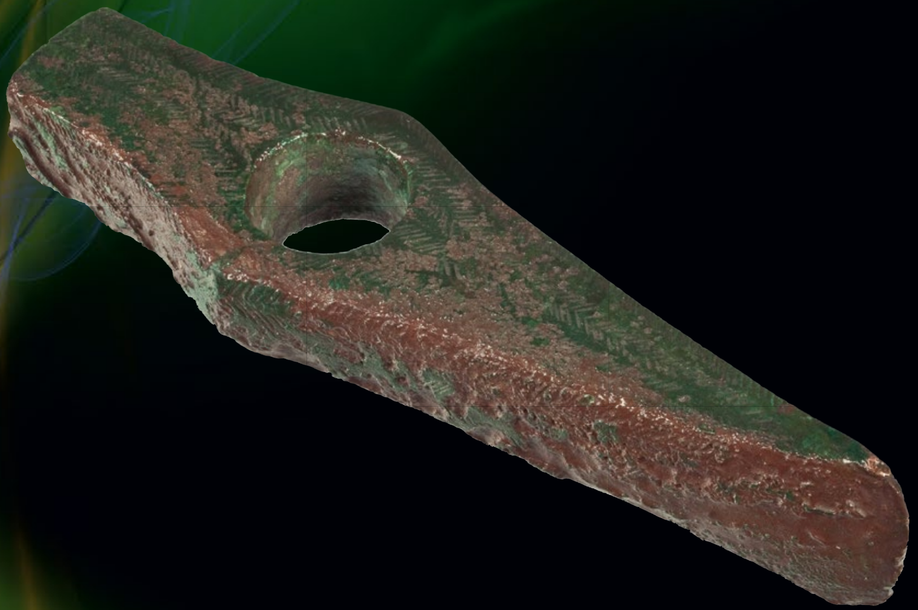
The Rise of Metallurgy in Eurasia

Evolution, Organisation and Consumption of Early Metal in the Balkans



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Cover: Ljiljana Dinić; Copper hammer-axe, type Pločnik, c. 4600 BC
(from Pločnik, Serbia) - Julka Kuzmanović Cvetković.
Inner back cover: Reconstruction of the world's earliest copper smelting. Green flames come from the extraction of metal from malachite. Experiments at Pločnik, Serbia (2013) - Marko Djurica

Copyeditor: Kate Sharpe

To the memory of Borislav Jovanović, our colleague, friend and inspiration

(1930 – 2015)

Contents

Chapter 5

Introduction to Belovode and results of archaeometallurgical research 1993–2012

Miljana Radivojević

The site of Belovode (44°18’42.34”N, 21°24’27.09”E) is located near the village of Veliko Laole, *c.* 140 km southeast of Belgrade (MAP) and lies on a windy plateau with the eponymous spring running through the settlement. The location is typical for a Vinča culture settlement: a large rolling plateau of ellipsoidal shape at an altitude of *c.* 200 m, suitable for agricultural activities as well as cattle breeding in the dense forests and pastures (Šljivar *et al.* 2006: 251–252). The nearby Mlava River runs deep into the volcanic mountain range called Homolje, which lies within a zone of primary copper mining and metallurgy (Krajnović and Janković 1995).

The site has been excavated since 1993 by the National Museum of Belgrade and the Museum in Požarevac (Šljivar and Jacanović 1996a, 1996b, 1997a; Jacanović and Šljivar 2003; Šljivar *et al.* 2006; Šljivar 2006). Given that the publication record for Belovode has been mainly limited to attempts to interpret and explain archaeometallurgical activities, a more detailed account of the history of research at the site will be the focus of this chapter.

In 2010, the site of Belovode received wide international recognition following a study of five copper slag pieces, identified as the earliest in the world (Radivojević *et al.* 2010a). Further analyses of archaeometallurgical materials excavated up to 2009 (Radivojević 2012, 2013; Radivojević and Kuzmanović Cvetković 2014) led, in 2012, to the establishment of one of the largest ever international collaborative projects focusing on Eurasian archaeometallurgy[1].

During almost two decades of excavations at Belovode, led by the National Museum in Belgrade, four building horizons were recognised within *c.* 3 m of cultural stratigraphy (Belovode A–D, Figure 1). These were found to correlate well with the entire Vinča culture sequence: Vinča Tordoš (A to B1) and the Gradac Phase (I–III) (Jovanović 1994; Šljivar 1993–2009; Šljivar and Jacanović 1996b) (see Chapter 4).

The internal phasing of Belovode was established on the basis of distinctive ceramic typology (Garašanin 1951, 1973), including locally recognised pottery variations (Arsenijević and Živković 1998; Šljivar and Jacanović 1996b). At its earliest stages, Belovode was most likely inhabited by Starčevo groups, as indicated by occasional finds of late Starčevo pottery. Several excavated dwellings yielded a great abundance of pottery sherds, covering all Vinča culture phases, along with stone tools, obsidian blades, and decorative items made of malachite (copper carbonate), bone and precious stones. Malachite, pottery and bones with green stains, and numerous ‘fired surfaces’ are also characteristic of this settlement and will be explored in detail below. The later occupation of this site by the Vinča culture ended by fire in *c.* 4650 BC, a practice that appears to be common not only for this culture, but across the Balkans at the time (Stevanović 1997). By the end of the 4th millennium BC, a section of the site was briefly re-occupied by the Late Chalcolithic Kostolac culture, recorded both in the excavated materials as well as by C14 dating (Jacanović and Šljivar 2003: 298; Radivojević and Kuzmanović Cvetković 2014). By 2011, *c.* 430 m^2 of the site had been excavated through 17 trenches, usually 25 m^2 in size (Figure 1).

Nine accelerator mass spectrometry (AMS) radiocarbon dates have recently been obtained from animal bones from Belovode, confirming the expected Vinča culture chronological span (cf. Radivojević and Kuzmanović Cvetković 2014; Whittle *et al.* 2016; also see Chapter 37, this volume). The probability distribution for the beginning of the Vinča occupation of Belovode indicates a date of *c.* 5350 BC, while the boundary for the end of the Vinča culture use of the site is set at *c.* 4650 BC. Of particular significance here is the dating of the earliest stratigraphic evidence for the extractive metallurgy in Belovode, which starts at around 5000 BC; this is currently the earliest secure date for copper metal production anywhere (Radivojević *et al.* 2010a). Importantly, it coincides with the intensive mining activities in Rudna Glava, which culminated in around *c.* 5000 BC in the vicinity of this site (*c.* 50 km).

The mining site at Rudna Glava does not, however, appear to have been exploited by the inhabitants of

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[1] https://gtr.ukri.org/project/D208DC64-842F-4E99-9C9E-248D8185D75A

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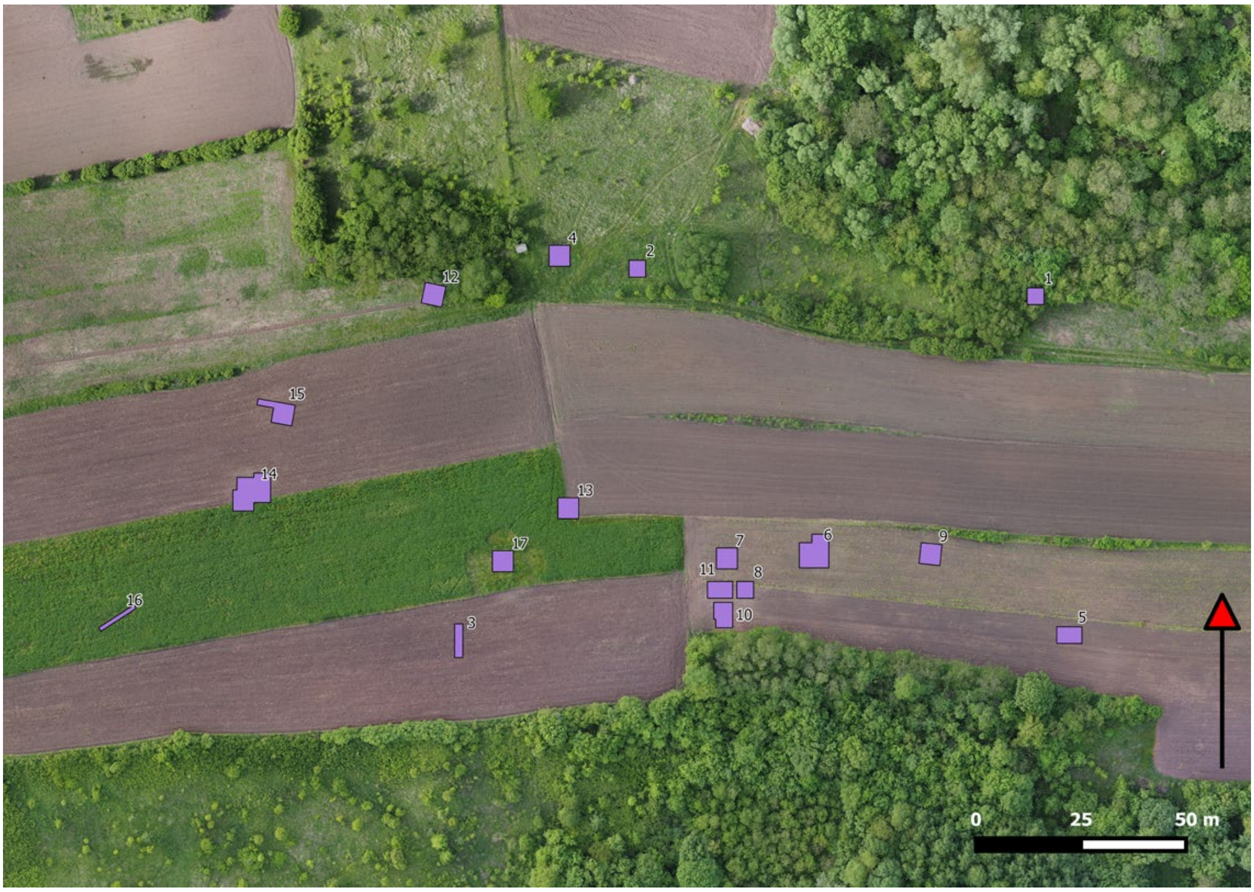

Figure 1. Plan of the Belovode settlement with locations of excavation trenches by 2011. Prepared by J. Pendić.

Belovode; another copper source, discovered at Ždrelo (Figure 2), *c.* 10 km away from Belovode, has been argued to be the most likely candidate according to lead isotope analysis (Radivojević *et al.* 2010a: 2781, Figure 10). It is notable that a total of nine Vinča culture settlements have been found in the wider catchment area of Belovode, prompting scholars to propose their association with the mining and metallurgical activities both at Belovode and in the wider area (Šljivar and Jacanović 1996b). This notion remains to be explored in future research.

Archaeometallurgical materials up to 2009

All materials considered below were discovered at the site of Belovode before 2009 (see Table 1). The majority of the studied collection consists of copper minerals and malachite beads, while the rest of the assemblage includes individual slag samples, slagged ceramic sherds and copper metal artefacts. The sample size appears small in comparison with the amount of technological debris (and slags in particular) in later prehistoric periods; however, it covers a crucial period in the evolution of metallurgy in Europe and, as a coherent assemblage, is unprecedented in size, quality and resolution. In order to address metal production in the Vinča culture, activities related to copper mineral use and pyrometallurgical activities are described here in three distinctive stages of the process: copper mineral processing, (s)melting debris, and the making and working of finished metal objects.

All copper minerals studied here are recognised as archaeological since they originated from archaeological sites (in contrast to geological minerals from the mines). Although bead minerals and ores in this study are both typically malachite, a rationale to distinguish between these was developed in a previous study of material from Belovode (Radivojević *et al.* 2010a) and relates to their differentiation in the technological treatment applied during processing. Thus, minerals most likely used for bead making at Belovode (i.e. 'cold' processing; Figure 3a) are referred to as 'copper minerals', while those most likely used for copper smelting (or 'hot' processed) will be termed 'copper ores'.

In this study, copper ores are assumed to include significant manganese content, as first indicated by previous chemical analyses of copper production evidence from Belovode (Radivojević *et al.* 2010a). Macroscopically, these ores appear green and black, where green comes from the colour of malachite and black from the manganese content (Figure 3b).

Figure 2a) Open-cast mine (?) at Ždrelo; b) Entrance at the shaft-hole in Ždrelo (photo by M. Radivojević).

Table 1. Study material from Belovode, arranged by analytical number. All samples starting with M (except for M3, M6, M10 and M35) have already been studied and presented in Radivojević 2007).

No.	Analytical No.	Year	Field label	Field context	Type of Material
1	Belovode 3	2007	Trench 13, spit 14	Household	Copper mineral
2	Belovode 9	1995	Trench 3, spit 12	Household	Malachite bead
3	Belovode 10	2001	Trench 8, spit 22	Household	Malachite bead
4	Belovode 12	2007	Trench 13, spit 10	Household- workshop?	Malachite bead
5	Belovode 13	2003	Trench 10, spit 27	Household	Malachite bead
6	Belovode 18	2007	Trench 13, spit 10	Household	Copper mineral
7	Belovode 23	2001	Trench 8, spit 23	Household	Malachite bead
8	Belovode 30a, 30c	1995	Trench 3, spit 5	(Building) waste pit	Slagged ceramic sherd
9	Belovode 31a, 31b	1995	Trench 3, spit 6	(Building) waste pit	Slagged ceramic sherd
10	Belovode 33b	2008	Trench 14, spit 15, surface 4	Household	Copper mineral
11	Belovode 34a	2008	Trench 14, spit 3	Household	Copper minerals from an amphora (3 pieces)
12	Belovode 131	1995	Trench 3, spit 6	(Building) waste pit	Copper slag
13	Belovode 134	1995	Trench 3, spit 7	(Building) waste pit	Copper slag
14	Belovode 136	1995	Trench 3, spit 5	(Building) waste pit	Copper slag
15	Belovode 154	1999	Trench 7, spit 6	Household - workshop?	Malachite bead
16	Belovode M3	1995	Trench 3, spit 8	(Building) waste pit	Copper mineral
17	Belovode M6	1995	Trench 3, spit 10	(Building) waste pit	Copper metal droplet
18	Belovode M10	1995	Trench 3, spit 19	Household	Copper mineral
19	Belovode M14	2002	Trench 9, spit 11	Household	Copper metal droplet
20	Belovode M32	1995	Trench 3, spit unknown	Household	Malachite bead
21	Belovode M35	1995	Trench 3, spit 17	Household	Copper mineral
22	Belovode M12	1998	Trench 6, spit 10	Household	Copper mineral
23	Belovode M13	2000	Trench 7, spit 18	Household	Copper mineral
24	Belovode M17	2004	Trench 10, spit 8	Household	Copper mineral
25	Belovode M20	1995	Trench 3, spit 2	(Building) waste pit	Copper slag
26	Belovode M21	1995	Trench 3, spit 4	(Building) waste pit	Copper slag
27	Belovode M22 (a,b)	1995	Trench 3, spit 5	(Building) waste pit	Copper slag
28	Belovode M23	1995	Trench 3, spit 7	(Building) waste pit	Copper slag

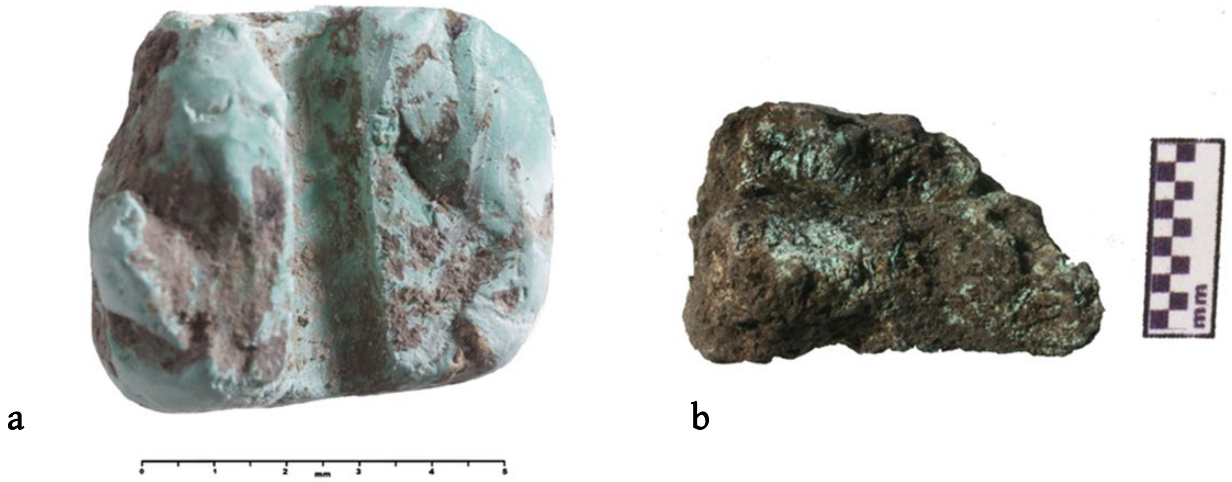

Figure 3. a) Typical bead malachite from Belovode; b) Typical black and green copper mineral from Belovode (Radivojević and Rehren 2015: Figure 2)

‘Smelting’ of copper ores refers to the primary extraction process, where the produced metal was usually further purified by refining or melting. Fundamentally, copper smelting can be separated into two discrete steps: the reduction of copper ore to copper metal, which requires reducing conditions and temperatures from *c.* 700 °C upwards (Budd and Ottaway 1991), and the melting of the copper metal, which requires temperatures around and in excess of 1083 °C. The melting or second step in this process has fewer constraints on the redox (reduction-oxidation) conditions, while smelting requires an oxygen partial pressure of less than $10^{-5,5}$ atm (cf. Elliott 1976). Thus, the balance between the two opposing tendencies, high temperature and reducing conditions, is the key to the successful smelt.

In the first step, the mineral (typically malachite) decomposes:

$$Cu_2CO_3(OH)_2 \rightarrow 2CuO + CO_2 + H_2O \ (< c.\ 500\ ^\circ C)$$

and the incomplete burning of charcoal provides the carbon monoxide which drives the reduction of copper oxide to copper metal:

$$2CuO + CO \rightarrow Cu_2O + CO_2$$

$$Cu_2O + CO \rightarrow 2Cu + CO_2$$

At temperatures < 1083 °C, copper precipitates in the solid state and forms a porous, spongy, powdery mass, still mixed with gangue minerals (Hauptmann 2007: 222; 2020). However, although relatively low temperatures are unlikely to allow for the separation of slag, there may well have been pockets of higher temperature directly in front of the blow pipes that led to localised fusion and partial melting of slag and metal. Thus, copper that formed during the first step was only transformed chemically and needed higher temperatures for a physical change from the solid powdery mass into liquid metal. The higher temperatures also facilitated full separation from gangue minerals, resulting in the formation of slag.

For the full melting of copper, temperatures of around *c.* 1100 °C needed to be maintained for a period sufficient to allow fully molten conditions to be reached throughout the charge. Such temperatures require an installation which retains and concentrates the heat generated from the fuel within the reaction container (Rehren 2003). This container can, for both stages, be a ceramic vessel such as a crucible or furnace, or a simple hole in the ground leaving little identifiable structure. Significantly, it is possible that both steps were performed in the same container or structure, facilitating the blending of one step into the other. They may also have been conducted separately, depending on various constraints originating from the physical, social or environmental spheres.

The common forms of smelting debris found at archaeological sites are installations, slagged ceramics and slags. Technical ceramics (crucibles, furnace remains, or *tuyères*) are particularly interesting for studying past metallurgical processes as they reflect technological choices. The most informative part of a crucible or an installation fragment is the slag attached to its walls, as is present on several 'slagged sherds' from Belovode (Figure 4b). One of the most valuable materials for studying metal production at the site are the small slag pieces (eight in total), discovered together with slagged sherds in a single trench (No. 3) (for a typical example see Figure 4a).

Slag is a waste product of high-temperature metallurgical processes, which 'collects' all unwanted substances or impurities from ores, the furnace lining and the charcoal ash within the smelting, casting or alloying systems. Chemically, it is a solution of molten oxides. Two major oxide components of metallurgical slags are silica (SiO_2) and iron oxide (typically the more reduced ferrous oxide, FeO), which are combined in wide varieties with elements such as manganese, magnesium or zinc. Other common constituents are lime (CaO) and alumina (Al_2O_3), followed by a number of minor compounds which contribute to the overall chemical composition. A slag of optimum composition has a low energy of formation, a low melting point and a high degree of fluidity (e.g. low viscosity) (Bachmann 1980: 118, 1982: 10). Slag can represent a single phase (glass), although ancient slag is most commonly constituted of a wide range of crystals. The newly formed compounds in the ancient slag are usually referred to as 'phases' or 'crystals' rather than minerals, even if they are structurally identical, since they are formed by anthropogenic processes.

Slag is ideal for studying past pyrometallurgical activities because it typically contains information about all components affecting its formation. The slag chemistry preserves details of the conditions of the smelt and composition of ores, indicates which metal was extracted, the contribution of fuel ash, potential fluxes, and even the design of installations (Rehren *et al.* 2007). Moreover, slags are ideal for study because they are highly resistant to weathering conditions and are usually accessible for invasive analysis by archaeological scientists. In the archaeological record, early slags are typically formed on top of the metal in a crucible or lined container (Tylecote *et al.* 1977: 307) and may thus be found as free pieces or attached to the walls of an installation. However, the free slag samples can be easily overlooked or mistaken for clay or minerals, as was the case with copper slags from Belovode (Radivojević *et al.* 2010a: 2779). Also, the slags from early periods were often crushed in the search for metallic prills for further refining, making them archaeologically less visible.

The artefact group at Belovode consists of malachite beads and copper metal items. Malachite beads were commonly processed in a series of 'cold' shaping techniques, applied

Figure 4. a) Belovode slag (sample 134); b) Belovode slagged sherd (sample 31b)

in several technological steps. The ornament (bead) making process starts with a raw nodule of mineral which, once roughly shaped (but not perforated), is recognised as a roughout. Subsequent fine chipping creates a preform or blank, and this is commonly followed by drilling to produce the final bead shape (Lankton *et al.* 2003: 16). Notably, the order of this sequence varies both culturally and in relation to the materials used. The variety present within the copper metal artefacts group is particularly informative for the metallurgical *chaîne opératoire* in the Vinča culture, allowing the investigation of different sequences of production and their interpretation within wider cultural, environmental and physical surroundings.

All technological analyses were carried out at the Wolfson Archaeological Science Laboratories at the UCL Institute of Archaeology, by the primary author of this paper, and under the supervision of Professor Thilo Rehren (then UCL Institute of Archaeology, currently at The Cyprus Institute in Nicosia, Cyprus). Below I present the main results, while the full dissertation paper is available in Appendix B_Ch5.

Results

Copper mineral use

Malachite beads, pendants and 'green' copper minerals are present from the earliest occupation of Belovode and continue throughout all building horizons. These most numerous finds at the site are usually uncovered and mixed with ash and pieces of charcoal. Other contexts include house floors, storage jars, ceramic sherds (with malachite adhered to them), or workshops within a household. Two such areas in Trenches 12 and 13 together yielded *c.* 2.5 kg of copper minerals throughout all building horizons, equivalent to almost one third of the total weight of malachite finds discovered at this site[2].

Ten samples from the copper minerals group were analysed, three of which came from the so-called 'metallurgical' Trench 3 (Radivojević *et al.* 2010a); the rest originated from various household contexts (Radivojević and Rehren 2016) (see Table 1). All samples, barring M3, M13, M17, 33b and 3, were confirmed to be malachite (copper carbonate) with significant levels of manganese in their composition (Radivojević 2013 18 ff., 2015). The remaining copper mineral samples were sourced from a vein containing cuprite with copper sulphides. The small size of all mineral samples (*c.* 1–3 cm) may imply that they were beneficiated, i.e. crushed to facilitate smelting.

Significantly, both oxidic and sulfidic minerals present a common feature: their colour is distinctively black (or dark) and green (see Figure 3), which is argued to reflect an intentional choice of a colour that appealed to the Belovode metallurgists (Radivojević and Rehren 2016). The only group of minerals from Belovode that were not black-and-green were those used for malachite bead making, implying that colour-coding was a very important aspect for copper mineral selection. Although the geological environment of eastern Serbia offers more than one kind of copper mineral (Krajnović and Janković 1995), the Belovode copper craftsmen seem to have selected only pure green or black/dark and green minerals.

Figure 5. Stone mallet from Trench 7 at Belovode (after Šljivar *et al.* 2006: plate I/4)

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[2] The author weighed all malachite from Belovode for the purpose of her PhD research completed in 2012 at the UCL Institute of Archaeology.

Two stone mallets, each with a central groove, were discovered in the context of workshops in Trenches 1 and 7 (Figure 5) (Šljivar *et al.* 2012: 259, Plate I/4), and could offer clues about tools used during processing. Similar finds from Rudna Glava (Jovanović 1982) suggest such tools may also have been used for mining.

(S)melting debris and installations

Charred surfaces with malachite, copper mineral powder, adhering to fragmented ceramic sherds and grooved stone mallets are common in household contexts in Belovode. A few small pottery vessels of conical shape and coarse fabric from Trenches 7 and 8 (Vinča B1 horizon, Figure 6) also had green minerals attached to the outer surface however analyses have shown that these were not crucibles (Radivojević 2007). In addition, a fragment of ceramic mould discovered on the site surface, is thought to originate from the latest layer of occupation (Šljivar *et al.* 2006: 259, Plate I/3).

Two shallow pits rimmed with ceramic sherds and a burnt layer of clay from Trenches 10 and 13, a Vinča B1 building horizon, are identified as early furnaces by the excavator (Figure 7), based on comparison with similar small hearths discovered at Durankulak in Bulgaria from the late 5th millennium BC (Šljivar *et al.* 2006: 253, 260, Plate II/4; cf. Dimitrov 2002). Elongated cylindrical ceramic forms (so-called 'chimneys') with a diameter of about 20 cm, a reconstructed height of up to 80 cm, and open at both ends (Figure 8) have been tentatively linked to these rimmed pits in the ground and, thus with the smelting operation (Šljivar 2006; Šljivar *et al.* 2006: 253). Nevertheless, neither of these 'chimneys' showed convincing traces of use in the smelting process after compositional analyses (Radivojević *et al.* 2010a: 2779).

Metal production evidence

Pyrometallurgical activities at Belovode are represented by only eight individual copper slag samples, four slagged ceramic sherds, and a copper droplet (metal) sample from Trench 3, all of which demonstrate sustained smelting activities taking place at the outskirts of the site. Another pyrometallurgical situation has been recovered in Trench 9; both will be

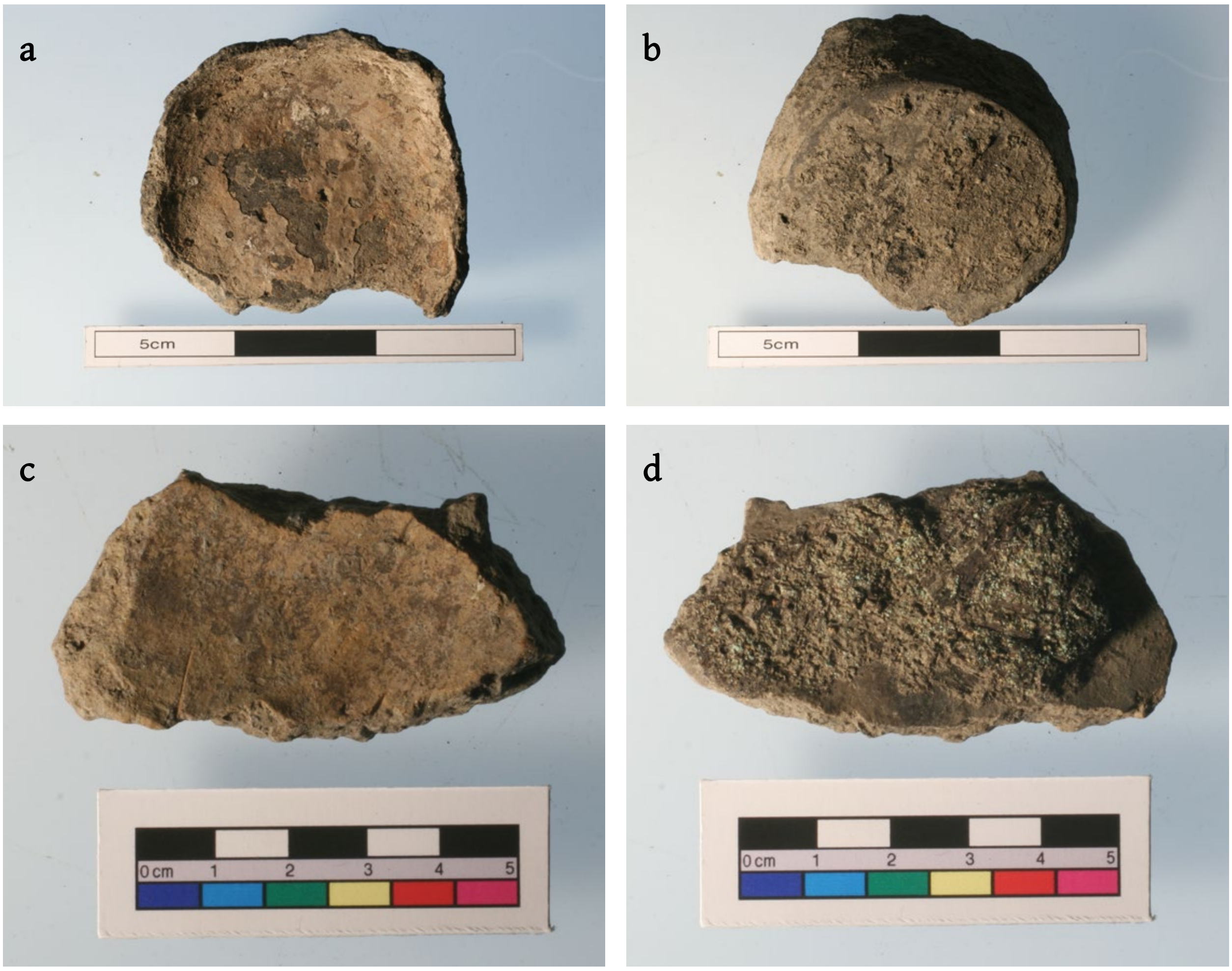

Figure 6. a, b) Fragment of ceramic vessel with green powder adhering to its bottom (labelled as Belovode 30ch, Trench VIII, spit 12, Belovode 2001 campaign, reported in Radivojević 2007); c, d) Bel29 Fragment of ceramic vessel with light green and brown matrix adhered to the outer walls (labelled as Belovode 26, Trench VII, spit 12, Belovode 2000 campaign, reported in Radivojević 2007).

Figure 7. Shallow pit lined with ceramic sherds from Trench 10 (upper left corner) (Photo courtesy of Duško Šljivar, National Museum Belgrade, Serbia).

presented in more detail below. These materials were usually discovered in areas filled with ash, charcoal, charred wood or stone constructions and mainly represent an outdoor activity.

Trench 3

Trench 3 (dimensions 8 m x 2 m) yielded evidence for copper smelting activities throughout the final, D horizon of occupation i.e. across the entire Gradac Phase sequence starting in *c.* 5000 BC (Radivojević *et al.* 2010a). This phase of the Vinča culture is known to have lasted longer in the Morava Valley settlements than in those situated nearer to the Danube (Jovanović 1994) and, at this site, most likely covers the late Vinča culture sequence, dated to *c.* 5000–4650 BC.

The Belovode D horizon, represented in this trench by materials from a waste pit (Šljivar and Jacanović 1996a), includes all finds from spits 1–11, including various archaeometallurgical debris (Table 2). Thousands of ceramic finds were unearthed in this horizon alone, many of which are diagnostic of the Gradac Phase in general, for example, the conical bowls with a thickened rim channelled on the interior, or tri- and four-partite vessels with a cone-shaped neck and protruding shoulder, usually accompanied by ornaments with typical incised ribbon decoration (Arsenijević and Živković 1998; see also Schier 1996; Schier 2000).

The slag pieces collected from this trench are vitrified, strongly magnetic and green-stained droplets, not exceeding 1 cm in length (example in Figure 4a). They appear to have been highly viscous and very rich in copper metal, however with no signs of crushing in pursuit of copper metal prills. This may have been due to their small size and weight, since all eight samples weigh, in total, less than 10 g. Given that, in appearance, these slag pieces resemble (green) malachite as a result of the corrosion of the copper metal prills entrapped in them, it is possible that the green colour facilitated their recognition in the field excavations, leading to a biased recovery in favour of more copper-rich pieces, and that those without green staining were more likely overlooked, as mentioned above (cf. Radivojević *et al.* 2010a: 2779).

The green staining on fragmented slagged sherds comes from the contact of these samples with the metallurgical process. The slagged mass is surrounded by heavily vitrified areas, which appear along the edges of the studied samples (Figure 4b), but also extend across their cross-sections. The latter implies that these sherds were most likely used in a fragmented form during the metallurgical process. A copper metal droplet (M6), found in addition to this assemblage, provides a more complete account of the types of metallurgical activities conducted in the workshop in Trench 3.

All metallurgy-related materials were discovered sealed with building waste such as the remains of house daub, domestic pottery and animal bones. Notably, in spit 10, which belongs to this building horizon, two shallow, rock-lined constructions, indicated as fireplaces were identified as potentially linked with metallurgical debris in excavation records. The stratigraphic evidence related to the earliest slag piece is dated to *c.* 5000 BC; the smelting evidence, according to the excavation reports, continued until the abandonment of the site in *c.* 4650 BC.

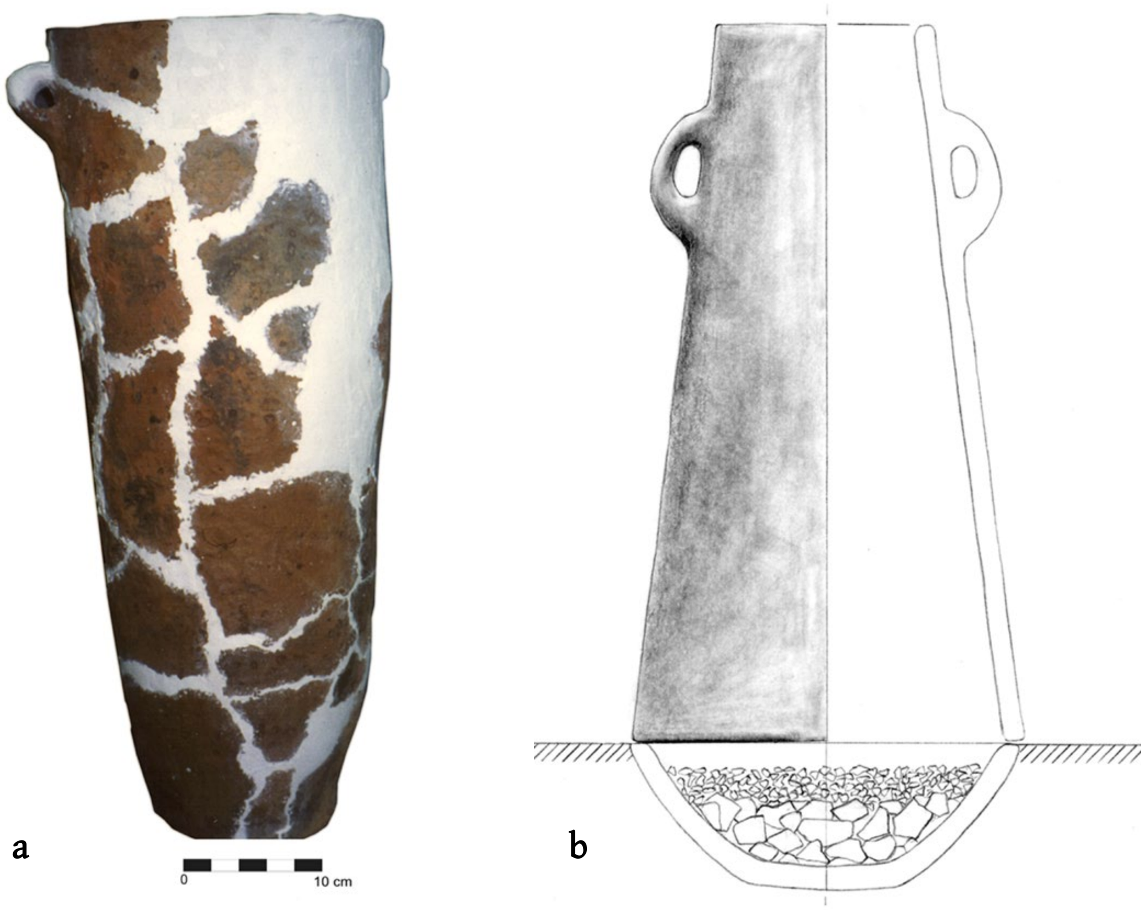

Figure 8. a) Reconstructed ceramic 'chimneys' from Belovode. Note the marks of secondary burning. (Photo courtesy of Duško Šljivar, National Museum Belgrade, Serbia); b) Tentative reconstruction of the smelting installation at Belovode (after Šljivar *et al.* 2006: plate II/5).

Table 2. Study materials from Belovode 'metallurgical' Trench 3, arranged by spits.

No.	Analytical No.	Year	Context	Type of Material	Chronology/building horizons
1	Belovode M20	1995	Trench 3, spit 2	Copper slag	Gradac Phase, Belovode D
2	Belovode M21	1995	Trench 3, spit 4	Copper slag	Gradac Phase, Belovode D
3	Belovode M22 (a, b)	1995	Trench 3, spit 5	Copper slag	Gradac Phase, Belovode D
4	Belovode 136	1995	Trench 3, spit 5	Copper slag	Gradac Phase, Belovode D
5	Belovode 30 (a,c)	1995	Trench 3, spit 5	Slagged ceramic sherd	Gradac Phase, Belovode D
6	Belovode 31 (a,b)	1995	Trench 3, spit 6	Slagged ceramic sherd	Gradac Phase, Belovode D
7	Belovode 131	1995	Trench 3, spit 6	Copper slag	Gradac Phase, Belovode D
8	Belovode 134	1995	Trench 3, spit 7	Copper slag	Gradac Phase, Belovode D
9	Belovode M23	1995	Trench 3, spit 7	Copper slag	Gradac Phase, Belovode D
10	Belovode M3	1995	Trench 3, spit 8	Copper mineral	Gradac Phase, Belovode D
11	Belovode M33b	1995	Trench 3, spit 8	Malachite bead	Gradac Phase, Belovode D
12	Belovode M6	1995	Trench 3, spit 10	Copper metal droplet	Gradac Phase, Belovode D
13	Belovode 9	1995	Trench 3, spit 12	Malachite bead	Vinča B1, Belovode C
14	Belovode M35	1995	Trench 3, spit 17	Copper mineral	Vinča B1, Belovode C
16	Belovode M10	1995	Trench 3, spit 19	Copper mineral	Vinča B1, Belovode B

Compositional and contextual analyses of archaeometallurgical assemblage from Trench 3 indicated that they were probably part of a minimum of three smelting episodes (Radivojević and Rehren 2015: 22 ff.). This assumption is further strengthened by the compositional analyses, which indicate strong correlations in the ore signature for production evidence, i.e. manganese and zinc. Manganese is specifically known for the advantageous chemical-physical properties of its oxides, which enable an easier reduction of copper ores to metal and slags (Huebner 1969: 462, Figure 3). The aesthetic aspect of copper and manganese rich ores (being black and green) has been argued to be particularly important, as this colour code appears to dominate the Vinča culture metallurgy of both copper and tin bronze making (Radivojević *et al.* 2013).

All slag-based samples appear very heterogeneous under the microscope with dross areas (copper-oxide-based phases) dominating the microstructure (Radivojević and Rehren 2015: 16). Other major phases are metal prills, spinels and delafossite, all of which are embedded in a glassy matrix (for comparison and detail, see Chapter 11). The co-occurrence of these phases suggests that the gas atmosphere had a partial oxygen pressure that was low enough to reduce cuprite to copper metal (Elliott 1976) and was therefore sufficient to smelt copper. The working temperatures of the smelting systems across all slag-based samples is estimated to have been just over 1083 ºC, according to the fully molten state of copper metal prills embedded in them.

Trench 9

Pyrometallurgical activities are also recorded in Trench 9 (dimensions 5 m x 5 m) (Jacanović and Šljivar 2003). In this trench, spits 21–7 yielded typical Vinča culture material that corresponds with all four building horizons at Belovode (A–D). Archaeological materials found in spits 6–1 belonged, most probably, to the latest manifestation of the Vinča D phase (cf. Jovanović 1994).

The use of copper minerals occurred regularly throughout the building horizons Belovode A–C, excluding Belovode D and the succeeding, not yet well-defined, horizon. Of particular note here is spit 11, which yielded a copper metal droplet (M14, see Table 1). This was discovered in the context of the regular appearance of ceramic pedestal bowls, typical of the Vinča A to Gradac Phase. Chronologically, and in relative stratigraphic terms, it can be correlated with the early Gradac Phase and, effectively, with the start of metallurgical activities in Trench 3, dated to *c.* 5000 BC (cf. Radivojević *et al.* 2010a). Interestingly, early Belovode horizons in this trench yielded Vinča culture figurines with modelled appliques: necklaces with perforated disc-pendants (Šljivar *et al.* 2012: 31, Plate III/1-4) resembling gold applications from the late 5th millennium BC burials and settlements in Bulgaria and Ukraine. Similar examples have also been discovered at the site of Vinča-Belo Brdo (Tasić 2008a: 151, Figure 58).

An unusually large, round slag cake was unearthed in spit 18 (Belovode 40) and argued as firmly contextualised within the Vinča B1 Phase (Šljivar *et al.* 2012: 33–34, Pl. VIII/1)(Radivojević and Kuzmanović Cvetković 2014) (see also Chapter 3, this volume). Preliminary analyses revealed that this artefact has significant concentrations of lead (Radivojević and Rehren: 2019), which may suggest the production of this metal even before copper; future analysis is expected to shed more light on this unusual archaeometallurgical evidence. In terms of absolute dating, the metal droplet (M14) could be dated to *c.* 5000 BC, while the slag cake (Belovode 40) may be ascribed the date of *c.* 5200 BC (Radivojević 2013: 17; also Chapter 3, this volume).

Copper mineral and copper metal artefacts

Malachite beads occur throughout all horizons at Belovode and vary in size from 4 mm to 1.5 cm in diameter. One exception is a deltoid pendant with perforation found in Trench 1, Vinča A1 horizon. Beads selected for this study were found related to various copper minerals, slag pieces, workshop activities or dwelling structures. They occur as whole artefacts or fragments, implying the potential presence of a bead workshop at the site. Some Belovode beads have undergone mineralogical study, which confirmed the presence of malachite, with traces of tenorite and kolwezite (Jović 1996). Compositional analyses further suggested the use of rather pure malachite for bead making, with main impurities being low contents of iron and zinc oxides, as well as manganese in one instance (Radivojević 2012: 306, Table 48).

Both copper metal droplets (M6 and M14) were initially thought to be copper minerals until their cross-sections revealed a dark red phase (copper metal) surrounded by a thick, light green layer of corrosion (Radivojević *et al.* 2010a; Radivojević 2013: 28, Figure 18). The amorphous shape of the metal phase in both samples indicates that it was once molten copper, however it is now heavily corroded due to post-depositional processes. Both droplets most likely cooled rapidly, as indicated by porosity holes and cracks throughout the investigated polished sections.

In addition to these metal droplets, two further copper artefacts have been identified as belonging to the Vinča culture occupation of Belovode: a copper chisel and a bun-shaped metal ingot (Šljivar *et al.* 2006: 252, 269, Plate I/1, 2). Since these were found in the vicinity of the site, they could belong to the Late Chalcolithic occupation, as already indicated by the late 4th millennium BC dates from the top horizon in a defined area of this settlement.

Discussion

Extensive microanalytical examination of copper minerals and production debris indicate that, together, they form a coherent assemblage, largely linked through significant manganese and zinc content, which came from the gangue minerals in an exploited copper deposit.

It is noteworthy that, regardless of how insignificant the small amount of slag from Belovode may appear, it fits the overall picture of rather ephemeral production evidence in pre-Bronze Age metallurgy (Craddock 2001: 152). The early slags of the 5th and 4th millennium BC mostly look the same: they weigh hardly more than a few grams each and reach nut size at most (cf. Hauptmann 2007: 158); it is not surprising that, size-wise, the earliest documented slags sit at the lower end of the range of slag 'heaps' identified thus far (Bourgarit 2007: 4, Table 1).

The Belovode smelting installation debris also fit well the 'ephemeral model' of Chalcolithic metallurgy. Slagged sherds from the site suggest the presence of a hole-in-the-ground installation lined with broken pottery. However, none were discovered in situations related to a hearth or a similar detectable feature in the field. Hence, it may be hypothesised that the copper smelting installation was too ephemeral to survive *c.* 6000–7000 years of post-depositional processes and took the form of a shallow indentation in the soil, lined with ceramic sherds. Such an installation was possibly operated by using blowpipes or *tuyères*, where five to six blowpipes would normally suffice to bring the temperature to around the 1100–1200 ºC needed to (s) melt copper (cf. Rehder 1994: 221).

The chemical fingerprint of the ore used for smelting copper at Belovode indicates a strong chemical association of Mn and Zn, with some Co, Ca and Fe (Radivojević 2013: 21 ff.). It is likely, therefore, that the ores used could have been a paragenesis of copper-zinc-manganese, with some other elements coming from the attached primary copper sulphide mineralisation (such as S, Fe) or gossan. Their inclusion in the ore charge may not have been intentional, but most likely evolved as a natural consequence of the stratigraphy of weathered copper sulphide ore bodies (cf. Rostoker *et al.* 1989: 85).

With regard to the copper droplets M6 and M14, the structural difference between them results from M6 containing a sulfur-rich phase. Thus, it may be assumed that the smelted ore in M6 was originally a mineral combination of copper sulphides (chalcocite) and oxides (or carbonates). This assumption is corroborated by the presence of a similar mineral combination at Belovode (3 and 33b), which suggests that it was these types of 'mixed' minerals that were included in smelting activities at the site (Radivojević 2013: 30).

Conclusion

The overview of activities related to copper mineral use and extractive metallurgy at the Vinča culture site of Belovode suggests that metal smiths at the settlement were covering several stages of the metallurgical *chaîne opératoire*. The analytical highlights underlying this narrative are the compositional connection of copper minerals and production debris from Trench 3 through manganese-rich copper ores, the potential presence of a minimum of three separate smelting events in Trench 3 and identification of both slagging and non-slagging events in the earliest context of metallurgical development at the site.

The black and green minerals (both oxidic and sulfur-rich) selected by the Belovode miners predate the earliest documented smelting event at this site, indicating that they were potentially experimented with during the first centuries of occupation. This experimentation, although probably unsuccessful, could be recognised in sample M6, which contains molten copper, but also some residual sulfur-rich phases. The distinctively coloured black and green copper minerals became copper ores only at the dawn of the 5th millennium BC, and their smelting is attested by the strong presence of manganese and zinc in the chemical signatures of glassy slag matrices as well as in other newly formed phases in the metal production samples.

The combination of analytical results and the available fieldwork data facilitated recognition of potentially three separate smelting events in Trench 3, one of which, represented by M6, was most likely unsuccessful. Firstly, the data demonstrates sustainable smelting activities during at least the latest building horizon in Belovode (*c.* 5000–4600 BC), while sample M6 was produced somewhat earlier. Secondly, the data shows a similar technological principle for the slagging events, but also highlights that the early beginnings of metallurgy were not producing slag, as previously assumed by Craddock (1995). Still, it was not long before the Belovode metal smiths optimised metal extraction by producing minute concentrations of slag, documented at the turn of the 5th millennium BC (Radivojević *et al.* 2010a).

It appears that the Belovode metal smiths were aware of the properties of black and green copper minerals, knowledge that possibly developed over the course of a few centuries. This understanding related not only to manganese-rich copper minerals, but also to those rich in sulfur, indicating that it was the distinctive colour of the minerals that prompted their selection and subsequent smelting. The colour appeal of copper ores has already been argued to be the most significant sensory aspect for the invention of early metallurgical activities (Radivojević 2015) and the Belovode example stands out as the earliest currently known in the line of evidence assembled for the Vinča culture.

Small-scale smelting reactors emerge as the principal technological choice in the copper metal production, not only for Belovode but also throughout the Vinča culture. The minute size of slags and ephemeral evidence of smelting installations has also been discussed in Radivojević and Rehren (2016), who pointed towards a similar technological principle of copper production being present across the Vinča culture settlements of Belovode, Vinča, and Gornja Tuzla. The smelting process at all three sites can be generally characterised by the use of mixed copper ores selected for their colour, which were smelted in moderately reducing/partially oxidising conditions in an ephemeral 'hole-in-the-ground' installation. The process was conducted at all three Vinča culture sites with remarkably similar mastery, resulting in copper metal probably being produced in globules together with small quantities of highly viscous slag; some of this slag formed in direct contact with already-broken pottery sherds.

Although the Vinča culture production evidence studied here represents only very few of these episodes, the replication of the production pattern across all three sites within different occupational sequences indicates that the level of expertise achieved remained relatively unchanged and potentially stagnant across an estimated period of six centuries. The process slowly evolved into smelting of more complex copper ores only towards the end of this culture, as attested by the use of colourful complex copper-tin bearing ores for making tin bronze objects at the site of Pločnik around the mid-5th millennium BC (Radivojević *et al.* 2013). The distinctive role of black-and-green ores in early Balkan metallurgy therefore emerges as a key factor to support arguments for its independent development, as well as supporting claims about its unique technological trajectory.

One of the most significant outcomes of the results discussed here is to demonstrate the potential of a materials science approach for incorporating insufficiently contextualised materials into an integrated technological and archaeological narrative. The lack of precise contextual information is a result of insufficiently detailed documentation for the Belovode excavation. Despite careful excavation, records are not always clear and, unfortunately, do not offer sufficient information regarding the formation of the site. Since the majority of field documentation between 1993 and 2012 remains unpublished, relevant contexts for this study are limited to relative spits within individual trenches. This has provided only limited information on the spatial distribution of metallurgical debris and its relation to the spatially closest settlement features. Importantly, most of these features originated from arbitrary units, where the relationships are obscured and not always straightforward. This chapter therefore focused on the material properties of a variety of excavated artefacts, highlighting important connections between them from this particular perspective, rather than relying on a distribution plan of small trenches scattered across the site.

A further problem in relation to the state of research at Belovode arises from the lack of AMS data for specific contexts related to diverse metallurgical activities. AMS data for this site, recently synthesised in Whittle *et al.* (2016), do not offer sufficiently high resolution to distinguish copper mineral use and pyrometallurgical processes at Belovode (and other Vinča culture settlements), nor do they provide direct dating evidence for the majority of metallurgy-related finds. Therefore, the temporal analysis of metallurgical activities presented here has been mainly dependant on the relative chronology— based on specific pottery forms—and the conventional periodisation of the Vinča culture across the entire region. Such an approach, although potentially not without errors, currently provides the only feasible contextual framework for most metallurgy-related samples considered here.

An understanding of the relationship between the Gradac Phase and the rise of metallurgy is crucial for our interpretation of this process within the Vinča culture: the earliest stratigraphic evidence for copper smelting is discovered at the beginning of this phase and is also contemporary with the intensified activities in Rudna Glava, as well as the earliest dated copper implement from Pločnik. The changes in material culture that follow this phase are a phenomenon common across the whole Balkan region (cf. Garašanin 1994/1995) and will require particular attention for the interpretation of the origins of metallurgy in this part of Eurasia in future discussions.

The introduction of metallurgy evidently influenced other aspects of material culture at this time. The most important association is, beyond doubt, with pottery production, such as the conjunction of the appearance of black burnished ware and graphite painted decoration with the emergence of metallurgy, which has been discussed at length (e.g. Renfrew 1969; Gimbutas 1976a), and studied in detail within this volume (see Amicone *et al.* Chapters 14, 29 and 43, this volume) and in Amicone *et al.* (2020). Another valuable observation comes from Belovode, where modelled applications on figurines resemble contemporaneous metal jewellery from sites located along the lower Danube and further towards the northern Black Sea coast (see examples in Dumitrescu 1961; Sergeev 1963). This highlights the importance of the Danubian communication route for the spread of metallurgy across the Balkans, also highlighted in the most recent application of complex networks methodology by Radivojević and Grujić (2018).

The following chapters will explore in greater detail both the technological and archaeological circumstances for the appearance of the world's earliest metallurgy, providing a more nuanced contextualisation and chronological framework for the invention widely argued as the trigger for broader material and social changes, both in the Vinča culture and beyond.

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Appendix

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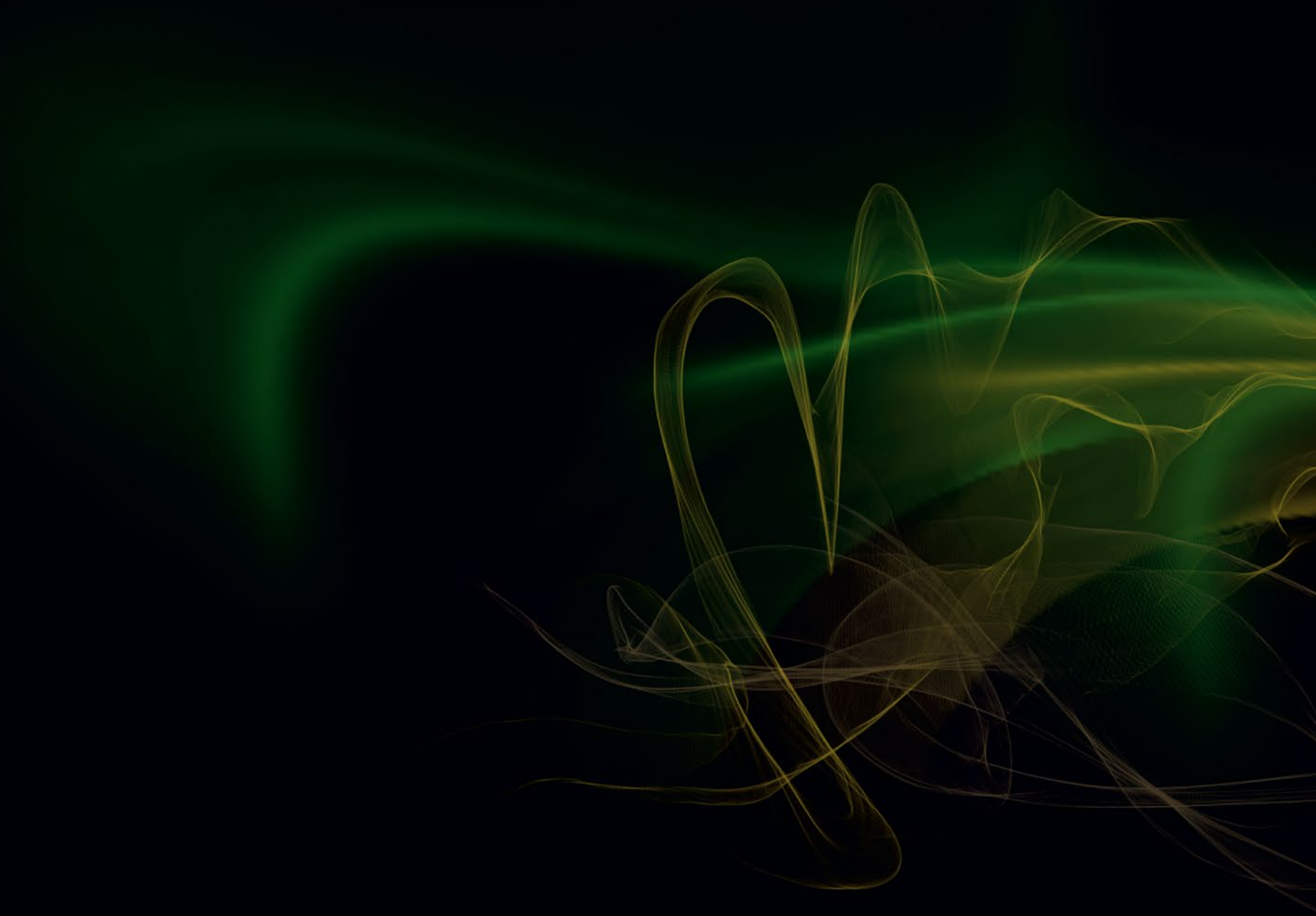

The Rise of Metallurgy in Eurasia is a landmark study in the origins of metallurgy. The project aimed to trace the invention and innovation of metallurgy in the Balkans. It combined targeted excavations and surveys with extensive scientific analyses at two Neolithic-Chalcolithic copper production and consumption sites, Belovode and Pločnik, in Serbia. At Belovode, the project revealed chronologically and contextually secure evidence for copper smelting in the 49th century BC. This confirms the earlier interpretation of c. 7000-year-old metallurgy at the site, making it the earliest record of fully developed metallurgical activity in the world. However, far from being a rare and elite practice, metallurgy at both Belovode and Pločnik is demonstrated to have been a common and communal craft activity.

This monograph reviews the pre-existing scholarship on early metallurgy in the Balkans. It subsequently presents detailed results from the excavations, surveys and scientific analyses conducted at Belovode and Pločnik. These are followed by new and up-to-date regional syntheses by leading specialists on the Neolithic-Chalcolithic material culture, technologies, settlement and subsistence practices in the Central Balkans. Finally, the monograph places the project results in the context of major debates surrounding early metallurgy in Eurasia before proposing a new agenda for global early metallurgy studies.

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