## Supplementary Information

## Community structure of copper supply networks independently evaluates the archaeological record of the 7th – 4th millennium BC Balkans

M. Radivojević\*

<sup>1</sup>McDonald Institute for Archaeological Research, University of Cambridge, Downing Street, CB2 3ER Cambridge, UK \*Corresponding author: <u>mr664@cam.ac.uk</u>

J. Grujić<sup>\*</sup>

<sup>2</sup>Vrije University Brussels, Artificial Intelligence Lab, Pleinlaan 2, Building G, 10th floor & MLG, Département d'Informatique, Université libre de Bruxelles, 1050 Brussels, Belgium \*Corresponding author: jgrujic@vub.ac.be

**Data.** For networks analyses we used published and unpublished sets of compositional data for 410 copper-based objects under consideration, spanning c. 6200 to c. 3200 BC. These were assembled from publications of Pernicka et al. [1, 2], Radivojević et al. [3], Radivojević [4] and UK's AHRC-funded "Rise of Metallurgy in Eurasia" project (hosted by the UCL Institute of Archaeology, no. AH/J001406/1)[5]. All data originate from the analytical set up of a single laboratory, Centre for Archaeometry in Mannheim, Germany, led by Professor Ernst Pernicka. The data from 410 objects are presented with a unique laboratory number (given by Centre for Archaeometry) and include the following types of materials (Table S1): copper mineral (30 in total), mineral ornament (17 in total), production evidence (smelting/casting, 22 in total), metal ornament (99 in total), and metal implement (242 in total). The metal implement type some instances contained information on the type of axe, included in a separate column (no data labelled as: *unk*). Besides a unique geographical location (given as latitude and longitude in degrees), sites are ascribed the following regional codes: SRB (Serbia), W (West Bulgaria), THR (Thrace), RHD (Rhodope), NC (North-central Bulgaria), NE (North-east Bulgaria) and BSC (Black Sea Coast).

Chronological and cultural attribution of studied materials was ascribed based on available relative and absolute dating in the area under consideration [1-3, 6-26]. Seven cultural periods were designated for this study: Early/Middle Neolithic (Period 1, 6200-5500 BC), Late Neolithic (Period 2, 5500-5000 BC), Early Chalcolithic (Period 3, 5000-4600 BC), Middle Chalcolithic (Period 4, 4600-4450 BC), Late Chalcolithic (Period 5, 4450-4100 BC), Final Chalcolithic (Period 6, 4100-3700 BC) and Proto Bronze Age (Period 7, 3700-3200 BC). We would like to emphasise that there is not a general consensus on the relative vs. absolute chronology of existing cultural phenomena observed here amongst (Balkan) archaeologists; thus, the entries on chronology in Tables S1 and S3 should be taken as tentative interpretations based on the latest chronological update in the field.

<u>Period 1. Early/Middle Neolithic (EN, 6200-5500 BC).</u> This period is represented only with the finds belonging to the Neolithic occupation of sites in eastern and western Serbia (proto/Starčevo culture) (see Figure 7a, Table S3). The use of malachite at the time of the introduction of agriculture or domestication is not uncommon, and similar examples have been documented in the Near East. However, the minerals listed here are unique since their function remained unknown, although the provenance analyses indicate their origin from local, eastern Serbian sources [18].

Period	C14 dates	Vojvodina	Central Balkans	West Bulgaria	South Bulgaria	Muntenia	North-east Bulgaria	Black Sea Coast (west)
Proto Bronze Age Final Chalcolithic	3200 3700	Boleráz Bodrogkeresz	Cernavoda III Salcuța IV ztúr KSBh	Galatin	Yagodina	Cernavoda III Cernavoda I	Usatovo Cernavoda I/Pevets	Cernavoda I
Late Chalcolithic Middle	4100	Tiszapolgár / KSBh Vinča D		Krivodol-Salcuţa- Bubanj hum (KSBh)	Karanovo VI Marica IV	Kodžadermen- Gumelniţa- Karanovo VI Boian-Spanţov	Kodžadermen- Gumelniţa- Karanovo VI	Varna III Varna I
Chalcolithic								Varna II Hamangia IV
Early Chalcolithic	4600	Vinča D Vinča C		Gradešnica Dikilitash-Slatino	Marica III- Karanovo V	Boian-Vidra	Poljanica	Sava / Hamangia III
Late Neolithic	5000	Vinð Vinð	ia B ia A	Kurilo/Akropotamos Topolnica	Karanovo IV Karanovo III	Boian III	Hotnica	Hamangia II Hamangia I
Early Neolithic	5500 6200	Star Lepensk	čevo :i Vir III					

Table S3. Relative and absolute chronology of malachite and metal-using cultures in the Balkans,  $7^{th} - 4^{th}$  mill BC. Green font stands for using copper minerals (e.g. malachite beads), red for metallurgical materials (e.g. metal artefacts, slags). The shaded fields indicate the periods and regions covered in this study.

<u>Period 2. Late Neolithic (LN, 5500-5000 BC).</u> This period is linked with the emergence of archaeological cultures that would grow into large metal producing and consuming phenomena (like Vinča in Serbia or Karanovo in Bulgaria) during the  $5^{th}$  millennium BC [27]. While Vinča culture occupied most of the central Balkans, the Karanovo phenomenon emerged in central Bulgaria and expanded significantly in the second half of the  $5^{th}$  millennium BC, including territories from the Black Sea coast to Thrace. With settlements rising on river plateaux across the region, exceptional craftsmanship in pottery and stone industry started to develop. Copper minerals and malachite beads found in settlements and cemeteries at the time became more numerous, although no thermal treatment has been recorded prior to c. 5000 BC. In eastern Serbia, the Vinča culture communities commenced mining activities in the currently earliest known copper mine, Rudna Glava [28]. Although no metal artefacts from this period (or later) were found to compositionally and isotopically match this source, there were other mines in otherwise copper-abundant region of eastern Serbia, such as Majdanpek, which were particularly prolific during the  $5^{th}$  millennium BC [1].

<u>Period 3. Early Chalcolithic (EC, 5000-4600 BC).</u> The start of the copper smelting activities is set around 5000 BC [3], which corresponds with the earliest known metal artefacts appearing in the Vinča culture site of Pločnik in south Serbia, followed by similar finds along the Black Sea Coast and in south Bulgaria [29]. Settlements grew in size, particularly along the lower Danube, which was

probably the easiest and quickest means of transport for the emerging long-distance trade of prestigious commodities, such as spondylus, obsidian, malachite beads and metal artefacts, amongst others [30, 31]. Many of these were found in the first organised cemeteries at the time, probably designating high status of buried individuals (i.e. Durankulak). The 'metal effect' is seen in the occurrence of graphite-painted decorations on pottery at the time, possibly imitating one of the most desired materials of the 5<sup>th</sup> millennium BC in the Balkans. Towards the very end of this period, archaeologists have recorded the first mining activities in Bulgaria, at the site of Ai Bunar, started by the bearers of the Marica culture [32, 33] (Table S3). It grew to be the most important source of eastern Balkan region throughout the later 5<sup>th</sup> millennium BC.

Period 4. Middle Chalcolithic (MC, 4600-4450 BC). This period is difficult to separate out from what appears to be an uninterrupted evolution of metal making cultures in eastern Balkans (Bulgaria) and slow disintegration of the Vinča culture in Serbia. It is generally characterised with the rise of two large cultural complexes and one culture in northeastern Bulgaria. While northern Vinča culture sites were rapidly being abandoned and conflagrated, a few southern ones (like Pločnik) continued to live until the very end of the Vinča culture in south Serbia (c. 4450 BC). Some scholars argued that it was the late Vinča culture in this region and Gradešnica in west Bulgaria that gave impetus for the formation of the Krivodol-Salcuta-Bubanj Hum (KSBh) I cultural complex [21]. The other large cultural complex was formed by the merging of Marica, Karanovo V and Boian Spantov cultures in south Bulgaria and Muntenia, and is known under the name of Kodžadermen-Gumelnita-Karanovo (KGK) VI (Table S3). Varna culture, named after the eponymous burial site with the world's earliest gold objects, occupied the (western) Black Sea coast. This is the time when large tell-sites dotted both riverbanks along the lower Danube, but also other regions in Bulgaria (like Karanovo tell-settlement, for instance). Metal production enters its peak production, where diversification in copper hammeraxe design was most likely due to communities seeking for a personal stamp in then fast-expanding metalmaking industry.

<u>Period 5. Late Chalcolithic (LC, 4450-4100 BC).</u> While in some parts of Bulgaria the transition from the previous period into this one is hardly recognisable in material culture, the Late Chalcolithic period has been supported with absolute dates. The material culture and domestic architecture are developing in this period together with the extensive burial evidence for the rising wealth of individuals, and hence a potential social stratification and emergence of an elite [34]. A rapid climate change towards the end of this period is seen as the major cause of disappearance of any record of the communities in the east and central Bulgaria. The disintegration of the communities seemingly started with the coming of the steppe population, although the complete cultural caesura must have been a combination of several factors [24]. In the west, KSBh cultural complex spreads over a vast space between Oltenia and the Aegean (Thassos). The material expression and settling habits differ from the developments in the east, with settlements mostly established at higher altitudes or caves. In Serbia, this cultural complex borders with the Tiszapolgár culture (Table S3).

<u>Period 6. Final Chalcolithic (FC, 4100-3700 BC).</u> This period was characterised by the shift in metalmaking industry towards the west of the observed area. Metallurgy intensified in the KSBh IV cultural complex, potentially due to the decline of the Thracian mining centres [21]. Evolving domestic architecture, settlements established on inaccessible paths, and innovations in pottery making were all part of this new phase of the KSBh cultural complex evolution. The mining and metal production was revived in eastern Serbia, particularly with the massive production of Jászladány type hammer axes, related isotopically to the Majdanpek mine, and culturally to the Bodrogkeresztúr culture [2]. This culture emerges east of the Tisza river, with sites dotted along its lowlands and into the Serbian Banat [15]; its southern spread is a matter of content, however, the spread of the Jászladány hammer axes indicates strong social and economic ties with area south of Danube. Gold objects occur for the first time in this part of the Balkans.

<u>Period 7. Proto Bronze Age (PB, 3700-3200 BC).</u> This period saw the final disintegration of all cultural complexes formed during the 5<sup>th</sup> millennium BC. Small-scale settlements with rare metal artefacts are recorded throughout Bulgaria, with new metal tools, like daggers, making the appearance for the first time, presumably echoing the Eurasian Steppe influence.

Each node in our network was followed by the designated time-period in our analyses in order to clarify which occupational horizon within a site (node) yielded which type of artefacts. Barring seven exceptions (see Table S1), all sites (or nodes) were ascribed a relative cultural affiliation based on the current state of research.

**Community structure (modularity) analysis.** Our network was built in two discrete steps: 1) we grouped the data in ten distinctive chemical clusters (*Artefacts Network*); 2) placed a connector between the sites that contain pairs of artefacts from the same cluster and analysed the final network for community structures (*Sites Network*). In both steps we used the Louvain algorithm [35] to obtain community structures (modules) and bootstrapping to test the significance of gained results.

**Artefacts Network – clustering the copper objects by trace element chemistry.** For each of 410 artefacts (Table S1) we used the readings for the following seven trace elements: arsenic (As), antimony (Sb), cobalt (Co), nickel (Ni), silver (Ag), gold (Au), and selenium (Se), since they are the ones that are commonly thought to survive the hot temperature treatment from the copper ore to the copper metal in our case [36-38]. We therefore extracted only these values (presented in Table S1) and then performed the following course of actions that led to obtaining the number of chemical clusters in our dataset:

1) transforming several compositional readings in our dataset with zero (0) value into a small positive number (0.0001); this number was smaller than the detection limit of any of the analysed elements;

2) calculating logarithms of all 7 trace elements;

3) running principal component analyses of the logged values and obtaining principal component scores;

4) determining Euclidean distance between all pairs of artefacts;

5) designing the Artefacts network with artefacts as nodes and links defined as  $1/d^2$  (d = Euclidean distance), and

6) obtaining the number of chemical clusters after conducting modularity analyses with the Louvain algorithm.

To rationalise this sequence, we will start with justifying the modularity approach to chemical clustering. Theoretically, the goal of chemical clustering is to detect groups of copper artefacts whose compositional signature (a string of 7 trace elements) is more similar within a group (or a cluster) than with compositional signature of copper artefacts - members of other groups (or clusters). In other words, the links that join copper artefacts of the same chemical cluster are based on compositional similarity, and they are comparatively stronger within a cluster of chemically similar artefacts than the links connecting these artefacts to other clusters. Since this compares closely to the definition of network modularity [39, 40], we designed the cluster analyses based on the principles of community

structure research in networks. There are other methods that can be used for determining the number of chemical clustering, however, we developed this one for two main reasons:

- 1) it offers a clear criterion for obtaining the number of modules by maximising the value of modularity (unlike, for example, hierarchical clustering);
- 2) it gives us an option to test the significance of the obtained clustering structure with bootstrapping, by using comparison between the value of modularity and the value of randomized networks.

Hence, the nodes of our network for obtaining chemical clusters were artefacts, while we defined links using Euclidean distance of the vectors of transformed trace element values. Namely, calculating Euclidean distance with the original trace element values proved challenging for two reasons: a) they showed lognormal, instead of Gaussian distribution in our case (Fig. 1) and b) they were correlated to begin with (Fig. 2a). Starting with the former, the lognormal distribution of our data indicated that small values are predominant (Fig. 1), and computing distances between the original data would lead to losing information on variation in smaller values. For instance, the difference between the values of 0.001 and 0.002 would make much smaller contribution in comparison to the difference between the values of 100 and 101. Hence, in order to account for these variations on the same scale, or same relative differences, we transformed the original values into logarithms. The logarithms of original data brought out clearly the correlations between chemical elements, like Sb and As, Au, Ag and Se, or Sb with Ag/Au/Se (Fig. 2b). This took us to acknowledging a particular (mathematical) property of compositional datasets, known as the constant-sum constraint (CSC), which refers to a constant sum of 1 or 100% for all variables in a measured sample [41, 42]. It means that individual variables in the compositional data do not vary independently -i.e. if one variable decreases, the proportion of the remaining must increase. Such an induced correlation may easily hinder the true relationships among variables (in our case trace elements), which is why the next step in our data processing was to eliminate these correlations. For this, we ran principal component analysis (PCA), a statistical procedure used to reduce the dimensionality of a dataset consisting of a large number of interrelated variables, while retaining the variation present in the dataset. The output are uncorrelated variables (principal components), ordered in a way that the first few keep most of the variation present in all of the original variables [43]. The PCA is the same procedure as eigenvalues decompositions from linear algebra. The PCA removed these correlations (Fig. S1), preparing the output, now calculated as principal component scores (Table S1), for network analysis. The logarithmic transformation, PCA, and visualisation in Figures 1, 2 and S1 were all computed in R (we used the corrplot library for correlations in these figures).

The straight approach to PCA with original compositional data has already been known as fraught with difficulties for the reasons mentioned above [41, 42]. Aitchison [41] proposed a way around these constraints by arguing that the best way to compute principal components out of restricted types of data (e.g. in allometry, or compositional data) is to use logarithms of the original data. This supports the treatment of our original data, although it was also in our case evident as a necessity from lognormal distribution (Fig. 1). A disadvantage of his approach was in that it could not handle zeros (0), which in our case was about to lead to losing a small handful of objects where particular trace elements were not detected (or were below the detection limit of the analytical instrument). An alternative, however, was to replace zero values with a small positive number, which is what we did before transforming the original values into logarithms. Our small positive number was smaller than the detection limit of any of the analysed elements (0.0001), as mentioned above.



Figure S1. The principal component analysis yielded the uncorrelated variables (compare with Figures 1 and 2).

In the following step, the principal component scores (Table S1) were used to calculate the Euclidean distance between all pairs of artefacts. For this, we followed the rationale below: if  $\vec{a}$  is a principal component vector of one artefact and  $\vec{b}$  is a principal component vector of another artefact, the distance between the two artefacts will be defined as Euclidean distance between these two vectors as:

$$d(\vec{a},\vec{b}) = \sqrt{\sum_{i=1}^{n} (a_i - b_i)^2}$$

Thus, the network we formed has artefacts as nodes and links defined as  $1/d^2$  (d = Euclidean distance). The number of clusters was obtained with the Louvain algorithm [35]. We used the original implementation of the code written in C++ by E. Lefebvre, and later adapted by J.-L. Guillaume; it is also freely available for download on https://sites.google.com/site/findcommunities/ (the current version is maintained on https://sourceforge.net/projects/louvain/).

Louvain method is based on the maximization of modularity Q, which measures the quality a certain partitioning of a network and is defined as:

$$Q = \frac{1}{2m} \sum_{ij} \left[ A_{ij} - \frac{k_i k_j}{2m} \right] \delta(c_i, c_j)$$

where  $A_{ij}$  is the weight of the link,  $k_i$  and  $k_j$  are weighted degrees (also known as strengths – the sums of the weights of all the links coming from that node) of the nodes *i* and *j*, *m* is the half sum of all the weights in the network, and  $\delta(c_i, c_j)$  is delta function, which will be 1 if the nodes *i* and *j* belong to the same cluster  $c_i$  ( $c_j$ ). Modularity *Q* can result in values between -1 and 1, and the larger the value, the better the partitioning of the network. This is because more links exist between the nodes of the same cluster in contrast to the links between the nodes of different clusters. Louvain algorithm includes an additional benefit in that it maximizes the partitioning of the entire network (ie level 1) but also produces alternative partitioning (level 2, level 3 etc), where modularity reaches a local maximum. Out of two levels of results, one with 6, and the other with 10 clusters, we opted for 10 and tested the significance of our results with network randomization (bootstrapping).

*Network randomization (bootstrapping).* We performed the bootstrapping in the following way: we used the obtained partitioning of the network and then randomized it, keeping only the properties that were important (here we only preserved the weights of the links, but we shuffle the nodes they were connected to). The result was the unique partitioning of the randomized network and the corresponding modularity value. This process was repeated 1000 times, and it yielded the distribution of 1000 modularity values in our randomized network, which we then compared with the modularity value of our Artefacts Network (Fig. 3a). The calculations were run in Python using the code for the Louvain algorithm (written in C++) for obtaining the modularity values. Histograms in Fig. 3 in the main text were produced in Gnuplot.

The modularity of the Artefacts Network is 0.3088 and the mean of the distribution of modularities of the randomized networks is 0.1012. The latter has the standard deviation of 0.0008, making the value of the original network 280 standard deviations larger than the mean of the randomized networks values (see Figure 3a). This corresponds to the p value of <0.001, since we randomized the network 1000 times over.

## *Clustering method – consistency with the previous research.*

In the previous study on the provenance of the 5<sup>th</sup> millennium BC Balkan copper metallurgy, Pernicka et al [1, 2] conducted average-link analyses (a type of hierarchical clustering) in order to group more than 300 copper artefacts into cohesive clusters. They initially transformed the trace element concentration of As, Sb, Ag, Co, Ni, Au, and Se into logarithms and then applied the average-link cluster analysis with Euclidean distances using the SAS (Statistical Analysis Software) program package. This program uses the cubic clustering criterion [36, 44] as the parameter for determining the optimum number of clusters, which is how Pernicka et al. [1, 2] arrived to defining nine chemical clusters in their research. They then used discriminant analysis to calculate the probability of each sample to belong to the cluster it was assigned to with the average-link procedure, and applied the 50% rule: where cases (objects) had less than 50% probability of belonging to the assigned cluster, they were re-assigned to the cluster they had the highest probability for membership.

In order to check the consistency of our clustering method (modularity) with the one described above, we tested the data from our two largest clusters, cluster 2 and cluster 4 (Table S1), against the trace element patterns of the two most prolific prehistoric copper mines in the Balkans, Majdanpek and Ai Bunar (Figures S2 and S3). Namely, Pernicka et al. [2, 117, Fig. 20] managed to identify the chemical correlation between the Majdanpek mine and their cluster 2 (58 artefacts), and the Ai Bunar mine and their cluster 3 (43 artefacts), hence providing support for the argument that these two mines/copper deposits were exploited to make the observed sets of copper artefacts from the 5<sup>th</sup> millennium BC Balkans. We performed a similar test by plotting the trace element values of our cluster 2 artefacts (161 objects) with the trace element signature of Majdanpek (Fig. S2), and the trace element values of our cluster 4 artefacts (129 objects) with the trace element signature of Ai Bunar (Fig. S3). We chose these clusters since the former relates mostly to sites in Serbia and western Bulgaria, while the latter shows similar preferred associations with the sites in central and east Bulgaria. Also, these clusters (2 and 4) largely represented expanded versions of Pernicka et al.'s clusters 2 and 3 respectively; we were not, however, expecting the exact overlap between these given that we were working with a larger dataset than these authors.

The plot on figure S2 shows a general consistency of cluster 2 artefacts with the Majdanpek ore field (grey), with the notable exception of three samples in total (labels: MA-071499, L354, L355, see Table S1). While Ni and Ag values in the Majdanpek ore and cluster 2 artefacts appear most correlated in Fig. S2, the greatest fluctuations are noticed in the Sb, Co and Au values. The plot on figure S3 also presents a tight pattern of cluster 4 artefacts matching closely the trace element pattern of Ai Bunar ores (grey field). The trace element values in this plot are highly correlated, barring As and Sb readings.

Chemical fluctuations can be explained with several factors, both from the perspective of designated ore fields or the nature of artefacts making. Namely, when it comes to potential chemical variability in the ore fields, noteworthy is that the grey (mine) patterns in figures S2 and S3 stand for the 10<sup>th</sup> and 90<sup>th</sup> percentile of the maximum and minimum recorded trace element values for Majdanpek and Ai Bunar. Although it does not mean that the grey patterns are incorrect, there is always a possibility that the sample size representing this ore field was not representative to begin with.

Speaking of the chemical fluctuation of trace element patterns of artefacts against the original ore background, the lower readings of As and Sb in copper artefacts (in fig. S3 and partly in fig. S2) may imply the possibility of loss during metal extraction or recycling, particularly since the former has been known as volatile. The extent of volatility of As during arsenical copper recycling has been hotly debated in archaeology and archaeometallurgy, with discussions mostly concentrating on the redox conditions of the (s)melt and the compositional threshold below which As in copper becomes less volatile [45, 46]. In this light, and given that we are addressing here traces of both As and Sb (in ppm, not in percentages), we propose the recycling hypothesis only as an assumption that needs further probing. If the loss of As and Sb was indeed related to recycling in our cases, then such practice must have occurred within regionally (and potentially culturally) defined spaces. This conclusion follows neatly our modularity research, and is also addressed in the main manuscript.



Figure S2. Trace element signatures of 161 copper artefacts belonging to cluster 2 (lines) plotted against the trace element signature of Majdanpek (grey field), a copper mine in eastern Serbia.

The third potential explanation for the observed chemical fluctuations is that both clusters 2 and 4 reflect chemical signatures of several deposits adjacent to Majdanpek and Ai Bunar respectively. This is not improbable given that nowadays the preserved prehistoric mining commonly represents copper deposits that survived the later exploitation (and hence destruction) as not economically feasible investments in modern terms. While Ai Bunar might represent such a case, the exploitation of Majdanpek has only been confirmed through provenance analyses thus far [1], and not through verified traces of prehistoric exploitation beyond a few chronologically indistinctive grooved hammerstones kept in the Mining Museum in Majdanpek in Serbia. Thus, the best-case scenario for the surviving ancient mining is the poor ore quality, which may provide some grounds to presume that our two prolific mining sites in Serbia and Bulgaria may be only reflecting the less rich remnants of the actual copper mineral vein that had been mined in their vicinity.

Chronology of the plotted artefacts may also help understand the chemical fluctuations. Cluster 2 is dominated by copper artefacts from two distinctive chronological 'block periods': 5500-4450 BC and 4100-3700 BC, while cluster 4 includes mainly artefacts from 4450-4100 BC. The fluctuating pattern of cluster 2 artefacts may indicate the use of different ore sources in 5500-4450 BC and 4100-3700 BC respectively, although regionally constrained to eastern Serbia. On the other hand, the tight pattern of cluster 4 may indicate exploitation of a source in the vicinity of Ai Bunar with lower As and Sb content, or Ai Bunar itself followed by extensive recycling that took place within the constrains of the cultural / social boundaries of the KGK VI and related cultural complexes. All options presented here will be addressed in detail in future research.

Overall, figures S2 and S3 exhibit noticeable correlation of cluster 2 and cluster 4 artefacts with Majdanpek and Ai Bunar. Our clustering method shows good consistency with the cluster analyses of Pernicka et al. [1, 2], which along with bootstrapping (Fig. 3a), verifies its reliability.



Figure S3. Trace element signatures of 129 copper artefacts belonging to cluster 4 (lines) plotted against the trace element signature of Ai Bunar (grey field), a copper mine in central Bulgaria.

**Sites Network – community structure analyses of archaeological sites**. In this step, the archaeological sites represented nodes, and links between them were based on sharing the same chemical cluster for pairs of copper artefacts found in those sites. This relationship was established under the assumption that two artefacts belonging to the same chemical cluster could have ended up from the places of exploitation or production in two different sites through either direct or indirect contact (i.e. various types of intermediaries); we encompass both options under the term 'supply network'. Thus, the link between the sites in out network practically works in the following way: artefact A and artefact B from two different sites belong to (chemical) cluster 1, and therefore these two sites (nodes) have a link placed between them. If these two sites contain more artefacts from the same cluster, the weight of the link is larger. For example: if site *i* contains artefacts from clusters [0,1,1,1,1,2,2,2,3] and site *j* has artefact from clusters [0,1,1,2,2,8,9], then the weight of the link is 5 (one for each artefact of the common type). We analysed the final network with Louvain algorithm [35] and gained only one level with three distinctive community structures.

When randomizing the network, we cut each link and randomly reconnected it to a different node while saving only the information of the degree of each node for this type of network. We took into consideration, for instance, that the link with weight 5 is actually 5 links. We repeat the randomization procedure 1000 times. The modularity of the original network (Sites Network) was 0.276, which is 57 standard deviations larger from the mean of the modularities of the randomized network ( $0.078 \pm 0.004$ ) (Figure 3b). Geographical coordinates of archaeological sites/nodes (Table S1) were used solely for illustrative purposes in this paper. Visualisation of Sites Network (Figures 4-7) was produced in Python from scratch, using Matlibplot package and the background map with kind permission of Prof. M. Milinkovic (University of Belgrade, Serbia). The Sites Network is the final outcome of our network design, and the only one whose modularity we discuss in the article.

Since some of the observed sites (nodes) were active throughout multiple time-periods, and we wanted to observe their position in each of them, we regarded the same site in a different period as a separate node (site-period), and added the chronological span to the site name for easier navigation through results (see Table S2). Most importantly, apart from chemical cluster number we did not use any archaeologically relevant information in our network. In total, we have 79 sites and 93 site-periods. The sites (nodes) that appear in more than one period are listed below (Figure S4):

- Ai Bunar 4600-4450 BC, 4100-3700 BC
- Belovode 5500-5000 BC, 5000-4600 BC, 4600-4450 BC
- Durankulak 5000-4600 BC, 4600-4450 BC, 4450-4100 BC, 3700-3200 BC
- Goljamo Delcevo 4600-4450 BC, 4450-4100 BC
- Gomolava 5000-4600 BC, 4600-4450 BC
- Hotnica 4450-4100 BC, 3700-3200 BC
- Pločnik 5500-5000 BC, 5000-4600 BC, 4600-4450 BC
- Smjadovo 4450-4100 BC, 3700-3200 BC
- Tell Ruse 4600-4450 BC, 4450-4100 BC
- Zlotska pecina 4100-3700 BC, 3700-3200 BC



Figure S4. The sites (nodes) that exist throughout multiple time periods. Pločnik, Zlotska pecina and Goljamo Delcevo change the module over the observed time frame (c. 6200 to c. 3200 BC).

The importance of archaeological sites (nodes). We initially tested the importance of our nodes with three different node centrality measurements: degree centrality (based on number of links each node includes), PageRank [47] and betweenness centrality [48]. All three yielded meaningful results for determining the importance of the specific archaeological sites. The degree centrality of the node (in this case weighted degree or strength) tells us with how many other sites the observed site had some kind of communication. The PageRank takes into account how important the observed sites are. However, given that our network is not directed, these two properties appear significantly correlated (see Figure S5), and hence both presented similar results for our study. On the other hand, the betweenness centrality is defined as a number of shortest paths that go through an observed node. In order to calculate it we defined the weights as 1/w or  $1/w^2$ , where w is the weight in the original network; this procedure ensured that if there were more connections between the sites, it was easier to travel between them. Once we compared the betweenness centrality and the PageRank we observed that barring the large difference for nodes of smaller PageRank values, the more important nodes were still more important by both measures (see Figure S6). Also, the betweenness centrality measure is not very robust and by removing only one artefact from the original input, the values change substantially, although again the more important nodes still come out the same. To conclude, using any of the importance measure yielded very similar results, which is why we give all three in Table S2. For the purpose of illustration in our maps (size of the nodes) we opted for PageRank; these are, again, not robust, which is why we use them only for visualisation.



Figure S5. PageRank vs. weighted degree (strength). The two measures are strongly correlated, as expected in undirected networks, which makes both useful for measuring the importance of the site (node).



Figure S6. PageRank vs. betweenness centrality. Please note that except for the values with small PageRank, the two measures are correlated, which makes both suitable for measuring importance of the site.

## Bibliography

- 1. PERNICKA, E., BEGEMANN, F. & SCHMITT-STRECKER, S. (1993) Eneolithic and Early Bronze Age copper artefacts from the Balkans and their relation to Serbian copper ores. *Prähistorische Zeitschrift* 68, 1-54.
- 2. PERNICKA, E., BEGEMANN, F., SCHMITT-STRECKER, S., TODOROVA, H. & KULEFF, I. (1997) Prehistoric copper in Bulgaria. Its composition and provenance. *Eurasia Antiqua* **3**, 41-180.
- 3. RADIVOJEVIĆ, M., REHREN, TH., PERNICKA, E., ŠLJIVAR, D., BRAUNS, M. & BORIĆ, D. (2010) On the origins of extractive metallurgy: new evidence from Europe. *Journal of Archaeological Science*, **37**, 2775-2787.
- 4. RADIVOJEVIĆ, M. (2012) On the Origins of Metallurgy in Europe: Metal Production in the Vinča Culture. *PhD Thesis*, UCL Institute of Archaeology, London.
- 5. RADIVOJEVIĆ, M., ROBERTS, B. W., MARIĆ, M., KUZMANOVIĆ-CVETKOVIĆ, J. & REHREN, TH. (eds) *The Rise of Metallurgy in Eurasia: The Archaeology of Early Metallurgy and Society in the Central Balkans*. London: UCL Press.
- 6. ANTONOVIĆ, D. (2014) *Kupferzeitlichen Äxte und Beile in Serbien. Prähistorische Bronzefunde, Vol 9, Band 27* Mainz: Akademie der Wissenschaften und der Literatur.
- 7. BOJADŽIEV, J. (2002) Die absolute Datierung der neo- und äneolithischen Gräberfelder von Durankulak. *Durankulak, Band II - Die prähistorischen Gräberfelder von Durankulak, Teil 1* (H. Todorova, ed.). Berlin-Sofia: Publishing House Anubis Ltd. Sofia, pp. 67-70.
- 8. FORENBAHER, S. (1993) Radiocarbon dates and absolute chronology of the central European Early Bronze Age. *Antiquity*, **67**, 218-256.
- 9. GEORGIEVA, P. (2012) *The Neolithic and Eneolithic in Southeast Europe*. Sofia: University Press "St. Kliment Ohridski".
- 10. GÖRSDORF, J. & BOJADŽIEV, J. (1996) Zur absoluten Chronologie der bulgarischen Urgeschichte. *Eurasia Antiqua*, **2**, 105-173.
- 11. HIGHAM, T., CHAPMAN, J., GAYDARSKA, B., SLAVCHEV, V., HONCH, N., YORDANOV, Y. & DIMITROVA, B. (2007) New perspectives on the Varna cemetery (Bulgaria) AMS dates and social implications. *Antiquity*, **81**, 640-654.
- 12. KRAUSS, R. (2008) Karanovo und das südosteuropäische Chronologiesystem aus heutiger Sicht *Eurasia Antiqua*, **14**, 117-149.
- KRAUSS, R., ZÄUNER, S. & PERNICKA, E. (2014) Statistical and anthropological analyses of the Varna necropolis. *Metals of Power-Early Gold and Silver. 6th Archaeological Conference* of Central Germany, October 17-19, 2013, Halle (H. Meller, R. Risch & E. Pernicka, eds.). Halle: Landesmuseum für Vorgeschichte, pp. 371-387.
- LAZAROVICI, C.-M. (2006) Absolute chronology of the late Vinča culture in Romania and its role in the development of the Early Copper Age *Homage to Milutin Garašanin* (N. Tasić & C. Grozdanov, eds.). Belgrade: Serbian Academy of Sciences and Arts and Macedonian Academy of Sciences and Arts, pp. 277-293.
- 15. LUCA, S. A. (1999) *Sfârșitul eneoliticului pe teritoriul intracarpatic al României cultura Bodrogkeresztúr*, vol. 11. Alba Iulia: Bibliotheca Musei Apulensis.
- 16. PATAY, P. (1974) Die hochkupferzeitliche Bodrogkeresztúr-Kultur. Bericht der Römisch Germanischen Komission, **55**, 1-71.
- 17. RADIVOJEVIĆ, M., REHREN, TH., KUZMANOVIĆ-CVETKOVIĆ, J., JOVANOVIĆ, M. & NORTHOVER, J. P. (2013) Tainted ores and the rise of tin bronze metallurgy, c. 6500 years ago. *Antiquity*, **87**, 1030-1045.
- 18. RADIVOJEVIĆ, M. (2015) Inventing metallurgy in western Eurasia: a look through the microscope lens. *Cambridge Archaeological Journal*, **25**, 321-338.
- 19. RADIVOJEVIĆ, M. & REHREN, TH. (2016) Paint It Black: The Rise of Metallurgy in the Balkans. *Journal of Archaeological Method and Theory*, **23**, 200-237.
- 20. TODOROVA, H. (1981) Die kupferzeitlichen Äxte und Beile in Bulgarien. Prähistorische Bronzefunde, Vol 9, Band 14. München: C. H. Beck'sche Verlagsbuchhandlung.

- 21. TODOROVA, H. (1995) The Neolithic, Eneolithic and Transitional Period in Bulgarian Prehistory. *Prehistoric Bulgaria* (D. W. Bailey, I. Panayotov & S. Alexandrov, eds.), vol. Monographs in World Archaeology No. 22. Madison: Prehistory Press, pp. 79-98.
- 22. SCHIER, W. (1996) The relative and absolute chronology of Vinča: new evidence from the type site. *The Vinča Culture, its Role and Cultural Connections* (F. Draşovean, ed.). Timişoara: National Museum, pp. 141-162.
- 23. SCHIER, W. (2014) The Copper Age in southeastern Europe historical epoch or typochronological construct? *New Approaches to Dating and Cultural Dynamics in the 6th to 4th Millennium BC. Prähistorische Archäologie in Südosteuropa 28* (W. Schier & F. Drașovean, eds.). Rahden/Westf.: Verlag Marie Leidorf Gmbh, pp. 419-435.
- WENINGER, B., LEE, C., ROHLING, E., BAR-YOSEF, O., BÖHNER, U., BUDJA, M., BUNDSCHUH, M., FEURDEAN, A., GEBE, H. G., JÖRIS, O., LINSTÄDTER, J., MAYEWSKI, P., MÜHLENBRUCH, T., REINGRUBER, A., ROLLEFSON, G., SCHYLE, D., THISSEN, L., TODOROVA, H. & ZIELHOFER, C. (2009) The impact of rapid climate change on prehistoric societies during the Holocene in the Eastern Mediterranean. *Documenta Praehistorica*, 36, 7-59.
- 25. TASIĆ, N., MARIĆ, M., PENEZIĆ, K., FILIPOVIĆ, D., BOROJEVIĆ, K., RUSSELL, N., REIMER, P., BARCLAY, A., BAYLISS, A., BORIĆ, D., GAYDARSKA, B. & WHITTLE, A. (2015) The end of the affair: formal chronological modelling for the top of the Neolithic tell of Vinča-Belo Brdo. *Antiquity*, **89**, 1064-1082.
- 26. TASIĆ, N., MARIĆ, M., FILIPOVIĆ, D., PENEZIĆ, K., DUNBAR, E., REIMER, P., BARCLAY, A., BAYLISS, A., GAYDARSKA, B. & WHITTLE, A. (2016) Interwoven Strands for Refining the Chronology of the Neolithic Tell of Vinča-Belo Brdo, Serbia. *Radiocarbon*, **58**, 795-831.
- 27. TODOROVA, H. (1978) *The Eneolithic Period in Bulgaria in the Fifth Millennium B.C.* Oxford: Archaeopress, British Archaeological Reports 49.
- 28. JOVANOVIĆ, B. (1980) The origins of copper mining in Europe. *Scientific American*, **242**, 152-167.
- 29. RYNDINA, N., INDENBAUM, G. & KOLOSOVA, V. (1999) Copper production from polymetallic sulphide ores in the northeastern Balkan Eneolithic culture. *Journal of Archaeological Science*, **26**, 1059-1068.
- 30. BAILEY, D. W. (2000) Balkan Prehistory. London, New York: Routledge.
- 31. CHAPMAN, J. (2007) Engaging with the exotic: the production of early farming communities in south-east and central Europe. *A short walk through the Balkans. The first farmers of the Carpathian Basin and adjacent regions. Quaderno 12* (M. Spataro & P. Baigi, eds.). Trieste: Società per la preistoria i protostoria della regione Friuli-Venezia Giulia, pp. 207-222.
- 32. CHERNYKH, E. N. (1978) Aibunar a Balkan copper mine of the fourth millennium BC. *Proceedings of the Prehistoric Society*, **44**, 203-217.
- 33. CHERNYKH, E. N. (1978) Gornoe Delo i Metallurgiya v Dreivneishei Bolgarii. Sofia: Bolgarskoi Akademii Nauk.
- 34. RENFREW, C. (1986) Varna and the emergence of wealth in prehistoric Europe. *The Social Life of Things: Commodities in Cultural Perspective* (A. Appadurai, ed.). Cambridge: Cambridge University Press, pp. 141-168.
- 35. VINCENT, D. B., JEAN-LOUP, G., RENAUD, L. & ETIENNE, L. (2008) Fast unfolding of communities in large networks. *Journal of Statistical Mechanics: Theory and Experiment*, **10**, P10008.
- 36. CHRISTOFORIDIS, A. & PERNICKA, E. (1988) Gruppierung von Metallanalysen mit Hilfe der Clusteranalyse. Die Endneolithische und Frühbronzezeitlichen Grabfunde auf der Nordstadt Terrasse Von Singen am Hohentwiel. Forschungen und Berichte zur Vor- und Frühgeschichte in Baden-Württemberg, Band 32. (H. R. Krause, ed.). Stuttgart, pp. 252-262.
- 37. PERNICKA, E. (1990) Gewinnung und Verbreitung der Metalle in prähistorischer Zeit. Jahrbuch des Römisch-Germanischen Zentralmuseums Mainz 37, 21-129.
- PERNICKA, E. (2014) Provenance determination of archaeological metal objects *Archaeometallurgy in Global Perspective* (B. Roberts, W. & C. P. Thornton, eds.). New York: Springer, pp. 239-268.
- 39. FORTUNATO, S. (2010) Community detection in graphs. *Physics Reports*, **486**, 75-174.
- 40. NEWMAN, M. E. J. (2010) *Networks: An Introduction*. Oxford: Oxford University Press.

- 41. AITCHISON, J. (1983) Principal component analysis of compositional data. *Biometrika*, **70**, 57-65.
- 42. KUCERA, M. & MALMGREN, B. A. (1998) Logratio transformation of compositional data: a resolution of the constant sum constraint. *Marine Micropaleontology*, **34**, 117-120.
- 43. JOLLIFFE, I. T. (2002) Principal Component Analysis (2nd edition). New York: Springer.
- 44. SAS INSTITUTE INC. (1983) *SAS Technical Report A-108, Cubic Clustering Criterion*. Cary, NC.: SAS Institute Inc.
- 45. MCKERRELL, H. & TYLECOTE, R. F. (1972) The working of copper-arsenic alloys in the Early Bronze Age and the effect on the determination of provenance. *Proceedings of the Prehistoric Society*, **38**, 209-218.
- 46. SABATINI, B. (2015) The As-Cu-Ni System: A Chemical Thermodynamic Model for Ancient Recycling. *Journal of Metals*, **67**, 2984-2992.
- 47. BRIN, S. & PAGE, L. (1998) The Anatomy of a Large-Scale Hypertextual Web Search Engine Seventh International World-Wide Web Conference (WWW 1998), April 14-18, 1998. Brisbane, Australia.
- 48. FREEMAN, L. C. (1977) A Set of Measures of Centrality Based on Betweenness. *Sociometry*, **40**, 35-41.