A lead-isotope database of copper ores from the Southeastern Alps: a tool for the investigation of prehistoric copper metallurgy

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Abstract

The Southeastern Alps were an important source of copper metal in prehistory, at least from the Eneolithic and through the Bronze Age, as documented by the abundant and substantial presence of smelting slags. Evidence of mining activity is scarce, because of limited ad hoc investigation and because of the subsequent systematic erasing by post-Medieval exploitation. Moreover, until recently the profusion of archeometallurgical and archaeological investigations focusing on the prehistoric exploitation of Northern Alpine, Central European, and Balkan ore sources has somehow obscured the early role of the Italian Southern Alps as a major copper producing area. The recent advances in the systematic characterization of the copper ores in the Southeastern Alps (including Alto Adige, Trentino, Veneto, and nearby regions) by lead isotope analysis, supported by mineralogical and geochemical interpretation, offer now the appropriate tools to re-evaluate the extent of prehistoric mining and the local patterns of ore exploitation. The developed database is a powerful tool to identify the metal derived from local production. It is suggested that (1) based on the abundance and chronological distribution of smelting slags evidence, two major periods of mining exploitation took place, the first in the middle of the 3rd millennium BC and the second during the Late Bronze Age; and (2) based on the discrimination of copper sources and the available analyses, most of the metal circulating in Northern Italy and in the greater Po Valley region was actually produced from Southern Alpine ores.

Keywords: copper metallurgy; copper ores; Eastern Alps; Eneolithic; Bronze Age; Lead isotope analysis.

1. Introduction

The Italian Alps do not contain large copper deposits in modern industrial terms; nevertheless, they are host to a large number of relatively small copper and polymetallic copper-bearing deposits of different genetic type and geological age. Most of these copper occurrences would be of limited economic interest in the present global resource market, but many of them have been extensively exploited since prehistory, possibly since Late Neolithic times. As a matter of fact, copper metal objects were circulating in the area well before the Bronze Age, as the archeological evidence clearly shows (e.g. Pedrotti 2002: p. 213, Pearce 2007, Angelini et al. 2013), also through spectacular finds such as the Iceman's copper axe (De Marinis 1992, Fleckinger 2003, Sperl 2005). The latter finding stands as a landmark as it is the only directly datable Eneolithic copper axe in the Southern Alpine region.

The copper deposits which have allegedly been exploited in prehistory are mainly located in the Southeastern Alps (Nimis et al. 2012, Artioli et al. 2013), namely in the Trentino and Alto Adige/South Tyrol regions, which show substantial evidence of Copper Age (Perini 1989, Pedrotti 2002, Pearce 2007, Angelini et al. 2013, Artioli et al. 2015) and Bronze Age (Weisgerber and Goldenberg 2004, Cierny 2008) smelting activities. Despite the intense prehistoric metallurgical activity, surprisingly few systematic investigations were carried out in order to define the geochemical and isotopic character of the Southern Alpine deposit for archaeometric and provenancing purposes. The attention to date has been mostly focused on the more northerly Austrian Alps (e.g. Lutz and Pernicka 2013 and references therein). Significantly, in a recent review of available lead isotope data for the Alpine region (Ling et al. 2014), the authors clearly state: "The Eastern Alps in particular are known for large and rich deposits of copper (also lead and silver). There is a well-documented exploitation throughout the Bronze Age of copper ores, mainly in Tyrol, ... Over 300 lead isotope data that can be used for comparisons have been published for the Alpine copper ores, mainly from Austria (Höppner et al. 2005; Köppel and Schroll 1983; Schroll 1997). Unfortunately, there are no lead isotope data published in numerical format for the mines in Mitterberg, the only published information about these ores is plotted as graphs (for example Pernicka, 2010, Fig. 8, p. 729). There is also archaeometallurgical evidence of ancient copper mines in the Italian Alps, but there are no lead isotope data available for these ores (Weisgerber and Goldenberg 2004)." Actually, several LI data for the Italian Western and Eastern Alps do exist in the literature (Cumming et al. 1987, Artioli et al. 2009, Nimis et al. 2012), nonetheless it is true that no systematic, archaeometry-oriented report of these data has yet been published. Small and

very small copper outcrops were likely ubiquitous in the Alps. Wherever small-scale investigations were undertaken, it was demonstrated that literally hundreds of localities were accessed in the Bronze Age, for many of which Pb isotope data are available (e.g. Valais: Cattin et al. 2011; Austria: Höppner et al. 2005; Köppel and Schroll 1983; Schroll 1997; Eastern Italian Alps: Artioli et al. 2013, Angelini et al. 2015; Western Italian Alps: Artioli et al. 2009). The present review is meant (a) to release in a complete and organic form the lead isotope (hereafter LI) data collected in the last decade within the AAcP project (Alpine Archaeocopper Project: geo.geoscienze.unipd.it/aacp/welcome.html) pertaining to copper deposits in the Southeastern Alps, (b) to define reference isotopic groups for the area, (c) to provide a general geological and geochemical interpretation of the discrimination potential of the LI data for these deposits with respect to other European and Mediterranean copper sources exploited in antiquity, and (d) to show examples of provenancing application to prehistoric copper and bronze objects from Northern Italy. It is believed that these exercises will greatly contribute to clarify the picture of ancient metallurgical exploitation in the Alpine region.

2. The development of the database: ore selection and characterization

Copper-bearing ore samples from the Southeastern Alps were systematically collected during the last decade based on geological, historical, and mining information. The early investigations (Artioli et al. 2008a) focused on the geochemical discrimination of the ore sources, starting with a few samples of the most well-known deposits in the region. The Agordo mining area (Belluno, Veneto) in the heart of the Dolomiti Bellunesi, which used to be one of the fundamental metal producing areas of the Republic of Venice in the XVII and XVIII centuries, was first selected in order to calibrate the sampling and analytical protocols (Artioli et al. 2008b, Artioli et al. 2012). The Agordo area in fact offers excellent occurrences of ores, smelting slags of various ages, and copper metal of unequivocal local origin, besides a wealth of historical information concerning mining sites and metallurgical activities. Agordo hosts one of the most famous Italian mining schools, founded in 1867, and preserves a wealth of information on historical mining and ore processing. The Agordo area was extensively surveyed and sampled in close collaboration with personnel of the mining school, mineral collectors and the local archaeological group (ARCA, Gruppo Archeologico Agordino). A large number of mineralogical, geochemical and isotopic analyses were performed on the samples from the Agordo mines (Valle Imperina, Sasso Negro, Valle del Mis, Passo di Vallés) and compared to the data obtained on local copper smelting slags related to Medieval and pre-industrial extraction activities and on raw copper fragments associated with the slags (Artioli et al. 2008b, Artioli et al. 2010, Giunti 2011, Artioli et al. 2012). This preliminary test investigation allowed us to optimize the ore sampling and separation protocols, and to understand in detail the potentials and limits of the tracing parameters in linking the minerals to the extracted raw copper, through the smelting slags.

The research was then extended to the other known copper districts in the Trentino, Alto Adige/South Tyrol and Veneto regions. Recently, the main copper occurrences in the more easterly Friuli Venezia Giulia (Carnia area) and a few deposits from the more westerly Valcamonica area (Brescia, Lombardia) were also sampled and analysed. The reported data virtually include all occurrences of known copper mineralization in the explored regions. In fact, care was taken in covering all the different genetic typologies of deposits in a given area or mining district, so that the geochemical signature of any presently inaccessible deposit can be predicted from the present data on solid geological grounds. The project now plans to extend the survey further westward to the Central Alps.

Whenever possible each deposit was directly investigated in the field, and representative samples were collected from outcrops or mining dumps. Only in a few cases, when little or no copper ores were still accessible in the field, we had to rely on local private collections or museum specimens from a given locality. In a few cases, core samples from past mining exploration projects were made available.

Fig. 1 shows the location of the sampled mining sites, and Table 1 reports the detailed list of the analysed samples, together with geographical coordinates, locality names, as well as essential geological and mineralogical information.

Fig. 1. Map of the Southeastern Alps with main geological units and location of sampled copper deposits.

3. Lead isotope analyses

The ore samples were mineralogically and petrographically characterized in polished sections by optical microscopy under reflected light and by X-ray powder diffraction (XRPD). Representative portions of the ore containing primary or secondary copper-bearing mineral assemblages (Table 1) were selected and then gently crushed. An adequate amount of mineral grains was then separated by handpicking under a binocular microscope, and their mineralogical composition checked by XRPD. An aliquot of the separates (2–10 mg) was dissolved in aqua regia by high-pressure microwave digestion in sealed PTFE vessels. The dissolved lead was purified using the SrSpecTM resin

(EIChroM Industries; Horwitz et al. 1992), following the same procedure described in Villa (2009). About 100 mL of resin are filled in a 3-mm diameter hand-made PTFE column. The height to width ratio is approximately 4. The sample solution is loaded in 0.5 mL 1M HNO₃, 1.5 mL of which is also used to wash out the matrix metals, while Pb is very strongly retained on the resin. Pb is then eluted with 3 mL 0.01M HNO₃ and is ready for analysis. Lead isotope analyses were performed with a Nu InstrumentsTM Multi-Collector-ICP-MS at the Institut für Geologie, University of Bern (Switzerland). The sample solution was ionized by introducing it into a 9000 K plasma. All elements were ionized simultaneously. Mass fractionation was monitored by adding a small quantity of Tl, which has a known ²⁰³Tl/²⁰⁵Tl ratio, is ionized together with and fractionated by the same mechanism as Pb, and does not interfere with Pb isotope measurements. Calibration was carried out using the NIST SRM 981 international standard. The results are reported in Table 2. Typical in-run relative uncertainties (2 SE of the mean) on ²⁰⁶Pb/²⁰⁴Pb, ²⁰⁷Pb/²⁰⁴Pb, and ²⁰⁸Pb/²⁰⁴Pb isotope ratios were smaller than 0.02 %. The measured isotopic composition for SRM 981 were indistinguishable from the certified value and the recent, more precise literature measurements (Rehkämper and Mezger 2000), so that no adjustment of the measured ratios was necessary. The external reproducibility on the SRM 981 reference material amounted to ± 0.015 % (2 σ), very similar to the individual in-run precision on unknown samples.

4. Results and discussion

The geology and geochemistry of the deposits will not be discussed in detail in this context, since they are of strict geological interest. However, we argue here that the solid geological interpretation of the LI data (Nimis et al 2012) allows a more robust definition of the major isotopic groups in the investigated region, and that such groups provide a very useful basis for discrimination of metals extracted from the Southeastern Alps from those derived from other copper sources in continental Europe and around the Mediterranean region. When ambiguities arise because of overlapping LI fields, then the mineralogical character of the deposit (i.e. chalcopyrite-pyrite vs. tetrahedrite-tennantite vs polymetallic Cu-Zn-Pb sulphides) or the chemistry of the ores themselves (e.g. the contents of different chalcophile elements) can be efficiently used as additional discrimination parameters. An analogous combination of geochemical and isotopic parameters has been used to resolve isotopic ambiguities in specific cases (Baron *et al* 2013; Villa 2016). The fundamental difference with previous approaches, mostly limited to trace element concentrations (see discussion in: Pernicka 2014) and which has in some cases given rise to serious misunderstandings (see

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discussion in: Cattin *et al.* 2015), is the use of common-denominator ratios of geochemically related trace elements, as discussed in more detail by Villa (2016).

Definition of the Southeastern Alpine isotopic groups

The measured LI data are graphically reported in Fig. 2, using the preferred representation based on the ²⁰⁶Pb/²⁰⁴Pb, ²⁰⁷Pb/²⁰⁴Pb, and ²⁰⁸Pb/²⁰⁴Pb ratios. Such representation allows a good visual discrimination of the isotopic fields and at the same time maintains a readily interpretable significance from the geological and geochronological points of view (Nimis 2010; Artioli et al. 2013). The binary diagrams clearly show that the ore deposits from the Southeastern Alps can be described by two major fields.

The first field includes the deposits in the Late Ordovician-Early Silurian crystalline basement located along the Valsugana thrust (Calceranica, Vetriolo, Valle Imperina), which are characterized by ²⁰⁶Pb/²⁰⁴Pb ratios <18.10. These deposits consist of metamorphosed volcanogenic massive sulphide (VMS) deposits sharing similar genesis and features. We therefore define the field as *Valsugana VMS* (marked with red dots in Fig. 2). The copper-bearing mineral assemblage typically includes small amounts of chalcopyrite in a pyrite-dominate mineralization, with accessory sphalerite, galena and arsenopyrite (locally abundant at Calceranica) (Andreatta, 1928; Frizzo 2004). Higher concentrations of chalcopyrite, which were the object of prehistoric mining (Preuschen 1973; Šebesta 1992), are known to have occurred locally at least at Vetriolo. A few other samples from geographically scattered deposits of various origin also fall in this isotopic field: Rifugio Borromeo/Paradiso (Val Martello, Alto Adige), Pian delle Loppe (Val del Mis, Agordo, Veneto), Cinque Valli and Pamera (Valsugana, Trentino); this genetically heterogeneous group includes deposits which were either genetically similar to those of the Valsugana VMS deposits (Rifugio Borromeo; Casari, 1986) or were probably formed at least in part by metamorphic and hydrothermal remobilization of earlier VMS deposits (Nimis et al. 2012).

The other major field, with ²⁰⁶Pb/²⁰⁴Pb ratios >18.26, contains most of the ore deposits in the Southalpine units of the Alto Adige, Trentino and Veneto (AATV) regions, so that we define it *Southalpine AATV* (marked with orange dots in Fig. 2). These deposits are geologically younger and, for the major part, genetically related with post-metamorphic Early Permian and Triassic magmatism. They are mainly formed by polymetallic (Pb, Zn, Cu; with variable, generally minor Ag, Sb, Co, Bi, As) sulphide-rich veins and the mineralization generally consists of chalcopyritesphalerite-galena-pyrite. Other less common mineral assemblages are tetrahedrite ± galena (Pattine– Pian della Stua and Montagiù, Trentino), chalcopyrite-bearing magnetite and pyrrothite (Pamera, Trentino), bornite-chalcopyrite-chalcocite (Sasso Negro and Passo di Vallés, Veneto) and chalcopyrite-pyrite-bismuthinite (Duadello–V. delle Volte, Lombardia). Many of the deposits falling in this LI field, including the most voluminous ones of Val dei Mocheni (Trentino) and Montefondoli/Pfundererberg (Alto Adige), have very similar geochemical characters and mineralogical compositions, so that it is virtually impossible to distinguish the single mines on the basis of isotopic, chemical, and mineralogical parameters. The copper deposits located in the Carnic Alps (Monte Avanza, Casera Pramosio, Comeglians; yellow dots in Fig. 2) also have an isotopic character very similar to that of the Southalpine AATV deposits; however, they have a distinctive tetrahedrite-rich composition, so that a low-Sb content in the metal is prima facie evidence to exclude them as potential sources.

Fig. 2. Plot of LI data for copper ores from the Southeastern Alps. See text for the definition of the isotopic groups.

The deposits which fall well outside the two major ore fields (some of the pink and dark yellow dots in Fig.2) include most of the deposits located in the Austroalpine units of Alto Adige north of the Insubric Line (Stelvio, Oris), and denominated *Austroalpine AA*, or hosted in the Glockner Nappe of the Tauern Window (*Predoi*, Alto Adige). In particular, the latter deposit shows rather low ²⁰⁷Pb/²⁰⁴Pb and ²⁰⁸Pb/²⁰⁴Pb ratios for a given ²⁰⁶Pb/²⁰⁴Pb value, consistent with its very distinct oceanic (ophiolitic) origin. To date none of these copper deposits show evidence of prehistoric exploitation; for this reason and because of their distinct LI signature, they will not be considered for the provenancing application of the database.

It is worth noting that a few samples of secondary copper minerals show LI compositions falling well outside the fields of the primary minerals from the same group of deposits (cf. for instance samples AF-s1 from Montefondoli and TIL_As from Tiles). When this occurs, the non-sulphidic minerals typically show more radiogenic compositions, characterized by higher ²⁰⁷Pb/²⁰⁴Pb, ²⁰⁸Pb/²⁰⁴Pb and/or ²⁰⁶Pb/²⁰⁴Pb ratios, indicating mixing with lead derived from nearby high-(U, Th) rocks. This observation should be cause of caution when provenancing objects on the basis of LI data derived from non-sulphidic ores.

Discrimination of the Southeastern Alpine deposits from other deposits

If we plot the LI data of the Southeastern Alpine deposits together with the available LI data on European and Mediterranean copper ores (such as the OXALID database at the Oxford Isotrace

Laboratory, http://oxalid. arch.ox.ac.uk/; and the vast literature cited in Ling et al. 2014) there is an enormous amount of overlap between the various groups of deposits (see Fig 3 in Artioli et al. 2013). Because of the extent in the data overlap, searching for the provenance of a given object against all available ore data using a quantitative statistical approach can only provide a preliminary discrimination of the possible sources. This approach is applied, for instance, when calculating the Euclidean distances in the 3D space from the measured value to all data in the database (Stos 2009), or when using more sophisticated approaches such as the kernel density estimates (Baxter et al. 1997). Without dwelling in complex statistical treatment, here we limited our analysis to the graphical comparison of the LI data for the Southeastern Alpine region to those published for the copper deposits which are in close geographical proximity and are therefore the most interesting for archaeometric problems related to metal provenance and diffusion. Specifically, for discriminating purposes we here compared the Alpine LI data with those from the Iberian peninsula (including Portugal), Sardinia, Central Europe from France to Slovakia, the Balkans from Romania to Greece, Turkey (including Taurus and Pontic mountains), and the Aegean area (including Cyprus and Crete). For the time being we are not discussing the LI signal of the British Isles and the Levant. It will be shown that this simple approach can often be sufficient for a robust discrimination of the copper sources. All data from the literature were screened and only analyses performed on copper ores were considered. This screening excluded, for instance, many important Pb-Zn(Ag) deposits in which copper is only a minor accessory metal, but included other polymetallic deposits in which Pb, Zn and Cu were all present in significant amounts.

Fig. 3 shows the distribution of LI data available for the whole Eastern Alps (Italy and Austria) and Tuscany. The available data for the Austrian ores (Köppel and Schroll 1983a, Weber 1997, Horner et al. 1997, Höppner et al. 2005) were subdivided in fahlore-based (i.e. containing significant to dominant tetrahedrite–tennantite) and chalcopyrite-based (i.e. with only accessory or no tetrahedrite–tennantite) types, the latter being mostly located in the Stubai–Ötz (Tyrol) and Drau (Carinthia) valleys with a few further examples in Salzburg and Styria. It should be noted that the few available data for the chalcopyrite-rich but fahlore-bearing Mitterberg ores (Köppel and Schroll 1983a,b) indicate high LI ratios, mostly lying well outside the shown limits of the employed diagrams (²⁰⁶Pb/²⁰⁴Pb = 19.14–20.07, ²⁰⁷Pb/²⁰⁴Pb = 15.70–15.79 and ²⁰⁸Pb/²⁰⁴Pb = 39.30–42.08). The Austrian ores show some overlap with the Southeastern Alpine ores in the ²⁰⁶Pb/²⁰⁴Pb vs ²⁰⁷Pb/²⁰⁴Pb plot, but they can be readily discriminated in the ²⁰⁶Pb/²⁰⁴Pb vs ²⁰⁸Pb/²⁰⁴Pb plot, as they show systematically lower ²⁰⁸Pb/²⁰⁴Pb values for a given ²⁰⁶Pb/²⁰⁴Pb ratio. The only serious overlap resides in the lower left part of both diagrams, where the Calceranica and Vetriolo deposits of the Valsugana VMS group substantially overlap with a few of the chalcopyrite-based mines from

Carinthia (Kaser Wieserl, Knappenstube, Plaiken), Salzburg (Rettenbach), and Styria (Walchen). To date there is no indication that any of these Carithian mines were active in prehistoric times. However, their discrimination from some of the Southeastern Alpine deposits remains challenging and should be pursued further, possibly on the basis of chemical tracers.

The main body of the Southern Tuscan ores (pale grey diamonds in Fig. 3) is well discriminated from the Southeastern Alpine ores, whereas virtually all data for Northern Tuscany (Alpi Apuane) overlap with the field of the Southalpine AATV and Carnia ores. Again, there is yet no archaeological indication of prehistoric mining or ancient copper metallurgy in the Alpi Apuane area, but the existence of such deposits should be taken into account in future work.

Fig. 3. Comparison of the LI data of the copper ores from the Southeastern Alps, Tuscany, and Austria. Austrian deposits are distinguished into fahlore-based (Fhl) and chalcopyrite-based (Cpy) types, the latter being mostly located in the Stubai–Ötz (Tyrol) and Drau (Carinthia) valleys. Source of literature data: Tuscany – Lattanzi et al. (1992), Stos-Gale et al. (1995); Austria – Köppel and Schroll (1983a), Weber (1997), Horner et al. (1997), Höppner et al. (2005).

Expanding the comparison to geographically farther deposits, perusal of the database shows that most of the available data for copper ores in other European regions, including the French Massif Central (Cevennes, Mont Lozere, etc.) and Languedoc (Cabrières), Central Europe (Erzgebirge, Harz Mountains, Bohemian Massif), the Balkans (Serbia, Romania, Bulgaria), and Switzerland (Valais), are well distinct from the Southeastern Alpine data. All available data for the Eastern Mediterranean ores (Greece, the Aegean, Turkey, Cyprus) are also well distinct. The only regions that have some degree of overlap with the Southeastern Alpine ores are: the central and South-western Sardinia (Fig. 4) and part of Iberia, including mines located in Andalusia, Alcudia Valley, and the Iberian Pyrite Belt (IPB) (Fig 5).

Fig. 4. Comparison of LI data for copper ores from the Southeastern Alps and Sardinia. Source of data for Sardinia after Swainbank et al. (1982), Boni and Koeppel (1985), Ludwig et al. (1989), Stos-Gale et al. (1995), Begemann et al. (2001).

Fig. 5. Comparison of LI data for copper ores from the Southeastern Alps and Iberia. Source of data for Iberia after Arribas and Tosdal (1994), Stos-Gale et al. (1995), Marcoux (1998), Santos Zalduegui et al. (2004), Klein et al. (2009).

Keeping in mind that Sardinia is mainly host to lead-zinc mineralization, the copper ores of Sardinia can be rationalized into four major isotopic groups (Artioli et al. 2013). Only two of these groups show a substantial overlap in the ²⁰⁷Pb/²⁰⁴Pb vs ²⁰⁶Pb/²⁰⁴Pb plot with the Southeastern Alpine deposits: in particular, the deposits located in central Sardinia (Barbagia, Sulcis, Ogliastra; among

which the most important deposit is the famous one of Funtana Raminosa, whose name significantly means "cupriferous water source") partly overlap with the Southalpine AATV field, and the ores of South-west Sardinia (Iglesiente) overlap substantially with the Valsugana VMS field (Fig. 4a). Fortunately, because of their very distinct geological origin, the great majority of Sardinian ores has lower ²⁰⁸Pb/²⁰⁴Pb ratios, so that in general they can be well discriminated from the Alpine ones (Fig 4b).

As for Iberia, the wealth of data that recently became available in the literature thanks to the effort of Spanish researchers has remarkably increased the database for the Iberian mines, although further work is needed to interpret them on a geologically and geochemically robust basis. Taken at face values, some of the Iberian LI fields do show some overlap with the Southeastern Alpine field. In this respect, the most important Iberian mines that should be taken into account when interpreting the provenance of metals of potential Alpine derivation are (Fig 5a) a few of the mines in Andalusia (Sierra de Gador) and some of the mines in the Alcudia Valley (Virgen del Socorro, Navalahiguera, Santa Rita, Las Minillas, Los Diegos, Santa Isabel, San Justo). These deposits cannot be distinguished from the Southeastern Alpine deposits from their LI signal alone. More work on the chemical tracers is needed for adequate discrimination of these ores. Note that a few of the mines forming the high ²⁰⁷Pb/²⁰⁴Pb tail of the Iberian Pyrite Belt field (IPB: Los Molares, Minas de Cala) overlap the Southeastern Alpine deposits only in the ²⁰⁷Pb/²⁰⁴Pb vs ²⁰⁶Pb/²⁰⁴Pb plot, but they systematically have lower ²⁰⁸Pb/²⁰⁴Pb ratios and can thus be safely distinguished from the Alpine ones.

5. Applications

The following examples will show the potential of the LI database for the Southeastern Alps for the interpretation of the ancient copper metallurgy in the region.

Eneolithic copper metallurgy in the Southeastern Alps

Table 3 reports all available isotopic data for Eneolithic objects from northeastern Italy. Some of them were taken from the literature, whereas others are presented here for the first time. The data are graphically reported in Fig. 6, together with available data for Alpine Eneolithic copper smelting slags (Artioli et al. 2015).

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	Sample label	locality	Type of object	²⁰⁶ Pb/ ²⁰⁴ Pb	²⁰⁷ Pb/ ²⁰⁴ Pb	²⁰⁸ Pb/ ²⁰⁴ Pb	reference				
1	CBU_AX MR	Col del Buson	flanged-axe	17.934±2	15.653±3	38.168±6	Angelini et al. 201				
2	CBU_AXO	Col del Buson	shaft hole-axe	17.937±2	15.656±2	38.174±7	Angelini et al. 201				
3	CBU-AXFR	Col del Buson	axe fragment	18.457±3	15.674±2	38.661±7	Angelini et al. 201				
4	CBU-Per	Col del Buson	copper ring	18.851±2	15.681±2	38.850±6	Angelini et al. 201				
5	CBu-L	Col del Buson	copper awl	18.209±3	15.681±3	38.471±7	Angelini et al. 201				
6	CAM-AxO	Montebelluna	Cu axe (As-Ag)	18.524±2	15.673±2	38.700±5					
7	Peri	Peri	Cu wire (Cu)	18.097±2	15.658±2	38.342±5					
8	Sp-Al	Spessa	halberd (Cu-As)	18.50±3	15.70±2	38.45±5					
9	Gamb	Gambarella	halberd (Cu-As)	18.65±1	15.70±1	38.92±3					
10	Sp-Pg	Spessa	dagger	18.097±7	15.647±5	38.25±1					
11	BFO60-15	Millan	raw Cu fragment	18.265±1	15.690±1	38.540±4	Artioli et al. 2015				
12	Mil1	Millan	raw Cu fragment	18.279±3	15.693±3	38.545±7	Artioli et al. 2015				
13	bg29n	Bongiovanna	rolled Cu sheet	18.243±2	15.674±2	38.498±6					
	-	_	(interpreted as an awl)								
14	MA-073090	Cellore, loc Arano	Cu awl	<mark>17.920</mark>	<mark>15.655</mark>	<mark>38.159</mark>	Pernicka and				
							Salzani 2011				
15	MA-073084	Cisano	Cu ingot	17.909	<mark>15.631</mark>	38.092	Pernicka and				
							Salzani 2011				

Table 3. LI data for Eneolithic objects from Northern Italy. Uncertainties (2σ) refer to the last digits. The sample number in the first column corresponds to the plotted points in the diagrams of Fig. 6.

Fig 6. LI data for Eneolithic objects from Northern Italy (Table 3), and coeval copper smelting slags from Trentino and Alto Adige (Artioli et al. 2015), compared with data for the Southeastern Alpine copper deposits.

The LI data plots (Fig. 6) show that most objects are perfectly compatible with a provenance from Southeastern Alpine ores. Only two objects remain of dubious interpretation: the copper ring fragment (CBU-Per) from Col del Buson (Belluno), which shows a much higher ²⁰⁶Pb/²⁰⁴Pb ratio than any known Southern Alpine deposit; and the halberd from Spessa (Verona), made of arsenical copper, whose ²⁰⁸Pb/²⁰⁴Pb ratio is too low for a Southeastern Alpine derivation. Some deposits from nearer Carnia are isotopically close (Fig. 6), but incompatible with the arsenical copper composition of the object, given the high antimony contents in the ores, which are invariably rich in tetrahedrite. Clearly for this object the isotopic data must be confronted with additional chemical and archaeological data to reach a satisfactory interpretation.

The available LI data for Eneolithic copper smelting slags from Trentino and Alto Adige (Artioli et al. 2015) are also plotted in Fig. 6. The excellent match of the LI signals clearly indicates a relationship between the objects and the local copper ores. A detailed analysis of the LI data allowed to decode the local patterns of exploitation of the deposits, by evidencing close links between the individual smelting sites and the original mines (Artioli et al. 2015). Here, it is sufficient to remark the capability of LI data to delineate a very dynamic picture of copper mining and ore processing in the Southeastern Alps during the Copper Age. Given the large amount of

slags found at several sites (up to several hundred kilograms at Millan, Alto Adige, and tens of kilograms at La Vela Valbusa, Trentino) it is not unreasonable to talk about a substantial copper production, at least during the second half of the 3rd millenium BC.

Middle Bronze Age swords

As another example, we tested available LI data for a few Middle Bronze Age (MBA) swords. The data are reported in Table 4. Two of the data (the sword and rivet from Olmo di Nogara) are presented here for the first time. The other data are from Pernicka and Salzani (2011).

Table 4. LI data for MBA swords from Northern Italy. Uncertainties (2σ) refer to the last digits.

Sample label	locality	Type of object	²⁰⁶ Pb/ ²⁰⁴ Pb	²⁰⁷ Pb/ ²⁰⁴ Pb	²⁰⁸ Pb/ ²⁰⁴ Pb	reference
ont95p	Olmo di Nogara	rivet	18.352±4	15.683±4	38.565±7	
ont95s	Olmo di Nogara	sword (tomb 95) BMII	18.503±4	15.692±4	38.707±7	
MA-073064	Bor di Pacengo	sword	<mark>18.777</mark>	<mark>15.692</mark>	<mark>39.037</mark>	Pernicka and Salzani 2011
MA-073047	Bacino Marina	Sword (Rixheim)	18.092	<mark>15.644</mark>	<mark>38.290</mark>	Pernicka and Salzani 2011
MA-073048	Imboccatura Mincio	spada (Pepinville)	18.095	<mark>15.649</mark>	<mark>38.302</mark>	Pernicka and Salzani 2011

Fig 7. LI data for for MBa swords from Northern Italy (Table 4) compared with data for the Southeastern Alps and Southern Tuscany.

The LI diagrams (Fig. 7) clearly indicate that three of the swords and the rivet are made with Southern Alpine copper. The sword from Bor di Pacengo is instead clearly from Southern Tuscany. All the Sardinian ores can be ruled out as candidate sources on the account of the ²⁰⁸Pb/²⁰⁴Pb values. It is unfortunate that Jung et al. (2011) did not publish the LI data for their analysed objects from Northern Italy, which included several swords. The discussion in their paper mostly relies on the conventional ²⁰⁷Pb/²⁰⁶Pb vs ²⁰⁸Pb/²⁰⁶Pb diagrams, which are much less discriminating that the conventional geological plots used here. As a consequence, the interpretation of the data in the paper is rather inconclusive. In the same paper, however, the authors report a very small-size plot of ²⁰⁶Pb/²⁰⁴Pb vs ²⁰⁷Pb/²⁰⁴Pb (Jung et al. 2011, Fig. 23.8), which shows that several of the objects are clustered around ²⁰⁶Pb/²⁰⁴Pb=18.1-18.3 and ²⁰⁷Pb/²⁰⁴Pb=15.63-15.67. Since, as shown above, ores from Sardinia, Southeastern Alps, and Tuscany can be discriminated by LI data with sufficient confidence, the data indicate that most of these objects are made out of Southern Alpine ores.

Again, the above example shows that in the MBA and presumably through the Recent Bronze Age the copper sources in the Southern Alps were still very active and supplied most of the copper circulating in Northern Italy, despite the scarce archaeological evidence in terms of mining and metallurgical sites. We know, however, that during the Late Bronze Age (LBA) there was a resurrection of copper exploitation in the area linked to the Luco/Laugen culture, which is testified by the huge amount of LBA copper smelting slags found in archaeological sites (Metten 2003, Weisgerber and Goldenberg 2004, Cierny 2008, Addis et al. 2014). Based on the widespread occurrence of the slags in the territory and their substantial volume, it can be argued that during the LBA the Southern Alpine copper production reached a peak. The subsequent major wave of mining activity occurred in the Middle Age, when large groups of German miners moved to several of the Alpine valleys to organize and carry out the mining operations (Šebesta, 2000; Zammatteo, 2009).

6. Conclusions

The reference LI database for the Southeastern Alpine copper deposits (AAcP database) is presented, together with the essential geological and geochemical interpretation of the data. The reported data fill a long-noted gap in the characterization of Alpine copper ore resources. Groupings are proposed that encompass genetically related copper ore deposits, which are characterized by homogeneous LI signals and, at least in some cases, restricted geographic distribution. Such groupings may serve as references for a robust discrimination of potential metal sources. The elevated discriminating power of the database is demonstrated by applications to Eneolithic objects and MBA swords for Northern Italy. It is believed that the database will be a very useful tool for future provenancing of metals and related materials (slags), which will no doubt contribute to clarify the emerging picture of ancient copper metallurgy in the Alpine and Central Mediterranean area (Dolfini 2013, Angelini et al. 2013, Perucchetti et al. 2015).

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