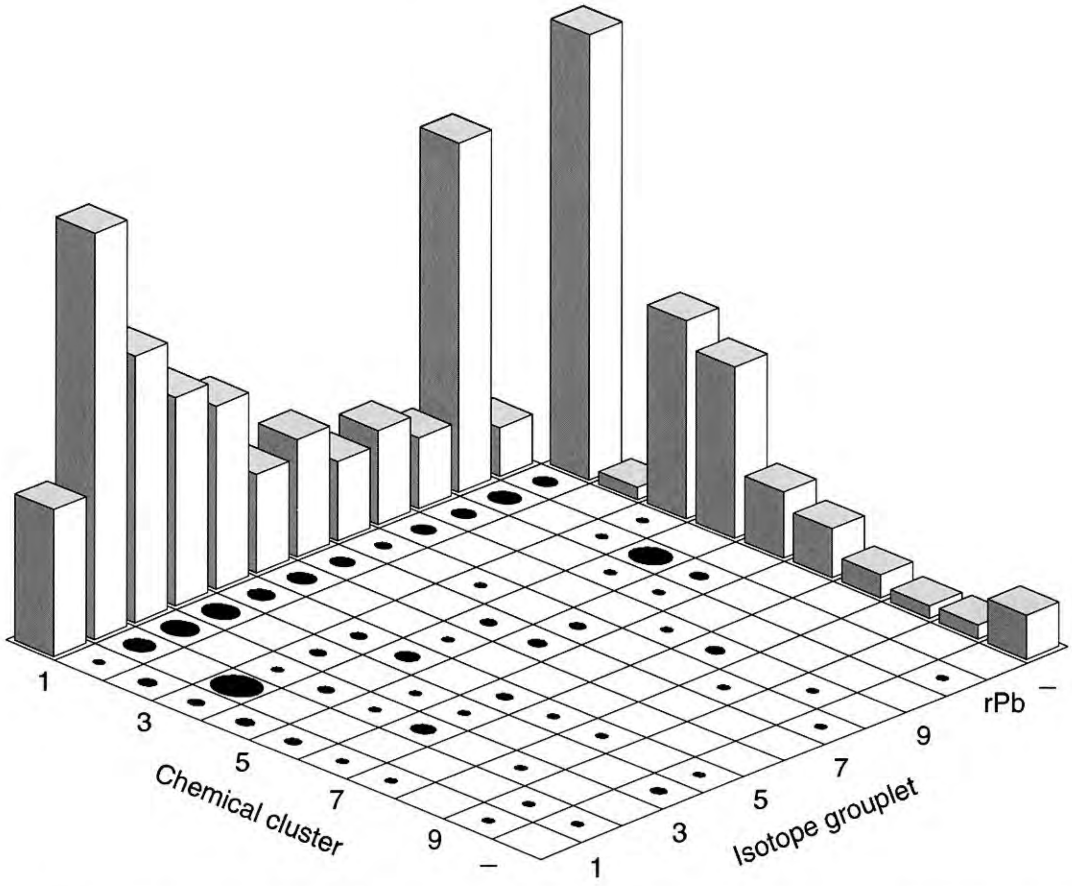

Fig. 27. Classification matrix based on composition of Late Chalcolithic artifacts from Bulgaria (190) and Serbia (2). The bar graphs give the total number of objects per isotope grouplet (left) and per chemical cluster (right), respectively. The size of the black circles is a measure of the number of objects in each individual box; the exact numbers can be found in Table 4. Cluster #1, characterized by low concentrations of all trace elements except silver, is particularly heterogeneous in the isotopic composition of its lead suggesting a multi-source origin of the metal the objects in this cluster are made from. Among Bulgarian copper ores the combination grouplet #2/cluster #3 is characteristic of Ai Bunar while ores with radiogenic lead (rPb) belonging to cluster #1 and #4 occur at Rosen-Medni Rid in the extreme southeast of Bulgaria.

centrations that agree well with what is predicted on the basis of analyses of the ores themselves[284]. According to these data arsenic, antimony, cobalt, nickel, and gold are all too high to meet the specifications of cluster #4. The most blatant case is the cobalt content of "Rudna Glava copper" of a few tenths of one percent which is actually higher than found so far in any of the artifacts from Serbia and Bulgaria. At Rosen/Burgas and Devenköy-Çatak Tepe the situation looks at present more favorable in this respect but the very few samples analyzed so far makes, of course, a positive assignment tentative at best. It may be noted in passing that the geographic distribution of the objects from the box gr/cl rPb/4 is very uneven; with only one exception (HDM 1922 from Tell Ruse) all of them come from Durankulak at the Black Sea coast some hundred kilometers northeast of Varna.

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[284] Pernicka u. a. 1993.

The non-uniformity of the trace element patterns and the lead isotope signature unequivocally shows that the ores utilized during the Bulgarian Late Chalcolithic "metal boom" must have come from a variety of sources. But then this period lasted some 300 years, from about 4400 to 4100 B.C. The question is, therefore, whether a finer resolution in time would show that there have been occasional shifts within this time span and that, at any given time, there was present copper with a narrowly constrained lead isotopic composition and that the wide range actually observed simply might be due to the superposition of many such narrow populations. To look into this possibility we may compare artifacts from the same graves because such funeral offerings are presumably rather rigorously "contemporaneous". As a rule, one would hardly expect to find, in the same grave, metal objects that are different in age by more than a few decades.

At Durankulak there are 18 instances where two or more metal objects were retrieved from the same grave. (In addition we analysed three malachite beads from grave #643, two from grave #626 and one each from graves #611 and 648.) For easy reference we have compiled, in *Tab. 6*, some of the essential material features of these objects. As to the lead isotopy there are several cases where artifacts from the same grave are completely different in their signature. The most pronounced examples are two pairs of bracelets from grave #253 and grave #660, respectively. From both pairs one bracelet each falls at the extreme low end of all measured ^{206}Pb-normalized abundance ratios and the other at the extreme high end. Not quite as distinct from one another in their isotopy are two bracelets in the pairs retrieved from grave #29 (HDM 1976 and 1977) and grave #533 (HDM 2014 and 2015) and the four and five artifacts, respectively, from graves #477 and 514. In all cases, however, is there again one object each with very low abundance ratios and one with high ratios. We interpret this to mean that the diversity among Late Chalcolithic artifacts of the lead isotopic composition is not an apparent effect due to having pooled objects from too long a period of time but that it rather reflects the availability to the same people of objects made from copper with a diverse lead isotopy.

As a rule, for objects from the same grave a difference in isotopy goes parallel with grossly different contents of at least some trace elements, usually arsenic and/or antimony but occasionally also nickel, cobalt, or silver *(Tab. 6)*. We take this as a convincing argument that such artifacts, contemporaneous to within perhaps a few decades, cannot have been produced at the same site. We have argued before[285] that differences in the contents of trace elements by factors of more than several hundred or even several thousand are improbable to have been maintained in artifacts from any one workshop because it requires that material from different charges be kept rigorously separate and that residues, scrap or miscasts and spills from one charge never be mixed with the other. While this may have been possible in principle there is no reason apparent why people should have desired to do so. An inference of this conclusion that objects with very low trace element concentrations cannot have been produced in the same workshop where also high-impurity metal was utilized is, of course, that either the low-impurity or the high-impurity artifacts, or both, must have been imported as such.

In contrast to the pronounced heterogeneity in some multiplets of the lead isotopy and trace element contents there are other pairs of artifacts from the same grave for which both features agree remarkably well. Particularly noteworthy in this respect are the pairs of bracelets from graves #390, 447, 501, 756 and, with somewhat poorer agreement, that

[285] Tadmor u. a. 1995.

Grave	Object	HDM	$^{208}Pb/^{206}Pb$	$^{207}Pb/^{206}Pb$	$^{204}Pb/^{206}Pb$	As	Sb	Co	Ni	Ag	Au	Se
29	bracelet	1976	1.9927	.8069	.05133	< 1	29	6	150	150	29	38
	bracelet	1977	2.0825	.8439	.05402	2490	590	1	< 15	620	21	177
217	bracelet	1979	2.0730	.8382	.05350	< 4	0.2	8	26	4	0.08	1
	bracelet	1982	2.0555	.8306	.05308	4	7	1	26	640	10	40
251	bracelet	2141	2.0838	.8444	.05398	2790	760	3	< 40	800	14	95
	bracelet	2142	2.0816	.8429	.05391	20	330	4	30	770	11	120
253	bracelet	1965	1.8130	.7402	.04701	310	4	40	80	156	18	30
	bracelet	1978	2.0831	.8441	.05402	10100	1820	1	< 20	800	12	68
275	bracelet	1968	2.0191	.8194	.05224	5	2	90	2400	180	27	43
	bracelet	1981	2.0583	.8344	.05327	< 5	8	1	107	910	36	16
390	bracelet	1944	2.0749	.8448	.05399	1	1	10	82	104	19	21
	bracelet	1945	2.0754	.8449	.05402	1	1	9	71	111	20	26
447	spiral ring	2009	2.0589	.8363	.05339	1	3	1	96	240	33	47
	bracelet	2023	1.9966	.8102	.05174	< 3	2	6	118	280	40	52
	bracelet	2024	1.9977	.8103	.05168	< 2	2	3	100	276	37	55
	finger ring	2027	2.0825	.8441	.05403	6300	1310	1	< 25	860	10	279
455	finger ring	2013	2.0799	.8412	.05374	1	0.2	1	27	720	3	400
	finger ring	2029	2.0831	.8441	.05400	1100	320	3	95	1980	15	33
496	finger ring	2002	2.0198	.8192	.05234	640	12	78	98	500	13	66
	finger ring	2137	2.0292	.8230	.05249	11	0.7	18	132	145	20	58
	finger ring	2138	2.0775	.8461	.05405	69	2	10	99	170	9	18
501	bracelet	2025	2.0675	.8325	.05310	340	15	30	284	450	0.6	100
	bracelet	2026	2.0679	.8325	.05310	360	15	32	310	470	0.6	102
506	finger ring	2031	1.7515	.7169	.04536	23	1	5	105	205	55	27
	spiral ring	2036	2.0063	.8145	.05196	10	9	13	132	215	100	72
514	finger ring	1962	2.0260	.8216	.05250	78	2	9	34	1520	31	19
	finger ring	2008	2.0492	.8306	.05309	41	31	6	53	330	7	105
	bracelet	1963	2.0850	.8445	.05395	3200	1010	1	< 23	810	11	89
	bracelet	1964	2.0835	.8442	.05399	8400	1490	2	< 23	720	12	57
	spiral ring	2007	2.0838	.8443	.05401	720	650	1	< 12	500	11	56
533	bracelet	2014	1.8107	.7396	.04699	266	3	36	74	167	17	28
	bracelet	2015	2.0649	.8347	.05323	13	11	2	< 27	600	8	53
660	bracelet	1984	2.0827	.8435	.05393	234	1550	3	47	880	20	310
	bracelet	1993	1.9036	.7749	.04928	13	3	35	350	144	56	12
672	bracelet	1994	2.0394	.8248	.05258	< 3	0.6	4	156	580	34	33
	bracelet	1995	2.0482	.8303	.05308	53	8	107	64	180	3	96
732	bracelet	1986	2.0766	.8414	.05380	66	120	7	57	330	100	50
	bracelet	1996	2.0833	.8439	.05401	240	1500	2	< 105	650	8	78
756	bracelet	1989	2.0833	.8442	.05402	4	122	0.5	< 41	440	3	235
	bracelet	1998	2.0823	.8441	.05407	4	112	< 0.5	< 34	420	3	221
763	bracelet	1991	2.0831	.8441	.05402	56	370	0.5	< 63	720	17	81
	bracelet	1992	2.0819	.8439	.05407	5100	830	< 0.5	< 61	580	14	256

Table 6. Lead isotope and trace element data for multiplets of Late Chalcolithic artifacts retrieved from graves at Durankulak. The members of each multiplet are supposed to be contemporaneous to within a few decades at most. Trace element concentrations (in ppm ≡ 1/10.000 of 1%) have been rounded; the exact data are listed in Table A1. In some graves (#390, 447, 501, 756) the isotopic and trace element signatures are so similar that it is almost certain that such pairs of bracelets have been cast simultaneously from the same metal charge. As a rule, however, the material features of members of the same multiplet are quite diverse suggesting that metal from different sources must have been available concurrently.

from grave #514. For these objects it is almost certain that each pair was cast at the same time from a single charge of metal. Actually, for each pair the thickness of the rods the bracelets are made from is virtually identical also, which might suggest that the bracelets were made by cutting, and shaping, pieces of suitable length from longer rods[286].

Final Chalcolithic

Altogether, we now have data for 82 artifacts, 30 from the present study and 52 from Serbia published previously[287]. The eastern two thirds of Bulgaria are only poorly represented; there are just three axe-adzes (two stray finds and one from a hoard at Ai Bunar) but none of them comes from anywhere near the Black Sea coast. The reason is not so much a bias in the selection of objects available to us for sampling; it rather reflects the absence of Late Chalcolithic artifacts from this region as we have discussed above in connection with the chronology of the use of copper in Bulgaria. As also discussed there most of the artifacts from this period are heavy implements like axes and massive chisels. Only from Teliš-Redutite in Bulgaria and Zlotska pečina in Serbia do we have some of the small items (borers, awls, pins) that were dominant at Tell Ruse; personal ornaments like finger rings and bracelets as they made up most of the Late Chalcolithic artifacts from Durankulak are completely absent.

Chemically, and isotopically even more so, the composition is fairly uniform. According to the classification matrix *(Fig. 28)* more than half (47 out of 82) of the artifacts belong to chemical cluster #2, even two thirds (55 out of 80) to isotope grouplet #1. Fourty-four of the 80 objects analyzed for both their trace elements and their lead isotopy are gr/cl 1/2, a combination we have tentatively assigned to have originated from Majdanpek ores. Of the Bulgarian objects it is 11 (out of the total of 30) that show this combination. The essential reason for this fraction being low are the small items from Teliš-Redutite that are scattered all over the matrix. Only one out of fourteen of these objects is gr/cl 1/2, of the remaining Bulgarian samples it is ten out of sixteen.

Proto Bronze Age

This period is represented by 17 Bulgarian objects, in addition to two Serbian ones that were analyzed previously[288]. The typology reflects the first appearance during this period of daggers although we find it peculiar that daggers should be so dominant (11 objects out of 19). Also quite different from that of the previous periods is the chemical composition *(Tab. A1)*. While there is as yet only one (Sn) bronze these Proto Bronze Age artifacts are set apart from the previous periods by their high arsenic content; it ranges up to 8.4 percent (in a dagger from Durankulak, HDM 1966) and is above one percent in 10 of the 19 artifacts[289]. At the same time there are also a few cases still where As is in the ppm range so that, in the number frequency histogram of *Fig. 22*, objects from this period come to

[286] This suggestion is in agreement with the observation by Рындина 1994 that the bracelets were bent and brought into shape by hammering.

[287] Pernicka u. a. 1993. Lead isotope data exist for only 80 artifacts because one object each from Serbia (HDM 1338) and western Bulgaria (HDM 2213) was not analyzed isotopically.

[288] Pernicka u. a. 1993.

[289] The distribution of early arsenical copper in southeastern Europe and possible reasons for its introduction are discussed by E. Schubert 1981.

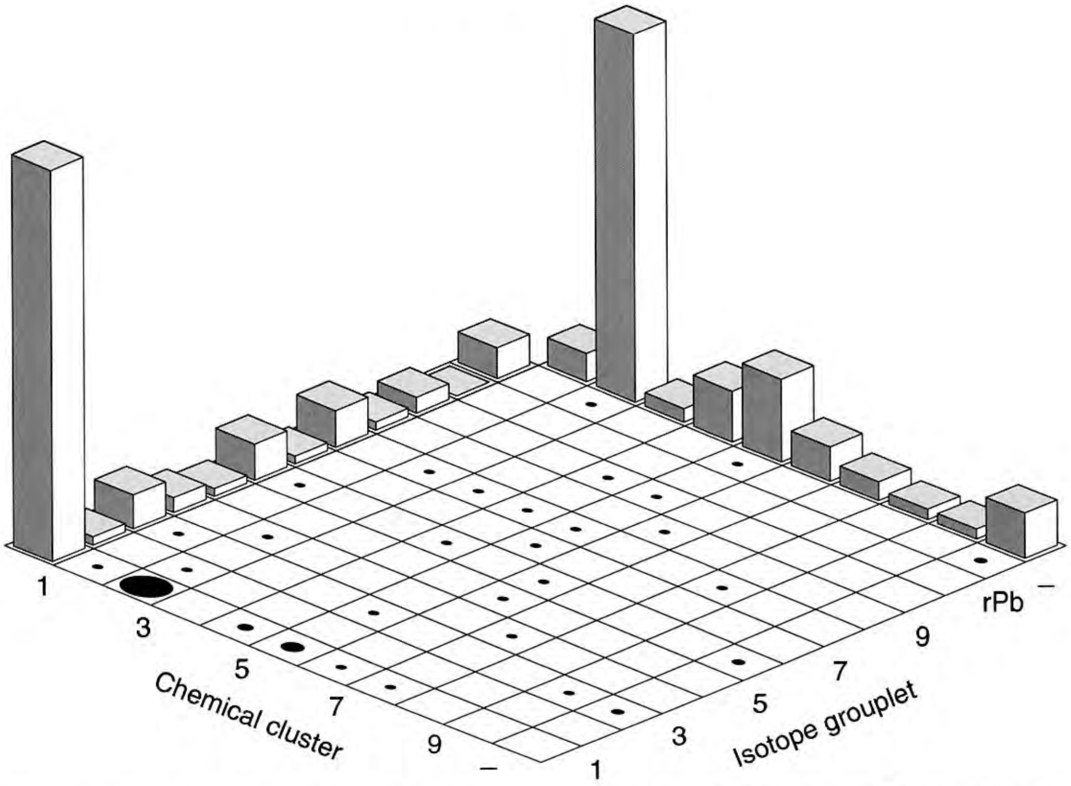

Fig. 28. Classification matrix of Final Chalcolithic copper artifacts from Serbia (51) and Bulgaria (29). There is a clear dominance of a single type of metal, of combination grouplet #1/cluster #2, that is also characteristic of copper ores from Majdanpek in east Serbia.

fall at the extreme right end of the distribution as well as at the extreme left end. A peculiar case is the dagger, type Jagodina, from Haramijska Dupka in the central Rhodopes (HDM 2740). It is the only one made from pure copper (As = 80 ppm, Sn < 80 ppm), a soft and easily deformable material that would appear not to be very suitable for this type of implement, at least not if it was intended to be used as a tool or a weapon. Finally, there are three obtrusive chemical mavericks, namely the stray find dagger from Malorad (HDM 2736) with 2.5 percent nickel and two borers (HDM 2147 and 2742) with almost 2 percent silver[290].

The general appearance is as if a variety of new ores were being experimented with. Perhaps this was by necessity, because the old sources were exhausted. But it is possible as well that, for what reasons ever, a change in cultural orientation occurred as it is also suggested by the appearance at the same time of new ceramic technologies and vessel shapes. In this connection it is an intriguing observation that there are contemporaneous metal artifacts from Ilıpınar near Iznik in northwest Anatolia that also have arsenic contents between 1.4 and 8 percent[291]. Moreover, their contents of all other trace elements

[290] Curiously, the highest mercury contents of all artifacts are found in three borers from Kačica (HDM 2147, 2148, 2149). It is very unlikely that this should indicate the use of native copper. We think it more probable that this Hg is a contaminant acquired from the environment either during burial or at some later stage.

[291] Begemann u. a. 1994. The objects in question derive from what at Ilıpınar is Late Chalcolithic, Phase IV (Roodenberg/Thissen/Buitenhuis 1989/90). In absolute terms, however, they date to between 3500 and 3700 B.C. (calibrated ^{14}C) and are thus contemporaneous with the Bulgarian Proto Bronze Age.

and the isotopic signature of their lead are sufficiently similar to those in the Proto Bronze Age objects from Bulgaria that a common origin is entirely possible.

Late Bronze Age

Most of the objects have tin contents of more than one percent. They clearly are made from intentional alloys although the range in Sn content between 1.1% and 12.3% indicates that there was as yet no strict control over the alloy composition. At the same time the arsenic contents are much more narrowly constrained than in any of the preceding periods (in 12 out of 14 cases it falls between about 0.1 percent and 0.5 percent) and the Sb/As ratios now tend to be ten times higher (≥ 0.3) than during the Proto Bronze Age.

There is again one object, a double axe from Vodnjanci (HDM 2038) in the northern Black Sea region, where the chemical composition would appear not to fit the intended purpose. The axe is made of rather pure copper; arsenic and tin are present in such small concentrations that the properties one expects of an axe will not have been improved much over those of pure copper. One might have presumed that, by this time, the craftsmen should have known better than to make a double axe from soft, malleable metal. If indeed the object is correctly dated, and was intended to be used as a tool, the superior qualities of the new alloy appears not to have been generally known.

Two objects are anomalous in the content of at least one element, a stray find metal fragment from Hotnica-Vodopada (HDM 2136) with 5% silver, and a stray find borer from Gradešnica (HDM 2731) with 4.5% zinc. In both instances the find circumstances do not warrant a detailed discussion. Other obvious anomalies that affect a large fraction of the artifacts are the high concentrations of cobalt and nickel. Among all Chalcolithic metal objects from the present study, including the ones from Serbia published previously[292], there are just two cases (HDM 1974 and 1995) where the cobalt content is higher than 100 ppm; of the ones from the Late Bronze Age it is in nine out of fourteen. Similarly, the nickel content is above 0.1% in eight out of the fourteen objects while in all Chalcolithic objects from Bulgaria there is just one case (HDM 1968) and among the ones from Serbia there are only three more[293].

Isotopically, half the objects have ^{206}Pb-normalized abundance ratios higher than any observed among the artifacts from all previous periods[294] *(Fig. 25)*. There is no correlation between Sn content and isotopy but there appears to be some grouping of the objects. HDM 2037 and 2730, a sickle from Kozlodujci (BSC) and a knife from Krivodol in Western Bulgaria, respectively, are indistinguishable from one another, as are HDM 1955 and 1958 from the tell settlement at Durankulak. Note also that the three objects that are clearly *not* Sn bronzes are all "normal" in the isotopic composition of their lead.

Regional groups, production centers, and distribution zones

Based on the chemical composition of its copper artifacts the Chalcolithic of southeastern Europe has been divided into three metallurgical provinces. Two of these, in the

292 Pernicka u. a. 1993.

293 The anomalous Chalcolithic objects from Bulgaria (HDM 1968, 1974, 1995) are all ornaments from Durankulak; their lead is in all cases of the radiogenic variety (rPb with ^{208}Pb/^{206}Pb < 2.050).

294 Objects with ^{208}Pb/^{206}Pb > 2.090 are designated grouplet "h".

west the "east Serbian[295]" or "north Balkan[296]" province and, in the east, the "Thracian[295]" or "Western Black Sea[296]" province were suggested to have covered present-day Bulgaria and Serbia, with possibly a small, southwest to northeast running, corridor between the two, some small distance east of present-day Sofia. Lead isotope data presented here to some extent confirm this general picture but they require also major modifications.

A break-down of the isotope data by geographical regions reveals some distinct differences. It shows in particular that the wide spread in lead isotope abundance ratios observed for the total suite of samples is due in large part to objects from Durankulak and other west-Pontic sites like Devnja and Goljamo Delčevo *(Fig. 29)*. West Bulgaria, on the other hand, is the region where the artifacts show very little variation in their lead isotopy. Together with five heavy implements from the Middle Chalcolithic (HDM 1903, 1910, 1911, 2703, 2726), and two more from the Late Chalcolithic (HDM 2727 and 2728), the nine Final Chalcolithic objects define a particularly tight isotope grouplet (#1). The same narrow isotope field is populated by about 2/3 of the fifty-five "Intermediate period (Bodrogkeresztúr and Bubanj Hum Ia)" Serbian artifacts analyzed previously[297]. Since the trace element abundance patterns of the two sets of samples agree also, the combination gr/cl 1/2 is actually the most abundant one, with 52 cases out of the total of 335. As both material properties agree there can be but little doubt that the metal in these artifacts came from the same ore deposit(s). As we have shown above Majdanpek would be a viable source to have yielded such metal (gr/cl 1/2). But we also emphasized that this assignment might turn out not to be unique because now, with the additional Bulgarian ore data at hand, there are also some Bulgarian ores where the lead isotope abundance ratios match those of grouplet #1 *(Fig. 19)*. Such ores are the mixed sulfide variety at Čelopeč (BG-3a) and others from several occurrences in the south-east of present-day Bulgaria (BG-6b and A&V 42.7 from Zidarovo, BG-15 from Kiten, and TG 188B-4 in Turkish Thrace). Plausibility arguments make it unlikely that south-east Bulgaria should have been the source region for a kind of copper that dominates in the western parts of Bulgaria and in Serbia but is not found anywhere at the Black Sea coast or in eastern Bulgaria. For Čelopeč, some 40 km east of Sofia, this argument does not apply. Here, the ores need to be chemically more thoroughly characterized in order to evaluate their potential role. Also field studies are warranted, because today mining is from a depth that will have been inaccessible in prehistoric times. What is mined here today certainly would not qualify because the ores from Čelopeč available to us (BG-3a,b) contain way too much arsenic, antimony, gold, and selenium as to make an assignment to cluster #2 possible. Only if there should have been outcrops, and the contents in their ores of these trace elements been quite different, might Čelopeč have yielded copper with the specifications of cluster #2. For the time being we maintain the tentative identification of this source with Majdanpek.

While there is a good match between West Bulgarian Final Chalcolithic artifacts and such from the Bodrogkeresztúr-Bubanj Hum Ia period in Serbia, such agreement does not exist with objects from the Pločnik hoard that date to Vinča-Pločnik IIb. Of the 17 implements analyzed[298] none fits the combination gr/cl 1/2. Also quite distinct in their chemical and isotopic composition are the only other two Middle Chalcolithic artifacts

[295] Todorova, 1981 a.
[296] Chernykh 1992, 49.
[297] Pernicka u. a. 1993.
[298] Pernicka u. a. 1993.

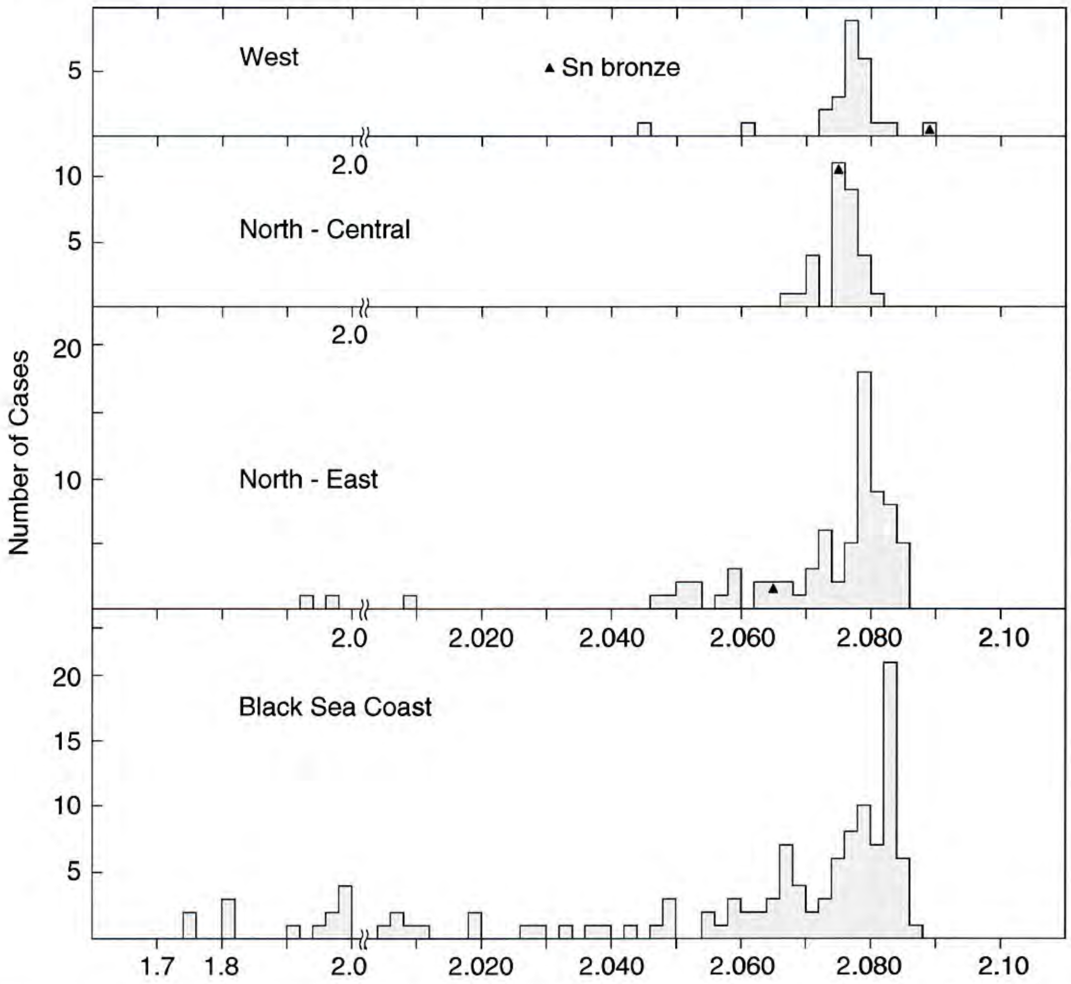

Fig. 29. $^{208}Pb/^{206}Pb$ abundance ratios of the traces of lead in Bulgarian copper artifacts from the Chalcolithic and Early Bronze Age show an extreme scatter for objects from the Black Sea coast, a very tight grouping for north-central and west Bulgaria, and an intermediate distribution for northeast Bulgaria. Quite obviously, different regions were supplied with copper from different sources.

from Serbia for which analyses are presently available, a hammer axe from Šumrakovac (HDM 1308, gr/cl 10/7) and a bracelet from Gomolava (HDM 1496, gr/cl 5/8). To the extent that agreement in lead isotopy and trace element signatures allows chronological inferences the data are in support of the contemporaneity of the Bulgarian Final Chalcolithic with the Bodrogkeresztúr-Bubanj Hum Ia period in Serbia while there is apparently no overlap with Vinča-Pločnik IIb.

Of the nine objects from western Bulgaria that are not members of the gr/cl 1/2-subset, one is a Late Chalcolithic tin bronze (HDM 2733) and three are arsenic-rich daggers from the Proto Bronze Age. For these we have no convincing source to offer. The same is true for the five remaining specimens although it is possible to find an occasional ore with an isotopic composition of its lead matching that in an artifact. Since none of these ores are sufficiently well characterized, however, such an exercise is not very meaningful at present.

For the non-gr/cl 1/2 objects from *Serbia* the situation is different in so far as they are not randomly scattered over the composition matrix but show a preference to belong to clusters #8, 5, or 1. Most extreme in this respect is cluster #8; with the exception of three Late Chalcolithic objects (HDM 1939, 1979, 2012) all its metal artifacts derive from Serbia (9) and west Bulgaria (1). Although clusters #5 and #8, just like native copper, are characterized by low concentrations of all trace elements the artifacts in these clusters in all probability are not made from native copper. For the members of cluster #5 the high nickel contents of around 100 μg/g argue against this, for the members of cluster #8 it is their high cobalt content that makes such a relation rather improbable.

Turning to the Black Sea coast and other West Pontic sites we have analyzed 114 Chalcolithic metal objects, most of them from the Late Chalcolithic *(Tab. 1)*. Of these not a single one belongs to gr/cl 1/2. Prominent instead is the combination gr/cl 2/3 that we have shown to characterize the ores from Ai Bunar and, at least as far as the lead isotopy is concerned, such from other ore occurrences in northern Thrace like, e.g., the Panagjurište district, to the west, and Prochorovo some distance east of Ai Bunar.

Also conspicuous is the large number of cases with radiogenic lead (rPb) that are almost completely absent elsewhere in Bulgaria and also in Serbia. Chemically, with but two exceptions these objects belong to cluster #1 (11 cases) or cluster #4 (17 cases). These two clusters are quite similar in their concentrations of all trace elements, however, so that a common origin of the copper in all 28 artifacts is feasible. As to possible sources of ore that would yield copper characterized by gr/cl rPb/1,4 we have pointed out already that the Eneolithic mine at Rudna Glava in Serbia does not qualify; there the concentrations in the ores of arsenic, antimony, cobalt, nickel and gold all are too high by far. We rather suggest that the region south of Burgas in the extreme southeast of Bulgaria should be considered to have yielded such copper although we emphasize that much more field work as well as more chemical and lead isotope data are required to make a really convincing case. At present we have just a single ore from Rosen (BG-7b) containing radiogenic lead[299] that points in this direction. Such an ore, however, is presumably not an exceptional outlier but must be expected to be rather common. Until the mid-seventies of this century uranium was mined in the Medni Rid region and waste rock was dumped at the nearby seashore to an extent that, because of potential radiation hazards, access to the beach is still forbidden. Moreover, the Medni Rid ridge is easily accessible, ancient mines are clearly visible and several submerged Chalcolithic settlements in this area testify to old activities there. Finally, the regional distribution of metal artifacts with this pattern (gr/cl rPb/1,4) is clearly oriented along the Black Sea coast suggesting sea-bound transport of near-coastal material. It is interesting to speculate that a large percentage of the Cucuteni-Tripolye metal from Moldavia and neighbouring regions may also derive from the Medni Rid area, in contrast to the previous assumption that it should be associated with Ai Bunar. This distribution pattern follows the same trade routes as e.g. those of Spondylus shells, namely along the Black Sea coast and the lower Danube. There is one notable difference compared with Thrace, however: So far no unworked pieces of copper minerals have been found at any northeast Bulgarian site (Ovčarovo, Poljanica, Vinica, Goljamo Delčevo, Radingrad, Omurtag, Tărgovište, Ruse, Durankulak) although most of these consist of multiphase settlements that have been completely excavated and the excavations near Stara Zagora (Azmaška Mogila, Berekecka Mogila, Čatalka) abound with such unworked ores.

The composition matrix for the artifacts from northeast Bulgaria *(Tab. 4)* shows no resemblance with that for west Bulgaria or Serbia but has some features in common with that for the Black Sea coast although there are also some distinct differences. Most notable is the almost complete absence in northeast Bulgaria of objects with radiogenic lead

[299] For the present purpose "radiogenic" lead is defined by $^{208}Pb/^{206}Pb < 2.050$.

and, vice versa, the absence of cluster #5 at the Black Sea coast that is the second-most frequent cluster in northeast Bulgaria. Moreover, objects characterized by gr/cl 2/3 (≡ Ai Bunar) are present but their fraction of the total is lower. Very much alike in northeast Bulgaria and at the Black Sea coast is the large number of artifacts with similar trace element concentrations but very dissimilar lead isotopy: they all belong to cluster #1 but are distributed among almost all isotope grouplets. Actually, northeast Bulgaria does not appear to be homogeneous in this respect because at Smjadovo, more than half way between Ruse and the Black Sea, such objects are particularly common, with 10 out of 14 falling into cluster #1 being distributed among five different grouplets. Of course, this regional distribution is suggestive of an ore source in the eastern part of Bulgaria. We note that, with the exception of grouplet #7 for which no ore source in Bulgaria has as yet been identified, isotopically fitting ores occur in southeast Bulgaria (Prochorovo, Medni Rid, Strandža, eastern Rhodope mountains). Clearly, this again points to the need for a more comprehensive study of ores from this region.

For north-central Bulgaria we find most striking that there is not a single object with the signature of Ai Bunar metal. Since most of these artifacts are *Late* Chalcolithic, however, we take this to be a chronological effect, not a regional one, suggesting that, by this time, Ai Bunar had been exhausted or for other reasons been abandoned.

As a final observation we note that there is no evidence in our data that, among the heavy implements, there should be any correlation between typology and provenance of the metal they are made from. Depending on the region where they were found different Late Chalcolithic flat axes (Kamenar) and heavy axes (Coteana) as well as Final Chalcolithic axe-adzes (Tîrgu Okna, Ariuşt, Jászladány) are all different in their grouplet/cluster characteristics *(Tab. 7)*. Regionally, however, there seems to be more coherence. From west Bulgaria a number of Jászladány axe-adzes, two of type Ariušt, a hammer axe type Mezökeresztes, a heavy axe type Pločnik and one type Sălcuţa, and also a flat axe type

Implement	Type	Region	Period	grouplet	cluster
Flat axe	Kamenar	W	Late Chalcolithic	3	1
	"	NE	"	7	4
Heavy axe	Coţeana	BSC	"	2	1
	"	NC	"	8	8
Axe-adze	Tîrgu Okna	NC	Final Chalcolithic	1	2
	"	THR	"	4	7
	Ariuşt	W	"	1	2
	"	NE	"	9	6
	Jászladány	W	"	1	2
	"	NC	"	1	7
	"	NE	"	6	–
Axe-adze	Jászladány	W	Final Chalcolithic	1	2
	Ariuşt	"	"	1	2
Hammer axe	Mezökeresztes	"	"	1	2
Heavy axe	Pločnik	"	Middle Chalcolithic	1	2
	Sălcuţa	"	"	1	2
Flat axe	Altheim-Ete	"	Proto Bronze Age	1	2
Axe-adze	Jászladány	NC	Final Chalcolithic	1	7
Flat axe	Altheim-Ete	"	Proto Bronze Age	1	7

Table 7. Material features (assignment to lead isotope grouplet and chemical cluster) of Chalcolithic implements show a dependence on find region (lower part) but not on type of implement. Apparently, several regional production centers each produced the whole typological range of objects rather specializing in the manufacture, and trading over long distances, of any particular type.

Altheim, var. Ete, are all made from metal characterized as gr/cl 1/2, while from north-central Bulgaria a Jászladány axe-adze as well as an Altheim flat axe, are both gr/cl 1/7. Apparently, there were no single centers during either the Late or the Final Chalcolithic that produced implements of a certain style or type and traded them to places all over present-day Bulgaria and Serbia. Shapes and types, once in fashion, rather were manufactured locally or at least regionally.

Summary and conclusions

One goal of the present investigation was to extend to Bulgaria our previous study[300] into the role Rudna Glava and Ai Bunar might have played in providing the ores from which the chalcolithic copper of the Balkans was eventually derived. For Rudna Glava the evidence is still negative; in Bulgaria, as in Serbia before, we again have not a single artifact that would fit the lead isotope and trace element fingerprints as they are predicted[301], and measured[302], for metal smelted from Rudna Glava ores. How ever seminal and important Rudna Glava may have been in the development of early extractive copper metallurgy in the Balkans, quantitatively it apparently was of no significance.

For Ai Bunar the situation is quite different[303]. From the Middle Chalcolithic in the third quarter of the fifth millennium B.C., i.e., from the period into which the earliest mining activities at Ai Bunar have been dated[304], we have five (out of a total of 36) artifacts where all material properties are fully compatible with a provenance of their metal from Ai Bunar. Four of these are heavy implements from Pločnik (HDM 1556, 1557, 1567, 1571), one is a bracelet from Durankulak on the Black Sea coast. Another 27 objects whose metal can be retraced to Ai Bunar are from the „metal boom" Late Chalcolithic. In this case they all originate from the Black Sea coast (19) and from north-eastern Bulgaria (8). Presumably, this regional prevalence simply reflects the fact that the geographic coverage of our Late Chalcolithic objects is very uneven, with more than 90% of them coming from north-east Bulgaria and the Black Sea coast *(Tab. 1)*. As to the type of objects that belong to the combination gr/cl 2/3, they are mostly ornaments from the graves at Durankulak and non-descript items like borers from Tell Ruse. But there are also two chisels from Tell Ruse (HDM 1921 and 2053) and two hammer axes, both of type Čoka-Varna, from Devnja (HDM 1934 and 1936).

From the Final Chalcolithic there is not a single artifact (out of a total of 82) whose metal might possibly be attributable to Ai Bunar. Of course, a trivial explanation would be that, by this time, the mines of Ai Bunar had been exhausted or been abandoned for some other reason. But it is also possible that we are again seeing a regional phenomenon, similar to that discussed for the Late Chalcolithic. It has been conjectured[305] that on the Balkans there should have been a different regional metallurgical orientation of western

300 Pernicka u. a., 1993.

301 Pernicka u. a., 1993.

302 Tylecote 1982.

303 We reemphasize that, for the time being, we have assigned to Ai Bunar all objects that show the combination (chemical) cluster #3 and (lead isotope) grouplet #2. For other grouplet #2-ores from the Srednogorje metallic zone (see above) it remains to be shown whether they also meet the trace element specifications of cluster #3 in order to make them fully competitive with Ai Bunar. Of particular interest will be the mine at Prochorovo, some 50 km west of Ai Bunar, because pottery found there dates to Karanovo V and VI suggesting that the mine might have been worked contemporaneously with Ai Bunar (Černych 1978).

304 Черных 1978; Černych 1978.

305 Todorova 1981 a.

Bulgaria and Serbia, on the one hand, and the central and eastern parts of Bulgaria, on the other, and that the metal needs of the latter were served by Ai Bunar. If this were so the absence of Ai Bunar metal during the Final Chalcolithic might simply reflect the almost complete absence of Final Chalcolithic objects from northeastern Bulgaria and the Black Sea coast. An analysis of more Late Chalcolithic artifacts from Serbia and western Bulgaria might reveal whether or not there is scientific evidence for such a regional differentiation of the ore sources. The present data just tell us that during the Middle Chalcolithic it had not yet been fully established because four of the heavy implements from Pločnik in Serbia meet all the specifications we have argued above to be characteristic of Ai Bunar copper. But then Ai Bunar might have been the only mine in operation at some early time in the Middle Chalcolithic and would, of course, have to have supplied all the metal in Bulgaria *and* Serbia.

An interesting observation concerning the importance of Ai Bunar is that from Middle Chalcolithic Pločnik as well as from Late Chalcolithic NE-Bulgaria and the Black Sea coast there are a number of artifacts that meet the lead isotope specifications of Ai Bunar (gr 2) but whose trace element contents puts them into cluster #1 rather than #3 of Ai Bunar proper. The distinction is most pronounced for arsenic; of the 28 cases in cluster #3 all but one contain more than 100 ppm arsenic while all 14 objects from cluster #1 have less than 5 ppm. Could the metal in the latter cluster possibly also come from Ai Bunar and thus make Ai Bunar an even more prominent ore source? We do not think so. It does not appear very probable that at Ai Bunar there should have been available copper ores with such a pronounced bi-modal distribution in their arsenic content. But even in case two such populations should have existed then it does not appear probable that the two groups of ore should have been kept so well separated that no mixing of the two should have occurred. More plausibly the copper of the low-As artifacts came from a different ore deposit in which case the keeping apart of low-As ores form high-As ones would have posed no problem. A likely location of such a deposit could be Prochorovo; according to Amov and Văkova[306] the lead isotopic composition of its ores is indistinguishable from that in the ores from Ai Bunar and, according to Černych[307], the concentration in the ores from Prochorovo of arsenic is very low.

An inference of the observed dichotomy of the arsenic contents in contemporaneous artifacts is, of course, that even during the Late Chalcolithic mixing of metal due to remelting cannot have been common. Moreover, if indeed the gr/cl 2/1 metal should derive from an ore deposit in the vicinity of Ai Bunar, like Prochorovo, the bimodal distribution of the arsenic concentrations also suggests that there cannot have been a joint smelting center nor one where the copper ingots were worked into the final objects. Ores and ingots rather appear to have been well separated all the way.

Another instance where objects with the same isotopy almost certainly derive from more than one ore deposit is grouplet #5. Its 18 members form a well-defined isotope field separate from all the others *(Figs. 17; 18)* and 16 of them come from Late Chalcolithic northeast Bulgaria or the Black Sea coast. Nevertheless, there are artifacts high in silver, arsenic, and antimony, all from Tell Ruse, that point to an ore source different from the one that has supplied the metal of objects low in these trace elements. As enumerated above, ores with a fitting lead isotope abundance pattern are known to occur at Čuprene in northwest Bulgaria, in the vicinity of Vraca north of Sofia, and at Medni Rid. For geographical reasons Čuprene and Vraca are unlikely candidates. Medni Rid, how-

[306] Amov/Văkova 1994.
[307] Черных 1978, 40 Fig. 21.

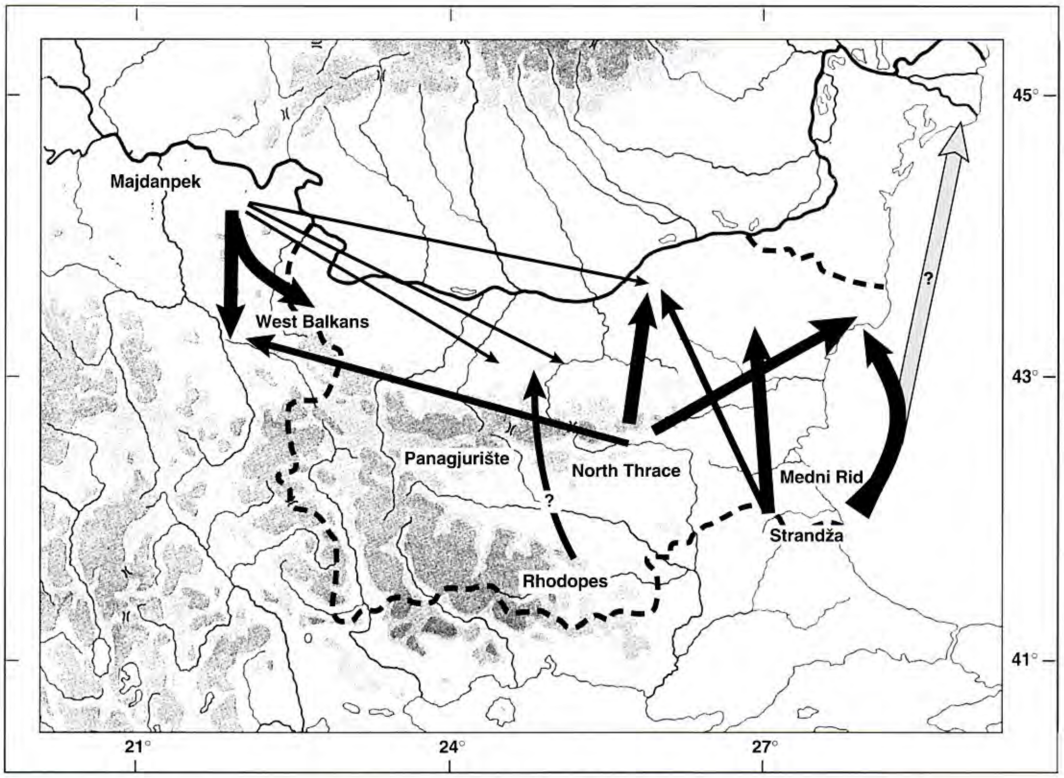

Fig. 30. Supply routes of Bulgarian and east Serbian Chalcolithic and Proto Bronze Age Copper.

ever, would well accord with the regional distribution of the artifacts made form such copper. Judged from the chemical analysis (BG-1c) such ores could very well have supplied the metal high in silver, arsenic, and antimony.

We summarize, by way of *Fig. 30*, our present evidence as to where, during the Chalcolithic, different geographical regions in Bulgaria received their metal from. Most obvious is the dominance of Ai Bunar and Medni Rid in eastern Bulgaria and of Majdanpek in west Bulgaria and in Serbia. The objects with the metal specifications of Medni Rid ores essentially come from a narrow distribution belt along the Black Sea coast in a northerly direction. There is at present no evidence that this type of copper penetrated into the interior of the country. On the other hand, we think it likely that such copper will be found among artifacts from the Cucuteni and Tripol'e cultures to the northeast where hitherto Ai Bunar appears to have been suggested to have been the sole supplier.

Since most of the eastern Bulgarian artifacts in our suite of samples date to the Late Chalcolithic, but those from the west and from Serbia to the Final Chalcolithic, the results can also be interpreted to indicate a shift with time of the importance of the mining regions, away from Medni Rid and Ai Bunar, during Late Chalcolithic times, to Majdanpek in the Final Chalcolithic. A follow-up study with emphasis on well-dated objects from regions underrepresented in the present investigation should allow one to resolve this problem. Moreover, there is still a considerable fraction of artifacts from all regions whose metal, at the present time, cannot be assigned to any mining region with any degree of certainty. What can be stated with a high degree of certainty, however, is that native copper cannot have been the starting material of these objects in particular nor, for that matter, that of any large number of Bulgarian metal artifacts in general. Although native copper is well known to have occurred in Bulgaria, to the extent that cart loads of it were delivered at copper smelters even at the beginning of this century, its trace element contents, in particular of nickel and cobalt, do not match those observed in the artifacts *(Fig. 23)*.

In addition to the major flows of material there are some minor points in *Fig. 30* that warrant comment. First, with four heavy implements from West-Bulgaria (3) and Serbia (1) we have evidence that Majdanpek apparently was exploited in the Middle Chalcolithic already, i.e., more or less contemporaneously with Ai Bunar. Where ever on the Balkans extractive copper metallurgy was invented the new technology did not take long to become disseminated. Of course, this does not argue against the possibility just mentioned that Majdanpek might have been dominant during the Final Chalcolithic only. Dominance during a later period does not preclude small-scale production during the Middle Chalcolithic already.

Another point to be noted in *Fig. 30* is the occurrence in north-central and north-east Bulgaria (at Hotnica Vodopada, Teliš-Redutite, and Tell Ruse, respectively) during the Late and Final Chalcolithic of objects with the signature of Majdanpek metal. Until now, trade along the lower Danube had only been documented upstream, via spondylus shells[308] and, possibly, gold[309]. We may now see the first evidence of what might have been exchanged. This accords well with the more general observation that, from the present study, a complex picture of early copper production and distribution emerges with several production areas with overlapping distribution zones. The model that single production centers supplied very large areas with copper, combined with more or less unidirectional trade, has to be abandoned in favor of a more complex pattern that almost resembles the modern situation. It seems that already in the Middle Chalcolithic several production centers were active in southeast Europe and at least in the present study we can find no evidence for technological differences in time and space. There is little we can contribute to the question where the knowledge of metallurgical techniques ultimately originated, whether it was introduced from other regions or was based on an indigenous development. We can, however, state that the increased need for metal during the "metal boom" of the Late and Final Chalcolithic could be entirely satisfied by the exploitation of regional ore sources that were abundantly available.

Acknowledgments

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[308] Willms 1985.
[309] Hartmann 1982.

Table A1

	Cu %	Sn [µg/g]	Pb [µg/g]	As [µg/g]	Sb [µg/g]	Co [µg/g]	Ni [µg/g]	Ag [µg/g]	Au [µg/g]	Fe [µg/g]	Zn [µg/g]	Se [µg/g]	Te [µg/g]	Hg [µg/g]
Late Neolithic and Early Chalcolithic malachite samples														
HDM 1481*	39	<34	3	16.5	1.7	173	16	<0.9	0.02	6700	250	<0.4	94	0.3
HDM 1482*	18	<44	2	24	2.7	6.1	208	1.3	0.27	24100	3000	<0.3	<3	0.2
HDM 1484*	55	<69	15	15	1.0	13	53	<0.6	0.19	4900	9500	<0.6	<6	–
HDM 1485*	37	<46	390	43	3.2	54	115	<0.8	0.132	9000	5000	<0.4	<4	–
HDM 1486*	55	<50	2	14	0.6	790	40	1.0	<0.02	1980	1370	0.6	<5	0.7
HDM 1487*	55	<23	120	77	1.1	0.8	8	4.5	0.02	1690	1300	43	<3	–
HDM 1488*	57	<23	1	6.2	0.3	6.1	29	<0.2	0.011	3500	870	0.3	<2	0.2
HDM 1489*	35	<35	50	60	2.8	7.8	196	0.9	0.43	8300	5200	<0.4	<4	–
HDM 1490*	55	<49	1	7.3	1.1	215	71	1.1	0.054	1710	2000	3.4	<3	–
HDM 1491*	51	<23	2	38	0.8	8.6	48	<0.5	0.027	52500	980	0.9	<2	0.6
HDM 1492*	56	<32	1	10	0.3	45	70	0.6	0.058	3200	4100	<0.3	<2	–
HDM 1493*	9	<45	13	17	2.6	4.9	46	6.1	0.37	25700	92	<1.8	<4	1.8
HDM 1495*	27	<54	50	95	2.8	110	149	1.3	0.43	15900	3020	<0.5	<5	40
HDM 1969	53	<10	2	7.2	1.32	1.5	13	0.7	0.026	6500	330	<0.5	<4	<0.3
HDM 1970	59	<20	2	2.9	<0.25	4.0	25	<4	0.76	960	2490	<0.6	<5	<0.3
HDM 2004	38	<40	3	45	0.90	500	152	<0.7	0.56	32000	1270	6.6	<9	<1
HDM 2209	55	<11	1	1.5	0.09	20.7	37	2.3	0.031	550	2000	<0.8	<4	1.5
HDM 2210	59	<7	<1	34	0.08	4.0	97	0.7	0.012	<40	300	<0.5	<2	1.1
HDM 2211	50	<45	16	18.0	1.90	280	157	<0.9	0.022	15700	330	<0.5	<6	1.3
HDM 2212	55	<12	7	18.9	0.40	320	228	<1.0	<0.016	2480	690	<0.9	<6	<1
Early Chalcolithic														
HDM 1904	100	<57	27	<1	68	1.0	52	540	13.2	110	6	67	18	<1
HDM 1912	100	<52	11	<2	14.7	1.5	130	530	10.2	<130	<7	70	<38	<1
HDM 1975	100	<90	250	770	770	2.0	39	420	8.1	850	<16	83	<70	<1
HDM 2005	88	<25	20	6.3	0.44	11.1	20	370	4.6	150	4	28	<10	<1
Middle Chalcolithic														
HDM 1903	100	<15	17	0.9	4.9	1.0	85	12	0.212	60	3	0.5	<2	<1
HDM 1910	100	<19	4	0.8	7.6	1.5	72	39	0.79	480	3	3.1	11	0.1
HDM 1911	100	<8	6	0.8	2.68	1.0	84	12	0.35	<50	7	0.8	<6	<0.3
HDM 1940	100	<52	62	1.3	1.55	1.5	41	850	12.0	<120	<7	320	124	<1

Table A1 (continued)

	Cu %	Sn [µg/g]	Pb [µg/g]	As [µg/g]	Sb [µg/g]	Co [µg/g]	Ni [µg/g]	Ag [µg/g]	Au [µg/g]	Fe [µg/g]	Zn [µg/g]	Se [µg/g]	Te [µg/g]	Hg [µg/g]
HDM 1976	100	<28	8	<1	29	5.6	150	150	29.2	<130	5	38	<15	<1
HDM 1977	99	<36	1020	2490	590	1.0	<15	620	20.8	<200	8	177	32	<1
HDM 2011	98	<35	510	3400	38	5.0	73	244	0.248	<450	34	56	<18	<1
HDM 2025	94	<18	90	340	15.2	30	284	450	0.61	<110	172	100	6	<1
HDM 2026	100	<20	90	360	15.2	32	310	470	0.65	<170	174	102	<11	<1
HDM 2041	96	<115	60	127	16.7	2.0	153	3400	53	2000	39	204	89	<1
HDM 2049	83	<43	90	15.3	6.0	5.5	23	147	14.8	2800	38	135	39	<1
HDM 2064	100	<10	20	<2	1.80	3.5	125	54	0.83	<80	6	42	<4	<1
HDM 2080	100	<35	42	10.9	3.1	39	890	11	0.46	5300	21	<1	<16	<1
HDM 2124	100	<95	40	92	2.75	11.2	67	360	2.94	610	23	274	196	<1
HDM 2702	103	<7	<1	<0.8	0.11	12.6	34	9	0.083	35	17	2.5	<3	0.2
HDM 2703	102	<10	5	10.9	2.15	*(0.1)*	52	12	0.275	45	5	1.0	<6	<0.2
HDM 2726	107	<12	25	6.3	5.1	*(0.1)*	89	16	0.47	<90	9	1.4	1	<0.3
Late Chalcolithic														
HDM 1905	100	<70	18	<2	4.9	1.5	104	1350	7.8	<150	<9	110	13	<1
HDM 1915	99	<25	33	2.0	2.69	0.5	157	1200	89	<160	<9	58	<23	<1
HDM 1920	100	<20	18	4.8	1.09	11.6	230	79	12.9	35	<4	30	11	<1
HDM 1921	100	<40	390	246	1210	1.5	<19	690	14.2	<270	<15	430	<43	<1
HDM 1922	100	<8	7	4.5	1.12	19.9	93	141	20.2	<10	5	14.6	<12	<1
HDM 1923	100	<60	250	<2	169	1.0	<32	620	5.1	<430	<28	106	<16	<1
HDM 1924	98	<42	70	3.1	27.4	1.0	10	560	13.3	110	<6	55	<18	<1
HDM 1925	100	<35	6	<2	1.26	0.5	14	1480	42	<170	<8	72	35	<1
HDM 1926	96	<40	24	<1	1.34	<0.5	66	292	11.1	<80	<4	33	37	<1
HDM 1927	97	<28	20	<1	0.50	<0.5	<41	240	4.1	<80	<4	141	<185	<1
HDM 1928	99	<40	2770	57	760	1.0	14	490	4.1	<180	20	276	23	<1
HDM 1929	100	<17	3	<1	0.68	0.5	8	252	10.5	<80	3	92	20	<1
HDM 1930	100	<13	6	<1	1.64	2.0	56	114	29.8	<60	7	9.3	<5	<1
HDM 1931	99	<12	83	<1	11.0	0.5	36	570	7.1	<390	<28	74	<16	<1
HDM 1932	100	<17	66	16.7	8.1	0.5	<4	1230	5.3	<160	<8	4.2	<15	<1
HDM 1933	100	<9	63	<2	128	0.5	21	271	4.6	45	<6	133	<11	<1
HDM 1934	100	<43	820	510	760	2.0	14	490	3.9	<180	83	300	28	<1

Table A1 (continued)

	Cu %	Sn [µg/g]	Pb [µg/g]	As [µg/g]	Sb [µg/g]	Co [µg/g]	Ni [µg/g]	Ag [µg/g]	Au [µg/g]	Fe [µg/g]	Zn [µg/g]	Se [µg/g]	Te [µg/g]	Hg [µg/g]
HDM 1935	100	<17	3	2.3	1.66	2.5	72	500	5.6	<100	<6	52	<15	<1
HDM 1936	100	<23	920	380	275	2.5	17	360	4.8	<120	<8	81	14	<1
HDM 1937	100	<10	10	<1	4.3	2.5	113	450	0.87	<90	<6	47	<10	<1
HDM 1938	100	<10	12	24.9	7.5	11.4	179	640	1.11	<110	13	72	8	<1
HDM 1939	100	<5	2	1.3	0.59	1.0	7	35	0.072	<30	6	0.7	2	0.2
HDM 1942	96	<80	2000	2.3	6.3	1.5	22	1210	2.26	<130	55	750	390	4.5
HDM 1943	82	<30	300	39	5.8	9.9	77	19	44	205	13	32	10	2.8
HDM 1944	100	<12	540	0.7	0.97	9.7	82	104	19.4	<60	6	21.3	<23	<1
HDM 1945	100	<23	310	0.6	1.10	8.6	71	111	20.0	30	6	26.4	7	<1
HDM 1946	100	<48	5	1.4	0.73	2.5	46	193	9.8	<110	<15	26.4	<9	<1
HDM 1947	95	<35	80	188	9.2	98	152	233	4.78	720	18	102	14	<1
HDM 1948	100	<35	950	5.9	5.7	15.3	162	191	10.3	<100	93	27.6	<5	<1
HDM 1949	100	<37	80	132	3.4	21.0	68	142	19.5	120	14	12.6	<8	<1
HDM 1956	100	<40	85	560	142	0.5	57	440	0.38	<150	8	55	<33	<1
HDM 1959	88	<30	80	37	82	4.5	31	400	12.8	130	5	114	33	<1
HDM 1960	100	<80	760	8700	217	5.0	78	430	0.57	700	94	24.4	<53	<1
HDM 1961	96	<20	39	<2	100	1.0	12	430	7.8	180	<10	222	<20	<1
HDM 1962	73	<110	15	78	1.56	8.8	34	1520	31	2500	24	19.3	<16	<1
HDM 1963	100	<115	240	3200	1010	1.5	<23	810	10.8	<370	<15	89	<52	<1
HDM 1964	100	<115	510	8400	1490	2.5	<23	720	11.8	<360	<15	57	<55	<1
HDM 1965	100	<50	33	310	4.2	40	80	156	18.5	430	22	30	<23	<1
HDM 1967	100	<1170	26	173	320	5.6	75	520	13.1	1100	<90	60	<50	<1
HDM 1968	100	<53	23	5	1.96	90	2400	180	27.2	15300	19	43	<25	<1
HDM 1971	100	<10	82	1.9	1.53	4.5	36	460	4.3	<140	<7	46	<8	<1
HDM 1972	100	<10	270	2240	72	1.0	18	171	0.075	<90	54	17.8	<5	<1
HDM 1973	100	<23	20	33	1.71	31	83	165	8.4	<140	<10	38	<10	<1
HDM 1974	100	<77	25	820	7.5	660	250	230	12.9	6200	34	22.5	16	<1
HDM 1978	100	<48	710	10100	1820	1.0	<20	800	11.6	<260	<15	68	<24	<1
HDM 1979	100	<25	11	<4	0.16	7.9	26	4	0.078	<180	7	0.7	<9	<1
HDM 1980	98	<20	13	5.4	4.8	12.5	82	330	6.8	<80	9	68	<18	<1
HDM 1981	99	<60	4	<5	8.0	1.0	107	910	36	<120	40	16	41	<1
HDM 1982	100	<40	<1	4.4	7.1	1.0	26	640	9.5	<140	<8	40	<38	<1

Table A1 (continued)

	Cu %	Sn [µg/g]	Pb [µg/g]	As [µg/g]	Sb [µg/g]	Co [µg/g]	Ni [µg/g]	Ag [µg/g]	Au [µg/g]	Fe [µg/g]	Zn [µg/g]	Se [µg/g]	Te [µg/g]	Hg [µg/g]
HDM 1983	110	<45	92	<2	0.27	0.5	48	240	13.1	<190	16	13.8	<30	<1
HDM 1984	110	<120	200	234	1550	3.0	47	880	20.0	<500	<25	310	<68	<1
HDM 1985	100	<65	23	<6	7.4	2.5	56	590	11.6	<320	<17	62	<40	<1
HDM 1986	97	<24	47	66	120	7.5	57	330	100	<90	<5	50	<50	<1
HDM 1987	100	<60	57	2.6	7.6	6.1	61	460	35	<150	8	181	<69	<1
HDM 1988	100	<69	17	<4	1.50	19.4	128	330	60	<350	<39	36	<38	<0.2
HDM 1989	100	<48	300	3.9	122	0.5	<41	440	3.4	<280	<31	235	<26	<1
HDM 1990	100	<66	200	1930	310	0.5	<61	720	13.0	390	<38	185	<37	<0.7
HDM 1991	100	<68	86	56	370	0.5	<63	720	16.9	<310	<41	81	<37	<0.3
HDM 1992	100	<65	1140	5100	830	<0.5	<61	580	14.5	<350	<50	256	<35	<1
HDM 1993	100	<51	33	12.8	3.4	35	350	144	56	<200	1	11.8	<28	<0.1
HDM 1994	100	<42	3	<3	0.60	4.0	156	580	34	<370	<27	33	<22	<0.1
HDM 1995	100	<33	28	53	7.8	107	64	180	3.0	2640	<31	96	<17	<0.4
HDM 1996	100	<118	480	240	1500	2.0	<105	650	7.9	<500	<87	78	<64	2.9
HDM 1997	100	<27	930	9.4	5.7	14.6	242	234	37	<150	20	36	26	<0.2
HDM 1998	100	<37	390	3.6	112	<0.5	<34	420	3.0	<260	7	221	<21	<1.3
HDM 1999	100	<29	100	<2.5	63	0.5	22	350	9.1	<200	<17	67	<15	<0.2
HDM 2000	83	<25	<1	162	2.20	7.2	60	99	25.0	2300	16	20.1	<14	<0.1
HDM 2001	90	<30	800	620	136	24.7	53	350	11.6	340	9	9.9	<13	<1
HDM 2002	95	<50	17	640	12.4	78	98	500	13.2	290	14	66	<18	<1
HDM 2003	84	<15	20	3.7	0.24	3.0	15	164	22.6	420	7	29.0	<13	<1
HDM 2006	84	<23	<1	9.0	23.4	0.5	48	1640	12.9	290	7	33	<17	<1
HDM 2007	89	<25	880	720	650	1.0	<12	500	11.1	<160	<6	56	<18	<1
HDM 2008	93	<28	23	41	31	6.1	53	330	7.4	50	<5	105	<15	<1
HDM 2009	92	<13	13	1.3	2.91	1.5	96	240	33	<50	20	47	<12	<1
HDM 2010	97	<30	90	31	3.5	6.6	50	157	2.95	<160	30	26.9	<24	<1
HDM 2012	97	<25	190	2.5	0.33	1.5	5	21	0.22	<150	16	1.7	<12	<1
HDM 2013	95	<55	4490	1.0	0.21	1.5	27	720	2.63	<200	8	400	52	<1
HDM 2014	95	<18	20	266	3.4	36	74	167	17.4	3900	24	27.8	7	<1
HDM 2015	100	<120	2	13.5	11.0	2.0	<27	600	7.7	<430	<20	53	86	<1
HDM 2016	100	<40	<1	0.9	1.01	2.0	<12	390	9.8	<200	<10	55	<18	<1
HDM 2017	100	<30	2	0.9	0.44	2.5	33	181	11.4	<140	<7	31	<12	<1

Table A1 (continued)

	Cu %	Sn [µg/g]	Pb [µg/g]	As [µg/g]	Sb [µg/g]	Co [µg/g]	Ni [µg/g]	Ag [µg/g]	Au [µg/g]	Fe [µg/g]	Zn [µg/g]	Se [µg/g]	Te [µg/g]	Hg [µg/g]
HDM 2018	100	<37	27	20.3	16.6	26.2	93	390	18.8	<170	10	61	27	<1
HDM 2019	100	<40	12	1.1	2.73	1.5	152	350	27.1	<190	47	37	<17	<1
HDM 2020	100	<60	5	<2	2.01	2.0	124	920	25.8	<240	30	38	30	<1
HDM 2021	100	<28	20	<1	19.0	3.5	39	217	2.27	<130	<6	64	12	<1
HDM 2022	100	<40	50	<1	52	35	30	266	15.4	<170	<8	98	<13	<1
HDM 2023	100	<28	20	<3	2.45	6.4	118	280	40	<180	13	52	<20	<1
HDM 2024	100	<38	23	<2	2.31	3.5	100	276	37	<160	9	55	14	<1
HDM 2027	100	<85	2010	6300	1310	1.0	<25	860	9.8	<420	<18	279	<73	<1
HDM 2028	88	<30	32	9.2	1.89	10.3	174	158	39	360	16	63	10	<1
HDM 2029	100	<65	170	1100	320	3.5	95	1980	15.4	<390	<20	33	55	<1
HDM 2030	89	<65	440	146	370	1.8	18	620	10.9	870	<12	249	27	<3
HDM 2031	95	<30	12	22.6	1.21	5.0	105	205	55	1120	10	26.8	<13	1.2
HDM 2032	98	<22	16	<2	5.5	1.5	294	630	7.9	<300	<7	80	13	<1
HDM 2034	98	<40	6	0.7	1.61	2.0	40	225	4.6	<100	6	227	113	<1
HDM 2035	97	<17	44	<2	27.2	1.0	22	330	8.7	<80	20	61	8	<1
HDM 2036	97	<30	45	9.9	9.3	13.0	132	215	100	<110	4	72	<17	<1
HDM 2039	96	<110	90	3500	870	3.5	58	5800	0.96	<700	<35	14.2	<65	<1
HDM 2040	100	<28	260	<1	2.68	2.5	22	300	4.6	<150	20	282	<30	<1
HDM 2043	100	<30	3	<2	14.3	3.0	143	520	0.36	<280	<15	117	<20	<1
HDM 2044	77	<70	2000	<4	750	1.5	<25	340	13.8	<360	19	113	<62	<1
HDM 2045	100	<55	110	330	450	2.5	18	670	26.3	<280	<15	104	<32	<1
HDM 2047	100	<130	33	<4	6.8	1.5	154	2600	14.5	<700	<33	87	<65	<1
HDM 2050	100	<25	150	32	87	5.5	117	126	0.97	<210	23	42	18	<1
HDM 2051	93	<38	43	8600	197	2.0	214	2840	15.9	<350	<18	18.8	<36	<1
HDM 2052	98	<40	70	9400	236	3.5	271	2560	14.4	<350	12	19.6	<37	<1
HDM 2053	99	<100	90	117	440	2.5	<17	430	9.6	<230	<12	490	103	<1
HDM 2054	100	<28	750	<5	3.1	1.5	81	600	17.8	<240	6	59	32	<1
HDM 2055	100	<100	17	<12	860	1.5	510	13100	16.2	<900	52	8.2	<68	<1
HDM 2056	99	<9	8	12.7	1.75	5.9	105	25	0.34	360	18	38	<3	<1
HDM 2057	100	<63	225	2430	197	3.0	<17	650	9.2	280	26	140	52	<1
HDM 2058	100	<10	36	1.3	3.1	2.0	64	11	0.29	<70	17	0.7	<5	0.1
HDM 2059	99	<12	2	4.1	0.78	1.5	41	64	1.15	100	3	152	9	<1

Table A1 (continued)

	Cu %	Sn [µg/g]	Pb [µg/g]	As [µg/g]	Sb [µg/g]	Co [µg/g]	Ni [µg/g]	Ag [µg/g]	Au [µg/g]	Fe [µg/g]	Zn [µg/g]	Se [µg/g]	Te [µg/g]	Hg [µg/g]
HDM 2060	100	<30	480	<2	1.53	2.0	17	852	5.3	190	11	15.0	<33	<1
HDM 2061	100	<10	3	<3	0.63	7.3	204	42	0.67	140	89	38	<4	<1
HDM 2063	98	<15	9	4.5	0.58	16.4	210	22	0.040	235	22	94	<7	<1
HDM 2065	100	<17	60	138	19.6	18.7	109	39	1.40	215	58	92	<6	<1
HDM 2067	100	<60	8	1.4	2.92	1.5	57	2210	8.5	<160	<10	129	<33	<1
HDM 2068	97	<40	<1	7.1	1.58	2.0	<8	340	12.8	410	<5	370	27	<1
HDM 2070	99	<30	75	1.7	122	1.5	34	600	10.2	<200	<10	134	<13	<1
HDM 2071	98	<100	230	4.7	249	3.5	11	560	8.7	<250	<9	98	<29	<1
HDM 2072	96	<40	26	167	2.71	5.3	65	320	2.76	115	10	284	205	<1
HDM 2073	93	<17	40	<2	10.9	2.0	125	540	4.6	<80	8	47	<7	<1
HDM 2074	97	<41	30	1.7	1.35	2.5	12	840	11.7	350	11	25.5	<42	<1
HDM 2075	100	<26	20	44	13.9	8.3	143	57	0.126	285	42	202	<18	<1
HDM 2076	95	<35	270	106	710	3.0	<13	600	8.0	<160	28	148	<17	<1
HDM 2077	100	<20	32	6.5	6.5	2.0	26	221	13.0	<70	4	32	19	<1
HDM 2078	92	<33	40	3.5	12.3	4.0	64	2000	10.3	<150	22	92	15	<1
HDM 2079	100	<63	8	1.1	7.9	2.0	38	510	7.9	<160	9	130	47	<1
HDM 2081	100	<60	10	<2.5	11.5	2.5	25	1600	12.4	<320	11	109	<43	<1
HDM 2082	98	<85	7	<2.5	3.8	3.0	132	2810	6.3	<420	68	85	44	<1
HDM 2083	97	<15	4	<8	4.9	1.5	23	270	17.4	65	<15	24.4	8	<1
HDM 2084	100	<75	30	196	2.26	6.4	68	330	3.3	<150	32	284	191	<1
HDM 2085	100	<13	2	1.7	0.28	5.5	118	16	0.098	<70	15	45	<12	<1
HDM 2088	94	<40	20	<3	11.1	1.0	37	1410	8.6	<230	29	113	31	<1
HDM 2089	88	<13	1	8.2	1.02	16.0	101	280	5.8	185	14	99	<9	<1
HDM 2090	89	<30	3	147	9.2	2.0	64	750	4.4	<120	<6	80	<20	<1
HDM 2091	93	<17	11	6.9	1.24	1.5	166	210	33	55	6	20.4	<16	0.6
HDM 2092	66	<135	3	24.8	164	2.5	186	11600	4.2	1490	49	17.8	<45	<1
HDM 2093	86	<20	10	19	25.4	2.5	97	670	23.2	95	19	60	<19	1.2
HDM 2096	100	<90	34	36	41	2.5	74	900	5.6	<270	<13	217	70	<1
HDM 2097	98	<75	8	<9	13.4	27.2	121	2580	760	350	22	119	<110	<1
HDM 2098	100	<75	1880	3700	1750	2.0	36	540	22	<290	<13	198	<70	<1
HDM 2099	100	<75	56	<5	12.9	4.0	72	2210	11.3	<400	<18	101	<55	<1
HDM 2100	95	<62	10	10.5	8.3	1.5	71	1400	50	<1500	<80	10.0	<34	<0.1

Table A1 (continued)

	Cu %	Sn [µg/g]	Pb [µg/g]	As [µg/g]	Sb [µg/g]	Co [µg/g]	Ni [µg/g]	Ag [µg/g]	Au [µg/g]	Fe [µg/g]	Zn [µg/g]	Se [µg/g]	Te [µg/g]	Hg [µg/g]
HDM 2101	100	<9	1	<6	0.44	7.8	101	13	0.096	100	12	22.8	<3	<1
HDM 2102	100	<25	2	<15	3.3	2.0	20	198	21.9	<90	72	88	12	<1
HDM 2103	84	<48	1	32	2.30	0.5	<29	234	48	1890	<40	120	<27	<0.6
HDM 2104	100	<17	22	<14	9.5	5.2	145	60	1.63	80	11	17.6	<7	<1
HDM 2119	100	<42	15	<7	9.7	1.5	<17	930	8.6	235	<13	42	29	<1
HDM 2120	100	<37	22	<7	1.29	2.0	35	950	8.3	300	<10	34	22	<1
HDM 2121	78	<29	140	48	137	0.5	<26	420	86	7800	21	137	26	<0.5
HDM 2122	100	<33	<1	31	13.5	2.0	<26	300	72	470	<19	310	<15	1
HDM 2123	86	43	390	3400	860	1.5	<44	890	54	<530	21	258	44	0.8
HDM 2125	100	<32	<1	990	162	15.8	117	740	38	7800	<24	46	23	<0.2
HDM 2126	99	<35	20	2740	14.4	2.0	177	930	14.4	2300	<26	52	21	<0.3
HDM 2127	100	<50	3	<10	4.0	1.5	169	119	21.1	3000	7	60	19	<1
HDM 2128	100	<35	320	84	450	4.0	42	460	6.6	460	<12	122	16	<1
HDM 2129	100	<25	180	<7	0.48	1.0	33	470	15.1	<150	<8	41	19	<1
HDM 2130	100	<28	<1	<7	19.3	4.0	360	300	6.5	720	10	490	<45	<1
HDM 2137	72	<40	5	11.5	0.65	17.9	132	145	20.5	8700	19	58	<35	<1
HDM 2138	54	<28	13	69	2.13	9.9	99	170	8.7	61000	28	18.0	<32	<1
HDM 2139	100	<34	12	2710	31	1.5	111	550	13.0	<370	<17	101	52	<0.4
HDM 2140	100	<28	16	13600	149	1.5	25	111	4.7	<340	<14	22.7	25	<0.1
HDM 2141	100	<52	110	2790	760	3.5	<40	800	14.1	<540	<29	95	28	<0.3
HDM 2142	100	<83	260	20.2	330	4.0	30	770	11.4	<230	<10	120	<55	<0.6
HDM 2143	63	88	8	257	380	1.0	<110	9500	14.3	1400	<73	154	209	0.5
HDM 2144	99	35	12	<1	0.90	0.5	52	271	5.5	<120	6	146	60	<0.6
HDM 2145	100	35	14	<2	1.00	0.5	42	290	5.7	<170	<12	154	62	<0.9
HDM 2146	98	<23	16	1.3	1.00	6.0	227	124	1.98	135	<11	66	12	<0.1
HDM 2150	98	26	17	22.8	37	1.0	159	1260	8.9	<380	<9	2.1	19	<0.9
HDM 2707	104	<26	23	26.7	52	0.6	144	1410	6.3	<190	11	48	<30	1.4
HDM 2709	98	<30	7	25.0	9.4	<0.5	31	66	0.84	290	10	360	273	<3
HDM 2710	106	<25	30	<4	0.85	0.5	22	370	9.4	<150	55	100	<26	<1
HDM 2711	92	<35	440	<3	11.4	2.9	125	289	10.9	<120	53	87	14	<1
HDM 2712	101	<50	3	43	12.4	*(0.4)*	26	276	6.2	<130	20	176	81	<2
HDM 2713	92	<50	2	41	6.9	*(0.1)*	16	290	6.6	220	51	221	108	<2

Table A1 (continued)

	Cu %	Sn [µg/g]	Pb [µg/g]	As [µg/g]	Sb [µg/g]	Co [µg/g]	Ni [µg/g]	Ag [µg/g]	Au [µg/g]	Fe [µg/g]	Zn [µg/g]	Se [µg/g]	Te [µg/g]	Hg [µg/g]
HDM 2714	94	<32	2	<3	6.4	*(0.2)*	28	790	9.4	<150	58	164	36	<2
HDM 2715	91	<15	40	287	227	<0.5	19	254	6.3	<110	19	20.9	<5	<0.6
HDM 2716	95	<40	2	<3	1.69	*(0.2)*	28	400	9.8	<100	30	164	15	<2
HDM 2717	101	<35	48	1.4	22.7	*(0.4)*	69	450	13.7	<165	39	71	7	<1
HDM 2718	103	<63	33	2.8	5.9	<0.5	40	520	11.0	<150	65	188	78	<2
HDM 2719	101	<38	4	<4	200	<0.5	24	610	8.6	<160	17	222	38	<2
HDM 2720	87	82800	450	3300	234	99	182	153	18.9	4200	182	24.8	31	1.5
HDM 2721	100	<60	2	2.7	5.7	1.9	24	550	16.6	<275	38	77	3	1.8
HDM 2722	97	<60	3420	4700	1500	*(0.2)*	16	650	38	<350	39	410	45	<5
HDM 2725	101	<85	110	142	145	*(0.1)*	180	980	149	<170	10	18.2	18	0.8
HDM 2727	109	<20	54	150	22.6	6.7	82	96	4.4	<180	11	7.7	5	0.7
HDM 2728	108	<30	110	<5	1.57	1.3	23	49	2.02	<240	16	2.3	<10	<0.5
HDM 2733	88	28600	1260	2380	3000	19.5	560	990	12.9	<420	30	24.1	<20	<2
Final Chalcolithic														
HDM 1901	100	<15	10	79	3.1	1.0	26	30	0.38	65	17	3.1	3	<1
HDM 1906	100	<19	30	95	5.6	4.0	94	22	0.35	55	19	0.9	<3	0.2
HDM 1907	84	<13	33	53	5.8	0.5	63	16	0.34	60	8	1.7	2	<1
HDM 1908	100	<17	9	35	5.4	1.0	68	9	0.23	75	7	0.4	<2	0.2
HDM 1909	100	<15	10	53	4.1	1.0	73	15	0.43	110	10	0.8	4	0.2
HDM 1913	100	<9	40	<0.8	4.0	3.5	78	35	4.8	100	19	35	4	<1
HDM 1914	106	<9	230	510	63	1.0	49	68	1.26	35	14	9.1	<9	<1
HDM 1916	100	<20	33	128	12.4	8.9	109	75	1.60	105	30	2.2	<15	0.4
HDM 1917	100	<25	55	5.0	5.6	1.0	73	27	0.45	40	8	0.5	<5	<1
HDM 1919	100	<7	<1	710	0.9	1.0	4	2	0.56	50	3	0.6	<4	<1
HDM 1941	100	<9	13	5400	28.6	2.5	91	87	1.54	<90	4	8.5	4	<1
HDM 2105	100	<57	8	860	79	3.0	116	155	23.6	<920	<53	28.1	<30	1.2
HDM 2106	100	<41	4	103	5.3	2.5	147	280	0.88	<760	<49	66	<22	<0.3
HDM 2107	100	<18	60	78	19.9	2.0	91	430	6.0	<90	17	15.6	<8	<1
HDM 2108	100	40	40	350	26.1	30	169	52	24.7	<320	40	54	7	<0.2
HDM 2109	100	<32	13	<12	7.7	2.5	142	920	16.3	<140	<7	51	25	<0.1
HDM 2110	100	<23	16	<10	43	3.0	42	650	14.4	<100	<7	18.7	<13	<0.1

Table A1 (continued)

	Cu %	Sn [µg/g]	Pb [µg/g]	As [µg/g]	Sb [µg/g]	Co [µg/g]	Ni [µg/g]	Ag [µg/g]	Au [µg/g]	Fe [µg/g]	Zn [µg/g]	Se [µg/g]	Te [µg/g]	Hg [µg/g]
HDM 2111	100	<23	14	7.6	4.8	1.5	51	32	1.13	<150	<16	6	<8	<0.1
HDM 2112	100	<16	5	4.1	4.2	1.0	115	28	1.20	<100	<10	27.2	5	<0.1
HDM 2113	100	<28	27	3	1.6	0.5	53	144	1.71	<190	<19	9.3	<10	<0.1
HDM 2114	98	137	220	4.5	32	1.0	430	1290	238	<540	<27	49	21	<0.2
HDM 2115	99	<21	590	430	44	4.5	73	129	4.3	<230	11	80	14	<0.3
HDM 2116	100	<14	2	2.8	7.9	2.5	164	20	0.79	<150	49	48	<5	<0.2
HDM 2117	100	<53	46	900	96	2.0	400	3200	9.3	<690	<37	59	33	<0.3
HDM 2118	99	<15	20	20.1	2.4	22.3	181	42	2.20	940	50	68	<6	<0.3
HDM 2213	100	151	–	193	15.9	85	95	32	1.01	<110	20	4.6	5	3.7
HDM 2705	100	<10	20	15.0	3.7	<0.5	88	11	0.22	<68	14	0.4	<7	<0.2
HDM 2706	106	<9	26	4.9	2.3	*(0.1)*	85	12	0.22	<100	10	0.5	<7	<0.2
HDM 2708	105	<18	5	18.1	24.1	3.5	610	299	10.7	<135	8	24.7	18	<0.6
HDM 2724	104	<15	110	53	26.9	<0.5	81	99	0.57	<100	10	7.3	6	<5
Proto Bronze Age														
HDM 1902	100	<37	37	14800	60	1.0	57	112	2.74	245	3	22.6	17	<1
HDM 1918	100	<105	8	22000	168	2.0	227	245	6.6	<300	10	28	18	2
HDM 1952	65	<260	1780	3800	3400	109	1200	48	11.4	800	28	5.2	<73	<1
HDM 1954	100	<110	20	<6	187	5.0	121	810	14.3	<400	<16	73	<40	<1
HDM 1966	98	<900	11	84000	145	6.9	1300	<44	5.9	<4400	<260	80	<200	–
HDM 2134	100	<110	20	37000	39	22.7	73	126	3.3	340	67	34	<35	<1
HDM 2135	99	<50	6	49000	5.8	2.0	<15	138	1.01	<210	4	4.3	<48	<1
HDM 2147	91	600	2450	44	12.3	34	63	18000	<1.50	940	109	<9	153	9
HDM 2148	91	23500	890	13000	430	26.9	296	500	35	2120	91	7	<21	7.7
HDM 2149	99	<23	11	259	18.8	1.0	214	228	9.2	<330	<13	47	<15	5.6
HDM 2701	109	<18	56	2	2.5	*(0.4)*	30	23	0.57	<100	8	4.0	<20	<0.4
HDM 2736	96	<100	460	18500	5000	99	25000	29	0.68	840	350	9.1	<53	<3
HDM 2737	109	<150	76	13200	390	5.7	134	17	5.0	<360	93	74	117	<2
HDM 2738	100	77	700	25800	273	14.1	134	400	4.8	11500	14	<2	<52	<1
HDM 2740	112	<80	4	80	8.7	<0.5	226	4400	36	<570	78	16.8	21	<2
HDM 2741	113	<85	60	22700	390	*(0.3)*	240	450	18.8	6700	22	12.6	<45	<4
HDM 2742	110	<130	8	550	37	1.6	85	19000	13.0	<870	94	26.7	<100	<5

Table A1 (continued)

	Cu %	Sn [μg/g]	Pb [μg/g]	As [μg/g]	Sb [μg/g]	Co [μg/g]	Ni [μg/g]	Ag [μg/g]	Au [μg/g]	Fe [μg/g]	Zn [μg/g]	Se [μg/g]	Te [μg/g]	Hg [μg/g]
Early Bronze Age														
HDM 2704	101	<70	120	8000	380	0.6	83	17	2.72	<135	21	5.6	73	<0.6
HDM 2739	104	<70	2	14800	104	*(0.1)*	520	238	4.7	<350	18	28.9	<60	0.9
Late Bronze Age														
HDM 1955	98	11000	3000	2790	8600	184	3200	1770	9.6	<800	63	<5	<170	<2
HDM 1957	90	79000	1820	3600	2500	154	2320	550	22	750	85	<55	<120	<1
HDM 1958	92	26700	1790	3400	1810	500	3300	930	11.7	390	113	<40	<65	<4
HDM 2033	86	57000	1000	5300	1660	133	1910	300	17.2	3800	24	11.9	<55	<1
HDM 2037	96	73000	330	960	470	910	1190	230	4.3	2900	250	22	<22	1.3
HDM 2038	100	520	100	2800	199	90	149	133	3.8	2310	22	119	45	<1
HDM 2131	92	<43	4	1430	32	<0.5	360	1190	18.6	<500	<25	28.2	17	<1
HDM 2132	100	48000	330	2290	970	330	1430	920	26.2	<600	59	18.2	26	1
HDM 2133	100	<75	135	14800	400	1.5	520	800	20.5	1060	<13	22.7	61	<1
HDM 2136	78	54000	250	1650	610	140	410	50000	390	<4500	290	<30	<25	<1
HDM 2730	96	123000	370	1970	2700	310	3000	640	7.3	1750	52	8.4	<60	<2
HDM 2731	94	21900	570	1400	350	54	380	1120	13.8	2130	45000	54	<30	<4
HDM 2732	85	123000	73	260	118	7	45	93	45	1350	29	17.6	<16	1.2
HDM 2734	98	31300	1740	1530	5700	210	3600	1730	8.8	<850	52	<7	<40	<4
Classical Period														
HDM 1953	93	63000	20930	1410	300	1850	2560	740	34	<580	230	16.7	<35	<1
Unstratified or uncertain														
HDM 1950	91	<830	980	860	2110	2.5	100	2470	15.3	<400	<22	38	<53	2.1
HDM 1951	100	<70	420	259	92	2.0	35	620	1.96	<150	12	81	16	<1
HDM 2042	66	<40	4	99	11.0	49	33	620	1.54	12500	38	34	17	1.1
HDM 2046	86	73000	570	3500	2810	164	2830	320	32	3100	43	9.7	<110	<1
HDM 2048	84	1750	1210	1230	430	90	1270	360	114	1700	191000	4.8	<46	–
HDM 2062	100	<10	6	10	2.33	10.7	178	30	0.31	90	4	350	13	<1
HDM 2066	100	22100	550	630	690	126	340	320	264	3000	18700	51	<25	<1

Table A1 (continued)

	Cu %	Sn [μg/g]	Pb [μg/g]	As [μg/g]	Sb [μg/g]	Co [μg/g]	Ni [μg/g]	Ag [μg/g]	Au [μg/g]	Fe [μg/g]	Zn [μg/g]	Se [μg/g]	Te [μg/g]	Hg [μg/g]
HDM 2069	100	81	790	830	530	6.9	127	400	7.8	<300	10	95	<37	<1
HDM 2086	88	<105	710	1450	1450	0.5	<16	910	75	1810	10	370	68	<1
HDM 2087	85	<53	1910	93	1100	1.0	<20	700	9.1	<310	17	370	<61	<1
HDM 2094	99	<63	140	3200	275	<0.5	450	10800	0.150	<340	<18	3	<25	<5
HDM 2095	98	<7	21	48	2.85	8.4	74	42	2.00	200	43	128	<4	<1
HDM 2723	94	61700	73	3400	320	700	920	98	3.1	1970	74	18	15	1.2
HDM 2729	108	<20	7	4	2.41	1.4	78	20	1.40	<275	7	23.4	<9	<0.7
HDM 2735	104	<85	390	9100	103	11.4	120	590	1.08	1900	10	1.9	<33	<1.5

Table A1. Results of neutron activation analyses of Bulgarian and Serbian malachite and metal objects. Ir was below the detection limit of about 0.005 μg/g in all samples. Co values below 0.5 μg/g are only approximate. They are given as italic numbers in brackets. –: element not measured. Malachite data marked with an asterisk are from Pernicka u. a. (1993).

Sample	Location	Cu	Pb	As	Sb	Co	Ni	Ag	Au	Se
BG-1 b	Medni Rid	20	5	9.9	0.87	17.3	<30	40	0.4	43
BG-1 c	Medni Rid	37	530	3600	82	226	131	995	0.03	<2.5
BG-2 b	Boboševo	6	4	104	0.9	2.4	12	4.6	0.035	1
BG-3 a	Čelopeč	36	230	330	7.7	2.2	20	45	2	151
BG-3 b	Čelopeč	30	8200	99 000	2140	4.9	<32	46	12.3	930
BG-4	Plakalnica	72	2	<7.5	6.8	5.2	<90	4780	0.12	36
BG-5	Kremikovci	36	n.m.	330	640	25.5	<35	321	<0.07	5.3
BG-6 b	Zidarovo	3	30	4.4	0.6	67	105	<2.5	0.05	2.1
BG-6 c	Zidarovo	18	5	24.1	4.1	7.9	<22	700	0.39	<6.5
BG-7 b	Rosen-C.	8	10	1450	1.1	340	<37	15.6	0.78	10.7
BG-7 c	Rosen-s. k.	38	45	300	1.5	10.7	<22	113	0.6	16.4
BG-7 d	Rosen-s. k.	28	85	11.7	0.54	81	45	37	3.8	52
BG-9 b	Elacite	57	<0.1	260	1.3	2.8	<4	0.4	0.07	4.8
BG-10 c	Car Assen	1	n.m.	7.5	0.4	3.1	<10	<0.6	0.12	0.7
BG-11 c	Radka	14	4900	17 200	330	15.7	<58	456	2.1	9.0
BG-12 d	Brădceto	3	2	1.4	0.22	209	101	<0.8	0.015	<2.5
BG-14	Silistar	1	n.m.	3.9	0.5	32	24	0.2	0.24	7.1
BG-15	Kiten	8	20	3.2	1	17.2	<30	23.5	<0.05	7.4
BG-17 a	Ai Bunar	9	n.m.	16 900	440	3.3	<18	143	4.5	81
BG-17 b	Ai Bunar	53	n.m.	1 200	67	2.9	<17	148	1.9	60
BG-17 c	Ai Bunar	13	n.m.	3 900	600	4.4	<25	103	0.22	176
BG-17 d	Ai Bunar	8	n.m.	960	147	4.0	<20	423	0.12	40
BG-17 e	Ai Bunar	12	9000	6 100	420	n.m.	<20	55	0.76	6.4
BG-17 f	Ai Bunar	7	8000	3 500	120	n.m.	9	9.2	1.46	9.6
BG-17 g	Ai Bunar	6	4500	4 500	570	n.m.	17	15.2	1.16	8.6
BG-17 h	Ai Bunar	3	2400	1 120	66	n.m.	27	8.5	0.16	5.8
BG-17 i	Ai Bunar	22	3700	11 400	3400	n.m.	<25	143	15.7	85
RO-1	Altintepe	1	570	27	1.56	26.2	14	6.8	0.19	15
TG253-K	Majdanpek	43	n.m.	1 630	1.97	13.9	10	<0.8	0.02	2.6

Sample	Location	Fe	Cr	Zn	Te	Hg	Th	U
BG-1 b	Medni Rid	17.8	53	1780	<10	<7	0.44	<1.3
BG-1 c	Medni Rid	5.3	190	2010	10	<12	0.59	1.5
BG-2 b	Boboševo	0.20	20	<9	<60	70	2.0	5.9
BG-3 a	Čelopeč	7.1	<6	26	<220	<11	<0.7	<1.3
BG-3 b	Čelopeč	8.2	<8	1950	142	1730	<0.5	<6
BG-4	Plakalnica	4.4	60	<12	9	<4	<0.3	<2.5
BG-5	Kremikovci	26.0	16	1090	29	3.1	<0.4	40
BG-6 b	Zidarovo	4.6	310	200	<13	0.08	3.0	0.9
BG-6 c	Zidarovo	3.6	18	<19	<12	<40	2.2	<2.8
BG-7 b	Rosen-C.	10.8	9	86	31	22	1.5	53
BG-7 c	Rosen-s. k.	14.2	7	56	65	<35	<0.2	101
BG-7 d	Rosen-s. k.	23.7	20	95	85	<190	0.26	132
BG-9 b	Elacite	0.03	5	3	2	77	<0.04	3.5
BG-10 c	Car Assen	3.3	4	3	4	55	3.6	6.8
BG-11 c	Radka	12.5	33	24	<18	9600	<0.4	<3
BG-12 d	Brdzeto	0.15	2	810	<4	<6	0.45	6.6
BG-14	Silistar	6.9	22	52	<5	168	2.2	1
BG-15	Kiten	5.0	37	26	<12	39	1.7	0.9
BG-17 a	Ai Bunar	2.12	16	8300	<10	<110	1.8	1.9
BG-17 b	Ai Bunar	0.43	12	13 300	7	<22	<0.1	1.9
BG-17 c	Ai Bunar	11.0	33	13 800	<13	<36	4.1	2.7
BG-17 d	Ai Bunar	3.5	37	<9	<8	<86	7.2	2.5
BG-17 e	Ai Bunar	2.92	7.1	27 200	31	4	n.m.	n.m.
BG-17 f	Ai Bunar	0.80	2.5	2 900	<7	n.m.	n.m.	n.m.
BG-17 g	Ai Bunar	3.6	3.1	5 600	<10	<1	n.m.	n.m.
BG-17 h	Ai Bunar	2.21	2.3	12 200	<7	<1	n.m.	n.m.
BG-17 i	Ai Bunar	1.97	0.7	5 200	<25	6	n.m.	n.m.
RO-1	Altintepe	22.9	41	8 100	5	2.2	n.m.	n.m.
TG253-K	Majdanpek	2.89	12	4 700	<20	0.8	n.m.	n.m.

Table A2. Element concentrations in ore samples from Bulgaria (BG), Romania (RO) and Serbia (TG 253-K, Majdanpek) as determined by instrumental neutron activation analysis. Copper (Cu) and iron (Fe) concentrations are given in percent. All other elements in µg/g; n.m. = not measured. With one exception (BG-6c with 0.18% Sn) the Sn content was below 260 mg/g in all cases. Ir was < 0.05 mg/g in all cases.

Sample	location	country	Cu	As	Sb	Co	Ni	Ag	Au	Fe	Zn	Se	Hg	Ir
BG-2a	Boboševo	W	91	230	0.6	0.6	<3	450	<0.014	<200	<12	3.4	8.5	<0.01
BG-6a	Zidarovo	BSC	97	<1.5	0.09	0.8	<3	1.3	<0.017	183	<6	<0.8	1.3	0.0032
BG-7a	Rosen-Central	BSC	97	0.9	0.18	*(0.3)*	<2.5	<0.4	<0.012	1330	6	<0.5	0.4	0.002
BG-8a	Panagjurište-Assarel	W	87	11	1	0.5	<3	21.8	4	<1400	98	3.1	0.8	<0.03
BG-9a	Panagjurište-Elacite	W	95	<1.5	1.7	0.5	1.8	<0.4	3.9	50	<5	12.5	1.6	<0.003
BG-10a	Panagjurište-Car Assen	W	99	80	9.1	1.2	<4.5	2.5	3.9	4100	<31	7.1	9.4	0.0027
BG-11a	Panagjurište-Radka	W	92	<4	<0.2	0.6	<32	<2.5	<0.15	<2450	<25	<8	<11	0.0071
BG-11b	Panagjurište-Radka	W	98	1.2	0.39	0.5	<2.5	0.5	<0.015	<100	<5	<0.6	10.4	0.0025
BG-12a	Malko Tărnovo	STR	91	<1	<0.2	0.7	<4	51.6	<0.013	370	<5	<0.7	34	0.0036
BG-12b	Malko Tărnovo	STR	100	4.5	0.77	5.7	<15	74.3	0.051	1500	<22	<3	8.1	0.016
BG-12c	Malko Tărnovo	STR	90	1.4	<0.2	*(0.3)*	3.3	5.8	<0.09	<1000	15	<0.7	<11	0.017
BG-12e	Malko Tărnovo	STR	100	<3	0.23	1.3	<13	122	<0.03	415	<18	3.3	16.9	0.014
BG-13a	Madžarovo	RHD	100	1.5	1.8	*(0.2)*	2.5	9.1	1.1	<90	38	<0.7	9.2	0.0022
BG-13b	Madžarovo	RHD	96	1.8	0.67	0.6	5.4	13.2	0.37	155	16	<0.6	1.8	0.0024
HDM 1589	Zlate Hory	CZ	94	9.2	1.10	*(0.3)*	1.3	0.2	<0.010	72	9.2	5.7	42	<0.001
HDM 1591	Přibram	CZ	96	11.1	53	0.6	<15	1680	<0.020	340	64	<3	400	<0.004
HDM 1594	Cvikov near Ceska Lipa	CZ	96	4200	159	<0.7	<25	800	<0.050	242	12.0	<3	125	<0.006
HDM 1577	Donnersberg	D	98	60	0.19	0.5	<3	31	0.009	29	2.7	<0.3	<1	0.152
HDM 1583	Recsk	H	97	109	0.27	1.1	<3	24.6	0.25	94	4.3	<0.5	228	<0.001
HDM 1585	Ruda Banja, Galyagos	H	98	7.5	3.2	0.8	<33	0.6	0.046	71	3.4	0.4	226	<0.001
HDM 1676	Recsk	H	96	108	<0.03	0.5	<2	22.7	<0.008	<28	<15	<0.2	390	<0.002
HDM 2598	Urvölgy	H	100	5.5	1.38	0.7	<5	0.7	<0.013	<60	3.9	<0.4	65	<0.001
HDM 2599	Rakosbanya	H	97	12.3	0.23	<0.5	<7	208	<0.015	<75	<4	24.2	400	<0.0015
HDM 2600	Rezbanya	H	100	2.0	1.95	<0.7	<6	181	<0.013	<65	7.4	<0.4	62	<0.001
HDM 1574	Saszka Banja	RO	100	<1	78	*(0.4)*	<6	162	0.064	52	<5	<7	610	<0.002
HDM 1584	Moldova	RO	99	7.0	0.16	0.7	4.2	0.5	0.073	750	2.2	<0.5	102	<0.001
HDM 1587	Saszka	RO	99	<1	0.14	0.8	<3	1.8	<0.020	87	2.2	<0.2	8	<0.001
HDM 1674	Deva	RO	93	4.1	<0.04	0.7	<3	0.3	0.003	2410	34	2.0	30000	<0.001
HDM 1575	Altsohl/Zvolen	SK	99	1	8.2	1.9	<4	9.8	<0.005	341	<5	<7	18	<0.001
HDM 1580	Schmöllnitz	SK	98	4.8	1.41	0.8	<5	1.3	0.29	500	8.6	<0.5	4200	<0.001
HDM 1586	Herrengrund	SK	100	5.6	1.52	0.7	<2	0.6	0.70	115	1.8	0.6	<2	<0.001
HDM 1675	Herrengrund	SK	95	5.0	3.0	1.2	4.7	1.0	0.024	370	<5	<2	91	<0.002
HDM 1395	Bor	SRB	96	9.8	0.5	0.7	13	0.2	<0.03	200	2.8	1.5	<1	<0.001
HDM 1396	Bor	SRB	97	7.0	1.0	*(0.2)*	20	0.2	<0.09	230	3.6	0.7	<1	<0.001
HDM 1471	Majdanpek	SRB	91	33	2.5	0.8	<3	1.3	0.005	250	1.7	8.9	<1	<0.001

HDM 1472	Majdanpek	SRB	93	<3	<0.4	0.5	<3	2.8	<0.002	10	<3	6.2	<1	<0.001
HDM 1476	Cadinje	SRB	98	<3	0.5	0.6	<3	7.2	<0.003	<50	<3	21.2	<1	<0.001
HDM 1497	Veliki Krivelj	SRB	96	<1	0.12	0.2	<2	0.5	0.02	250	7	0.1	<1	<0.001
HDM 1498	Veliki Krivelj	SRB	95	<1	0.12	0.5	1.9	0.5	0.31	30	3	<0.2	<1	<0.001
TG 199	Kiziltarla	TR	94	680	3	<0.5	4	10.3	0.38	n.m.	2	<0.2	n.m.	<0.001
207-G1	Kürtün/Cayircukur	TR	97	<3	<0.03	0.7	4	34	0.0068	n.m.	16	7.8	n.m.	0.0012
TG 210	Bakırtepe	TR	94	64	3	<0.5	4	46	<0.05	n.m.	3	<0.3	n.m.	<0.001
HDM 49	Ergani Maden	TR	100	<25	<7	*(0.3)*	<20	12.0	<0.60	<450	<20	<18.0	n.m.	0.0008
HDM 332	Ergani Maden	TR	100	<3	<1	8.3	<13	<0.8	<0.13	9000	35	<0.6	n.m.	<0.002
HDM 389	Kırmızı Tarla	TR	100	<5	1.0	*(0.2)*	28	37	<0.20	<80	<5	<0.5	n.m.	<0.002
HDM 390	Kırmızı Tarla	TR	97	4	<1	*(0.2)*	8	36	<0.20	<60	<4	<0.4	n.m.	<0.002
HDM 1500	Kırmızı Tarla	TR	96	42	<0.04	0.6	<3	73	<0.007	57	<2	<0.2	64	<0.001

Table A3a. Trace element concentrations in samples of native copper from Bulgaria as well as from the Czech Republic (CZ), Germany (D), Hungary (H), Romania (RO), Slovakia (SK), Serbia (SRB), and Turkey (TR), as determined by instrumental neutron activation analysis. All values are given in µg/g except for Cu which is given in percent; n.m. = not measured.

sample	location	region	Sc	Cr	Sn	Te	Cs	Ba	Ce	Hf	Th	U
BG-2a	Boboševo	W	0.05	<1.5	<17	<9	<0.26	<85	<2.4	<0.30	<0.16	<1.4
BG-6a	Zidarovo	BSC	0.4	0.8	<8	<3	<0.16	<1	<30	<0.09	0.04	0.6
BG-7a	Rosen-Central	BSC	1.8	0.5	<6	<2.5	<0.12	<23	12	<0.084	0.62	0.8
BG-8a	Panagjurište-Assarel	W	1.5	n.m.	<14	<3	<0.39	112	<3	<0.10	0.29	1.7
BG-9a	Panagjurište-Elacite	W	1.1	<0.8	<10	1.8	<0.14	<24	<1	<0.07	36.8	<0.7
BG-10a	Panagjurište-Car Assen	W	1.8	<1.5	<10	<4.5	<0.17	2.2	<35	<0.10	0.16	<0.8
BG-11a	Panagjurište-Radka	W	1.5	<1.1	<10	<47	<0.16	1.8	<15	<0.10	<0.3	n.m.
BG-11b	Panagjurište-Radka	W	0.03	2.4	<7	<2.5	0.05	0.9	<13	<0.08	0.21	<0.7
BG-12a	Malko Tărnovo	STR	0.3	3.4	<28	<4	0.07	<1.3	<34	<0.10	0.08	<0.6
BG-12b	Malko Tărnovo	STR	0.46	<3	<33	<15	0.47	<120	3.3	<0.40	0.16	<1.7
BG-12c	Malko Tărnovo	STR	0.29	<0.9	<8	3.3	<0.1	<26	0.9	<0.07	0.1	<2.3
BG-12e	Malko Tărnovo	STR	0.12	<4.3	<30	<13	0.56	<100	<2.5	<0.30	<0.25	<1.5
BG-13a	Madžarovo	RHD	3.6	<1.1	<8	2.5	<0.14	0.9	<27	<0.09	0.15	<0.7
BG-13b	Madžarovo	RHD	0.14	<1	<8	5.4	<0.13	0.7	<27	<0.09	<0.07	<0.7

Table A3b. Concentrations in Bulgarian samples of native copper of some elements that are not usually determined in ancient copper-based alloys, because they are efficiently separated from copper on smelting. They may, however, be of some use to compare with untreated or cold-worked copper artifacts from prehistoric sites. All values are given in µg/g; n.m. = not measured.

HDM no.	Location	Region	Description	lead concentration [μg/g]	$^{208}Pb/^{206}Pb$	$^{207}Pb/^{206}Pb$	$^{204}Pb/^{206}Pb$
Neolithic							
1969	Durankulak	BSC	malachite bead	2	2.0677	0.8351	0.05333
1970	Durankulak	BSC	malachite bead	2	2.0679	0.8337	0.05324
2004	Durankulak	BSC	malachite bead	3	2.0513	0.8298	0.05299
2209	Durankulak	BSC	malachite bead	1	2.0757	0.8390	0.05353
2210	Durankulak	BSC	malachite bead	<1	2.0729	0.8368	0.05333
2211	Durankulak	BSC	malachite bead	16	2.0692	0.8360	0.05337
2212	Durankulak	BSC	malachite bead	7	2.0754	0.8364	0.05332
Early Chalcolithic							
1904	Slatino	W	chisel	27	2.0835	0.8447	0.05397
1912	Marica (Devebargan)	THR	heavy axe (oval cross section)	11	2.0714	0.8369	0.05336
1975	Durankulak	BSC	finger ring	250	2.0824	0.8440	0.05401
2005	Durankulak	BSC	finger ring	20	2.0339	0.8241	0.05263
Middle Chalcolithic							
1903	Radlovci	W	hammer axe (Pločnik)	17	2.0770	0.8430	0.05409
1910	Dragoman	W	hammer axe (Pločnik A)	4	2.0771	0.8428	0.05401
1911	Dragoman	W	hammer axe (Pločnik A)	6	2.0751	0.8419	0.05396
1940	Ai Bunar	THR	hammer axe (Pločnik B)	62	2.0795	0.8384	0.05347
1976	Durankulak	BSC	bracelet	8	1.9927	0.8069	0.05133
1977	Durankulak	BSC	bracelet	1020	2.0825	0.8439	0.05402
2011	Durankulak	BSC	bracelet	510	2.0679	0.8325	0.05307
2025	Durankulak	BSC	bracelet	90	2.0675	0.8325	0.05310
2026	Durankulak	BSC	bracelet	90	2.0679	0.8325	0.05310
2041	Tell Ruse	NE	borer	60	2.0786	0.8409	0.05371
2049	Tell Ruse	NE	borer	90	2.0801	0.8415	0.05371
2064	Tell Ruse	NE	borer	20	2.0776	0.8442	0.05406
2080	Tell Ruse	NE	borer	42	2.0772	0.8428	0.05403
2124	Goljamo Delčevo	BSC	borer	40	2.0772	0.8404	0.05400
2702	Däržanica	W	heavy axe (Gumelniţa)	<1	2.0447	0.8258	0.05262*
2703	Däržanica	W	heavy axe (Sălcuţa)	5	2.0777	0.8431	0.05403
2726	Mezdra	W	heavy axe (Pločnik)	25	2.0774	0.8430	0.05399
Late Chalcolithic							
1905	Djakovo	W	flat axe	18	2.0753	0.8391	0.05356
1915	Sadovec	NC	hammer axe (Vidra A)	33	2.0768	0.8374	0.05338
1920	Tell Ruse	NE	flat axe (Kamenar)	18	2.0538	0.8322	0.05321
1921	Tell Ruse	NE	chisel	390	2.0835	0.8443	0.05401
1922	Tell Ruse	NE	chisel	7	1.9217	0.7814	0.04978
1923	Tell Ruse	NE	heavy axe (Gumelniţa)	250	2.0847	0.8446	0.05398
1924	Sava, cemetery	BSC	heavy axe (Coţeana)	70	2.0833	0.8441	0.05400
1925	Varna I	BSC	hammer axe (Vidra B)	6	2.0697	0.8375	0.05346
1926	Varna I	BSC	hammer axe (Vidra B)	24	2.0743	0.8397	0.05365

Table A4 (continued)

HDM no.	Location	Region	Description	lead concentration [µg/g]	$^{208}Pb/^{206}Pb$	$^{207}Pb/^{206}Pb$	$^{204}Pb/^{206}Pb$
1927	Varna I	BSC	flat axe (Sălcuţa)	20	2.0815	0.8413	0.05370
1928	Devnja	BSC	hammer axe (Čoka-Varna)	2770	2.0842	0.8458	0.05399
1929	Devnja	BSC	hammer axe (Varna)	3	2.0745	0.8387	0.05355
1930	Devnja	BSC	hammer axe (Devnja)	6	2.0052	0.8130	0.05185
1931	Varna I	BSC	hammer axe (Devnja)	83	2.0829	0.8424	0.05378
1932	Varna I	BSC	chisel	66	2.0662	0.8362	0.05341
1933	Devnja	BSC	heavy axe (Gumelniţa)	63	2.0843	0.8445	0.05399
1934	Devnja	BSC	hammer axe (Čoka-Varna)	820	2.0835	0.8442	0.05399
1935	Devnja	BSC	chisel	3	2.0702	0.8377	0.05346
1936	Devnja	BSC	hammer axe (Čoka-Varna)	920	2.0829	0.8442	0.05403
1937	Hotnica Tell	NC	hammer axe (Vidra A)	10	2.0756	0.8425	0.05394
1938	Hotnica Tell	NC	hammer axe (Vidra B)	12	2.0704	0.8377	0.05353
1939	Hotnica Tell	NC	heavy axe (Coţeana)	2	2.0712	0.8394	0.05378
1942	Durankulak	BSC	finger ring	2000	2.0794	0.8415	0.05377
1943	Durankulak	BSC	finger ring	300	2.0744	0.8449	0.05394
1944	Durankulak	BSC	bracelet	540	2.0749	0.8448	0.05399
1945	Durankulak	BSC	bracelet	310	2.0754	0.8449	0.05402
1946	Durankulak	BSC	bracelet	5	1.9534	0.7936	0.05053
1947	Durankulak	BSC	bracelet	80	2.0104	0.8154	0.05203
1948	Durankulak	BSC	bracelet	950	2.0789	0.8467	0.05415
1949	Durankulak	BSC	bracelet	80	2.0481	0.8276	0.05281
1956	Durankulak	BSC	bracelet	85	2.0675	0.8323	0.05305
1959	Durankulak	BSC	pin	80	2.0799	0.8410	0.05374
1960	Durankulak	BSC	bracelet	760	2.0677	0.8326	0.05308
1961	Durankulak	BSC	hammer axe (Čoka-Varna)	39	2.0846	0.8444	0.05396
1962	Durankulak	BSC	finger ring	15	2.0260	0.8216	0.05250
1963	Durankulak	BSC	bracelet	240	2.0850	0.8445	0.05395
1964	Durankulak	BSC	bracelet	510	2.0835	0.8442	0.05399
1965	Durankulak	BSC	bracelet	33	1.8130	0.7402	0.04701
1967	Durankulak	BSC	finger ring	26	2.0839	0.8441	0.05392
1968	Durankulak	BSC	bracelet	23	2.0191	0.8194	0.05224
1971	Durankulak	BSC	flat axe (Sălcuţa)	82	2.0591	0.8338	0.05330
1972	Durankulak	BSC	bracelet	270	2.0658	0.8326	0.05311
1973	Durankulak	BSC	bracelet	20	1.9864	0.8077	0.05150
1974	Durankulak	BSC	finger ring	25	1.8189	0.7421	0.04705
1978	Durankulak	BSC	bracelet	710	2.0831	0.8441	0.05402
1979	Durankulak	BSC	bracelet	11	2.0730	0.8382	0.05350
1980	Durankulak	BSC	bracelet	13	1.9644	0.7978	0.05082
1981	Durankulak	BSC	bracelet	4	2.0583	0.8344	0.05327
1982	Durankulak	BSC	bracelet	<1	2.0555	0.8306	0.05308*
1983	Durankulak	BSC	bracelet	92	2.0807	0.8475	0.05422
1984	Durankulak	BSC	bracelet	200	2.0827	0.8435	0.05393

Table A4 (continued)

HDM no.	Location	Region	Description	lead concentration [μg/g]	$^{208}Pb/^{206}Pb$	$^{207}Pb/^{206}Pb$	$^{204}Pb/^{206}Pb$
1985	Durankulak	BSC	bracelet	23	2.0762	0.8381	0.05347
1986	Durankulak	BSC	bracelet	47	2.0766	0.8414	0.05380
1987	Durankulak	BSC	bracelet	57	2.0567	0.8326	0.05314
1988	Durankulak	BSC	bracelet	17	1.9693	0.7996	0.05096
1989	Durankulak	BSC	bracelet	300	2.0833	0.8442	0.05402
1990	Durankulak	BSC	bracelet	200	2.0840	0.8444	0.05401
1991	Durankulak	BSC	bracelet	86	2.0831	0.8441	0.05402
1992	Durankulak	BSC	bracelet	1140	2.0819	0.8439	0.05407
1993	Durankulak	BSC	bracelet	33	1.9036	0.7749	0.04928
1994	Durankulak	BSC	bracelet	3	2.0394	0.8248	0.05258
1995	Durankulak	BSC	bracelet	28	2.0482	0.8303	0.05308
1996	Durankulak	BSC	bracelet	480	2.0833	0.8439	0.05401
1997	Durankulak	BSC	bracelet	930	2.0778	0.8455	0.05403
1998	Durankulak	BSC	bracelet	390	2.0823	0.8441	0.05407
1999	Durankulak	BSC	flat axe	100	2.0767	0.8410	0.05376
2000	Durankulak	BSC	finger ring	<1	1.7460	0.7135	0.04520*
2001	Durankulak	BSC	finger ring	800	2.0794	0.8427	0.05392
2002	Durankulak	BSC	finger ring	17	2.0198	0.8192	0.05234
2003	Durankulak	BSC	finger ring	20	2.0613	0.8390	0.05361
2006	Durankulak	BSC	spiral ring	<1	2.0699	0.8352	0.05321*
2007	Durankulak	BSC	spiral ring	880	2.0838	0.8443	0.05401
2008	Durankulak	BSC	finger ring	23	2.0492	0.8306	0.05309
2009	Durankulak	BSC	spiral ring	13	2.0589	0.8363	0.05339
2010	Durankulak	BSC	borer	90	2.0624	0.8355	0.05342
2012	Durankulak	BSC	bracelet	190	2.0795	0.8475	0.05424
2013	Durankulak	BSC	finger ring	4490	2.0799	0.8412	0.05374
2014	Durankulak	BSC	bracelet	20	1.8107	0.7396	0.04699
2015	Durankulak	BSC	bracelet	2	2.0649	0.8347	0.05323
2016	Durankulak	BSC	bracelet	<1	2.0551	0.8296	0.05301
2017	Durankulak	BSC	bracelet	2	2.0097	0.8168	0.05214
2018	Durankulak	BSC	bracelet	27	2.0428	0.8285	0.05294
2019	Durankulak	BSC	bracelet	12	2.0622	0.8386	0.05356
2020	Durankulak	BSC	bracelet	5	2.0733	0.8409	0.05363
2021	Durankulak	BSC	flat axe	20	2.0640	0.8361	0.05342
2022	Durankulak	BSC	hammer axe (Varna)	50	2.0781	0.8421	0.05384
2023	Durankulak	BSC	bracelet	20	1.9966	0.8102	0.05174
2024	Durankulak	BSC	bracelet	23	1.9977	0.8103	0.05168
2027	Durankulak	BSC	finger ring	2010	2.0825	0.8441	0.05403
2028	Durankulak	BSC	finger ring	32	2.0366	0.8287	0.05289
2029	Durankulak	BSC	finger ring	170	2.0831	0.8441	0.05400
2030	Durankulak	BSC	finger ring	440	2.0844	0.8445	0.05398
2031	Durankulak	BSC	finger ring	12	1.7515	0.7169	0.04536
2032	Durankulak	BSC	borer	16	2.0818	0.8433	0.05389
2034	Durankulak	BSC	pin	6	2.0745	0.8390	0.05356
2035	Durankulak	BSC	spiral ring	44	2.0827	0.8442	0.05400
2036	Durankulak	BSC	spiral ring	45	2.0063	0.8145	0.05196
2039	Tell Ruse	NE	borer	90	2.0794	0.8468	0.05412
2040	Tell Ruse	NE	borer	260	2.0808	0.8415	0.05374
2043	Tell Ruse	NE	borer	3	2.0514	0.8301	0.05317
2044	Tell Ruse	NE	borer	2000	2.0831	0.8442	0.05400
2045	Tell Ruse	NE	borer	110	2.0831	0.8439	0.05396

Table A4 (continued)

HDM no.	Location	Region	Description	lead concentration [µg/g]	$^{208}Pb/^{206}Pb$	$^{207}Pb/^{206}Pb$	$^{204}Pb/^{206}Pb$
2047	Tell Ruse	NE	double-spiral-headed pin	33	2.0807	0.8450	0.05393
2050	Tell Ruse	NE	borer	150	2.0774	0.8432	0.05405
2051	Tell Ruse	NE	borer	43	2.0809	0.8469	0.05413
2052	Tell Ruse	NE	borer	70	2.0814	0.8470	0.05411
2053	Tell Ruse	NE	chisel	90	2.0829	0.8440	0.05398
2054	Tell Ruse	NE	borer	750	2.0786	0.8372	0.05347
2055	Tell Ruse	NE	borer	17	2.0800	0.8466	0.05412
2056	Tell Ruse	NE	borer	8	2.0732	0.8404	0.05383
2057	Tell Ruse	NE	borer	225	2.0817	0.8436	0.05401
2058	Tell Ruse	NE	borer	36	2.0782	0.8430	0.05398
2059	Tell Ruse	NE	borer	2	2.0581	0.8316	0.05302*
2060	Tell Ruse	NE	borer	480	2.0667	0.8367	0.05343
2061	Tell Ruse	NE	borer	3	2.0628	0.8336	0.05330
2063	Tell Ruse	NE	borer	9	2.0465	0.8277	0.05303
2065	Tell Ruse	NE	borer	60	2.0760	0.8427	0.05407
2067	Tell Ruse	NE	borer	8	2.0838	0.8423	0.05372
2068	Tell Ruse	NE	borer	<1	2.0510	0.8300	0.05292**
2070	Tell Ruse	NE	borer	75	2.0849	0.8444	0.05396
2071	Tell Ruse	NE	borer	230	2.0843	0.8442	0.05390
2072	Tell Ruse	NE	borer	26	2.0730	0.8386	0.05356
2073	Tell Ruse	NE	double-spiral-headed pin	40	2.0790	0.8389	0.05346
2074	Tell Ruse	NE	borer	30	2.0652	0.8356	0.05332
2075	Tell Ruse	NE	loop-headed pin	20	2.0720	0.8403	0.05386
2076	Tell Ruse	NE	borer	270	2.0841	0.8443	0.05397
2077	Tell Ruse	NE	borer	32	2.0784	0.8408	0.05373
2078	Tell Ruse	NE	borer	40	2.0807	0.8417	0.05373
2079	Tell Ruse	NE	borer	8	2.0715	0.8382	0.05354
2081	Tell Ruse	NE	borer	10	2.0785	0.8421	0.05380
2082	Tell Ruse	NE	borer	7	2.0794	0.8389	0.05352
2083	Tell Ruse	NE	borer	4	2.0727	0.8385	0.05359
2084	Tell Ruse	NE	borer	30	2.0724	0.8385	0.05353
2085	Tell Ruse	NE	borer	2	2.0484	0.8277	0.05295
2088	Tell Ruse	NE	borer	20	2.0677	0.8367	0.05338
2089	Tell Ruse	NE	loop-headed pin	1	2.0586	0.8317	0.05314
2090	Tell Ruse	NE	borer	3	2.0754	0.8415	0.05376
2091	Tell Ruse	NE	borer	11	2.0090	0.8160	0.05203
2092	Tell Ruse	NE	loop-headed pin	3	2.0769	0.8433	0.05389
2093	Tell Ruse	NE	borer	10	2.0780	0.8392	0.05354
2096	Tell Ruse	NE	borer	34	2.0757	0.8388	0.05351
2097	Tell Ruse	NE	borer	8	2.0795	0.8420	0.05378
2098	Tell Ruse	NE	pin	1880	2.0832	0.8442	0.05400
2099	Tell Ruse	NE	double-spiral-headed pin	56	2.0823	0.8421	0.05363
2100	Tell Ruse	NE	double-spiral-headed pin	10	2.0794	0.8461	0.05404
2101	Tell Ruse	NE	borer	1	2.0529	0.8281	0.05292*
2102	Tell Ruse	NE	borer	2	2.0571	0.8319	0.05313
2103	Tell Ruse	NE	borer	1	2.0598	0.8325	0.05306
2104	Tell Ruse	NE	loop-headed pin	22	2.0784	0.8432	0.05400
2119	Goljamo Delčevo	BSC	borer	15	2.0679	0.8370	0.05346

Table A4 (continued)

HDM no.	Location	Region	Description	lead concentration [µg/g]	$^{208}Pb/^{206}Pb$	$^{207}Pb/^{206}Pb$	$^{204}Pb/^{206}Pb$
2120	Goljamo Delčevo	BSC	borer	22	2.0700	0.8379	0.05343
2121	Goljamo Delčevo	BSC	bracelet	140	2.0811	0.8435	0.05398
2122	Goljamo Delčevo	BSC	spiral ring	<1	2.0612	0.8323	0.05315
2123	Goljamo Delčevo	BSC	spiral ring	390	2.0837	0.8443	0.05399
2125	Goljamo Delčevo	BSC	borer	<1	2.0462	0.8269	0.05276
2126	Goljamo Delčevo	BSC	double-spiral-headed pin	20	2.0796	0.8430	0.05383
2127	Goljamo Delčevo	BSC	borer	3	2.0068	0.8143	0.05198
2128	Goljamo Delčevo	BSC	borer	320	2.0826	0.8438	0.05398
2129	Goljamo Delčevo	BSC	borer	180	2.0817	0.8477	0.05415
2130	Goljamo Delčevo	BSC	borer	<1	2.0694	0.8374	0.05360**
2137	Durankulak	BSC	finger ring	5	2.0292	0.8230	0.05249
2138	Durankulak	BSC	finger ring	13	2.0775	0.8461	0.05405
2139	Onogur castle	BSC	flat axe (Sălcuţa?)	12	2.0765	0.8386	0.05344
2140	Debrene/Prilep	BSC	flat axe (Goljamo Delčevo)	16	2.0786	0.8440	0.05415
2141	Durankulak	BSC	bracelet	110	2.0838	0.8444	0.05398
2142	Durankulak	BSC	bracelet	260	2.0816	0.8429	0.05391
2143	Hotnica Tell	NC	borer	8	2.0777	0.8439	0.05392
2144	Hotnica Tell	NC	borer	12	2.0769	0.8395	0.05350
2145	Hotnica Tell	NC	borer	14	2.0771	0.8393	0.05342
2146	Hotnica Tell	NC	borer	16	2.0749	0.8364	0.05336
2150	Karanovo Tell	THR	borer	17	2.0778	0.8381	0.05347
2707	Goljamo Delčevo	BSC	hooked wire	23	2.0782	0.8382	0.05348
2709	Smjadovo	NE	copper strip	7	2.0783	0.8406	0.05370
2710	Smjadovo	NE	chisel	30	2.0796	0.8412	0.05370
2711	Smjadovo	NE	borer	440	2.0797	0.8408	0.05363
2712	Smjadovo	NE	casting spill	3	2.0717	0.8383	0.05349
2713	Smjadovo	NE	casting spill	2	2.0727	0.8384	0.05352
2714	Smjadovo	NE	borer	2	2.0786	0.8409	0.05366
2715	Smjadovo	NE	ear ring	40	2.0843	0.8440	0.05383
2716	Smjadovo	NE	borer	2	1.9779	0.8018	0.05110
2717	Smjadovo	NE	borer	48	2.0787	0.8443	0.05391
2718	Smjadovo	NE	borer	33	2.0813	0.8390	0.05341
2719	Smjadovo	NE	borer	4	2.0780	0.8409	0.05373
2720	Smjadovo	NE	borer	450	2.0655	0.8342	0.05321
2721	Smjadovo	NE	borer	2	2.0639	0.8327	0.05318
2722	Smjadovo	NE	borer	3420	2.0829	0.8442	0.05397
2725	Zaminec	W	flat axe	110	2.0792	0.8424	0.05381
2727	Krivodol	W	borer	54	2.0797	0.8443	0.05416
2728	Zaminec	W	borer	110	2.0756	0.8426	0.05400
2733	Zaminec	W	borer	1260	2.0881	0.8453	0.05397
Final Chalcolithic							
1901	Hotnica	NC	axe-adze (Tîrgu Okna)	10	2.0766	0.8427	0.05408
1906	Plakuder	W	axe-adze (Jászladány)	30	2.0801	0.8435	0.05396
1907	Plakuder	W	axe-adze (Jászladány)	33	2.0780	0.8430	0.05403
1908	Plakuder	W	axe-adze (Jászladány)	9	2.0765	0.8428	0.05399

Table A4 (continued)

HDM no.	Location	Region	Description	lead concentration [μg/g]	$^{208}Pb/^{206}Pb$	$^{207}Pb/^{206}Pb$	$^{204}Pb/^{206}Pb$
1909	Plakuder	W	axe-adze (Jászladány)	10	2.0775	0.8430	0.05402
1913	Teliš	NC	axe-adze (Jászladány?)	40	2.0787	0.8432	0.05396
1914	Teliš	NC	axe-adze (Jászladány)	230	2.0758	0.8423	0.05400
1916	Kočan	W	hammer axe (Mezökerestes)	33	2.0786	0.8431	0.05397
1917	Stolnik/Elin Pelin	W	axe-adze (Jászladány)	55	2.0768	0.8428	0.05409
1919	Ivanovo	NE	axe-adze (Jászladány)	<1	2.0688	0.8357	0.05340
1941	Ai Bunar	THR	axe-adze (Tîrgu Okna)	13	2.0778	0.8409	0.05374
2105	Teliš	NC	double-spiral-headed pin	8	2.0754	0.8400	0.05362
2106	Teliš	NC	knife	4	2.0745	0.8407	0.05378
2107	Teliš	NC	knife	60	2.0690	0.8376	0.05351
2108	Teliš	NC	borer	40	2.0760	0.8425	0.05400
2109	Teliš	NC	pin	13	2.0792	0.8399	0.05358
2110	Teliš	NC	borer	16	2.0709	0.8371	0.05341
2111	Teliš	NC	borer	14	2.0771	0.8428	0.05402
2112	Teliš	NC	borer	5	2.0709	0.8397	0.05379
2113	Teliš	NC	borer	27	2.0769	0.8426	0.05397
2114	Teliš	NC	borer	220	2.0744	0.8369	0.05336
2115	Teliš	NC	borer	590	2.0753	0.8424	0.05402
2116	Teliš	NC	borer	2	2.0815	0.8447	0.05392
2117	Teliš	NC	borer	46	2.0798	0.8456	0.05401
2118	Teliš	NC	borer	20	2.0665	0.8373	0.05355
2213	Živlovci/Montana	W	axe-adze (Jászladány)	–	n.m.	n.m.	n.m.
2705	Plakuder	W	axe-adze (Jászladány)	20	2.0775	0.8431	0.05408
2706	Plakuder	W	axe-adze (Ariuşt)	26	2.0781	0.8434	0.05407
2708	Smjadovo	NE	axe-adze (Ariuşt)	5	2.0708	0.8356	0.05325
2724	Rebărkovo	W	axe-adze (Ariuşt)	110	2.0783	0.8433	0.05408
Proto Bronze Age							
1902	Hotnica, region	NC	flat axe (Altheim, var. Ete)	37	2.0772	0.8423	0.05394
1918	Durankulak	BSC	dagger (Nerušaj)	8	2.0776	0.8428	0.05400
1952	Durankulak	BSC	dagger	1780	2.0877	0.8448	0.05386
1954	Durankulak	BSC	pin	20	2.0799	0.8426	0.05389
1966	Durankulak	BSC	dagger (Usatovo)	11	2.0733	0.8365	0.05334
2134	Hotnica-Vodopada	NC	dagger (Mondsee)	20	2.0781	0.8433	0.05406
2135	Hotnica-Vodopada	NC	dagger (Mondsee)	6	2.0755	0.8410	0.05375
2147	Kačica	NC	borer	2450	2.0740	0.8367	0.05333
2148	Kačica	NC	borer	890	2.0741	0.8369	0.05343
2149	Kačica	NC	borer	11	2.0747	0.8368	0.05339
2701	Jasen	W	flat axe (Altheim, var. Ete)	56	2.0779	0.8431	0.05394
2736	Malorad	W	dagger	460	2.0610	0.8315	0.05301

Table A4 (continued)

HDM no.	Location	Region	Description	lead concentration [µg/g]	$^{208}Pb/^{206}Pb$	$^{207}Pb/^{206}Pb$	$^{204}Pb/^{206}Pb$
2737	Galiče	W	dagger (Cucuteni)	76	2.0725	0.8397	0.05374
2738	Lesura	W	dagger	700	2.0726	0.8357	0.05347
2740	Haramijska dupka	RHD	dagger (Jagodina)	4	2.0674	0.8364	0.05350
2741	Haramijska dupka	RHD	dagger (Sofievka)	60	2.0693	0.8356	0.05328
2742	Haramijska dupka	RHD	borer	8	2.0762	0.8381	0.05337*
Early Bronze Age							
2704	Vidbol	W	flanged axe	120	2.0718	0.8393	0.05362
2739	Lesura	W	dagger	2	2.0789	0.8411	0.05378*
Late Bronze Age							
1955	Durankulak	BSC	metal band	3000	2.0997	0.8511	0.05432
1957	Durankulak	BSC	pin	1820	2.0915	0.8474	0.05405
1958	Durankulak	BSC	spiral-headed pin	1790	2.0970	0.8511	0.05431
2033	Durankulak	BSC	spiral-headed pin	1000	2.0831	0.8423	0.05371
2037	Kozlodujci	BSC	sickle	330	2.1130	0.8620	0.05501
2038	Vodnjanci	BSC	double axe	100	2.0641	0.8316	0.05300
2131	Hotnica-Vodopada	NC	borer	4	2.0723	0.8356	0.05325
2132	Hotnica, region	NC	dagger	330	2.0966	0.8510	0.05425
2133	Hotnica, region	NC	dagger	135	2.0724	0.8350	0.05320
2136	Hotnica-Vodopada	NC	metal fragment	250	2.0671	0.8349	0.05320
2730	Krivodol/Oslen	W	knife	370	2.1131	0.8620	0.05502
2731	Gradešnica	W	borer	570	2.0805	0.8409	0.05366
2732	Gradešnica	W	borer	73	2.0850	0.8479	0.05447
2734	Gradešnica	W	loop-headed pin	1740	2.1025	0.8545	0.05456
Classical Period							
1953	Durankulak	BSC	stylo	20930	2.0685	0.8332	0.05315
Unstratified or uncertain							
1950	Durankulak	BSC	wire	980	2.0875	0.8460	0.05424
1951	Durankulak	BSC	wire	420	2.0671	0.8324	0.05308
2042	Tell Ruse	NE	hook	4	2.0754	0.8416	0.05373
2046	Tell Ruse	NE	borer	570	2.0865	0.8450	0.05390
2048	Tell Ruse	NE	bracelet	1210	2.0767	0.8390	0.05357
2062	Tell Ruse	NE	borer	6	2.0486	0.8288	0.05306
2066	Tell Ruse	NE	borer	550	2.0822	0.8423	0.05385
2069	Tell Ruse	NE	chisel	790	2.1016	0.8593	0.05498
2086	Tell Ruse	NE	borer	710	2.0829	0.8440	0.05405
2087	Tell Ruse	NE	borer	1910	2.0836	0.8442	0.05399
2094	Tell Ruse	NE	pin	140	2.0796	0.8469	0.05412
2095	Tell Ruse	NE	pin	21	2.0582	0.8335	0.05346
2723	Zlatarevo	NE	borer	73	2.0758	0.8417	0.05371
2729	Gradešnica	W	borer	7	2.0757	0.8420	0.05396
2735	Gradešnica	W	borer	390	2.0723	0.8352	0.05341

Table A4. Lead isotope abundance ratios in Neolithic malachite and copper-based Chalcolithic and Bronze Age metal artifacts from Bulgaria. Regions are abbreviated as follows: BSC = Black Sea coast, NC = north central Bulgaria, NE = north-east Bulgaria, RHD = Rhodope mountains, SRB = Serbia, THR = Bulgarian Thrace, W = western Bulgaria. Uncertainty (2σ) of all abundance ratios <0.1%, except when marked: *<0.2% or with **<0.4%.

Sample	Location	Region	Description	Lead content [μg/g]	$^{208}Pb/^{206}Pb$	$^{207}Pb/^{206}Pb$	$^{204}Pb/^{206}Pb$
BG-1 b	Medni Rid	BSC	chpyr, pyr	5	2.0645	0.8359	0.05341
BG-1 c	Medni Rid	BSC	chpyr, born, mal, pyr, fhl	530	2.0745	0.8450	0.05411
BG-2 b	Boboševo	W	mal, azur	4	2.0690	0.8371	0.05344
BG-3 a	Čelopeč	W	born, cov, pyr, chpyr	230	2.0797	0.8424	0.05392
BG-3 b	Čelopeč	W	born, enarg	8200	2.0802	0.8425	0.05384
BG-4	Plakalnica	W	born	2	2.0833	0.8476	0.05424
BG-6 b	Zidarovo	BSC	mal	30	2.0754	0.8421	0.05395
BG-6 c	Zidarovo	BSC	born, chpyr	5	2.0666	0.8377	0.05360
BG-7 b	Rosen-Centre	BSC	mal	10	1.9451	0.7928	0.05057
BG-7 c	Rosen-s. klad.	BSC	mal	45	2.0605	0.8352	0.05344
BG-7 d	Rosen-s. klad.	BSC	chpyr, born	85	2.0654	0.8371	0.05359
BG-9 b	Elacite	W	mal	<0.1	2.0596	0.8361	0.05367
BG-11 c	Radka	W	chpyr, pyr, gal, fhl, born	4900	2.0845	0.8447	0.05406
BG-12d	M. Tărnovo-Brădceto	STR	mal	2	2.0646	0.8355	0.05336
BG-15	Kiten	THR	mal, azur, cov, born	20	2.0801	0.8449	0.05428
BG-17	Ai Bunar	THR	mal	500	2.0829	0.8441	0.05400
BG-17 e	Ai Bunar	THR	mal	9000	2.0836	0.8444	0.05398
BG-17 f	Ai Bunar	THR	mal	8000	2.0836	0.8445	0.05400
BG-17 g	Ai Bunar	THR	mal	4500	2.0823	0.8442	0.05399
BG-17 h	Ai Bunar	THR	mal	2400	2.0830	0.8442	0.05400
BG-17 i	Ai Bunar	THR	mal, azur	3700	2.0824	0.8441	0.05403
RO-1	Altintepe	RO	chpyr, pyr	570	2.0901	0.8581	0.05457

Table A5. Lead isotope abundance ratios of copper ore samples from Bulgaria and Romania. Regions are abbreviated as follows: BSC = Black Sea coast, RO = Romania, SRB = Serbia, STR = Strandža mountains, THR = Bulgarian Thrace, W = western Bulgaria. Ore minerals are abbreviated as follows: azur = azurite, born = bornite, chpyr = chalcopyrite, cov = covelline, mal = malachite, enarg = enargite, fhl = fahlore, gal = galena, pyr = pyrite.

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Zusammenfassung

In Fortsetzung einer früheren Untersuchung zur Rolle der beiden kupferzeitlichen Bergwerke von Rudna Glava und Ai Bunar für die Kupferversorgung Südosteuropas wurden 290 Metallobjekte, sieben Malachitperlen, sowie 76 Proben von Kupfererzen und gediegen Kupfer, vorwiegend aus Bulgarien, hinsichtlich ihrer Spurenelementgehalte und Bleiisotopenverhältnisse analysiert. Von den Artefakten datieren 258 in die Zeit vom späten Neolithikum bis zur Proto-Bronzezeit und bilden zusammen mit 77 prähistorischen Kupferobjekten aus Serbien die Materialbasis für die folgenden Schlußfolgerungen:

Aus dem Vergleich der chemischen Zusammensetzung von gediegen Kupfer und Metallartefakten aus Südosteuropa läßt sich schließen, daß nur wenige Artefakte möglicherweise aus gediegen Kupfer, die überwiegende Mehrheit jedoch aus verhüttetem Kupfer hergestellt wurde. Leider sind zeitgleiche Verhüttungseinrichtungen bisher noch nicht belegt.

Während noch immer kein einziges prähistorisches Metallobjekt Rudna Glava zugeordnet werden kann, stimmen bei 32 Artefakten aus der mittleren und späten Kupferzeit alle gemessenen Parameter mit denen der Erze von Ai Bunar überein. Es ist besonders bemerkenswert, daß sich darunter vier Schwergeräte von Pločnik befinden. Dagegen stammt kein einziges von 82 analysierten endkupferzeitlichen Artefakten von Ai Bunar. Damit ist es erstmals gelungen, die prähistorischen Abbauphasen dieser Lagerstätte indirekt zu datieren.

Wegen der chemischen und isotopischen Einheitlichkeit von zwei Artefaktgruppen schließen wir auf mindestens zwei weitere spätkupferzeitliche Abbauzentren, von denen wir eines in Prochorovo, ca. 80 km östlich von Ai Bunar, vermuten. Das zweite Zentrum liegt mit hoher Wahrscheinlichkeit im Bereich um Medni Rid in der Nähe von Burgas am Schwarzen Meer.

Im westlichen Bereich unseres Untersuchungsgebietes haben wir bereits früher die noch heute im Abbau befindliche Kupferlagerstätte von Majdanpek als Ursprung der meisten endkupferzeitlichen Artefakte aus Serbien identifiziert. Die neuen Ergebnisse legen nun einen Beginn der Abbautätigkeit bereits in der mittleren Kupferzeit nahe. Kupfer dieser Zusammensetzung findet sich auch entlang der Donau bis Tell Ruse. Es deutet sich daher ein Handel mit Kupfer an, der vielleicht das Gegenstück zum Handel mit Spondylus- und Dentaliummuscheln donauaufwärts bildete.

Das Auftreten von Kupfer von Ai Bunar in Serbien einerseits und von Majdanpek an der unteren Donau andererseits läßt auf eine Verbreitung der Metallgewinnung in relativ kurzer Zeit in ganz Südosteuropa schließen. Obwohl das Kupfer aus Aibunar, Prochorovo (?) und Medni Rid vorwiegend in Ost- und Nordostbulgarien zu finden ist und das von Majdanpek vorwiegend in Westbulgarien und Serbien, hat es offenbar Austauschbeziehungen zwischen den beiden Regionen gegeben. Aus den analytischen Ergebnissen ist zusätzlich abzulesen, daß es noch weitere Produktionsstätten von Kupfer gegeben hat.

Резюме

Как продолжение предыдущих исследований о роли энеолитических рудников Рудна Глава и Ай Бунар в обеспечении металлом Южной Европы, были проанализированы на содержание микроэлементов и содержание изотопа свинца 258 металлических предметов, 7 малахитовых бусин и 76 проб рудной и самородной меди, происходящих, в основном, из Болгарии. Дополнительно к ним были проанализированы ещё 77 медных предметов из Сербии. Результаты исследования 258 артефактов, датируемых временем от позднего неолита до позднего энеолита, позволили сделать следующие выводы:

Сравнение химического состава самородной меди и металлических орудий Южной Европы показывает, что лишь немногие из них сделаны из самородной меди. Подавляющее большинство изготовлено из меди, выплавленной из руд. К сожалению, до сих пор не найдено никакого устройства по выплавке меди.

Ни одно из изделий не изготовлено из металла, происходящего из Рудна Глава. Все измеренные параметры 32-х орудий среднего и позднего энеолита соответствуют параметрам руды из Ай Бунара. Необходимо особо отметить, что 4 массивных орудия происходят из Плочника. Из других 82 х изученных предметов, датируемых поздним энеолитом, ни один не происходит из Ай Бунара. Таким образом, впервые удаётся косвенно датировать периоды разработок этих месторождений.

По химическому единству и идентичности состава изотопов в двух группах орудий выделяются, по крайней мере, ещё два местонахождения разработок периода позднего энеолита. Одно из них мы, предположительно, локализуем в Прохорово, примерно в 80 км восточнее Ай Бунара. Второе местонахождение, вероятно, расположено в районе Чёрного моря, на Медни Рид близ Бургаса.

Нами было уже установлено, что в западной части исследуемой территории большинство орудий из Сербии, датируемых поздним энеолитом, происходят из до сих пор разрабатываемого месторождения Майданпек. Новые результаты показывают, что начало разработок на нём относится к середине энеолита. Медь такого состава обнаружена вдоль Дуная до Тель-Русе. Это свидетельствует о том, что медь была предметом обмена на раковины Sponylus i Dentalium, поступавшие вверх по Дунаю.

Появление меди из Ай Бунара в Сербии, с одной стороны, и из Майданпека на Нижнем Дунае, с другой, позволяет определить границы распространения добычи металла в относительно короткое время по всей Южной Европе. Медь из Ай Бунара, Прохорово (?) и Медни Рид встречается, в основном, только в восточной и северо-восточной Болгарии, а медь Майданпека преимущественно в западной Болгарии и Сербии. Но, вероятно, существовал взаимообмен между этими регионами. Кроме того, результаты анализов показывают, что для выработки меди использовались и другие рудники.

Abkürzungsverzeichnis

Die in diesem Band vorkommenden Abkürzungen richten sich nach dem in der Archäologischen Bibliographie 1989 erschienenen Zeitschriftenverzeichnis sowie nach dem Sigel-Verzeichnis im Archäologischen Anzeiger 1992, 743–749. Die kyrillischen Kürzel folgen denjenigen der Советская Археологическая Литература – Библиография (Москва, Ленинград 1965–1986) bzw. den in der Советская bzw. Российская Археология gebräuchlichen. Außer diesen wurden die folgenden Abkürzungen und Sigel verwendet.

ActaInterdisA	Acta Interdisciplinaria Archaeologia
ABalt	Archaeologia Baltica
AEur	Archäologie in Eurasien
AHung	Archaeologia Hungarica
AmAntiquity	American Antiquity
ArcticAnthr	Arctic Anthropology
BAM	Beiträge zur Archäologie des Mittelmeer-Kulturraumes
BAR	British Archaeological Reports
BeitrAltertumSaarbrücken	Saarbrücker Beiträge zur Altertumskunde
EurAnt	Eurasia Antiqua
Geowiss	Geowissenschaften
GodSarajevo	Godišnjak Sarajevo
JAnthrA	Journal of Anthropological Archaeology
JbAKWien	Jahrbuch für Altertumskunde Wien
JRadioanalytNuclChem	Journal of Radioanalytical Nuclear Chemistry
MatAllgA	Materialien zur Allgemeinen und Vergleichenden Archäologie
PA	Photographisches Album der Prähistorischen und Anthropologischen Ausstellung zu Berlin 1 (Berlin 1880)
SSMM	Sak'art'velos saxelmcip'o muzeumis moambe
StudPrHist	Studia Praehistorica
VerDeutschGesBotanik	Verhandlungen der Deutschen Gesellschaft für Botanik

АОР	Археологически открития и разкопки
Археология	Археология. София
ГМСБ	Годишник на музеите в Североисточна България
ГНМ	Годишник на Народния музей
ИАИ	Известия на Археологическия институт при БАН (продължение на ИзБАИ)
ИАК	Известия Императорской археологической комиссии (Санкт-Петербург)
ИРОМК	Известия Ростовского областного музей краеведения (Ростов-на-Дону)
ИзНМВарна	Известия на Народния музей Варна
ИзНМТар	Известия на Народния музей Тарновско
КДСП	Курганные древности степного Поднепровья (Днепропетровск)
ПАСЕ	Проблемы археологии степной Евразии (Кемерово)
РП	Разкопки и проучвания (София)