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Physical Barriers, Cultural Connections: Prehistoric Metallurgy across the Alpine Region

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This paper considers the early copper and copper-alloy metallurgy of the entire Alpine region. It introduces a new approach to the interpretation of chemical composition datasets, which has been applied to a comprehensive regional database for the first time. The Alpine Chalcolithic and Early Bronze Age each have distinctive patterns of metal use, which can be interpreted through changes in mining, social choice, and major landscape features such as watersheds and river systems. Interestingly, the Alpine range does not act as a north-south barrier, as major differences in composition tend to appear on an east-west axis. Central amongst these is the prevalence of tin-bronze in the western Alps compared to the east. This ‘tin-line’ is discussed in terms of metal flow through the region and evidence for a deeply-rooted geographical division that runs through much of Alpine prehistory.

Keywords: Copper Age; Bronze Age; Alps; GIS; metal artefacts; early metallurgy; elemental composition

Introduction

Archaeometallurgical research in the European Alps has tended to engage with a narrow range of traditional questions. Although this has been crucial for developing our field of research and for creating a firm basis for future work, it is possible that such questions now hinder more than help. Instead of focusing on the provenience of raw material, we would like to focus our attention on flows of metal and, of course, of people, and on how this movement was related to the physical world. Our analysis aims to place the metallurgical evidence in a three-dimensional world where topographical elements play a key role alongside the cultural context for material culture production and use. The Alpine region is a perfect test area for this approach, because of its dramatic topography and the number of different cultures it contains, the distribution of which often reflects the location of the main river systems. In this paper, we explore the shifting perceptions and use of metal during the period 3800/3600 to 2000 BC using a novel approach to the modelling of the chemical data in association with Geographical Information Systems (GIS). One key, and indeed surprising, result of our research is that the circulation of metal was apparently not dictated by the geomorphology of the Alps, but was instead influenced by a number of strong cultural barriers that seem more dependent on the river systems, and possibly on prehistoric ethnic boundaries, than on the mountain range itself.

Datasets and Methodology

Designing the Database

This research encompasses the entire Alpine region, roughly corresponding to today's southeast France (the Rhône valley), Switzerland, southern Germany, northern Italy, Austria, and Slovenia (Figure 1). The first step of the research was to create a database of all the published chemical analyses of copper-alloy artefacts dating to 3800/3600–2000 BC, to which a further set of thirty-five unpublished determinations (kindly provided by Peter Northover) was added (Table 1). The

latter artefacts were dated with reference to Pedrotti's (2001) scheme, alongside further personal communication with A.L. Pedrotti. For further details on the dataset, see note 1¹.

The database (to be published as part of the doctoral thesis of the first author) records, for each object, information about the source (author) of the chemical analysis and its original identifying number, typology, find site, chronology and alternative dates (if applicable), chemical composition in weight percentage, the museum where it is held, inventory number, and bibliography. The database comprises 4760 entries. Most of the data are based on Krause (2003), plus some new analyses (e.g. Angelini & Artioli, 2007; Angelini, 2004, 2007; Pernicka, 2011; Cattin et al., 2011). Given that some objects have been analysed more than once, our statistical modelling was based on the mean value of the set of available data. The nature of the find context was also noted (i.e. settlement, hoard, burial, or single find) as well as its geographical coordinates. The coordinates were mostly obtained from Krause (2003) and were validated through cross-referencing with other sources (Bosio et al., 1990; Rossi & Bishop, 1991; Poggiani Keller, 1992; Casini, 1994), including Google Earth and the reference database for the Alpine stilt houses (<http://www.palafittes.org/>).

¹ Chemical data were acquired using Krause's database (Krause, 2003) and relative bibliography, which includes (Otto & Witter, 1952; Junghans et al., 1960, 1968a, 1968b, 1974; Barker, 1971; Ottaway, 1982; Rychner, 1995). Further information were acquired from (Marchesetti, 1889; Colini, 1907; Barfield & Broglio, 1966; Matteoli & Storti, 1974; Budd, 1991; Barfield, 1995; Artioli et al., 2003; Hook, 2003; Angelini, 2004, 2007; De Marinis, 2005: 200, 2006a, 2006b; Höppner et al., 2005; Angelini & Artioli, 2007; Pearce, 2007; Kienlin, 2008; Angelini et al., 2010; Cattin et al., 2011; Pernicka & Salzani, 2011). The compositional data published by Pittioni and co-workers (Preuschen & Pittioni, 1939; Pittioni, 1957; Neuninger & Pittioni, 1963) were not included in the database because their determinations are semi-quantitative, and the presence of elements is solely indicated by symbols (e.g. Sn: +++). The quality of these data is insufficient for the kind of geostatistical analysis employed in our research, which mathematically assesses the influence of elevation, distance and other topographical metrics on the dataset (see below).

The creation of such a broad database poses key methodological questions, including: a) the challenge of combining data produced by different analytical methods; b) how best to express chemical data with different inherent accuracies and precisions, due to the range of analytical methods employed; and c) how to reconcile the different chronologies and seriation sequences proposed by different authors.

One may question the practicality and wisdom of including incomplete and old data within a single database, since analyses undertaken with older techniques may not be as accurate and precise as those carried out with modern methods. To compound the problem, calibration and secondary standard data are usually not given in journal publications. Pernicka (1986) considered this question in the context of the feasibility of using the *Studien zu den Anfängen der Metallurgie* (SAM) data (Junghans et al., 1960, 1968a, 1968b, 1974), and concluded that they are comparable with data obtained by modern techniques. Rychner and Northover (1998) also undertook a comparative study of the several techniques used to analyse ancient metal artefacts. The results obtained with OES, ICP-AES, AAS, XRF, EPMA and NAA were compared and the authors concluded that ‘The modern analytical techniques are capable of producing accurate, reproducible data that can, with thorough standardisation, be used interchangeably with other data and behave similarly in cluster and classification’ (Rychner & Northover, 1998: 31). According to Rychner and Northover, the accuracy of XRF was more problematic, since the inter-laboratory comparisons showed quite a wide range of results. Lutz and Pernicka (1996) compared data obtained with a portable XRF and those obtained with NAA and demonstrated a general comparability of the two sets of results. It seems likely that the XRF issues found by Rychner and Northover were due more to human errors in different laboratories rather than an intrinsic deficiency of the technique. Finally, Merkl (2011: 89, figs. 8.4 and 8.5) undertook Principal Components Analysis (PCA) on data obtained by Otto and Witter, the SAM group, and Krause. He observed that analyses on the same objects by different laboratories are usually clustered together when plotted. The few outliers were identified as

individual errors of measurement. We may conclude that, with caution, data obtained by different methods, by different laboratories, and at different times, can be compared.

The second point, namely consistency of expression, is fundamental, given the way in which we wish to treat the data. In order to undertake geostatistical analysis, the data must be expressed purely numerically, i.e., without symbols intended to convey semi-quantitative observations, such as +, <, > or ~. For the data published by Otto and Witter and Junghans et al., the symbols have been converted into numerical values using the table of conversion proposed by Ottaway (1982, sec. XXIII) (Figure 2). In other cases, we have made a number of simple assumptions. Where the symbol indicates the presence of an element as ‘trace’ or ‘less than detection limits’, we have assigned a value of half the detection limit. So, if for any element the detection limit is 0.01 per cent, we have assigned 0.005 per cent to ‘trace’ or ‘<0.01’.

Chronological Systems

Another issue to be considered is the chronology of the artefacts. Working with such a wide geographical area forces us to merge, simplify and often struggle with different chronological schemes. The first major challenge lies in the terminology of the period in question, variously known as Late/Final Neolithic in northern Alpine terminology and as Copper Age (or Eneolithic) in the southern Alps (*c.* 3800/3600–2200 BC); it is followed in both regions by the initial Early Bronze Age (*c.* 2200–2000 BC), which is also included in our research. Scholarly tradition going back to the nineteenth century has it that the broadly equivalent terms Chalcolithic/Eneolithic/Copper Age were solely introduced in those regions in which a substantial metallurgical phase predating the Bronze Age was identified. This has generated an unhelpful terminological split across Europe whereby most southern and eastern countries (with the notable exception of Greece) now recognize a Copper Age phase wedged between the Neolithic and Bronze Age, while most northern and western countries do not (Lichardus, 1991; Kienlin, 2010: ch. 2; Heyd, 2013: fig. 10). The fault line slices neatly through the Alpine region, for the French, Swiss,

German, and Austrian scholarships define the earliest metallurgical phases as Late/Final Neolithic, while in Italy and Slovenia these are called Copper Age or Eneolithic (notwithstanding the recent attempts to identify a Chalcolithic phase in central and western Europe: Heyd, 2013). The terminological inconsistency is best exemplified by the metal-equipped Iceman, whose mummy was found on the border between Austria and Italy; following their respective scholarly traditions, the Austrians see him as ‘Stone Age man’ and the Italians as ‘Copper Age man’ (De Marinis & Brillante, 1998: 45–46).

A further, more severe, problem is encountered when one tries to define a set of parameters that could unambiguously identify the earliest metallurgical period in the entire region. Purely technological factors based on the presence/absence or amount of metal artefacts are plainly inadequate. For example, the north-alpine Pfyn, Altheim and Mondsee groups, which show precocious florescence of metal-using beginning *c.* 3800 BC, were followed by the Horgen and Cham groups, which were significantly poorer in metal objects (Strahm, 1994, 2005; Kienlin, 2010: 13–16); conversely, the initial stages of the Italian Copper Age were long thought to be poor in metal artefacts, although this reading has been tempered by recent research (Pearce, 2007: 51–52; Kienlin et al., 2009; Dolfini, 2013). Until recently, a seemingly useful distinction was made between metal use, which would occur in securely Neolithic contexts (*c.* 4500–3800/3600 BC), and metal production, which would define a Final Neolithic/Copper Age phase (*c.* 3800/3600–2200 BC; Skeates, 1993; Strahm, 2005). However, this reading must now be rejected given that the earliest evidence of smelting and metalworking both north and south of the Alps has been pushed back to the late fifth millennium BC (Höppner et al., 2005; Mazziere & Dal Santo, 2007; see Dolfini, 2013, for discussion). Likewise, cultural parameters that seek to correlate the emergence of metallurgy with sweeping changes in the social organization of society are not without their problems. Leaving aside the growing scepticism regarding the role played by early metalwork in triggering social inequality, one must note that, perhaps influenced by their differing terminologies and evidence, French and German-speaking archaeologists tend to be much more cautious than their Italian

colleagues in postulating structural links between pre-Bronze Age metallurgy and the emergence of social complexity (compare, for example, Kienlin, 2010: ch. 5, and Mille & Carozza, 2009, with Guidi, 2000: ch. 4).

A final problem is presented by the absolute chronology of the earliest metallurgical phases. Here, disagreement between scholars is aggravated by the existence of national sequences in all the countries concerned. Moreover, the respective phase boundaries are often placed at different times in the sequence, thus making correlations difficult (Table 2). The problem becomes intractable when one tries to discern sub-phases within the major phases. For example, early copper artefacts from northern Italy have been divided into several sub-phases by Carancini (2001) and De Marinis (1997, 2013), but they disagree on both the absolute chronology of the sub-phases and on which objects should be assigned to each of them. To make things worse, both chronological schemes have been criticized by Barfield (2007), Barfield and Kuniholm (2007), and Cocchi Genick (2012: 556–81). Furthermore, Dolfini (2010, 2013, 2014a) has recently argued that the entire sequence of early Italian metalwork must be revised considering the flawed methodological premises in which it is grounded, as well as its manifest mismatch with the radiocarbon dates available. Trying to synchronize any of these sequences with the no less controversial schemes proposed by Krause (2003), David-Elbiali (2000) and others for the northern and western Alps is an unachievable task.

Given the difficulties with disentangling such a veritable Gordian knot, we have decided to cut it clean through. In practice, this means that all the objects assigned to the Late Neolithic/Copper Age in the entire Alpine region have been grouped together within an all-encompassing category, which for the sake of consistency we call ‘Copper Age’. This phase begins with the first sustained production and use of metalwork *c.* 3800/3600 BC and ends with the inception of the Bronze Age *c.* 2200 BC. It excludes the earliest experiments with copper metallurgy in the late fifth and initial fourth millennia BC on the account that the objects belonging

to this phase are, both north and south of the Alps, relatively few, poorly dated, and not always analysed.

Although rather crude, our grouping has the distinct advantage of being grounded in conventional (and widely accepted) chronological and cultural boundaries for the period in question; it also makes our work less susceptible to later revisions, which would be all the more likely given the volatile artefact seriation sequences currently available for the region. By choosing to group together all metal artefacts which can be labelled Late Neolithic or Copper Age, we consciously reject the current possibility of arriving at a finer-grained chronological understanding of metal circulation and exchange in the long period of time from 3800/3600–2200 BC — an exercise that demands a concerted and extensive research effort.

Similar problems, albeit on a less dramatic scale, are raised by the chronology of the initial Early Bronze Age. In particular, technological or cultural parameters are just as inadequate to capture the emergence of tin-bronze metallurgy and the parallel development of Bronze Age Alpine society, as they are to define the Copper Age. From a metallurgical viewpoint, it has long been recognized that the adoption of tin-bronze probably occurred during the advanced Early Bronze Age, and that the new alloy would have coexisted for some time with *fahlerz* copper and other Copper Age compositional groups (Pare, 2000; Krause, 2003; De Marinis, 2006a). Instead of either seeing an alloy sequence, or parallel existence of alloys, below we will highlight the practice of adding tin to arsenical and antimonial composition groups. From a cultural viewpoint, the frantic search for elites that characterized so many prehistoric studies in the twentieth century has recently been questioned by new conceptual approaches, which stress the small scale of mining and smelting operations as well as the scarcity of Early Bronze Age ‘central places’ in the Alpine region (Kienlin & Stöllner, 2009; Kienlin, 2013). As with the Copper Age, a definition of the Alpine Early Bronze Age must be grounded in broadly accepted chronological parameters to be of any use. Luckily, the exercise is this time made easier by the near-synchronous appearance of Bronze Age cultural groups in the entire region c. 2200 BC (Krause, 1989; Pearce, 1998: 57; David-Elbiali, 2000: fig.14; Della

Casa, 2013: fig. 39.3; Nicolis, 2013: 694; Roberts et al., 2013: fig. 2.1), with a possible slight delay in south-eastern France (Strahm, 2005: 33; but this is now denied by Guilaine et al., 2001, and Mille & Carozza, 2009).

Another aspect of the chronological knot lies in the clear-cut subdivision of the Early Bronze Age into three major phases based on Reinecke's chronology, which has long provided a solid framework for dating central European prehistory (Table 2; see also, among others, David-Elbiali, 2000: fig. 14; Krause, 2003: fig. 34; Roberts et al., 2013). This subdivision has good correspondence to the chronological framework drawn by De Marinis (2005) for northern Italy (Table 2; see also Nicolis, 2013: 694), bearing in mind that our research is solely concerned with the beginning of the sequence in Reinecke's phase Bz A1. The main issue here is that, even when agreeing upon the same overall chronology, different authors propose different absolute dates for the same objects. This is the case with De Marinis (2005, 2006b) vis-à-vis Carancini (1996; see also Carancini & Peroni, 1999) for northern Italy, and with Krause (2003) vis-à-vis David-Elbiali (2000) with regard to western Switzerland. Considering that Carancini's framework has been questioned repeatedly (De Marinis, 2005, 2006a; Dolfini, in press), we have decided to disregard it for the purpose of this research. Moreover, De Marinis' chronology holds the distinct advantage of being comparable with Krause's, with the exception of certain objects from the Italian north-west. In this work, we have tried to take into consideration the different dates provided by these scholars, and to explain how our subsequent interpretations differ according to their systems.

Interpreting the Chemical Data

In this work, we have not analysed the chemical composition data using cluster analysis, which, since Ottaway (1982), has been the most common method used on metal chemical datasets. Cluster analysis results in the creation of a number of static metal compositional groups, each of which is commonly considered as coming from a specific source. We think that this obscures useful archaeological structures in the data associated with recycling, oxidative loss linked with use,

mixing, alloying, and so forth (Bray & Pollard, 2012). These changes have been demonstrated experimentally (McKerrell & Tylecote, 1972), and have long been recognized in industrial metallurgy (Hampton et al., 1965; Charles, 1980; Beeley, 2001). The chemical shifts over the course of a unit of metal's 'life-time' means that it could easily move between the very fine divisions defined in some classification schemes — see, for example, the mathematically defined categories of the SAM project: Junghans et al. (1960, 1968). Clustering places artificial boundaries on interpreting material flows over time and space. Instead, to aid interpretation, we place the data into simple pre-defined metal categories by considering the presence/absence of arsenic, antimony, silver, and nickel (see Table 3). These can be thought of as heuristic categories intended to broadly capture variation within the dataset, which is then further investigated in a number of ways (see below). To determine presence and absence, we first subtract tin from the total, and then normalize the data to 100 per cent. The aim of this procedure is to identify the main chemical signature of the copper-alloy base, free from the diluting effect of alloying. Characterizing the level and distribution of tin is of course crucial to understanding metal flow, and occurs in parallel with investigating the copper-base composition.

A threshold value of 0.1 per cent for 'present' was used for As, Sb, Ag, and Ni, after correcting for alloying (Bray & Pollard, 2012). These four elements were chosen firstly as all past chemical composition projects have analysed for them. We also need elements that have been consistently recognized as being diagnostic of copper identity in some way. As, Sb, Ag, and Ni, with their different chemical properties, mechanical properties and natural abundances fulfil this requirement, and they have been commonly used in previous chemical-typological work (such as the various SAM schemes, and Northover, 1980). These four elements capture much of the variation seen in copper mineral deposits, pass into the metal during the smelt, and then behave differently during technological processes — for example, different oxidation rates upon melting. Possible issues over the heterogeneity of metal, for example segregation of arsenic, may be resolved with a proper sampling strategy, which Pernicka describes as taking at least 3 mg of metal. This is a

threshold widely passed, at least for the SAM project (Pernicka, 1986: 25), which is the core of our database. The use of these four elements gives us a reliable, if broad, picture of the early history of metal flow. Of course, other elements could profitably be investigated, and will be in future research. However, our current approach has been tested in detail for the British Isles (Bray & Pollard, 2012), and has the distinctive advantage of making data processing and grouping manageable due to the number of elements involved. Bismuth was not taken in consideration, even if it is claimed to be a diagnostic element for the mineral deposits (Pernicka, 1999, 2011, 2014), because, when present, it is often found in very small quantities, well below our threshold (0.1 per cent). This is often close to the detection limit of analytical instruments, so that we may have too many false negatives in the dataset. More importantly, it was not analysed by all the authors and, had we decided to include it, the consistency of our database would have been affected. Similarly, lead was not considered in this phase of the research because it is completely insoluble in copper, thus leading to high segregation rates in the objects. In any case, less than 10 per cent of objects in our database pass the threshold of 0.1 per cent lead.

The resulting sixteen compositional groups (all possible combinations of presence/absence for the four trace elements) are outlined in Table 3. This approach aims to objectively capture variation within the whole assemblage — each compositional group is not seen as deriving from a single source, and each source is not necessarily expected to produce copper of just one compositional group. Also, through recycling, mixing and smithing, a unit of copper may pass through a number of groups in its ‘lifetime’ (Bray & Pollard, 2012). This kind of classification allows the metal chemistry to be considered from an archaeological rather than a purely statistical perspective, in order to characterize the flow of metal, rather than just as clusters of maximum difference in the final composition. In other words, the sixteen groups are a starting point to understand the process of metal flow, a process that encompasses the whole ‘lifetime’ of a unit of copper: from the extraction of the ore from a specific mine, through possible alloying, manipulation, mixing and recycling. Our approach aims to tease apart the complete sequence of

factors that affect the final chemical composition of a copper-alloy assemblage, rather than conflate them into one step of statistical analysis. The compositional data are linked with the coordinates of the find-spots using ArcGIS 10.2, which allows us to investigate the relationship between metal groups and topographical elements, in particular rivers and watersheds. We do not simply consider the raw number of objects with a certain composition per zone, but instead analyse the ‘percentage presence’ of the chemical groups. This approach is also known as Ubiquity Analysis, and has been fruitfully applied to datasets from a range of disciplines including archaeobotany and palaeoclimate reconstruction (Bray et al., 2006).

Results: A History of Alpine Metal

Copper Age: Copper Compositional Groups

In the Copper Age (*c.* 3800/3600–2200 BC), roughly 55 per cent of all the analysed metal artefacts were made of either pure copper (Group 1; Figure 3) or copper with traces of arsenic (Group 2; Figure 4). Artefacts with these compositions are distributed all over the Alps, but the map shows slight east-west differences that may be significant: whereas arsenical copper (Group 2) was slightly more dominant in the north-western part (33 per cent of all objects were made from arsenical copper versus 29 per cent of pure copper), pure copper (Group 1) predominated in the north-east (36 per cent versus 23 per cent of arsenical copper). In northern Italy, the prevailing composition was arsenical copper (Copper Group 2: 26.5 per cent). Interestingly, the second most common composition was not pure copper, but copper with arsenic and silver, in particular in the eastern Alps (Group 9; Figure 5). The well-known relationship between composition and artefact classes in the Copper Age should be remembered, namely that daggers tend to be made of arsenical copper and axes of pure copper (De Marinis, 2006b; Pearce, 2007; Dolfini, 2014b). As has been noted before, in copper chemical groups that contain both axes and daggers, for example, Group 9 (As and Ag), daggers have a higher average arsenic level than axes. Our new model can offer an

explanation for this, as arsenic is lost from axes as they are heated, re-smithed and recycled.

Meanwhile, daggers tend to be sharpened through abrasion at room temperature, with no associated chemical changes (see Bray & Pollard, 2012, for similar case studies).

Overall, however, what this brief survey underlines is the importance of a large number of different copper groups during the Copper Age across the Alpine region. Considering the entire zone, arsenical copper and pure copper together represent approximately 50 per cent of all artefacts: the remaining 50 per cent is made of several different copper groups, manifested as various combinations of arsenic, antimony, silver, and nickel. In the Alpine Copper Age, at least seven different copper groups, apart from pure copper and arsenic copper, are represented, in a range of 8 per cent (Group 4) to 2 per cent (Group 12). These seven groups often have clearly defined, restricted distributions. For example, group 4 is more ‘eastern’ (Figure 6), whereas groups 5 and 7 are more ‘western’ (Figures 7 and 8).

Group 4, defined as copper with low levels of silver, has a clear eastern distribution. In the north-east of Italy, it is particularly prevalent and is more common there than pure copper and arsenic copper, contrary to the general pattern of the alpine region. In particular, this copper composition is closely associated with the Bocca Lorenza type axes. These are common in the northern-west part of Italy, and are represented, besides the axes from the burial in cave of Bocca Lorenza, by one axe from Merendole (PD) and one in Tormičeva cave in San Canziano (TS) (Pearce, 2007: 45). In the northern zone of the Alps, grip-tongue daggers also show the copper group 4 signature (copper with low levels of silver). These artefacts are often related to Bell Beaker groups, for example the daggers found in Kirchheim (Krause, 2003), Wolfarshauser, and Moosinning.

Copper Group 9 (silver and arsenic as minor elements) also has a predominantly eastern distribution. Most of the artefacts from the Remedello cemetery have this composition signature, in particular the daggers of Remedello type, whereas the Beaker-style dagger found in the cemetery is

made of arsenical copper (Group 2). Many objects from Trentino are made of copper belonging to Group 9. This is the case for the beads and an awl from La Rocca di Manerba, Frana del Bersaglio, Riparo Gaban, Arco, and the axe from Col del Buson. In the northern zone of the Alps, this composition is mainly found in flat axes, as in the cases of Lieferind, Attersee, Rainberg, Mondsee.

Group 5, nickel as the only trace element, is typical of Switzerland, broadly corresponding to the SAM copper group FC (Junghans et al., 1960, 1968b, 1974). Its focussed distribution was recognized by SAM, and also, more recently, by Matuschik (2004) and Cattin (2008). Strahm (1994: 29, 2005: 32) hypothesized a west-Alpine source and notes its presence in France. This copper group is also found in the Southern part of the Alps, for example in the Copper Age cemetery at Sabbione (Pearce, 2007: 85).

The antimony-silver pattern of Group 7 is commonly linked with the early mines excavated near Cabrières in southern France (Ambert, 1995; Bourgarit & Mille, 2005; Prange & Ambert, 2005). As with many of the other copper patterns this type has clear regional hotspots and associations with particular object classes. In south-western France ornaments often show the Sb-Ag signature, in particular beads found in burials in dolmens (such as Dolmen des Cudières) and caves (for example Le Baume de Lan). In Switzerland, on the other hand, this composition is more often found in axes, such as those from La Graviers, Vallamand, Treytel, and Vinelz. Similarly in Italy, the axe from Fiesse is group 7, which was potentially found in a Beaker Culture context (Krause, 2003).

Overall, the picture that builds up is one of a diverse range of subtly different copper types in use across the Alpine region, many of which have clear and limited geographical ranges and associations with particular object classes. This seems to indicate societies in the Copper Age mainly extracted and smelted copper locally, and did not trade the metal widely outside of their territory. It is important to note that for this period, whether we classify a certain group of objects based on Krause's, De Marinis' or David-Elbiali's chronology, the overall result is the same. In

other words, even considering slight differences in the general chronology of the objects, the same metal groups and distributional patterns consistently stand out as significant.

The Early Bronze Age 1: Copper Compositional Groups

The transition between the Copper Age and the Early Bronze Age (*c.* 2200–2000 BC) is accompanied by a clear change in the trace element composition of the artefacts. Compared to the Copper Age, the chemical groups are far more consistent across the entire study area, suggesting the growth of fewer but larger-scale exchange networks. Almost 80 per cent of all the artefacts dating to this period fall into either Groups 12 (Figure 9), 16 (Figure 10) or 1 (Figure 11). Almost half of all the artefacts dated by Krause as Bz A1 belong to Group 12 (copper with As, Sb and Ag), and are mostly found in the northeast Alps. This group represents the famous Ösenringe chemical composition, and indeed 80 per cent of the Group 12 objects in the Alpine region are Ösenringinge from hoards. Many of these depositions are extremely large, containing more than a hundred artefacts. Axes and daggers in the north-east of our study zone have a similar composition.

Group 16 (copper with low levels of arsenic, antimony, silver, and nickel) is also a very significant composition pattern: comprising roughly 22 per cent of the artefacts dated to Bz A1. This group seems to have been more common in the western and central regions, with a particular focus on the Rhine valley and the northern parts of the Rhône, Oglio and Adda rivers. Outside of this principal distribution, there are occasional deposits of objects made from Group 16 copper. For example, the Wolnzach, Germany axe hoard from the north east of the study area. This collection of local, north-east Alpine style ‘Saxon’ axes (Kienlin et al., 2006: 462; Kienlin, 2010: 137) stands out in an area dominated by the use of Group 12 copper. Hence, we have a case of artefacts made of copper typical for another region but with the typology of the local zone. This might be interpreted as evidence for the movement of a coherent batch metal, which was then recast.

If we consider the Alps as a whole Group 16, copper was used in the production of all categories of artefact, and was less closely tied to a specific type, as with Group 12 and the Ösenringe. It has to be noted, though, that as we move from east to west, the number of hoards made of Ösenringe decreases and, at the same time, the number of hoards composed by axes goes up. These axes mainly belong to Group 16.

The third most important composition is pure copper (Group 1, approximately 15 per cent of the Initial Early Bronze Age assemblage), which is found most commonly at the western end of our study area, roughly corresponding to the Rhône valley. As in the Copper Age, the use of different chronologies does not change the general patterns of copper distribution (or is it composition) across the Alpine range. Awls and pins in French burials commonly have this copper signature, particularly assemblages found in caves, such as Grotte de la Carrière. In western Switzerland, again, awls, pins and lunulae from burials (e.g. in Les Places) are often Group 1 copper, though these are not cave depositions. In the eastern part of the Alps, away from the group's main distribution, there are occasional finds of very pure copper Ösenringe deposited in hoards, for example from Sirndorf, Stockerau, Bergen, and Eiselfing. Here again, we have an example of metal with a composition typical of one region (the west) used to make objects characteristic of another (the eastern Alps).

In northern Italy, the pure copper, Group 1 composition was related to axe-hoards deposits, which are reminiscent of western alpine behaviour, such as the Remedello Sotto and Serravalle assemblages (Perucchetti, 2008). The typology of these axes, similar to the Type Neyruz (Tecchiati, 1992), is also related to the western zone of the Alps.

The Appearance of Tin in the Early Bronze Age

The east-west pattern observable in the Copper Age copper groupings is echoed at the beginning of the Early Bronze Age (c. 2200–2000 BC), when copper-tin alloys make their first appearance.

Figure 12 shows a map of the mean percentage of tin per site based on Krause's chronology. Objects classified archaeologically as *Ösenringenbarren* and *Spangenbarren*, and which tend to contain only copper, were excluded from this analysis, because, if they were used as copper ingots (De Marinis, 2006b), then their overwhelming presence in the eastern region would have significantly underestimated the presence of tin. We are focussing here on finished objects, rather than copper in a putative trade form.

It is clear from Figure 12 that the mountains were not a barrier to the north-south distribution of tin, but there was a strong cultural barrier, a 'tin line', which divided the western part of the Alps, where tin was frequently used to alloy copper, from the east, where it was not. It is also important to note that the alloying process of adding tin to copper was not linked to any specific copper compositional group mentioned above. This strongly suggests that tin was moving independently of the copper.

If the chronologies of David-Elbiali and De Marinis are integrated in western Switzerland and northern Italy, the tin line is not evident in the first phase of the Early Bronze Age 1. As mentioned, David-Elbiali and De Marinis tend to postdate objects; hence, with their chronological framework, the tin line reappears in a second phase of the Early Bronze Age. Overall, if we take a broad approach and consider the entire Early Bronze Age (encompassing the different attributions of objects to one or another sub-phase of the Early Bronze Age), use the chronology proposed by De Marinis and David-Elbiali when possible, and exclude hoards (which were made of hundreds of objects that might be considered as copper ingots), we still have a picture of a higher percentage of objects made of bronze in the western part rather than the eastern.

Figure 13 shows the percentage of objects alloyed with tin in each zone in a second phase of the Early Bronze Age according to the chronology proposed by De Marinis and David-Elbiali. The blue-tinted squares indicate the zones where there is a greater percentage of objects containing tin. To the west of the tin line, there was a much higher percentage presence of bronzes: over 97 per cent in most places. Moreover, in particular in Switzerland, in a relatively small region there is a

concentration of sites whose assemblage had a significantly high level of tin (e.g. cemeteries at Hubel, Scloss, Thun, and Ecublens). On the other hand, in the east there was a zone of lower tin presence, with most areas having only 60–85 per cent of their metal assemblage as bronze. A particularly interesting result appears if we divide our chemical dataset by artefact category. Ornaments show a particularly strong division between coherent areas of tin-bronze use compared to the continuation of copper use (Figure 14). Tin addition would produce a brighter, more lustrous object, therefore the pattern of its use in ornaments perhaps supports the concept that display and colour, rather than mechanical properties, influenced the adoption and use of tin-bronze.

Overall, whichever chronology one adopts, we can conclude that in the central zone of the Alps, east of our tin line, there was a higher continuity of copper rather than bronze use. It is important to add to this that there is an area of high bronze use on the eastern edge of our study area. This was probably influenced by Central Europe and may be related to the alloying patterns of groups such as the Unetice Culture rather than the Alpine region. Finally, it is important to note that the Alps were not a north/south barrier to the presence or absence of tin, but that the same scenario appeared on both sides of the mountains.

Topography: A Key Element

As described above, the distribution of different groups of metal is not random, but shows a series of clear geographical patterns over time. Can we push this line of reasoning beyond the concept of two-dimensional geography and deal with topography? When we speak about metal flows, we are, ultimately, referring to the movement of people, which, of course, is influenced by the topography of the territory. One individual need not accompany objects through their entire journey, but access points and mutual meeting places must have been essential. In the Alps, attention naturally turns to the passes between the mountains, but it is important to connect these with their associated water and valley system. Rivers may reasonably be described as the ‘highways of the continent’ (Cunliffe,

2008: 38–47) in prehistory. Large rivers, such as the Danube, Rhine, Rhône, and Po would have been easy to navigate, even upstream, especially with the help of sails or human and animal traction. As Van de Noort (2013: 390) claims, ‘There is no doubting that the earliest boats in Europe were built for use on rivers and lakes, and that the types of craft that enable riverine traffic include hide- and skin-covered boats and logboats’. The oldest logboat from Italy is from Lake Bracciano, Central Italy, and is dated to the sixth millennium BC (Fugazzola & Mineo, 1995). However, most of the logboats found in Italy are from the north, from both lakes and rivers, and some of them have been dated to the Early Bronze Age. Unfortunately, there is a general lack of research on the chronology of these crafts as well as on their social role and technological development throughout the Bronze Age (Ravasi & Barbaglio, 2008). But even if they were not navigated, the importance of rivers is hardly diminished: for terrestrial travel, rivers work perfectly as reference points and as a secure marker of the path, and as a reliable source of potable water. The key role of rivers increases in the Alpine region, where glacial river valleys work as corridors between mountains, offering in energy terms the least costly transalpine paths.

Using GIS we tested the hypothesis that different metal groups were likely to follow the flow of different rivers. Using ArcGIS there is the facility to draw the watershed of each river, and since each artefact is linked to a site and, ultimately, to its coordinates, it was therefore possible to create a spatial join between each artefact and the watershed to which it belongs. Thus, for each artefact, two kinds of information are provided: the metal compositional group and the watershed to which it belongs. Therefore, it was possible to undertake a statistical analysis to verify the correlation between watersheds and compositional groups. The χ^2 test suggests that the distribution of different metal groups was not random with respect to watershed, neither in the Copper Age nor in the Bronze Age A1, with a p-value of less than 0.005 in both cases.

As summarized by Figure 15, in the Copper Age, the Rhône watershed was characterized by a dominance of arsenical copper (Group 2); the Adige and Po rivers have similar levels of pure copper (Group 1), but they differ in the percentage of Groups 5, 7 and 9 present. The Rhine, Danube

and Piave watersheds have significantly higher levels of pure copper than those of the Rhône, Adige and Po (Figure 15). Two points should be highlighted: the specific presence of group 5 (Ni) in the Rhine and the high values of group 9 (As, Sb) in the Piave, which, combined with the percentages in Po and Adige, reflect the previously-mentioned importance of this group in the north-east of Italy.

In the initial Early Bronze Age (Figure 16), the specific presence of different copper groups per watershed becomes more evident, with the exception of the Po and Rhine, which have extremely similar patterns. The importance of copper without impurities (Group 1) in the Rhône region is confirmed, whereas the Danube has a predominance of Group 12 (As, Sb, Ag), while Group 16 (As, Sb, Ag, Ni) dominates the Rhine and Po watersheds

It is interesting to note that the Rhône and Rhine had different patterns of copper group presence, because the tin line runs between these two rivers. In the Rhône, tin is present in all three dominant copper groups, 1, 12 and 16, while tin is generally absent from the Rhine's metal artefacts made of the same three copper groups. Copper was flowing across this line, Group 1 from the west, and Group 16 from the east into the west. However, tin does not accompany this copper movement, remaining prominent only in the western Alps. Moreover, we should note that in the initial Early Bronze Age the higher similarity of the patterns of the Rhine and Po compared to the Copper Age allows us to hypothesize close transalpine contacts and exchange of metal between these two areas.

Discussion

The transition from the Copper Age to the Early Bronze Age marked a change in Alpine metal production: from local production with a great variety of copper types in circulation, to a picture of large flows of metal of specific types. In particular, three types of copper were very common in the first phase of the Early Bronze Age: copper with As, Sb, Ag (Group 12), copper with As, Sb, Ag and Ni (Group 16), and copper with no trace elements (Group 1). This concurs with the conclusions of Krause (2002). Apart from Group 1, these are new dominant groups, indicating that there was a

new way of procuring metal from the Copper Age to the Early Bronze Age, with the possibility of new sources being exploited. Though each of these groups tend to have a zone where they dominate the artefact assemblage, it is interesting to note that there are several examples, especially in hoards, of objects that have a local typology but which are made from copper with the signature typical of another part of the Alps. These deserve more detailed study as they may provide the clearest case studies of how metal was moving through and around the Alpine zone.

The Early Bronze Age may also have seen the establishment of an important tin line running north-south across the Alps, from Oglio and Adda in northern Italy to Reuss and Rhine in Switzerland, with tin-bronze far more prevalent in the west than in the east. Although this line comes from a fresh analysis of the metal data, it mirrors well-known cultural boundaries and contact networks: in the north alpine region the tin line corresponds well to the boundary between the Early Bronze Age Rhône culture in the west and the Blechkreis culture in the east (Merkl, 2011: Figure 4.8). But it is also a witness to the importance of river systems: the western part was connected with the Rhône valley, whereas the eastern part communicated preferentially with the Rhine and, ultimately, the Danube watersheds. The question of the source of this tin is still open. Based on the analytical work of the SAM group (Junghans et al., 1960, 1968), we may postulate that there was an early movement of tin as an alloying material from France up the Rhône, but to identify the source of this flow requires further research.

Even more interesting is the fact that this tin line continues to the south of the Alps. In contrast with the river system model for the north of the Alps, the line neatly cuts the River Po, which is navigable, into two discrete areas. However, as in the northern Alps, the tin line marks the expression of cultural differences. In 1989, Peroni stated: ‘It may seem that, regardless of the Alpine barrier, and more important than it, a cultural barrier [...] cut Northern Italy transversely from North to South.’ (Peroni, 1989: 364–65). He was referring to the Middle and Late Bronze Age, and in particular to metallurgy from a typological perspective. Since then, the idea of a cultural barrier has appeared constantly in Italian studies discussing the Middle Bronze Age and later

periods. For example, two separate spheres of exchange, the first linking the south-western with the north-western Alps and the second linking the south-eastern with the north-eastern Alps, have been recognized by De Marinis (2006a) based on the find-spots of certain types of swords, and similar observations were put forward by Venturino Gambari (1998) and Baioni (2008). Two points are of special interest here: firstly, that the east-west separation of northern Italy was extremely long-standing, since it remained visible from the mid-second millennium to the mid-first millennium BC, and perhaps for longer; secondly, that it was initially marked by the manufacture and circulation of metalwork, but quickly became visible in other domains of social action, including pottery production and use.

The results of our research raise the question as to whether this ‘cultural barrier’ emerged in the Middle Bronze Age following the patterns of tin exchange highlighted above, or whether metal movement in the EBA was dependent on already existing barriers. Significantly, an east-west division first emerged in northern Italy during the Late and Final Neolithic (*c.* 4500–3600 BC), when the appearance of the Chassey-Lagozza pottery style in the north-west pushed the boundaries of the Square-Mouthed Pottery style, previously found all over the region, further east. Whereas the former style shows strong connections with southern France and the western Alps, the latter is linked to cultural development in the north-eastern Alps and Slovenia; the frontier between the two areas, which is not clear-cut, runs from the western shores of Lake Garda to the northern Apennines, slightly to the east of the cultural boundaries and circulation zones visible in the Bronze Age (Barfield, 1996: 67; Mottes & Nicolis, 2002; Pessina & Tiné, 2008: 99, fig. 3c–d). It is also worth noting that, in the same time period, the area encompassing the Rhône watershed, Piedmont and Liguria, lay at the core of the Europe-wide, westward-looking network of polished stone exchange (Barfield & Broglio, 1966: 63–65; Bouard, 1993; Bouard & Fedele, 1993; Venturino Gambari et al., 1996; Pétrequin et al., 2005, 2012).

The picture is less clear in the northern Italian Copper Age (*c.* 3600–2200 BC), which is characterized by the break-down of the extensive pottery styles of the Late Neolithic. At domestic

sites, these were replaced by a plethora of ceramic traditions and decorative motifs, which were often re-elaborated and re-combined in original ways even at sites lying at short distance from one another (Cocchi Genick, 2012: 498). In contrast, funerary sites feature significant super-regional similarities in burial behaviour and grave goods, which arguably reflect new, widespread ideas concerning the treatment of the dead and the importance of the ancestors (Dolfini, 2004, in press; Barfield, 2007; Fedele, 2013). Yet, even in the absence of clear markers of long-ranging exchange, the survival of the previous east-west division is hinted at by the directional movement of raw materials, goods and ideas across the Alps. In the western quadrant, certain shapes and decoration styles are found in pottery from Piedmont, eastern France and Switzerland (Venturino Gambari, 1998: 52–56); in the eastern quadrant, the trans-alpine networks emerging in the Final Neolithic for the exchange of the earliest metal objects were further developed in the Copper Age to include north-east Italy, Austria and Slovenia (Carancini, 2001; Visentini, 2009; Klassen, 2010; Dolfini, 2013). It is thus suggested that the circulation patterns of copper and tin highlighted in this study may reflect enduring cultural demarcations, which date back to the late fifth millennium BC.

We have shown here that the alloy of copper with tin seems to have been first adopted in the western part of the Alps, or, at least, contemporaneously with the far eastern edge of the study area, leaving a gap in the central part of the Alps, thus suggesting some level of continuity from the Neolithic through to the Iron Age and beyond. Remarkably, in the Alps tin was consistently added to all of the copper groups that were in circulation, including pure copper and copper with small percentages of arsenic, antimony silver and nickel. This is very important because on the one hand it suggests that tin was moving separately to the copper and was being alloyed locally (as opposed to copper-tin alloys arriving pre-made from a single remote source), and on the other hand it may say something about the reason why people decided to add tin. The presence of trace elements, especially when combined together, increases the ductility, malleability and also hardness of the finished products. In addition, it lowers the melting point of copper (Northover, 1989; Kienlin et al., 2006; Merkl, 2011: 77–78, 2011: 7.1). Nevertheless, the addition of tin seemed not to take account

of this, since it was added both to pure copper and to copper with significant levels of trace elements. It therefore seems likely that the addition of tin was made, at least in part, for non-functional reasons (a 'recipe', a 'ritual', or 'a good way to make metal'), implying that perhaps colour and display were influencing factors. One could speculate that it would have made a powerful statement to be able to obtain and use such a rare material, particularly one that could be displayed so strikingly. This reading is also supported by the fact that the most clear-cut difference in the adoption of tin between the western and eastern areas is seen in ornaments (Figure 14). Identifying these social choices, in part through artefact chemistry, underlines the importance of linking different datasets together. As Pearce (1998, 2007) laments, collections of archaeometallurgical data tend to be underused, often being limited to consultation when considering provenance or technology. In this case, however, we manage to have a hint of 'what actually happened' (Pearce, 1998: 53).

The idea that the use of copper-tin alloy spread throughout Europe from the Near East and Anatolia has long been the accepted model (e.g. Pare, 2000; Roberts et al., 2009). However, the early use of tin in the western part of the Alps perhaps reflects more detailed local patterns of metal movement, which may occasionally go against a simplistic radiating pattern from a single area or region. Without denying the overall westward-spreading model, Primas (2003) highlighted that in the south-east of Europe tin alloying was contemporary with lead alloying, whereas in central Europe tin alloying was absolutely predominant and lead was not used regularly until the Late Bronze Age, despite there being accessible sources of lead. According to her model, in central Europe there was a first phase in which 'selective use' of tin may have come from the south-east and involved a small number of objects. In a later phase, however, tin alloying became common practice in the British Isles, where abundant sources of the metal were available in Cornwall and Devon; thence the new technology would have spread to Central Europe, perhaps taking advantage of the old communication channels provided by the Bell-Beaker phenomenon. Primas' (2003)

model provides a plausible explanation for the patterns highlighted in this research, and in particular for the early appearance of tin in the western Alps.

Conclusion

In conclusion, through linking the chemical composition, location and typology of early metal artefacts in the Alpine region, we have shown that the Alps did not provide an insurmountable north-south barrier to the circulation of metalwork, as copper with similar impurity patterns are found on both sides of the range. Similarly, the spread of tin was not affected by the topography of the mountain range. Compositional copper groups were not randomly distributed, but reflected ore distribution, river flows and their watersheds. Hence, it seems that river systems deeply influenced the movement of raw materials and ideas, and, probably, of people. Occasionally though cultural barriers can be demonstrated to be a more powerful influence, as is the case with the tin line cross-cutting the Po river.

If the concept of the Alps as a physical barrier is rejected, we can recognize instead an important cultural barrier that created an east-west division in the flow of metalwork. In the north of the Alps, this reflects the river system, whereas in northern Italy, it cuts the Po valley in two, approximately between the Oglio and Adda rivers. This is important because the Po, which is a navigable river, becomes a remarkable exception to the river system transport network hypothesized by Cunliffe and seen north of the Alps. This barrier mirrors perfectly patterns seen in the typological study of Middle and Late Bronze Age metal artefacts, but the new synthesis presented here allows us to push its origin back in time to the Early Bronze Age and perhaps earlier. It may even be speculated that its origin is grounded in surprisingly long-standing Neolithic exchange networks and thus predates the emergence of metallurgy in the region.

A final observation must be made about the existence of different chronologies for the region in question. A unified chronological framework for the Alps is still work in progress: the present situation is not ideal for a synthetic study such as the one carried out here. Further research

is required, whose ideal outcome is an integrated chronology for the entire Alpine region, underpinned by new radiocarbon dates directly associated with metal objects. Nevertheless, we are confident that the major patterns described here are so fundamental to the flow of metals in the Alpine Copper and Early Bronze Ages that they will probably survive any future revision of the local sequences. It is only when a new, finer-grained chronology becomes available that the scientific, archaeological and geographical datasets discussed in this article can be cross-referenced to give a more nuanced insight into people's relationship with metal and their environment in the Alpine region.

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Table Captions

TABLE 1

PREVIOUSLY UNPUBLISHED COMPOSITION DATA COURTESY OF PETER
NORTHOVER

TABLE 2

CHRONOLOGICAL PHASES USED IN THIS WORK AND CORRESPONDENCE BETWEEN
THE AUTHORS

TABLE 3

DEFINITION OF COPPER GROUPS USED IN THIS PAPER

Figure Captions

FIGURE 1 Map showing the research area.

FIGURE 2 Table used to convert symbols in number.

FIGURE 3 Distribution map of Copper Group 1, copper without trace elements in the Copper Age. The size of the dots indicates the number of objects. The colour scale represents the percentage of each geographical square's metal assemblage that belongs to Copper Group 1.

FIGURE 4 Distribution map of Copper Group 2, arsenic copper in the Copper Age. The size of the dots indicates the number of objects. The colour scale represents the percentage of each geographical square's metal assemblage that belongs to Copper Group 2.

FIGURE 5 Distribution map of Copper Group 9, copper with arsenic and silver in the Copper Age. The size of the dots indicates the number of objects. The colour scale represents the percentage of each geographical square's metal assemblage that belongs to Copper Group 9.

FIGURE 6 Distribution map of Copper Group 4, copper with silver in the Copper Age. The size of the dots indicates the number of objects. The colour scale represents the percentage of each geographical square's metal assemblage that belongs to Copper Group 4.

FIGURE 7 Distribution map of Copper Group 5, copper with nickel in the Copper Age. The size of the dots indicates the number of objects. The colour scale represents the percentage of each geographical square's metal assemblage that belongs to Copper Group 5.

FIGURE 8 Distribution map of Copper Group 7, copper with antimony and silver in the Copper Age. The size of the dots indicates the number of objects. The colour scale represents the percentage of each geographical square's metal assemblage that belongs to Copper Group 7.

FIGURE 9 Distribution map of copper with Copper Group 12, arsenic, antimony and silver in the first phase of the Early Bronze Age (A1). The size of the dots indicates the number of objects. The colour scale represents the percentage of each geographical square's metal assemblage that belongs to Copper Group 12.

FIGURE 10 Distribution map of Copper Group 16, copper with arsenic, antimony, silver and nickel in the first phase of the Early Bronze Age (A1). The size of the dots indicates the number of objects. The colour scale represents the percentage of each geographical square's metal assemblage that belongs to Copper Group 16.

FIGURE 11 Distribution map of Copper Group 1, copper without traces elements in the first phase of the Early Bronze Age (A1). The size of the dots indicates the number of objects. The colour scale represents the percentage of each geographical square's metal assemblage that belongs to Copper Group 1.

FIGURE 12 Map of the mean percentage of tin per site in the first phase of the Early Bronze Age (A1), using Krause chronology.

FIGURE 13 Distribution map of tin-bronze presence, and the mean percentage of tin per site, in the second phase of the Early Bronze Age (A2a), using David-Elbiali and De Marinis' chronology. The colour scale represents the percentage of each geographical square's metal assemblage that is bronze.

FIGURE 14 Map of the mean percentage of tin per site for only ornaments. Second phase of the Early Bronze Age (A2a), using David-Elbiali and De Marinis' chronology

FIGURE 15 Percentage presence of each metal group per watershed in the Copper Age.

FIGURE 16 Percentage presence of each metal group per watershed in the first phase of the Early Bronze Age (A1).

Analys s	Item	Site	Context	sf	Period	Cu	Sn	Pb	As	Sb	Ag
MC10	Droplet	Monte Covolo	S1E1	M5	CA	99.74	0.01	0.02	0.28	2.56	1.24
MC11	Droplet	Monte Covolo	S3E1	M5	CA	96.04	0.02	0.03	0.16	0.79	1.25
MC12	Waste	Monte Covolo	S2W2		CA	99.72	0	0.26	0	0	0
MC13	?Bead	Monte Covolo			CA	99.82	0	0.04	0	0	0
MC6	Awl	Monte Covolo	N3W2	M2	CA	95.31	0.02	0.03	0.22	1.89	1.08
MC8	Droplet	Monte Covolo	S2E4	M6	CA	88.86	0.04	0	0.78	5.71	1.86
MC9	Wrought frag.	Monte Covolo		M4	CA	99.38	0	0.12	0.27	0	0.18
MC95/1	Copper strip or awl	Monte Covolo	MC92 F 46		CA	96.72	0.01	0.02	2.67	0.05	0.19
MC95/10	Tubular bead frag.	Monte Covolo	MC93 RF 60	2325	CA	94.28	0.01	0.05	0.1	0.01	0.03
MC95/11	Sheet fragments	Monte Covolo	MC93 RF53	1979	CA	51.12	0.04	0	0.27	0	0.01
MC95/12	Tubular bead frags.	Monte Covolo	MC94 RF 96	3469	CA	99.72	0	0.14	1.93	0.02	0.04
MC95/2	Tubular bead	Monte Covolo	MC93 RF 67	2511	CA	99.25	0	0.01	0.31	0.01	0
MC95/3	Awl/wire	Monte Covolo	MC93 RF 78	3091	CA	91	6.89	0.5	0.58	0.26	0.06
MC95/4	Tube fragments	Monte Covolo	MC92 RF 47	1421	CA	99.79	0	0.02	0.1	0.02	0
MC95/6	Tubular bead	Monte Covolo	MC92 RF 29	1459	CA	98.59	0	0.02	1.29	0	0.02
MC95/7	Fragment	Monte Covolo	MC94 RF 94	3597	CA	99.84	0	0	0.1	0	0.03
MC95/8	Large tubular bead	Monte Covolo	MC94 RF 96	3509	CA	98.55	0.01	0.21	1.03	0.02	0.08
MC95/9	Cast fragment	Monte Covolo	MC94 104		CA	99.57	0	0.01	0.25	0	0.02
TBC1	bar ingot	Valle di Non			A1		tr	0	0.92	0.01	0.02
TBC2	shaft-hole axe	Valle di Non			A1		3.21	0	0.06	0.02	0.21
TBC3	bar ingot	Valle di Non			A1		0.03	0	9.72	0.21	0.06
TNS1	wire fragm.	Moletta Pattone			CA		0	0.82	0.12	0.02	0.07
TNS10	wire, fragm.	Moletta Pattone			CA		0	0.19	tr	0.03	0.07
TNS11	spiral bead	Vela Vabusa			CA		0	0.03	3.2	0.03	0.04
TNS12	armlet	Bersaglio dei Mori			CA		tr	4.27	0.78	0.03	0.28
TNS15	needle	Riparo Gaban			CA		0	0.69	0.2	0.02	0.2
TNS16	riveted strip	Dos della Forca			CA		0	0.07	0.05	0	0.06
TNS2	large bead	Moletta Pattone			CA		0.02	0.13	0.11	tr	0.18
TNS3	spiral	Moletta Pattone			CA		0.01	0.34	0	tr	0.27
TNS4	strip	Moletta Pattone			CA		0	0.59	0	0	0.28
TNS5	strip	Moletta Pattone			CA		0	0.39	0.1	0	0.36
TNS6	strip	Moletta Pattone			CA		0	0.07	0.05	tr	0.19
TNS7	wire	Moletta Pattone			CA		0	0.36	0.06	tr	0.04
TNS8	wire	Moletta Pattone			CA		tr	0	0.07	0	0.35
TNS9	wire	Moletta Pattone			CA		0	0.21	0.07	0	0.09

Table 1: Previously unpublished composition data courtesy of Peter Northover

	Dates BC	Krause	David-Elbiali	De Marinis
CA	≈4000–2200	10, 12, 15, 20, 22, 25	–	Neolithic/CA
A1	2200–2000	3	A1	A1a
A2a	2000–1800	4	A2a	A1b/A1c
A2b	1800–1600	5	A2b	A2

Table 2: Chronological phases used in this work and correspondence between the authors.

Elements present at 0.1%	Group
Cu	1
Cu, As	2
Cu, Sb	3
Cu, Ag	4
Cu, Ni	5
Cu, As, Sb	6
Cu, Sb, Ag	7
Cu, Ag, Ni	8
Cu, As, Ag	9
Cu, Sb, Ni	10
Cu, As, Ni	11
Cu, As, Sb, Ag	12
Cu, Sb, Ag, Ni	13
Cu, As, Sb, Ni	14
Cu, As, Ag, Ni	15
Cu, As, Sb, Ag, Ni	16

Table 3: Definition of Copper Groups used in this paper