Eneolithic copper smelting slags in the Eastern Alps: Local patterns of metallurgical exploitation in the Copper Age

G. Artioli a,*, I. Angelini a, U. Tecchiati b, A. Pedrotti c

a Dipartimento di Geoscienze, Università degli Studi di Padova, Padova, Italy
b Ufficio Beni Archeologici, Provincia Autonoma di Bolzano, Bolzano/Bozen, Italy
c Dipartimento di Filosofia, Storia e Beni Culturali, Università degli Studi di Trento, Trento, Italy

ABSTRACT

A number of slags of all known sites in the Italian Eastern Alps showing occurrences of copper smelting activities in the Copper Age have been characterized by lead isotope analysis. All the investigated smelting slags from Trentino (Romagnano Loc, La Vela, Gaban, Acquaviva di Besenello, Montesi di Serso) and Alto Adige/Sud Tyrol (Millan, Gudon, Bressanone Circonvallazione Ovest) have been recently characterized by thorough mineralogical, petrographical and chemical analysis and demonstrated to be the product of copper smelting activities of chalcopyrite-based mineral charges, with an immature technological extraction process referred as the “Chalcolithic” smelting process. Revision of the available radiocarbon dates show that the metallurgical activities pertaining to the analysed slags can be attributed to the third millennium BC. The lead isotope analysis indicates clearly that the mineral charge used for the smelting process was extracted from nearby mineral deposits. The detailed analysis of the spatial distribution of ores and slags allows for the first time to define the local organization of the metallurgical operations.

© 2015 Elsevier Ltd. All rights reserved.

1. Introduction

The Italian Eastern Alps are a well-known source of copper metal that was exploited since prehistory, possibly since Late Neolithic times. Due to the large amount of archeological evidence, especially the widespread and abundant occurrence of copper smelting slags (e.g. Cierny et al., 2004; Cierny, 2008), the climax of the mining activities and copper production is currently attributed to the Recent and Final Bronze Age (Marzatico, 1997; Weisgerber and Goldenberg, 2004), and subsequently to Roman and Middle Age times, when large groups of German miners moved to some of the Alpine valleys to organize and carry out the mining operations (Sebesta, 2000; Zammateo, 2009). However the copper metal was circulating well before the Bronze Age, as the archeological evidence clearly shows (Pedrotti, 2002: p. 213): metal objects were circulating at least from the late neolithic (Angelini et al., 2013) and a number of Copper Age sites in the Trentino and Alto Adige areas yield evidence of smelting activities in the form of metallurgical slags, tuyeres, a multitude of copper objects including the Iceman’s axe, and a few occurrences of pyrotechnological installations (Perini, 1989; Pedrotti, 2002; Pearce, 2007).

The focus of the present investigation is to characterize the isotopic signal of the known Eneolithic smelting slags and to compare the measured lead isotope ratios with the signal of the copper deposits in the Eastern Alps (Nimis et al., 2012; Artioli et al., 2013), in order to pinpoint which deposits were actually exploited in the Copper Age, and possibly outline the local organization of the metallurgical activities.

2. Slag samples: selection and characterization

The slag samples to be investigated were selected based on (1) their secure occurrence in archaeological sites dated to the 3rd millennium BC, and (2) previous results of mineralogical, petrographic, and chemical studies on the slags confirming that they are indeed the product of copper smelting activities.

Table 1 lists the sites where the investigated copper smelting slags were located together with the related archeological literature. Fig. 1 shows the geographical distribution of the sites, all located in the Trentino and Alto-Adige areas.

The sites are clustered in three main areas:

* Corresponding author.
E-mail address: gilberto.artioli@unipd.it (G. Artioli).

http://dx.doi.org/10.1016/j.jas.2015.08.013
0305-4403/© 2015 Elsevier Ltd. All rights reserved.
a) Millan, Gudon, and the site of Circonvallazione Ovest are all located in the Isarco river valley near the city of Bressanone/Brixen

b) Romagnano Loc and La Vela are located in proximity of the Western bank of the Adige River, whereas Gaban, and Acquaviva di Besenello are located in proximity of the Eastern banks of the river in the outskirts of the city of Trento.

c) Montesei di Serso is located in the upper Valsugana Valley near the city of Pergine, again located in the Eastern area the Adige River.

The common feature of all these sites is the location at low altitude, near the bottom of the valley, in close proximity to the river and, presumably, to the coeval settlements. The archaeological occurrences of the Trentino slags and their dates have been extensively discussed by Pearce (2007): the critical revision of the available dates indicate that the start of the metallurgical activities at Gaban and Acquaviva di Besenello can be attributed to the early 3rd millennium BC, whereas the analysed slags from the other sites cluster around the second half of the 3rd millennium BC. The recent dates obtained on the Alto Adige sites (Millan, Gudon) confirm this chronology (Angelini et al., 2013).

All samples were previously thoroughly characterized by minero-petrographical and chemical analysis by X-ray powder diffraction, optical microscopy, and electron microscopy with energy dispersive spectroscopy (Artioli et al., 2009; Colpani et al., 2009). The common features of all slags are here summarized:

- Very heterogeneous and coarse texture (Fig. 2)
- Presence of primary sulphide relics (chalcopyrite) with only incipient reactions (Fig. 3)
- Abundant unreacted quartz
- Presence of typical slag minerals formed during smelting, especially fayalitic olivine, but also pyroxenes (see Colpani et al., 2009)
- Presence of abundant wuestite, frequently dendritic (Fig. 4) or agglomerated
- Presence of copper or matte droplets, frequently intermixed with magnetite

The overall features, such as the coarse texture, the presence of wuestite and magnetite even in the same slag and the occurrence of poorly reacted sulphides indicate an incomplete process of copper extraction and poorly controlled temperature and oxygen fugacity conditions. The slags never underwent a complete melting stage and the process of copper extraction was rather inefficient. These features altogether have been taken as evidence of a technologically non-standardized process of copper extraction, referred to as the “Chalcolithic” process (Pearce, 2007; Bourgarit, 2007). The observed mineralogical ad textural features are compatible with

Table 1
List of sites yielding the investigated copper smelting slags, with the related references of previous archaeological and archaeometric work.

<table>
<thead>
<tr>
<th>Locality</th>
<th>Area</th>
<th>References</th>
</tr>
</thead>
<tbody>
<tr>
<td>Millan</td>
<td>Isarco Valley</td>
<td>Tecchiati 2009, Colpani et al., 2009, Angelini et al., 2013</td>
</tr>
<tr>
<td>Gudon</td>
<td>Isarco Valley</td>
<td>Colpani et al., 2009, Angelini et al., 2013</td>
</tr>
<tr>
<td>Bressanone Circonvallazione Ovest</td>
<td>Isarco Valley</td>
<td>Angelini et al., 2013</td>
</tr>
<tr>
<td>Gaban</td>
<td>Adige Valley</td>
<td>Cattoi et al., 1995, Cattoi et al. 1997, D'Amico et al., 1998, Artioli et al., 2009</td>
</tr>
<tr>
<td>La Vela</td>
<td>Adige Valley</td>
<td>Fasani 1983, Perini 1989, Metten 2003, Artioli et al., 2009</td>
</tr>
<tr>
<td>Montesei di Serso</td>
<td>Valsugana, Trentino</td>
<td>Perini 1989, Metten 2003, Artioli et al., 2009</td>
</tr>
</tbody>
</table>
the age attributed to the sites, and are totally different from those observed in the Late Bronze Age slags found in the same area (Cierny, 2008; Anguilano et al., 2002; Addis et al., 2015).

One copper fragment was also recovered in the Millan site amidst the large amount of smelting slags (Fig. 5). Since it represents a very rare occurrence, and further evidence of the metallurgical activities, it was also analysed and compared with the slag data.

3. Lead isotope measurements

The slag samples were characterized in thin section by optical microscopy under transmitted and reflected light and by X-ray powder diffraction. A portion of each sample rich in fayalite-magnetite-sulphides was gently crushed and adequate amount of fragments was separated by handpicking under a binocular microscope. For the most part the selected fragments consist of fayalite-magnetite residues with microscopic inclusions of copper and sulphidic matte. In some cases small inclusions of partially reacted sulphide ores are present.

The separates (10–100 mg) were dissolved in aqua regia by high-pressure microwave digestion in sealed PTFE vessels. The dissolved lead was purified using the Sr_SpecTM resin (EICrhoM Industries; Horwitz et al., 1992), following the same procedure described in Villa (2009). About 100 mL of Sr_SpecTM 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 1 M 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
results are reported with Pb isotope measurements. Calibration was carried out using fractionated by the same mechanism as Pb, and does not interfere.

The external reproducibility on the NIST SRM 981 reference material amounted to

Table 2 were calculated by normal error propagation taking into account both in-run uncertainties and dispersion of repeated measurements on NIST SRM 981 during the same analytical session.

A small fragment of the copper fragment found at Millan (sample BFO60-15, Fig. 5) was also analysed in Bern using the same protocols. An earlier measurement performed at the Royal Holloway, University of London by Dr. Wolfgang Müller on the same object is also reported for comparison purposes (Table 2).

4. Results and discussion

The Pb isotope ratios measured on the Eneolithic copper smelting slags from Trentino and Alto Adige (Table 2) can be directly compared to the available data on copper ores from the Southern Eastern Alps (Nimis et al., 2012). The data are shown in Fig. 6a and b.

Apart from one slag sample from Gaban, which is located midway between the two major field of Southern Alpine copper deposits, as a first approximation all other slag data cluster in close proximity of the reported LI ore data. This is no surprise, since it is of course expected that the sulphidic ores used for metal extraction must derive from nearby sources. However, the availability of geologically and geographically well-resolved data for the ores permits a very detailed analysis of the pattern of mine exploitation in the areas around the smelting sites.

It can be clearly observed that the slags from Romagnano, La Vela, Gaban and Acquaviva sites, all located in the Adige Valley, show a close affinity to the ores of the Pre-Variscan massive deposits related to the Hercinian basement. These are mostly located in proximity of the Valsugana fault, and the major mine is that of Calceranica (Fig. 7). On the other hand the slags of Montesei di Serso and all the Alto Adige sites (Bressanone, Gaban, Millan) show a clear relationship to the Post-Variscan sulphide ores related to Permian and Triassic volcanics. Specifically, the Montesei slags show close affinity to the polysulphidic ores present in the Val dei Mocheni (Fig. 7), whereas the Alto Adige slags are evidently related to the ores of the Monte Fondoli area, near Chiusa (Fig. 8). The data obtained on the copper fragment associated to the Millan slags are also internally consistent and fit perfectly with the slags data and the Monte Fondoli ores.

The data indicate that in the Alto Adige area, where only one major deposit occurs (Monte Fondoli), these ores were supplying the chalcopirite charge for all the metallurgical activities along the Isarco Valley. Again it should be noted that all slag-producing sites are located not far from the river, at low altitude.

In the Trentino area, where many ore sources are available, a pattern of exploitation seem to appear: the metallurgical smelting sites located along the Adige River Valley obtained the chalcopirite charge essentially from the mine of Calceranica, easily reachable through at least three easy routes, the road though Folgaria, the Valsorda road, and the main entrance to the Valsugana Valley, just west of Trento. Interestingly, the slag sites are located almost
exactly at the outlet of these three roads into the Adige valley. Conversely the Montesei site, located near Pergine at the bottom of the Valsugana Valley, obtained the sulphidic ore exclusively from the Valle dei Mocheni, despite being conveniently located on the opposite side of the Valley with respect to the Calceranica mine. It looks that the Valsugana Valley acted as the boundary for the two independent metallurgical districts. This implies local control of the territory and of the ore resources.

It also proves interesting to compare the slag Pb isotope data with the available data on coeval objects (Fig. 9). The plots show clearly that the local ores linked to the smelting slags that were exploited for the copper production in the Southern Alps in the 3rd millennium BC show a consistent pattern relating the sulphidic ore sources and the smelting locations. The Monte Fondoli deposit is the only source supplying the smelting sites in Alto Adige, all located along the bottom of the Isarco River Valley. In Trentino the major copper ore deposits are located along or nearby the Valsugana Valley, and the valley itself seems to represent a major geographical boundary between independently managed mining districts.

Although copper-based objects were circulating in the area well before the mid-Eneolithic, as testified by several metal finds (Pedrotti, 2002; Pearce, 2007), the substantial amount of slags produced during the 3rd millennium (i.e. several hundred kilograms at Milland and La Vela) indicate the start of the massive exploitation of ores in the Southern Alps and the systematic and well organized production of copper metal. Correspondingly, many of the objects circulating in the region result to be produced from Southern Alpine copper.

5. Conclusions

The lead isotope analysis of the copper smelting slags from all known metallurgical sites in Trentino and Alto Adige during the 3rd millennium BC show a consistent pattern relating the sulphidic ore sources and the smelting locations. The Monte Fondoli deposit is the only source supplying the smelting sites in Alto Adige, all located along the bottom of the Isarco River Valley. In Trentino the major copper ore deposits are located along or nearby the Valsugana Valley, and the valley itself seems to represent a major geographical boundary between independently managed mining districts.
Acknowledgements

Ilaria Giunti, Anna Addis, and Caterina Canovaro helped at different stages in the sample preparation and MS measurements at the Institut für Geologie, Bern. The Museo di Storia Naturale, Trento kindly made the samples from Trentino available for the study. Dr. Wolfgang Müller allowed reporting of his measurement on sample BF060-15.

References


