COPPER TOOLS FROM KOZAREVA MOGILA. NEW DATA ON THE PROVENANCE OF COPPER ORES USED

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Cuvinte-cheie: *metalurgie, eneoliticul târziu, minereu de cupru, proveniență, depozite de cupru, izotopi de plumb, compoziția chimică.*

Keywords: *metallurgy, Late Eneolithic, copper ore, provenance, copper deposits, lead isotopy, chemical composition.*

Rezumat: *Articol prezintă un grup de șase unelte de cupru de la Kozareva mogila, care au fost examinate din punct de vedere al compoziției chimice și al izotopilor de plumb. Din punct de vedere cronologic, descoperirile sunt legate de eneoliticul târziu, astfel că acestea oferă informații despre metalurgia din această perioadă. Rezultatele analizelor de laborator indică că descoperirile examinate diferă unele de altele. Compoziția chimică sugerează faptul că acestea sunt realizate din cupru foarte pur. Cu toate acestea, pot fi observate unele diferențe în ceea ce privește concentrațiile de oligoelemente. Privitor la proveniența cuprului utilizat, se stabilește faptul că uneltele examinate sunt produse din minereuri cu origini diferite. Ca posibile surse de materie prime pot fi identificate mai multe zăcăminte/câmpuri de minereuri, care se află relativ aproape de sit.*

Abstract: *This paper presents a group of six copper tools from Kozareva mogila, which were examined for their chemical composition and lead isotopy. Chronologically, the finds are related to the Late Eneolithic, thus they give information on the metallurgy during this period. The results of the laboratory analyses show that the examined finds differ from each other. The chemical composition indicates that they are made of very pure copper. However, some differences regarding the trace element concentrations can be observed. On the provenance of copper used, it is established that the examined tools are produced from ores with different origins. As possible sources of raw material, several deposits/ore fields can be identified, that are located relatively close to the site.*

Introduction

Kozareva mogila is a well-known site that has been excavated for more than thirty years[1]. It is located south of the Balkan Mountains, close to the Black Sea coast-it is situated approximately 4 km from the modern seashore (**Fig. 1**). As a

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[1] GEORGIEVA 1991.

result of many years of excavation, different periods of habitation have been identified–Early and Late Eneolithic, Early Bronze Age and Hellenistic Period[2].

The present work provides new data on the Eneolithic metallurgy of the site. This paper discusses a group of copper tools found in Late Chalcolithic contexts for which it has been possible to be examined through interdisciplinary laboratory methods. Samples of the objects have been sent to "Curt–Engelhorn" Zentrum Archäometrie, Mannheim, Germany, where a chemical composition analysis as well as lead isotope analysis were performed. The results obtained allow for the provenance of copper ores used, thus providing information on the organization of metallurgical production in a region that is relatively poorly studied.

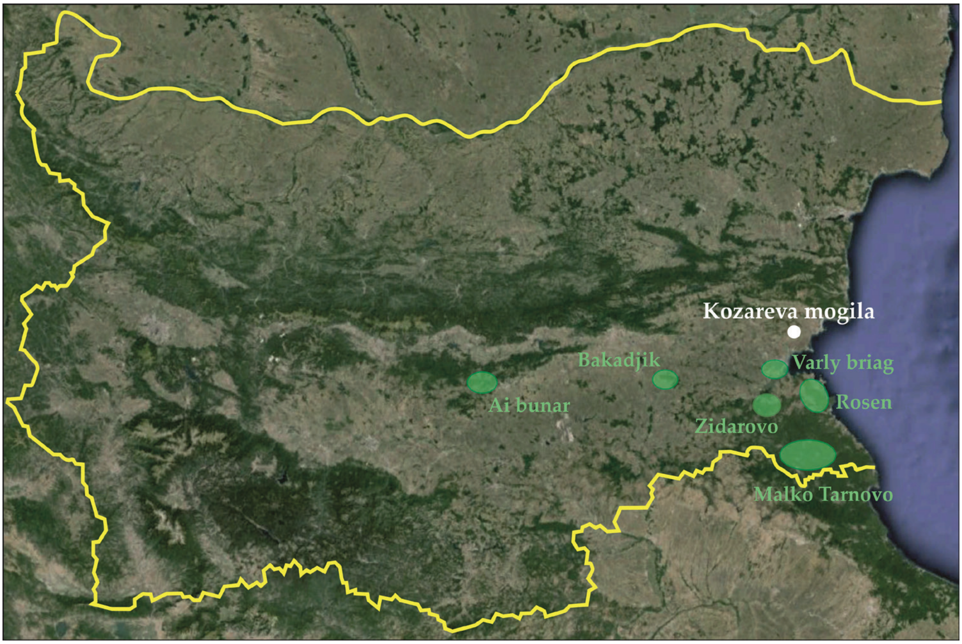

Fig. 1. Location of Kozareva mogila and the nearby copper ore fields.

The copper tools

A total of six copper tools are examined-two awls and four chisels (**Fig. 2**). One of the awls originates from the settlement mound (**Fig. 2.6**). It is found during the excavation of I burnt horizon. The awl has a quadrangular, close to square, cross-section, which is slightly rounded only at the tip. Its length is 9.9 cm, and its width and thickness are 0.2-0.3 cm.

The other tools originate from the necropolis. They are found in several Late Eneolithic graves, as part of the grave inventory – № 10, 12, 30, 64.

From Grave № 12 comes a copper awl with a quadrangular cross-section[3] (**Fig. 2.5**) In the middle part of the object, the section is close to square, while toward

[2] GEORGIEVA, POPOVA, DANOV 2018, p. 107.

[3] GEORGIEVA, DANOV 2021, p. 83, pl. VII.

the butt it narrows and becomes rectangular. At the tip, the cross-section is round. The length of the awl is 12.1 cm. Its width varies between 0.1-0.3 cm and its thickness is 0.1-0.2 cm. The tool is slightly bent at the upper part, which, according to the excavators, marks the place where its handle reached[4].

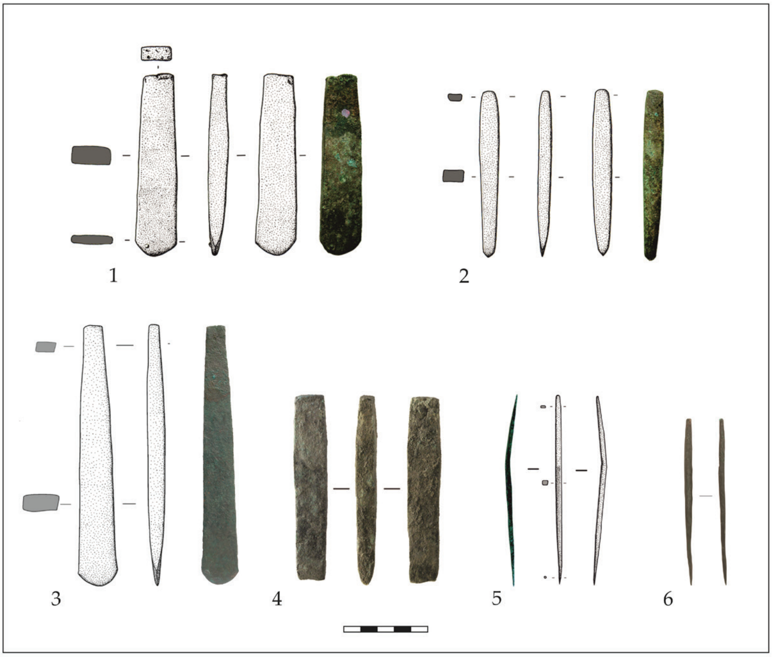

Fig. 2. Copper tools from Kozareva mogila: 1-big chisel from grave № 10[5]; 2-small chisel from grave № 10[6]; 3-chisel from grave № 30[7]; 4-chisel from grave № 64; 5-awl from grave № 12[8]; 6-awl from settlement mound (I burnt horizon) (drawings: Veselin Danov; photos: Petya Georgieva).

The four copper chisels come from graves №№ 10, 30 and 64. One chisel each is found in graves № 30[9] and № 64, while in grave № 10 there are two[10]. All the tools differ from each other.

Typologically, with close characteristics are the big chisel from grave № 10 (**Fig. 2.1**) and that from grave № 30 (**Fig. 2.3**). They have a trapezoidal shape with a rectangular cross-section and an arc – shaped edge. In both tools the maximum width coincides with that of the edge, and the maximum thickness is in the upper part of the tool-at a distance of 1/3 of the total length measured from the butt[11]. On

[4] GEORGIEVA, DANOV 2021, p. 60.
[5] GEORGIEVA, DANOV 2021, fig. 33, 1.
[6] GEORGIEVA, DANOV 2021, fig. 33, 2.
[7] GEORGIEVA, DANOV 2021, fig. 32; pl. XVIII, 4.
[8] GEORGIEVA, DANOV 2021, fig. 34.
[9] GEORGIEVA, DANOV 2021, p. 90–91, pl. XVIII.
[10] GEORGIEVA, DANOV 2021, p. 81–82, pl. V.
[11] GEORGIEVA, DANOV 2021, p. 57.

the other hand, they differ in their proportions. The chisel from grave № 30 has a more elongated shape – its length is 15.6 cm, while that of the chisel from grave № 10 is 11 cm. At the same time, the maximum width and thickness of the tools are in inverse proportion.

For the chisel from grave № 30 the maximum width is 1.9 cm and the thickness is 0.9 cm, while in the case of the chisel from grave № 10 they are 2.5 cm and 1.2 cm, respectively. The general impression of the two finds is that they share common typological characteristics, but the chisel from grave № 30 is more elegant than that from grave № 10, which is more massive.

From grave № 10 comes another chisel that differs in both shape and size (**Fig. 2.2**). It has a rectangular cross-section. The maximum width and thickness of the tool are in the middle and upper section, while towards the edge and the butt its walls narrow. The working edge is tapered and has an approximately triangular shape. This chisel is smaller. Its length is 9.7 cm. Its width varies from 0.7 to 1.0 cm and its thickness is between 0.5-0.8 cm[12].

Another chisel originates from grave № 64 (**Fig. 2.4**). It has a straight shape that narrows slightly toward the butt. It has a rectangular cross-section. The maximum width is measured in the upper half of the tool – at a distance of 1/3 of the total length, measured from the butt. The edge is straight but slightly deformed. The length of the chisel is 10.4 cm. Its width varies from 1.5 to 1.8 cm and its thickness is between 0.6–1.0 cm.

All the objects included in this study are found in a Late Eneolithic context and can be related to the late phases of Kodjadermen – Gumelnița – Karanovo VI culture. Most of them originate from the necropolis and represent part of the inventory of the buried individuals.

One awl originates from grave № 12, while more massive tools are found in graves № 30 and № 64. Of interest is grave № 10, from where there are two copper chisels. Also included is one find from the settlement mound – a small copper awl from I burnt horizon.

Chemical composition

The chemical composition of the objects is analyzed by Energy Dispersive X–ray Fluorescence (EDXRF). The results show that the tools are made of very pure copper in which the concentration of trace elements is very low[13] (**Tab. 1**). Of interest is the chisel from grave № 64, which differs significantly from the other finds (**Fig. 3**). In its composition the concentration of some elements like Bi and Pb is remarkably high. The levels of Ag are also higher compared to the other objects. At the same time, this is the only tool in which the concentration of Fe is high enough to be measured.

Another find with more noticeable differences in its chemical composition is the awl from grave № 12. This tool is characterized by relatively low levels of Ag and an extremely low Bi concentration that is below the detection limit.

[12] GEORGIEVA, DANOV 2021, p. 57–58.

[13] Most of the examined trace elements in the composition of the metal are very low in concentration and are below the detection limits.

Table 1. Chemical composition of the copper tools from Kozareva mogila

MA No.	Object	Context	Cu (%)	Mn (%)	Fe (%)	Co (%)	Ni (%)	Zn (%)	As (%)	Se (%)	Ag (%)
MA–227238	Chisel	Grave № 10	100	<0.01	<0.05	<0.01	0.03	<0.05	<0.005	0.040	0.087
MA–227239	Chisel	Grave № 10	100	<0.01	<0.05	<0.01	0.02	<0.05	<0.005	0.003	0.065
MA–227240	Awl	Grave № 12	100	<0.01	<0.05	<0.01	0.03	<0.05	<0.005	0.004	0.026
MA–227241	Chisel	Grave № 30	100	<0.01	<0.05	<0.01	0.02	<0.05	<0.005	0.026	0.079
MA–227242	Chisel	Grave № 64	99	<0.01	0.09	<0.01	0.02	<0.05	<0.005	0.000	0.133
MA–227243	Awl	I burnt horizon	100	<0.01	<0.05	<0.01	0.03	<0.05	<0.005	0.003	0.062
			Cd (%)	**In (%)**	**Sn (%)**	**Sb (%)**	**Te (%)**	**Au (%)**	**Hg (%)**	**Pb (%)**	**Bi (%)**
MA–227238	Chisel	Grave № 10	<0.002	<0.002	<0.005	<0.002	<0.005	<0.01	<0.01	0.011	0.036
MA–227239	Chisel	Grave № 10	<0.002	<0.002	<0.005	<0.002	<0.005	<0.01	<0.01	0.010	0.017
MA–227240	Awl	Grave № 12	<0.002	<0.002	<0.005	<0.002	<0.005	<0.01	<0.01	<0.003	<0.003
MA–227241	Chisel	Grave № 30	<0.002	<0.002	<0.005	<0.002	<0.005	<0.01	<0.01	<0.003	0.024
MA–227242	Chisel	Grave № 64	<0.002	<0.002	<0.005	<0.002	<0.005	<0.01	<0.01	0.264	0.278
MA–227243	Awl	I burnt horizon	<0.002	<0.002	<0.005	<0.002	<0.005	<0.01	<0.01	0.007	0.030

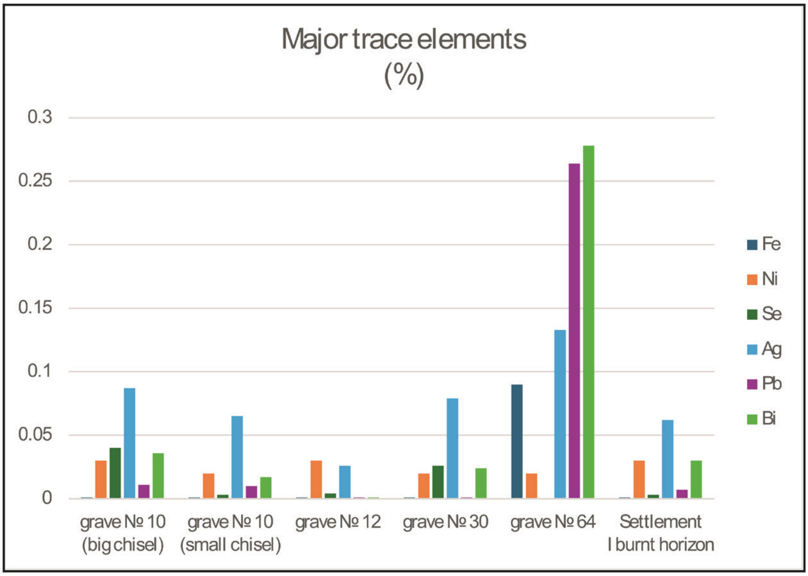

Fig. 3. Concentration of the major trace elements in the composition of the copper tools from Kozareva mogila.

For the rest of the objects, it is difficult to trace any particular differences. The values of the measured elements are relatively homogeneous in all the artifacts and vary within narrow limits. Only in the case of the big chisel from grave № 10 slightly higher levels are observed for some of the elements.

Regarding the information that the chemical composition can provide on the origin of the raw material, attention has been drawn to a group of elements that have been found not to change during the pyrometallurgical processes involved in the processing of the copper ore. Such elements are Ag, Au, Bi, Ir, Ni, etc[14]. These have been established to behave in a similar way to copper and follow it during the purification of the metal. For these reasons, they are preserved to a considerable extent in the composition of the finished product[15].

When examining these elements, it is necessary to note that not all of them are present in the composition of the tools from Kozareva mogila. For example, the concentration of Au is very low in all the artifacts, thus it was not possible to measure. Of most importance in this study are the elements Ag, Bi and Ni, which are present in sufficient quantities in each of the finds[16]. Based on these elements, a comparison analysis is made between the different artifacts, as a result of which some of the observations already presented have been confirmed (**Fig. 4**). It is well visible the separation of the chisel from grave № 64, which is at a great distance from all the other objects. Also remarkable is the isolated character of the awl from grave № 12. At the same time, this analysis provides additional insight into the other four objects.

[14] PERNICKA 2014, table 11.1.

[15] PERNICKA 1990, p. 76–78; PERNICKA 1999, p. 170–171; PERNICKA 2014, p. 250–255.

[16] Only in the awl from grave № 12 the concentration of bismuth (Bi) is very low and is below the detection limits.

As can be seen on the diagram the small chisel from grave № 10 and that from grave № 30 are clearly distinguishable from the other artifacts and are separated into a distinct group.

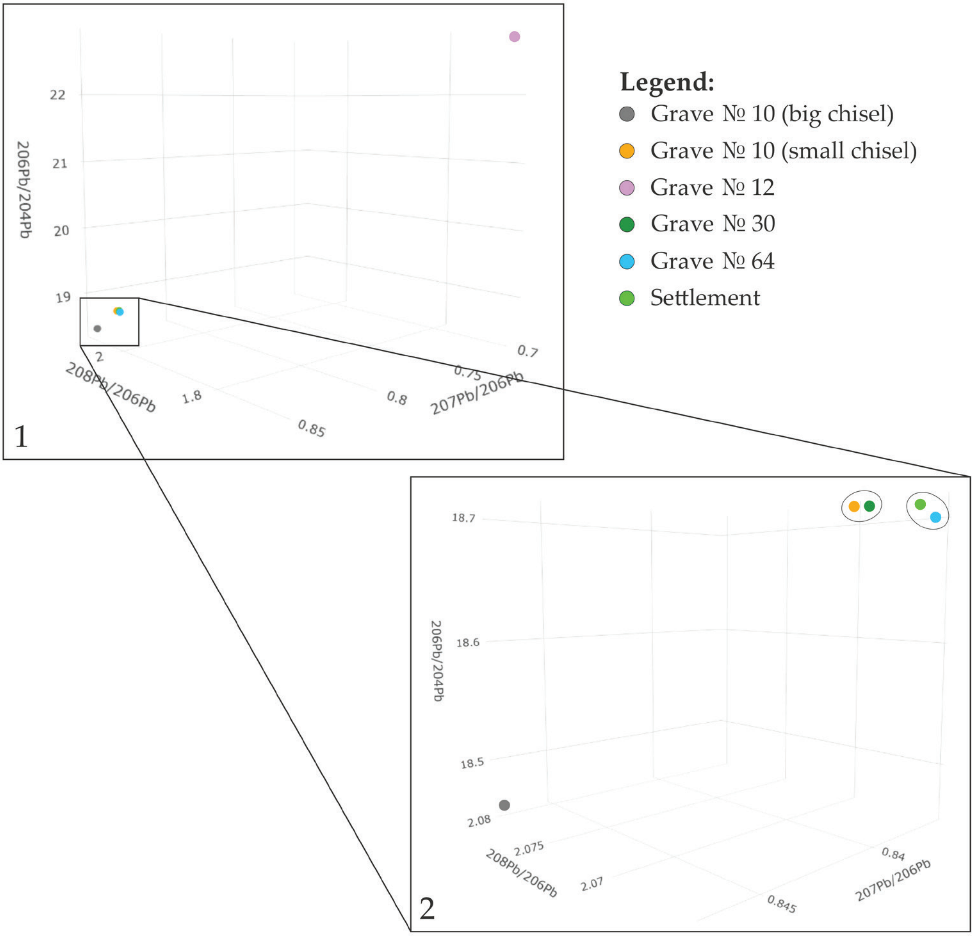

Fig. 4. Ni-Bi-Ag ratios in the composition of the copper tools from Kozareva mogila (an interactive 3D scatter plot is available at: https://y-milev.github.io/milev_pont/Fig_4.html).

Provenance study

To determine the provenance of the raw material from which the copper tools are made a lead isotope analysis is made (**Tab. 2**). The abundance ratios of the lead isotopes are measured by Inductively Coupled Plasma Mass Spectrometry (ICP-MS).

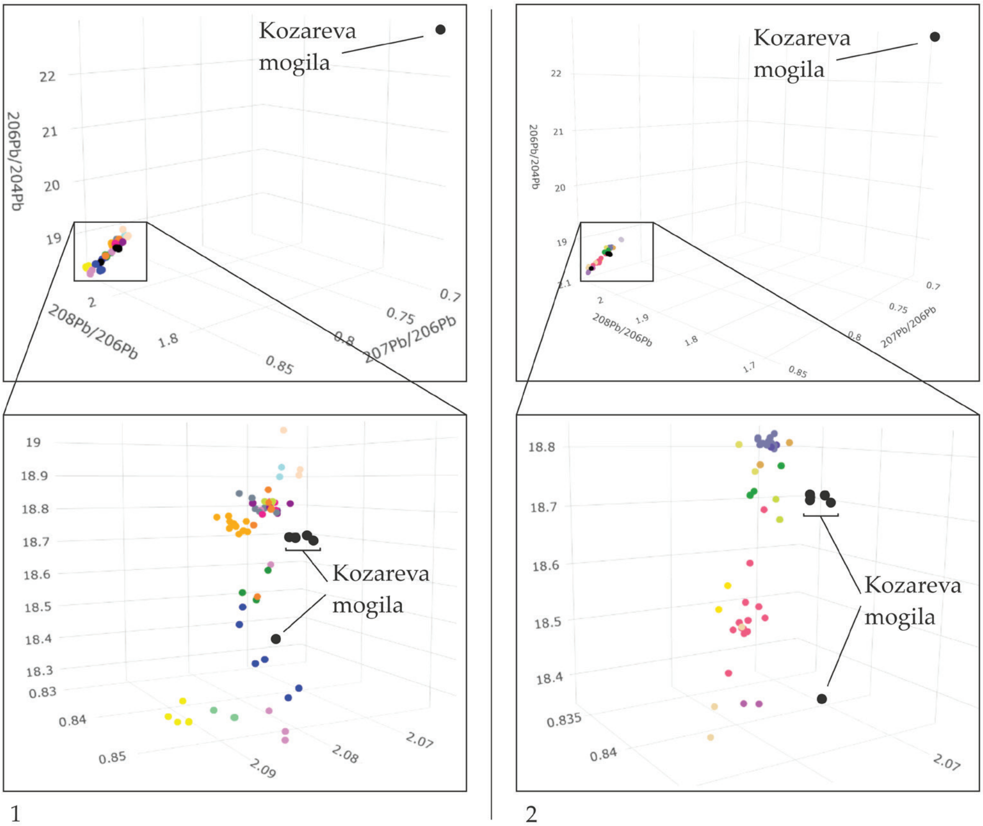

Fig. 5. Pb-isotope ratios of the copper tools from Kozareva mogila (an interactive 3D scatter plot is available at: https://y-milev.github.io/milev_pont/Fig_5.html).

Table 2. Pb-isotope abundance ratios of the copper tools from Kozareva mogila

MA No.	Object	Context	Orig. ID / Inv. №	^{208}Pb/^{206}Pb	^{207}Pb/^{206}Pb	^{206}Pb/^{204}Pb	^{208}Pb/^{204}Pb	^{207}Pb/^{204}Pb
MA–227238	Chisel	Grave № 10	KM_1 / 4.2 (2012)	2.0801	0.84689	18.461	38.401	15.635
MA–227239	Chisel	Grave № 10	KM_2 / 4.3 (2012)	2.0691	0.83732	18.712	38.716	15.668
MA–227240	Awl	Grave № 12	KM_3 / 4.5 (2012)	1.6887	0.69336	22.833	38.557	15.832
MA–227241	Chisel	Grave № 30	KM_4 / 4.3 (2014)	2.0685	0.83704	18.712	38.707	15.663
MA–227242	Chisel	Grave № 64	KM_5 / 4.1 (2021)	2.0653	0.83642	18.699	38.618	15.640
MA–227243	Awl	I burnt horizon	KM_6 / 4.1 (2020)	2.0662	0.83654	18.711	38.660	15.653

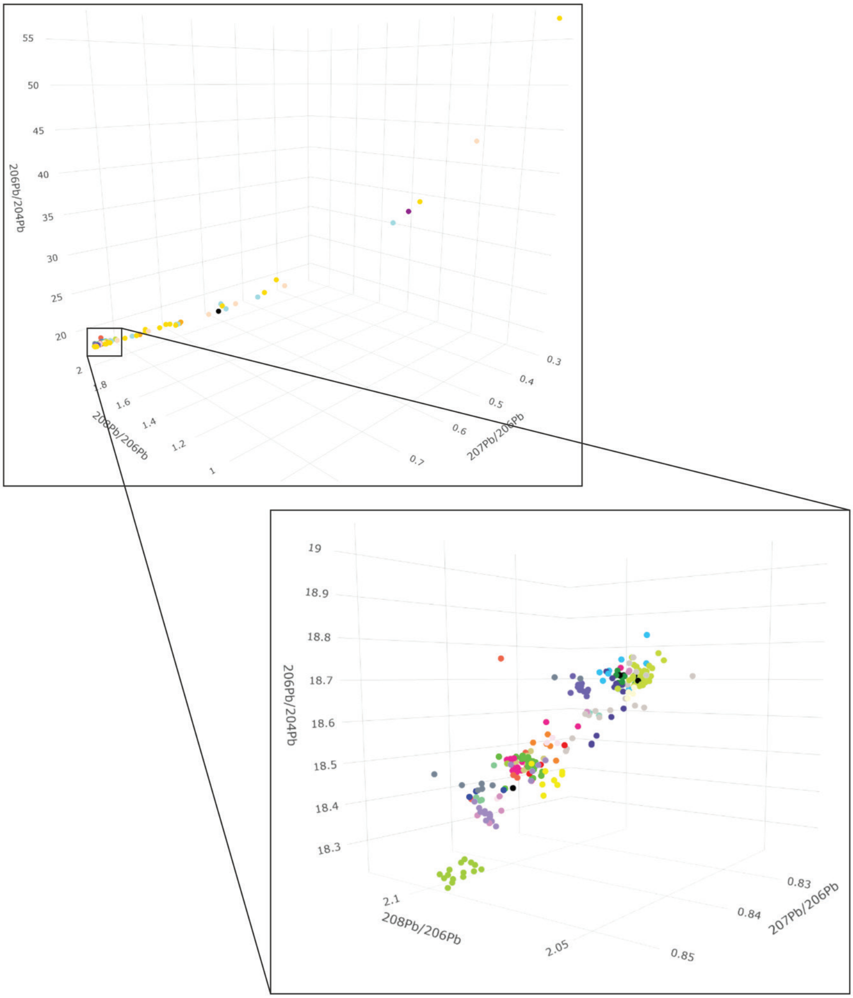

Fig. 6. Comparison of the Pb-isotope ratios of the copper tools from Kozareva mogila and copper ore deposits in: 1-Greece and Cyclades; 2-Anatolia.

The results show that the six artifacts differ from each other – four separate groups can be distinguished (**Fig. 5**). With most prominent differences in the isotope ratios is the awl from grave № 12, which is clearly distinguishable from all the other objects. This corresponds well with the observations on the chemical composition of the find and indicates that the tool is made of copper ore, different from that of the other artifacts. A similar situation is apparent for the big chisel from grave № 10, which also stands apart from the other tools and has an isolated character. The other four objects have more or less similar isotope ratios, which is why they are grouped close to each other. However, there are some differences, which allow them to be separated into two distinct groups. One includes the small chisel from grave № 10 as well as the chisel from grave № 30, in which the isotopic ratios are slightly higher. These two objects are also similar in their chemical composition, which further supports the possibility that they are made from copper ore of similar origin. The other group is represented by the chisel from grave № 64 and the awl from the settlement mound. With these two finds, however, it is noteworthy that, despite their similarities in terms of lead isotopes, they differ in their chemical composition-as already noted, the chisel from grave № 64 differs from the other artifacts due to the increased concentration of some of the major trace elements in its composition. This suggests that it is possible for the two objects, despite their similarities in the lead isotope ratios, to be made from copper ore that comes from different deposits with similar isotopic characteristics.

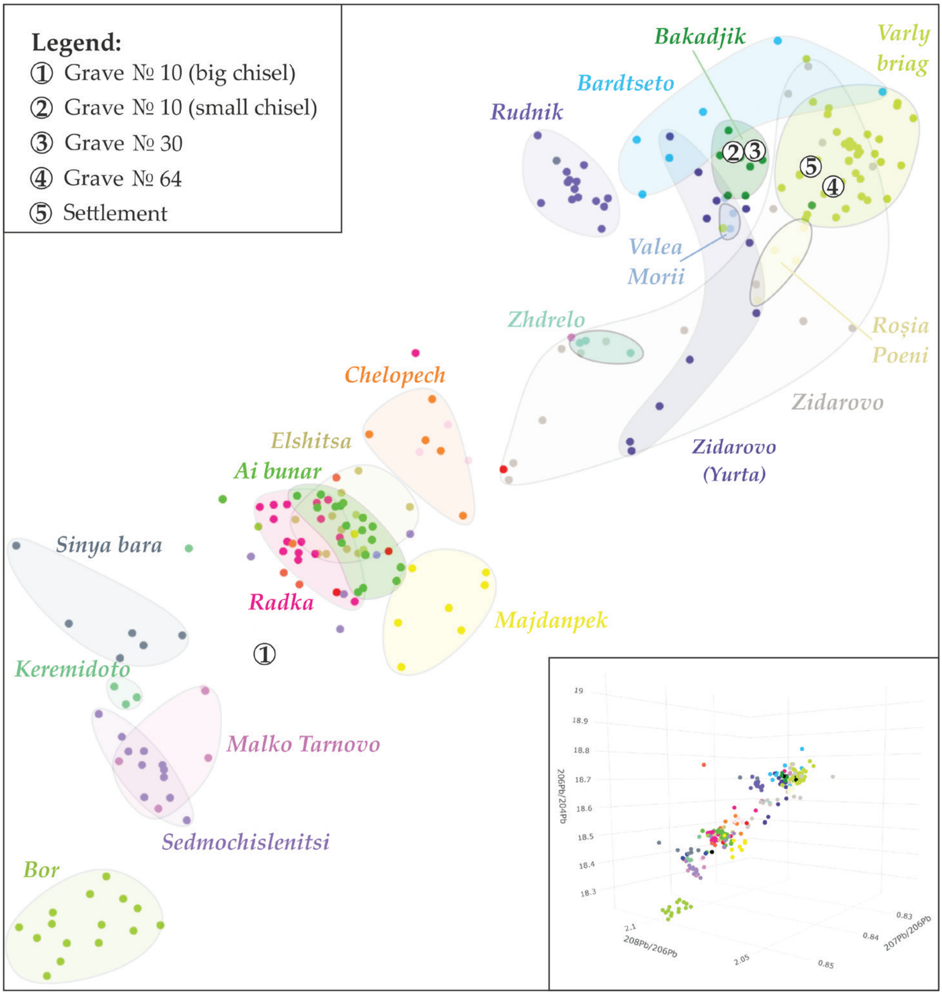

Fig. 7. Pb-isotope ratios of the copper tools from Kozareva mogila and copper deposits in the Balkans.

In order to determine the possible sources of raw material, a comparative analysis is made in which the copper tools from Kozareva mogila are compared with different copper and copper-based deposits[17]. To cover a larger geographical area, deposits from the Balkan Peninsula, Anatolia and the Cyclades are included[18].

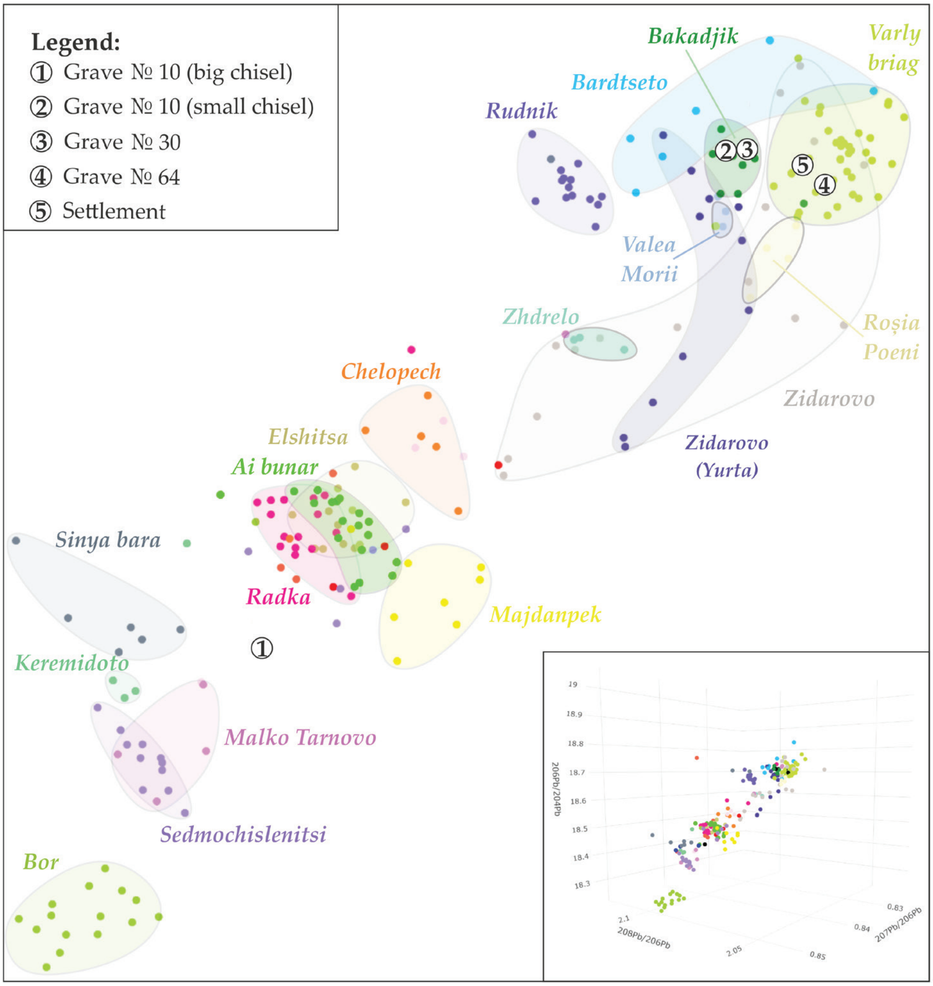

Fig. 8. Comparison of the Pb-isotope ratios of the copper tools from Kozareva mogila and copper ore deposits in Bulgaria, Serbia, and Romania (an interactive 3D scatter plot is available at: https://y-milev.github.io/milev_pont/Fig_8–9.html).

[17] The analysis is performed by comparing the isotopic ratios of $^{208}Pb/^{206}Pb$, $^{207}Pb/^{206}Pb$ and $^{206}Pb/^{204}Pb$. These are plotted in a three-dimensional scatter plot that provides a more detailed perspective (Fig. 7).

[18] Data on the isotopic ratios of the deposits is taken from already published studies – AMOV, BOGDANOV, BALDJIEVA 1974; AMOV, BALDJIEVA 1976; AMOV *et alii* 1976; AMOV *et alii* 1981; AMOV *et alii* 1985; WAGNER *et alii* 1986; DIMITROV *et alii* 1987; AMOV *et alii* 1989; WAGNER *et alii* 1989; GALE *et alii* 1991; PERNICKA *et alii* 1993; STOS-GALE, GALE, ANNETS 1996; PERNICKA *et alii* 1997; KOUZMANOV 2001; MARCOUX *et alii* 2002; BEGEMANN, SCHMITT-STRECKER, PERNICKA 2003; KOUZMANOV *et alii* 2009; KUNZE, PERNICKA 2020; RADIVOJEVIC, REHREN, PERNICKA 2021; TOMCZYK 2022.

The analysis shows that none of the tools examined is made of an ore that originates from Anatolia or the Cyclades (**Fig. 6**). The same can be said for Greece, which is generally poor in copper deposits[19]. Rather, the archaeological finds can be associated with copper deposits and/or ore fields that are located in relatively close proximity to Kozareva mogila.

For example, the two chisels with similar isotopic characteristics (the small chisel from grave № 10 and the one from grave № 30) could be associated with the area of Bakadjik ore field, Yambol region. The lead isotopes of the two objects are very similar to those of the Bakadjik deposit and fully overlap its signature field (**Fig. 8**). This suggests that they could be made of copper that originates from this deposit, or at least from this area. However, another possible source of raw material could also be the copper deposit Bardtseto, located in Malko Tarnovo region. As can be seen in the diagram, its signature field is close to that of Bakadjik. However, it should be noted that the two deposits differ considerably in their abundance ratios. In contrast to Bakadjik, where the lead isotope ratios are relatively homogeneous and thus form a very compact signature field, those of Bardtseto are very diverse and vary widely. This results in an isotopic field that is very extensive and heterogeneous, which in turn makes it difficult to work with this concrete deposit and relate it to finished copper products. For this reason, although Bardtseto cannot be excluded as a possible ore source, it is more likely that the ore from which the two chisels are made originates from the Bakadjik area, with which the two finds have very close similarities.

Of interest is the other copper chisel from grave № 10. Its lead isotope ratios are different from those of the small chisel, which shows that the two finds are made of different ore. At present it is difficult to identify the copper source used for the production of this artifact. This is because the lead isotopes of the find are very different from those of the deposits and do not match any of their signature fields (**Fig. 8**). This may be due to the uneven exploration of the different ore areas, resulting in missing data for part of the deposits. Another possibility is that the copper chisel could be made of mixed ores, originating from several deposits, which would result in the mixing of their lead isotopes[20].

The other two finds with similar isotopic ratios (the chisel from grave № 64 and the awl from the settlement mound) show similarities with the deposits at Varly Briag and Zidarovo, located in Burgas region (**Fig. 8**). The signature fields of the two deposits partially overlap, and the two finds fall into this exact zone. Because of this, the two deposits can hardly be distinguished from each other. It has already been noted that despite the similarities in lead isotope ratios, the two objects differ in their chemical composition. This suggests that it is possible for the two finds to be made from different ores. If this is true, it could be assumed that one of them is made of ore originating from the area of Varly Briag, while the other is made of copper from Zidarovo region.

For the awl from grave № 12, it can be said that it differs significantly from all the other tools. The values of its lead isotope ratios can be related to the presence

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[19] PERNICKA 1987, p. 620, 647–678.

[20] PERNICKA 1990, p. 103.

of “radiogenic lead” in the composition of the metal, which has an influence on the levels of lead isotopes[21]. This considerably reduces the number of deposits that can be assumed to be possible ore sources, because such an anomaly is relatively rare. However, there are several deposits where similar lead isotope behavior has been observed (**Fig. 9**). These are the deposits in the area of Rosen ore field[22] – Rosen, Medni rid and Propadnala voda, which are located near Kozareva mogila, as well as Rudna Glava and Crnaika[23], in eastern Serbia. Due to the proximity of Kozareva mogila to the Rosen ore field, it can be suggested that this could be the more probable source of raw material. However, Rudna Glava and Crnajka cannot necessarily be excluded since deposits of this type are difficult to distinguish.

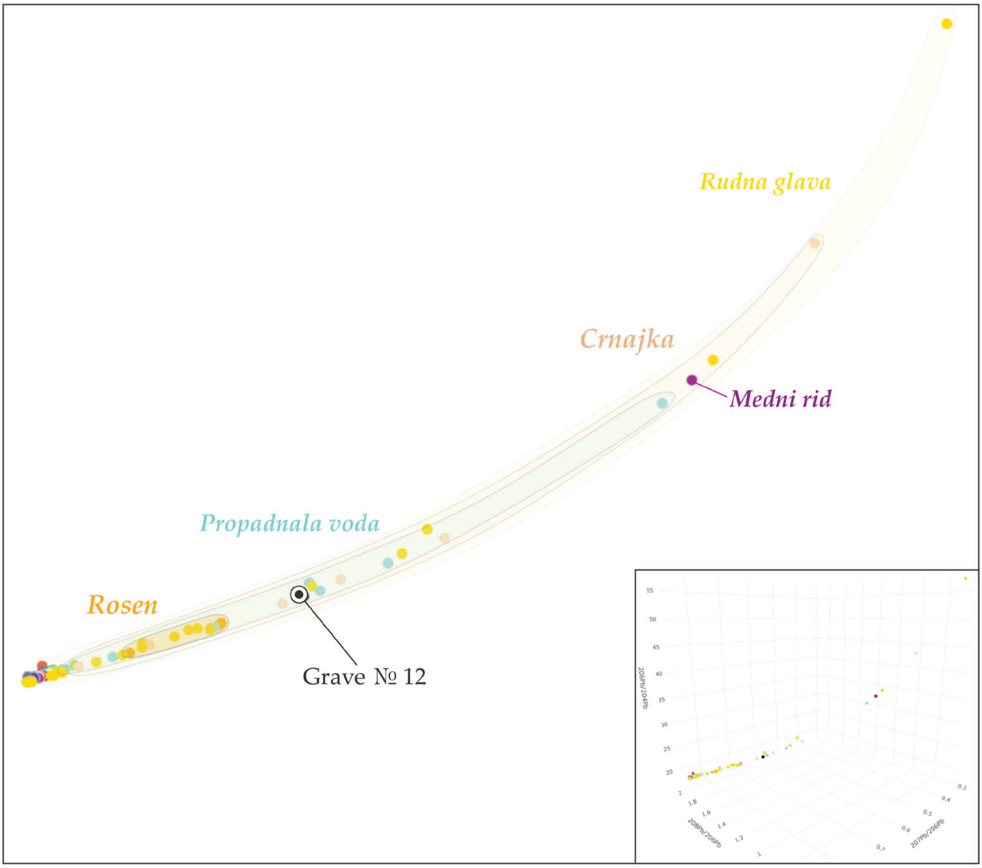

Fig. 9. Comparison of the Pb-isotope ratios of the copper awl from Grave № 12 and copper ore deposits with highly radiogenic lead (an interactive 3D scatter plot is available at: https://y-milev.github.io/milev_pont/Fig_8–9.html).

[21] PERNICKA 1990, p. 104; PERNICKA 1993, p. 29, 51–52; PERNICKA 2014, p. 249.

[22] KUNZE, PERNICKA 2020, p. 399–403.

[23] PERNICKA *et alii* 1993, p. 29.

Conclusion

Kozareva mogila provides interesting information on the organization of metallurgy and the distribution of copper in the Late Eneolithic. The results of the analyses show that the copper tools from the site are made from ore that originates from several different places. The data that we have for the moment comes mainly from the necropolis, where it can be seen that in the different graves, where copper objects are found, the latter are made from raw material from various sources. Exceptions are the small chisel from grave № 10 and that from grave № 30, which are made of copper with a similar origin. It is also established that the two copper tools found as part of the inventory in grave № 10 are made of different ores. Similar situations are known from other sites, such as the necropolis at Durankulak[24].

It is noteworthy that the presence of copper from Ai bunar has not been identified in Kozareva mogila, for the moment. The origin of the raw material used for the production of the tools can be related to copper deposits and ore fields, which are located in close proximity to the site-Zidarovo, Varly briag, Propadnala voda, Medni rid. The most distant region with which some of the finds could be related is the area of Bakadjik ore field.

Although the results presented here do not permit general conclusions, they provide new information on the Eneolithic metallurgy in a region that is relatively poorly studied, such as the area between the eastern slopes of the Balkan Mountains and the Burgas Bay.

Acknowledgments

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[24] PERNICKA *et alii* 1997, p. 134–135.

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