Journal of Archaeological Science 63 (2015) 78–83

Contents lists available at ScienceDirect

Journal of Archaeological Science

journal homepage: http://www.elsevier.com/locate/jas

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

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ARTICLE INFO

Article history:
Received 22 February 2015
Received in revised form
4 August 2015
Accepted 16 August 2015
Available online 19 August 2015

Keywords:
Copper metallurgy
Eastern Alps
Smelting slags
Eneolithic
Lead isotope analysis

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, Montesei 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 use 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.

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 (Šebesta, 2000; Zammatteo, 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:

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http://dx.doi.org/10.1016/j.jas.2015.08.013
0305-4403/

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

Locality	Area	References
Millan	Isarco Valley, Alto Adige	Tecchiati 2009, Colpani et al., 2009, Angelini et al., 2013
Gudon	Isarco Valley, Alto Adige	Colpani et al., 2009, Angelini et al., 2013
Bressanone Circonvallazione Ovest	Isarco Valley, Alto Adige	Angelini et al., 2013
Gaban	Adige Valley, Trentino	Cattoi et al., 1995, Cattoi et al. 1997, D'Amico et al., 1998, Artioli et al., 2009
La Vela	Adige Valley, Trentino	Fasani 1988, Perini 1989, Metten 2003, Artioli et al., 2009
Acquaviva di Besenello	Adige Valley, Trentino	Cattoi et al., 1995, Cattoi et al. 1997, D'Amico et al., 1998, Pedrotti 2002, Metten 2003, Artioli et al., 2009
Romagnano	Adige Valley, Trentino	Perini 1971, Perini 1989, Cattoi et al., 1995, Cattoi et al. 1997, Metten 2003, Artioli et al., 2009
Montesei di Serso	Valsugana, Trentino	Perini 1989, Metten 2003, Artioli et al., 2009

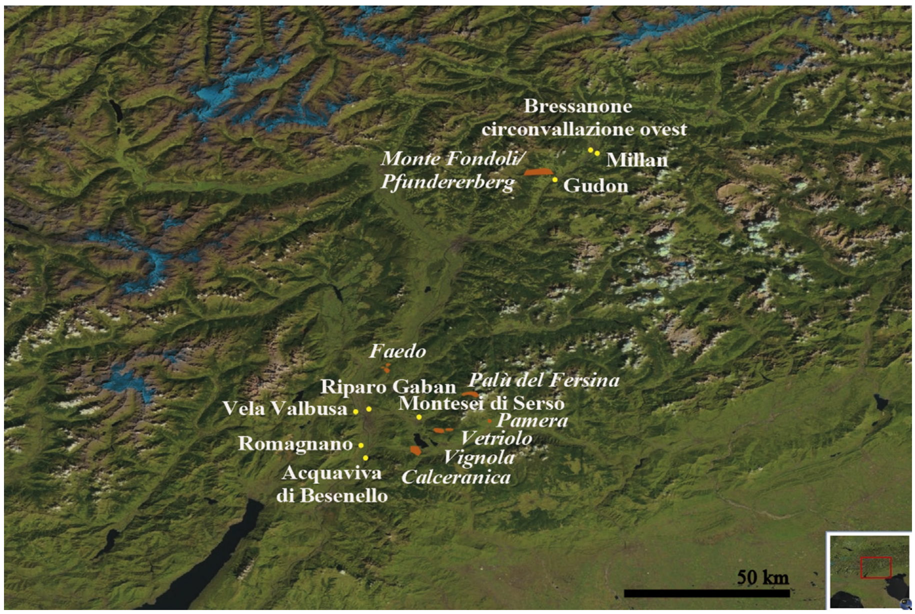

Fig. 1. Map of the location of the Eneolithic sites in the Eastern Alps showing the investigated copper smelting slags (yellow dots). Some of the nearby copper mining areas are also marked in the map (red fields, names in italics). Image courtesy of laboratoriobagolini.it/ais/. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

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 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

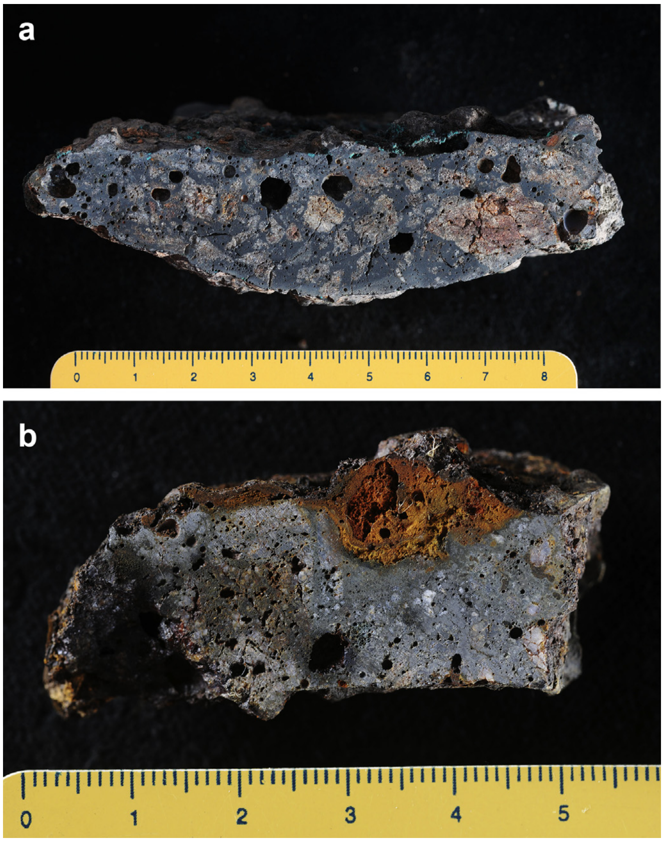

Fig. 2. Macroscopic images showing the coarse texture common of all the investigated Eneolithic slags: (a) slag sample from Riparo Gaban, Trentino (sett. IV C6); (b) slag sample from Millan, Alto Adige.

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 represent a very rare occurrence, and further evidence of the metallurgical activities, it was also analysed and compared with the slag data.

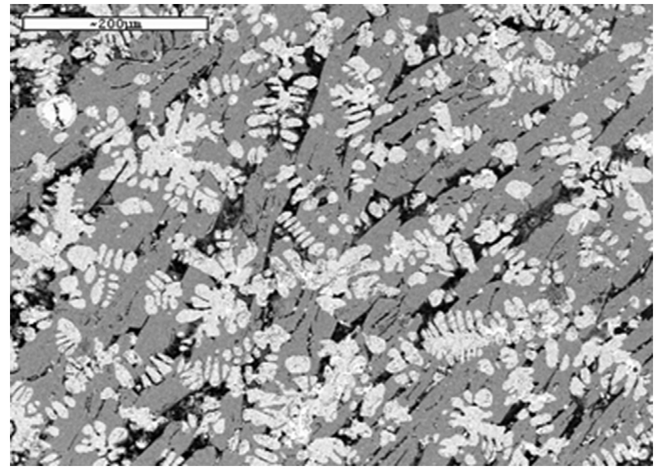

Fig. 4. Wuestite dendrites in the fayalitic matrix. Slag sample from Montesei di Serso, Trentino. Scanning electron microscopy, backscattered electron image, reference bar is 200 μm.

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 (EIChroM 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 HNO_3, 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

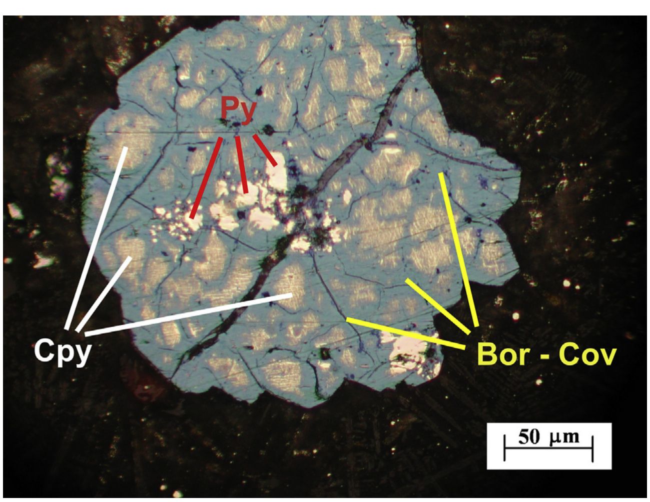

Fig. 3. Partially reacted primary sulphide in a slag sample from Riparo Gaban, Trentino (sett. IV C6). Optical microscopy, reflected light, the diameter of the sulphide grain is approximately 350 μm. Py = pyrite, Cpy = chalcopyrite, Bor-Cov = secondary sulphide of bornite-covelline composition.

Fig. 5. The copper fragment found in association with the copper smelting slags at the Millan site, Alto Adige (US-15). It is one of the rare pieces of metals ever found together with the copper smelting residues.

Table 2
Pb isotopic ratios measured on the Eneolithic copper smelting slags from Trentino and Alto Adige (Southern Italian Alps). The measurements performed on a copper fragment found in association with the Millan slags are also reported (sample BFO60-15).

Locality	Sample	$^{206}Pb/^{204}Pb$	2σ	$^{207}Pb/^{204}Pb$	2σ	$^{208}Pb/^{204}Pb$	2σ
Bressanone	BX-A4	18,298	0.001	15,682	0.002	38,539	0.006
Bressanone	BX-P1	18,271	0.002	15,682	0.002	38,516	0.007
Millan	BFO104-18	18,270	0.002	15,688	0.003	38,531	0.008
Millan	BFO60-15[a]	18,265	0.001	15,690	0.002	38,540	0.007
Millan	BFO60-15[b]	18,279	0.003	15,693	0.002	38,545	0.005
Gudon	US12A	18,265	0.003	15,685	0.001	38,525	0.006
Gudon	US14A	18,281	0.002	15,686	0.002	38,532	0.005
Gudon	US14C −2	18,287	0.002	15,699	0.002	38,573	0.006
Gudon	US14C −1	18,276	0.003	15,687	0.003	38,533	0.004
Gudon	US14D	18,276	0.003	15,690	0.002	38,543	0.006
Gudon	US15	18,265	0.002	15,684	0.002	38,517	0.007
La Vela	LV15	18,298	0.004	15,682	0.003	38,539	0.006
La Vela	LV 18	17,936	0.002	15,642	0.002	38,141	0.006
La Vela	LV 9	17,919	0.002	15,642	0.002	38,135	0.007
La Vela	LV 5	17,909	0.003	15,643	0.003	38,130	0.008
Montesei	MS 1	18,226	0.005	15,667	0.005	38,461	0.015
Montesei	MS 3	18,262	0.007	15,686	0.006	38,523	0.014
Montesei	MS 11	18,229	0.007	15,654	0.008	38,446	0.027
Montesei	MS 11 replica	18,238	0.001	15,664	0.002	38,473	0.005
Gaban	GAB 1 (C6)	18,110	0.001	15,658	0.001	38,344	0.004
Gaban	GAB 2	17,903	0.002	15,643	0.002	38,120	0.006
Gaban	GAB 3	17,896	0.002	15,643	0.002	38,114	0.007
Romagnano	ROM 1	17,988	0.003	15,647	0.002	38,191	0.006
Romagnano	ROM 13	17,921	0.002	15,641	0.002	38,131	0.007
Acquaviva	ACQ 1	17,911	0.002	15,640	0.002	38,119	0.007
Acquaviva	ACQ 2	18,195	0.002	15,661	0.002	38,431	0.007
Acquaviva	ACQ 3	17,940	0.001	15,650	0.002	38,171	0.003

[a] Measurement performed in Bern.
[b] Measurement performed in London (courtesy of W. Müller).

then eluted with 3 mL 0.01 M HNO_3 and is ready for analysis. Lead isotope analyses were performed with a Multi-Collector-ICP-MS (Nu Plasma II) 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 $^{203}Tl/^{205}Tl$ 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 (2s) on $^{206}Pb/^{204}Pb$, $^{207}Pb/^{204}Pb$, and $^{208}Pb/^{204}Pb$ isotope ratios were smaller than 0.2‰. The external reproducibility on the NIST SRM 981 reference material amounted to ±0.15‰ (2s), very similar to the individual in-run precision on unknown samples. Total errors reported in 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, Gudon, 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 chalcopyritic 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 chalcopyritic 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

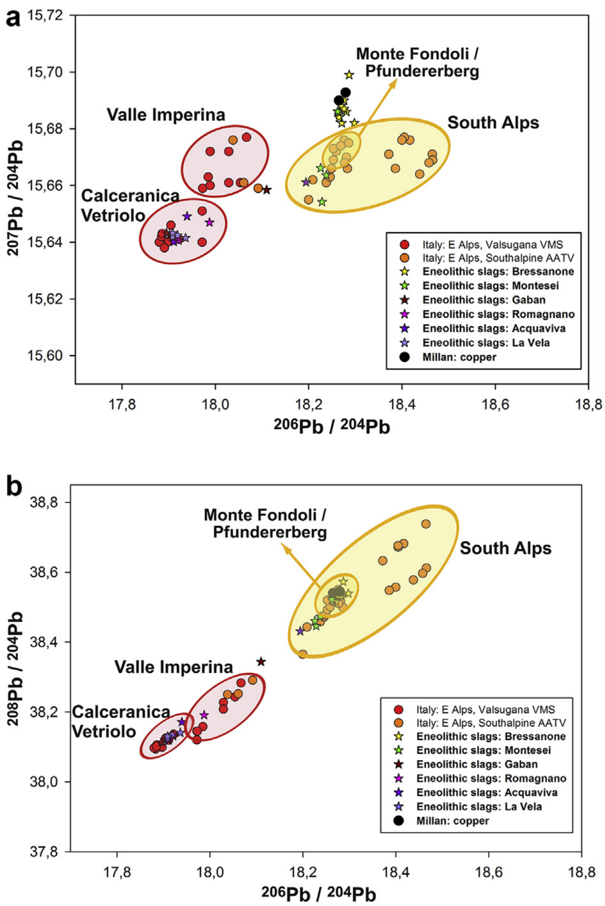

Fig. 6. Lead isotope ratios in the analysed Eneolithic copper smelting slags from Trentino and Alto Adige (stars), compared to the available data for copper ores (circles). The ore deposits have been divided into two main groups: the pre-Variscan massive ores related to the Hercinian basement and located in proximity of the Valsugana fault (red circles: valsugana VMS volcanogenic massive sulphides), the post-Variscan ores mostly constituted by polymetallic sulphides related to Permo-Triassic volcanics (orange circles: broadly labelled as Southalpine deposits from Alto Adige, Trentino, and Veneto AATV). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

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 are also the source for a substantial part of the circulating copper objects in the region. These include several of the objects found in the Col del Buson hoard (Angelini et al., 2011), the awl (from Cellore, Illasi) and the copper ingot (from Cisano, Bardolino) analysed by Pernicka and Salzani (2011), and the two axes from Serravalle (Tecchiati, 1991). The published objects from Tyrol and Serbia show a markedly different signal, mostly related to the Serbian deposits (Höppner et al., 2005; Pernicka et al., 1993, 1997). Only one of the Italian objects (a small metal ring from the Col del Buson hoard) seems to be related to the Balkan ores.

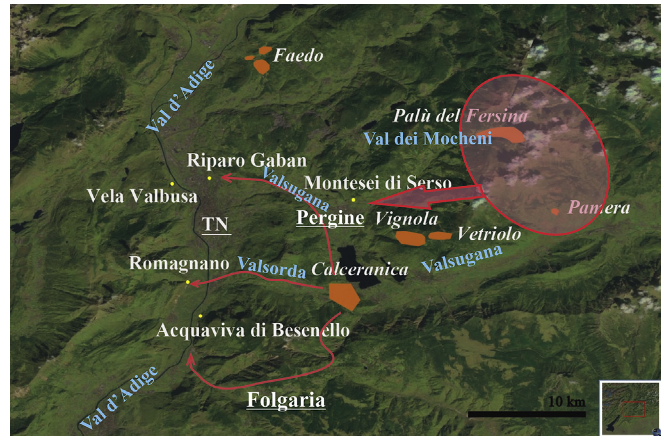

Fig. 7. Location of the occurrences of the analysed Eneolithic copper smelting slags in Trentino (yellow dots), together with the nearby ore sources matching the LI signal (orange fields, names in italics). The localities and geographical features cited in the text are reported (Town names are underscored). Image courtesy of laboratoriobagolini.it/ais/. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

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 seem 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.

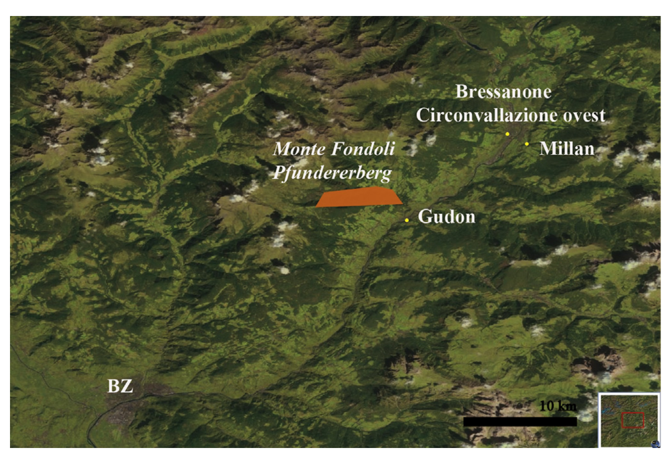

Fig. 8. Location of the occurrences of the analysed Eneolithic copper smelting slags in Alto Adige (yellow dots), together with the nearby ore source at Monte Fondoli (Pfundererberg, Chiusa) matching the LI signal (orange field, names in italics). Image courtesy of laboratoriobagolini.it/ais/. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

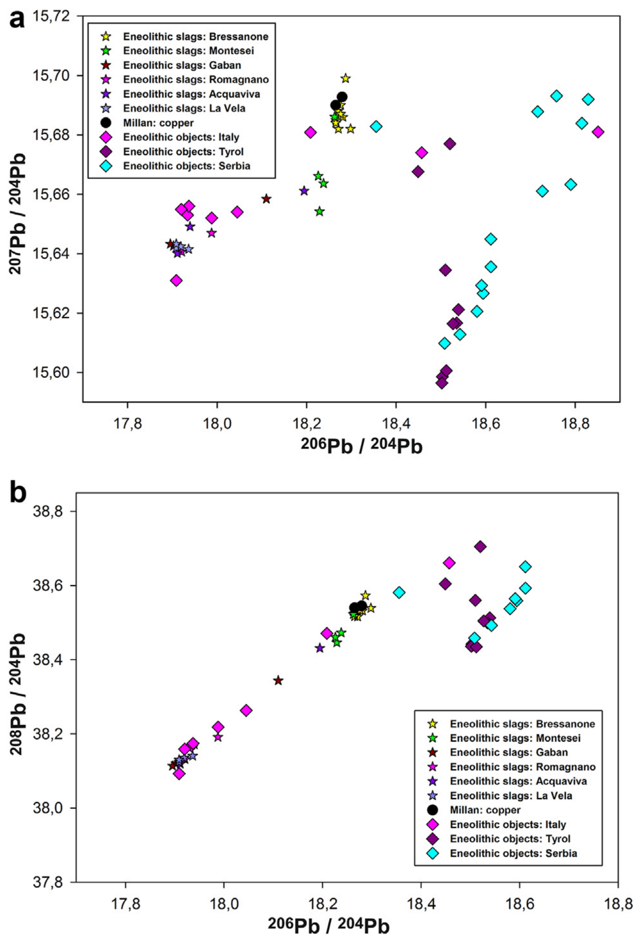

Fig. 9. Lead isotope ratios in the analysed Eneolithic copper smelting slags from Trentino and Alto Adige (stars), compared to the available data for late-Neolithic and Eneolithic objects (diamonds). The data for the objects from Northern Italy are from Angelini et al. (2011) and Pernicka and Salzani (2011). The data for the objects from Tyrol are from Höppner et al. (2005). The data for the Serbian objects are from Pernicka et al. (1993).

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 BFO60-15.

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