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METAL FLOW IN THE LATE BRONZE AGE ACROSS THE FRIULI PLAIN (ITALY): NEW INSIGHTS ON CERVIGNANO AND MUSCOLI HOARDS BY CHEMICAL AND ISOTOPIC INVESTIGATIONS.

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Abstract

Numerous metal hoards found in the Friuli-Venezia Giulia Plain, North Eastern Italy, are archaeologically dated to the Recent and Final Bronze Age. Here the results of an archaeometric study of the copper-alloy artefacts and ingots from two such hoards (Cervignano del Friuli and Muscoli, Udine) are presented. The mineralogical, metallographic and chemical analyses of the copper objects indicate that the metallurgical process employed copper derived almost exclusively from chalcopyrite (±sphalerite, galena) as ore charge. Distinct typologies of ingots (plano-convex and parallel-surfaces) are characterized by different degrees of metal refining. Tools and weapons are made of carefully refined and compositionally-controlled bronze alloys, containing specific proportions of tin. These observations point out a high level of technological knowledge in metallic copper extraction and use. The chemical and lead-isotope compositions of most analysed ingots and artefacts suggest exploitation of copper deposits from the Southeastern Alps, including deposits from the Valsugana area and other mining districts of the Trentino-Alto Adige region. One peculiar sample characterized by a significant Sb content suggests possible mixing with “local” fahlerz copper from Carnia and one other ingot, showing a highly radiogenic lead isotope composition may be interpreted as copper from southern Tuscany or as a mixed metal incorporating a small quantity of copper from northerly Alpine sources such as Mitterberg.

Keywords: Provenancing, Late Bronze Age copper-alloys, lead isotopes analysis, archaeometallurgy, North-Eastern Italy.

1. Introduction

The Italian Bronze Age is commonly subdivided into four periods: the Early Bronze Age (EBA: 2300 – 1700 BC), the Middle Bronze Age (MBA: 1700 – 1350/1300 BC), the Recent Bronze Age (RBA: 1350/1300 – 1150 BC), and the Final Bronze Age (FBA: 1150 – 950 BC) (Bietti Sestieri 2010; Borgna et al. 2018a, b). Differently to the English literature, which generally refers to RBA as the Late-BA, the Italian literature commonly use the term Late-BA in order to encompass RBA and FBA (Pearce 2004). Here the Italian convention will be adopted, i.e. the Late Bronze Age (LBA) will indicate both Recent and Final Bronze Ages.

Starting from the MBA, with a peak at the MBA-RBA transition, an important phenomenon of population increase and settlements diffusion took place in the area corresponding to the modern Friuli-Venezia Giulia, in Northastern Italy (Borgna et al. 2018a, b; Bettelli et al. 2018). Owing to its strategic position, this area played a key role as an economic hub between the Adriatic area and Central Europe and recent studies have recognized this region as an important connection route for the flow of metal between mainland Italy and the Balcanic area (Borgna 2009). Protohistoric sites located near Aquileia, on the fluvial axis Torre-Aussa-Natissa-Isonzo Rivers, coupled with numerous stray finds of bronzes, testify the importance of the territory in the diffusion of the metal during the Bronze Age, particularly in the MBA-RBA. Thus, an extensive study of copper ingots and weapons found in Friuli's metal hoards was proposed as fundamental for the understanding of the copper flow in the last part of the 2nd millennium BC (Vitri 1984, 1999; Borgna 2001, 2004). The project formed the core of a doctoral research project at the University of Padova (Canovaro et al. 2014; Canovaro et al. *in press*; Nardini et al. *in press*), carried out in collaboration with the Soprintendenza Archeologia, Belle Arti e Paesaggio of Friuli-Venezia Giulia. Here we focus on two important protohistoric hoards (Cervignano del Friuli and Muscoli, Udine) located in the Lower Friuli Plain (Fig. 1). The major goals are (i) to investigate the compositional variations between ingots and objects, as well as the working processes, (ii) to shed light on the nature and provenance of the Cu-ore charge used for the manufacturing of the objects, (iii) identify the ores exploitation areas, and (iv) try to reconstruct the circulation paths of copper metal in the North-Eastern Italy in the RBA-FBA.

Although the Friuli region has long been considered important for understanding the Bronze Age flow of copper metal (Pellegrini 1911), a systematic research program that could define the dynamics of metal flow between Northern Italy and the Balkans during the LBA has never been carried out. Only sporadic chemical analyses have been performed (Pigorini 1895; Tuniz et al. 1986; Casagrande et al. 1994; Giumlia-Mair 2000, 2003, 2005, 2009) and, until now, the interpretation of the copper provenance has not yet been supported by isotopic data.

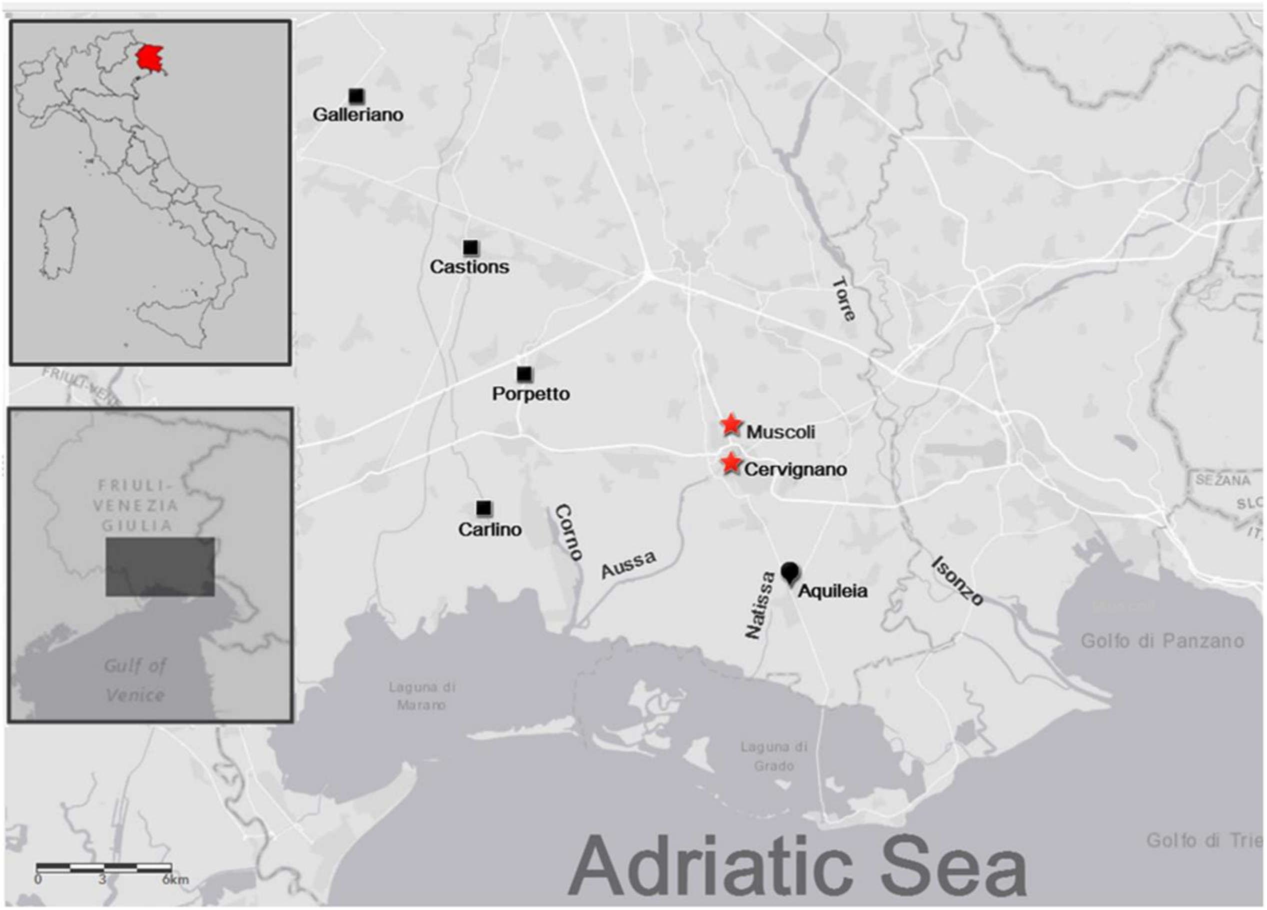

Fig. 1 Distribution map of the metal hoards discovered in in the Lower Friuli Plain. Red stars identify the hoards under study

The issue is even more intriguing if we consider that in the southern part of the Alps there are only modest copper ore deposits, with the notable exceptions of the Trentino and the South Tyrol regions, as discussed by Cierny (1992, 1995, 1997) and Zucchini (1998). In particular, copper ore deposits are scarcely present in the Friuli-Venezia Giulia and restricted to a few fahlerz and mixed chalcopyrite–fahlerz-rich occurrences in the Carnia region, along the Italy–Austria border (Artioli et al. 2016). Exploitation of these deposits is attested in the Middle Age (Giumlia-Mair 2005, 2009; Zucchini 1998), whereas prehistoric or protohistoric exploitation is yet dubious. Differently, in Veneto and Trentino, the presence of slags and isotopic data shows that the deposits in these areas were in use since the Chalcolithic (Artioli et al. 2015; Artioli et al. 2016). There are no analyses confirming the exploitation of these mining areas in the EBA, while there are data suggesting their use in the MBA-LBA both for local productions (Artioli et al.2016) and for "export" (Melheim et al. 2018) The origin of the copper circulating in Friuli-Venezia Giulia in the late 2nd millennium is therefore still a challenge.

In order to face this issue, copper-based objects and ingots were selected for investigation from the well-dated protohistoric hoards of Cervignano and Muscoli. The choice to include ingots in the study is a key factor: despite the likely eventuality of recycling or mixing of metal in the artefacts, the composition of raw metals (ingots) can be considered as close as possible to the composition of the fresh metal batch used by the metallurgical workshop. In the present investigation ingots were selected according to morphology and abundance in each hoard. The sample selection was extended to tools and weapons (especially axes and swords), whose production requires a carefully controlled metallurgical process in terms of composition and time/temperature path. They are assumed to be less prone to metal recycling and, thus, more suitable for provenance studies.

The aim of the study is to improve the knowledge about the flow and metallurgical processing of copper in the region during the MBA-RBA.

2. Cultural context

The last part of the MBA in Friuli-Venezia Giulia was characterized by a consolidation of the territorial organization of the settlements and by intense socio-economic changes: the archaeological record indicates a remarkable demographic explosion and the widespread diffusion of fortified settlements. These settlements were part of a structured and organized exchange system that linked the *Terramare* area, located approximately in the Plain south of the Po River (North Italy) (Bernabò Brea et al. 1997), to the Alpine and Danubian regions (Cassola and Vitri 1997). Despite the widespread diffusion of metallurgy, the phenomenon of metal hoarding was not frequent in this period and the archaeological discoveries are mostly attested by swords hidden in wet environments or by stray finds in settlements. This type of deposition is widely attested in Friuli-Venezia Giulia, especially in the Southern Plain, and the presence of swords having technical features and decorative style consistent with Central European models (De Marinis 2011) highlights the role of this area as a possible connection between the Po-Valley and the Danube areas (Ling et al. *submitted*).

In the RBA, the technological progress of metal mass production that stated in the MBA was further expanded and the circulation of techniques, ideas, models, types and materials across Europe encouraged the production of standardised objects and the formation of a common language, defined as a metallurgical *Koiné* (Peroni and Carancini 1997). From the archaeological and economical point of view, the main consequence of this increased demand of metallurgical productions was the growth of the exploitation of the ores in the Carpathian area and in the Alps. An indirect connected phenomenon is the presence of a large number of metal hoards that, regardless of the deposition motivation and praxis, testify the large amount of metal in circulation in this period.

At around 1200 BC an unstable situation occurred, leading to a serious crisis of the *Terramare* culture and to its definitive collapse. During the second part FBA, after this European- and Mediterranean-wide period of instability, the Friuli-Venezia Giulia Plain acquired a strategic importance as connection area for the commercial routes between the Alps (Slovenia and Urnfield spheres) and the Po Valley (Simeoni and Corazza 2011).

3. Materials

3.1 RBA: Cervignano

Accidentally discovered in 1984, the Cervignano del Friuli hoard was hidden on the border of a settlement probably during the first phases of the RBA (Vitri 1984, 1991; Borgna 2001; Tasca 2011). This hoard includes mainly ingots and a few objects, whose typology indicates active relationships with the Alpine area.

The deposit was interpreted as a founder's storage because of the prevailing presence of fragmented ingots and scarce tools, such as a winged-axe, a chisel and a sickle blade (Vitri 1984; Vitri 1991; Borgna 2001). For the purpose of the present study, 6 Plano-Convex (PC) ingots, 5 Parallel-Surfaces (PS) ingots and 1 Tongue-Shaped (TS) ingot were taken into account (Table 1). PC-ingots are sub-triangular or rectangular fragments, in which the flat section on top and the underneath curved surface are preserved (Fig. 2a); in most cases, the ingots fractures are net at the top and irregular at the bottom, reflecting a breaking action. The weight of each PC-ingot is fairly typical for European LBA plano-convex melting products deriving from small furnaces, and estimated 2–to–4 kg (Tylecote 1992). On the other side, the PS-ingots differ from the PC ones for the flat surfaces and the constant section, as depicted in Fig. 2b. Actually, this classification has not been formalized in any published work, but it refers to a provisional subdivision elaborated by Girelli (2013). Finally, a fragment having flat ends and exhibiting rounded margins on the tapering sides represents the unique evidence of TS-ingot; as can be seen from Fig. 2c, the fracture highlights the convex section.

Among the objects found in Cervignano, the attention was focused on the axe (Cer-Ax, Fig. 2d), which has a curved blade edge and large wings like flanges (Table 1). In antiquity, a wooden handle would have been inserted under the winged flanges and held in place by a thick rope, tied through the loop. Typologically, the winged-axe may be referred to a non-advanced stage of the RBA, likely to the Alpine *Freudenberg type* (Borgna, 2001; Tasca 2011).

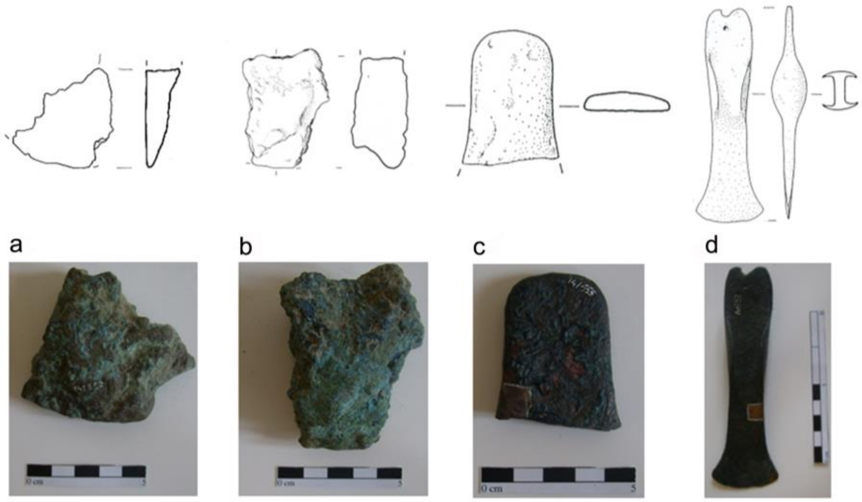

Fig. 2 Photos and archaeological drawings (Borgna 2001) of the different types of copper ingots and the axe belonging to the Cervignano hoard. a) Plano-convex ingot (Cer-PC-56); b) Parallel-surface ingot (Cer-PS-62); c) Tongue-shaped ingot (Cer-TS) and d) bronze winged-axe (Cer-Ax)

3.2 RBA-FBA1: Muscoli

The deposit of Muscoli is located close to Cervignano del Friuli (Fig.1). The hoard was found outside a settlement and it contains tools (10 sickles and 2 socketed-axes), weapons (2 swords deliberately broken) and fragmented PC-ingots (Marchesetti 1903; Pigorini 1904; Anelli,1949; Vitri 1990). Due to typological evidence of the enclosed items, it could be dated to a later period than Cervignano and definitively sealed at the beginning of FBA, but it includes more ancient materials attesting the circulation of metal in the end of the MBA and during the RBA (Borgna 2001, 2004).

The set of findings could be interpreted as a generic form of hoarding containing materials removed from the circulation for safekeeping and intended as reserve for commercial exchanges or supply of the local metallurgical activity. In addition, the preservation study of these artefacts reveals clear traces of use/wear that suggest a prolonged circulation and use (Borgna 2001).

Besides the 6 PC-ingots (Fig. 3a), comparable in weight and shape to those of Cervignano, two swords, both fragmented, and two axes were selected for this study (Table 1). One sword is associable to the type *Sacile* (Bianco Peroni 1970; Cupitò 2006) and it refers to the end of MBA (Fig. 3c, Mus-S-3), whereas the other, identified as a type *Cetona* (Bianco Peroni 1970), can be dated to the RBA (Fig. 3b, Mus-S-2).

The socketed-axes have different decorations. One axe (Mus-Ax-6, in Fig. 3d) shows an evident V-shaped decoration that commonly appears in weapons dated to the RBA. This specimen can be compared with similar finds discovered in Alpine and Transalpine areas, mostly in Slovenia and Northern Croatia (Turk 1996), besides the Italian axes of *Doss Trento* type (Borgna 2001; Carancini and Peroni 1999). Conversely, the sample Mus-Ax-7 (Fig. 3e) shows "pseudo-wings" decoration embossed on each sides of the body and obtained with ribs arranged in concentric circles. From a chrono-typological point of view, such axe should be dated to a later period, probably to the FBA, in the middle Urnfield context (Borgna 2001).

Fig. 3 Photos and archaeological drawings (Borgna 2001) of some artifacts belonging to the Muscoli hoard. a) Plano-convex ingot (Mus-PC-1); Muscoli swords; b) Mus-S-2; and c) Mus-S-3; bronze socketed-axes: d) Mus-Ax-6; and e) Mus-Ax-7

4. Sampling technique and analytical methods

The archaeometric study of the selected samples involved the employment of several characterization techniques, and the related analytical protocol was divided into different steps. Firstly, each specimen was observed under a stereo microscope and a few fragments (~5-6 mg) were detached using a fine steel blade to effectively avoid contamination. For each object, one micro-sample was embedded in epoxy resin and used to perform chemical and metallographic analyses (Scott 2012), while other 2-3 mg were employed for provenance studies.

Chemical analyses were performed using a CamScan MX 3000 Scanning Electron Microscope (SEM) equipped with a LaB_6 cathode, coupled with an energy dispersive spectrometer (EDS), working in high vacuum mode. The back-scattered electrons (BSE) images and the micro-chemical analyses were acquired using the CamScan Helios 5.2.22 software and the SEM Quant PhiZAF software, respectively. Bulk compositions were acquired analysing 3 to 5 dimensionally similar areas, whereas the inclusions dispersed within the α-phase were characterized using the spot-mode analysis.

The quantitative chemical analyses of minor and trace elements in the metal matrix, inclusions and segregations were performed by electron microprobe analysis (EPMA, CAMECA SX50) fitted with four vertical wavelength-dispersive spectrometers (WDS). The adopted working conditions for the selected suite of elements were accelerating voltage of 20 kV, beam current intensity of 20 nA and counting times of 5-10-5 s on background-peak-background, respectively. The results were processed using the PAP (CAMECA) software for the ZAF corrections.

On the basis of the analysis, only a selection of ingots, covering the different chemical compositions and typologies, and all the weapons were subjected to Lead Isotope Analysis (LIA). Acid digestion and chromatography of the specimens were carried out in an ultra-clean laboratory of the Department of Geosciences (University of Padova), tested in order to produce reliable isotopic samples (Villa 2009). The mass spectrometer used for the analysis is a Nu Instrument™ multicollector plasma-source (MC-ICP-MS) located at the Institut für Geologie of the University of Bern (Switzerland). The Pb setup uses eight Faraday collectors to measure all masses between ^{202}Hg and $^{209}PbH^{+}$. The sample solution was ionized by introducing it into 9000K plasma allowing for the simultaneous ionization of all elements. Mass fractionation was monitored by adding few nanograms of natural thallium, which possesses a known $^{203}Tl/^{205}Tl$ ratio, fractionated by the same mechanism as Pb and not interfering with Pb isotope measurements (White et al. 2000). Every five measured sample solutions, a calibration was carried out using the NIST SRM 981 international standard. The measured values of NIST SRM 981 matched favourably with those reported in literature (Hirata 1996; Belshaw et al. 1998; Rehkamper et al. 1998; Rehkamper et al. 2000; White et al. 2000), assuring the reliability of the performed isotopic analyses.

5. Discussion

5.1 Chemical and Microstructural Results

The characterization of the metal, both from a chemical and a structural point of view, is fundamental to make reliable considerations on the metallurgical process and on provenance. Especially for ingots, it is useful to not exclusively consider the analysed suite of elements and their concentrations, but also define the connections with the microstructural features, in order to achieve useful information on the different metallurgical steps.

According to the chemical analyses (Table 2), all the ingots from the Cervignano and Muscoli hoards are made of pure or almost-pure copper (92–98%), primarily associated with Fe, S, Pb and, in some cases, Zn. In only one case (Cer-PS-64) substantial concentrations of Sb (1.9%) were detected, in line with the presence of Sb-rich phases in the sample.

In all ingots, Pb is segregated and mixed with small amounts of other chalcophile elements such as Bi, As, Sb and Ag. In general, for ingots, the lead amount depends on the galena present in the extractive charge. Therefore, percentages up to 2.0–2.5% can be interpreted as deriving from the ores, and suitable for the LIA. In most of the PS-ingots, Pb is abundant and mainly arranged as grain boundaries segregations (Fig. 4a), whereas in PC-ingots Pb was detected in minor or trace concentrations and was almost absent in two samples from Muscoli (Mus-PC-6 and Mus-PC-7).

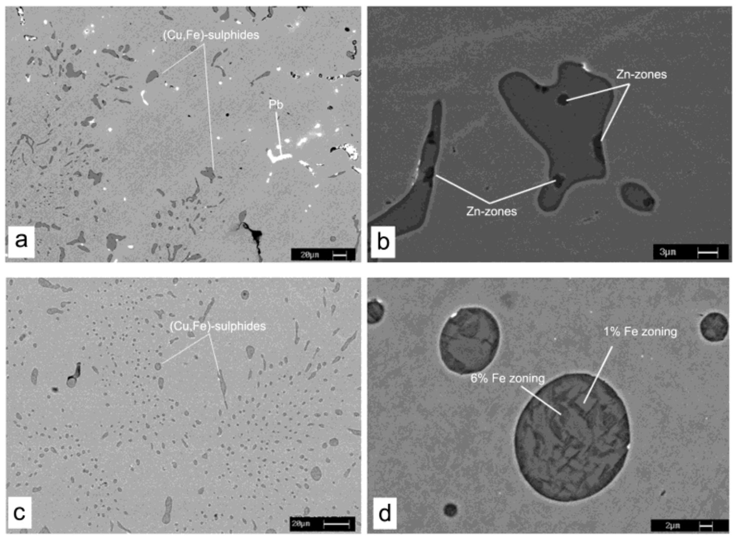

Fig. 4 SEM-BSE images of PC- and PS-ingots. a) PS-ingot (Cer-PS-61); b) detail of Cer-PS-61, showing Zn-rich inclusions (30% Zn in dark areas); c) PC-ingot (Mus-PC-1); d) detail of Mus-PC-1 showing Fe-zoned sulphide (light gray area 1 % Fe, dark gray area 6% Fe)

In principle, the variable concentrations of Fe detected by areal SEM-EDS analysis in the smelted copper (Table 2) could be related to the presence of residual (Cu,Fe)-sulphides, possibly due to the smelting temperature not being high enough to remove all the impurities, or to the use of an Fe-ore flux in the smelting process (Maddin et al. 1980; Craddock and Meeks 1987; Tylecote et al. 1977). The microstructural observations revealed the presence of chalcocite and bornite trapped in the intergranular or interdendritic spaces, which clearly indicate the sulphidic nature of the smelted ore (Hauptmann et al. 2002) (Figs. 4a and c). In such sulphides, in addition to the common Fe-zoning, the SEM observations revealed several Zn-rich inclusions (Fig. 4b), which were also observed in PC-ingots. They are of sub-micrometric size and considerably smaller than in PS-ingots (Fig. 4d). These observations indicate that the original ore-charge was made of (Cu-Fe)-sulphide, probably chalcopyrite, associated with minor sphalerite and galena. In the case of ingot Cer-PS-64, the significant Sb concentrations suggest the additional presence of tetrahedrite in the mix.

Variable contents of Fe and Zn were detected in the α-phase in the Cervignano and Muscoli ingots (EPMA data in Table 2). Fe and Zn are generally low and positively correlated in the PC-ingots from Cervignano (r^2=0.92), whereas they are highly variable and uncorrelated in the PS-ones. Furthermore, microstructural observations reveal higher amounts of Zn (especially in sulphides), Pb-segregations and Fe(Co)-inclusions in the PS-ingots supporting the typological distinction of ingots (Fig 4a and b). Since Fe and Zn decrease in quantity when they are subjected to subsequent stages of remelting (Tylecote et al. 1977; Merkel 1983) and basing on their higher concentrations of impurities, the PS-ingots can be interpreted as being less refined than the PC-ingots, suggesting the lack of the last refining step in a multi-stage production process (Tylecote 1981) or the use of a technologically less evolved process (Craddock and Meeks 1987; Hauptmann et al. 2002; Mangou and Ioannou 2000). As it will be argued in the paragraph dedicated to provenance, isotopes support the main thesis that PC and PS-ingots were smelted from the same metal source. Indeed, since ancient smiths should have been aware of different properties resulting after the smelting and the refining process, the different shapes of the ingots probably allowed for their proper identification and handling in the workshop. Thus, the PC-ingots could serve as raw metal (almost) ready for the production of the final objects, whereas the PS-ingots would not (yet) be appropriate for the creation of usable objects, since the high concentration of iron negatively affects casting and hammering steps (Craddock and Meeks 1987). In Muscoli, only PC typology is present but, in general, the situation is the same as the PC-ingots from Cervignano: very refined and with few inclusions.

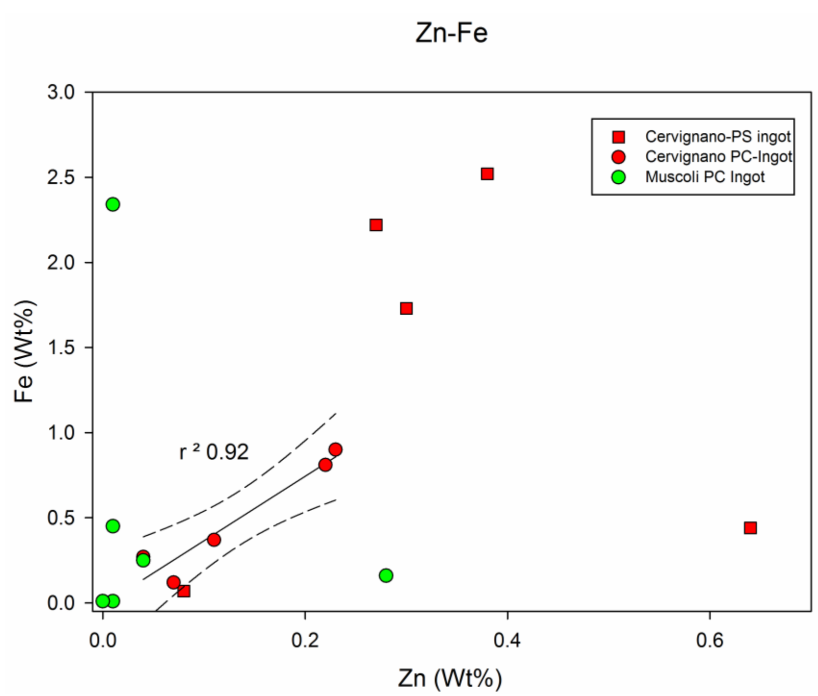

Fig. 5 Scatter plot of Fe and Zn concentrations detected in the α-phase of all the investigated ingots. In the diagram, the regression line calculated for PC-ingots and its coefficient of determination (r^2= 0.92) is reported

Cuprite (Cu_2O) inclusions were detected in almost every specimen, except in Cer-PC-72, Mus-PC-2 and Mus-PC-8. Cuprite forms as a result of the nearly complete immiscibility of oxygen in copper, both in liquid and solid states. In our specimens, cuprite occurs as spherical droplets showing eutectic intergrowths (Fig.6a). According to Hauptmann et al. (2002), this is evidence for a re-melting of the copper, as Cu_2O should not directly form in smelted copper. These oxide particles, along with sulphides, affect the malleability of the copper, making it brittle and facilitating the breaking of ingots, which is a useful feature for their exchange. In this regard, before casting, alloying or metallurgical working, most of the copper ingots should have been subjected to a further purification step in order to reduce or eliminate the cuprite and sulphide inclusions and improve the metal quality.

Among all ingots, the TS-ingot is peculiar, since it is a bronze ingot with 20% Sn content (SEM analysis, Table 2), significant levels of Ni (1.2%) and traces of As (0.3%), appreciable only by EPMA. The presence of coarse α-dendrites with an infill of (α+δ)-eutectoid phase (Fig. 6b) testifies for a slow ingot cooling (Scott, 2012). EPMA analysis detected small amounts of Ni (0.8%) in the δ-phase. The δ-phase is a hard and brittle constituent that considerably reduces the alloy ductility, making the metal easy to break, consistently with its ingot function. Moreover, since this alloy is not suitable to hammering, this ingot could not be a ready-to-use ingot for tools or weapon making. There are two possible explanations for the employment of such alloy. A high-tin bronze ensures a bright silver-like appearance, which could be visually attractive and simulate silver in the creation of pins or ornaments that only require casting. Alternatively, the alloy could have been used to produce a more workable alloy (~10%-Sn bronze) by adding nearly-pure copper in 1:1 proportions. The presence of widespread Cu- and (Cu,Fe)-sulphides dispersed in the matrix (Fig. 6b) indicates that this raw ingot was not obtained by recycling of refined bronzes and should have been necessarily refined before being profitably used by the craftsman. Hypothesizing that this ingot was obtained using finished objects recycling, certainly the sulphides would be smaller and finely dispersed in the matrix. In the future it might be fascinating to move the attention to the Sn isotopes for the cassiterite tracing. Concerning the tin supply in Friuli, several hypotheses may arise: one source could be identified in the mineral rich areas of southern Tuscany (Benvenuti et al. 2003), where cassiterite deposits were available. Otherwise, according to Giumlia-Mair (2000), another option is that tin could be easily been transported to the territory of the Caput Adriae from the deposits on the mountains of Bukulia and Cer (Western Serbia), located along the important fluvial arteries Danube and Sava (Huska et al. 2014). However, at the moment, it is not possible to further argue this matter, although it should be noted that the Tuscanian cassiterite is a very limited occurrence and there is no evidence of exploitation before the Middle Age.

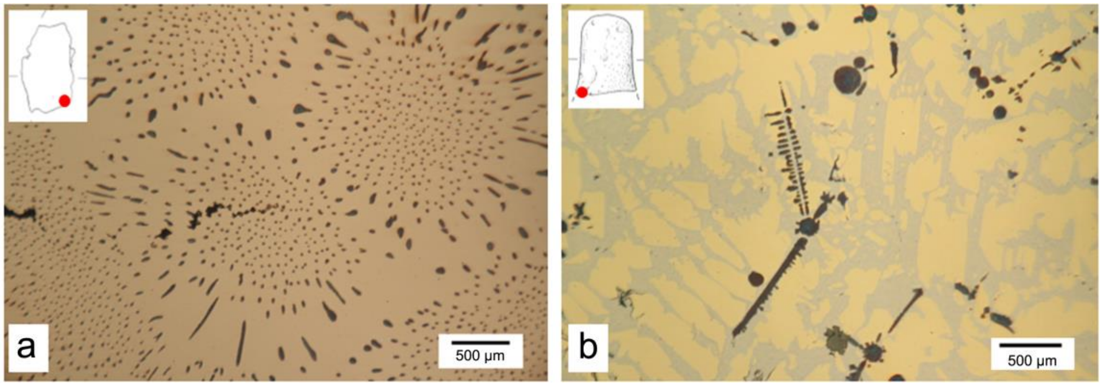

Fig. 6 a) Equiaxed grain of copper and cuprite intergrowth formed by a surplus of oxygen in the liquid copper during casting (sample Cer-PC-79). b) Optical image of the tongue-shaped ingot cross-section (TS-ingot). The image shows copper sulphides (dark gray) and (α+δ)-eutectoid phase (light gray) in copper matrix

For what concerns the objects, the chemical analyses revealed the employment of a quite pure bronze alloy (8-to-12% of Sn), characterized by As and Ni as major impurities. All samples show only minor Pb concentrations (<1.0%) , very low amounts of Fe and Zn and significantly less sulphide inclusions indicating an efficient purification of the metal (Craddock 2000). Such strictly controlled compositional ranges guarantee a proper castability and a good workability of the alloy, as required for the creation of swords and axes. Concerning the swords, the tin range was more carefully controlled (10–12%), as expected for quality weapons.

Both the objects and the TS-ingot are characterized by the presence of As and Ni in the α-phase, which are not detected in the other types of ingots. Arsenic is a volatile element and the smelting process leads to its reduction or elimination (Tylecote et al. 1977); therefore, its persistence in the objects would be justified only if the raw copper ingots were characterized by relatively high-As compositions. Instead, in our case the As concentrations in the objects (0.1–0.4%) are comparable to those in the ingots (Fig. 7). Moreover, Ni is also a distinguishing element of objects (0.3–0.6%) and exhibits a good correlation with As (Fig. 7), suggesting a common source for these two elements. The presence of traces of Ni, sometimes associated with minor As, is very common in Bronze Age objects (Pernicka and Salzani 2011; Melheim et al. 2018), although the origin of the As-Ni association is still debated (Liversage 1994). In fact, it is still unknown if such elements were present in the extractive charge and were thus derived from the ore mineral assemblage or if they were deliberately added to the mix for technological reasons (e.g., addition of Ni in order to retain the As; Sabatini 2015) or if their association is the result of several cycles of casting and recycling. However, the concentrations of such elements are so low that the last hypothesis seems unlikely. At the moment, “how” and “why” are still open questions.

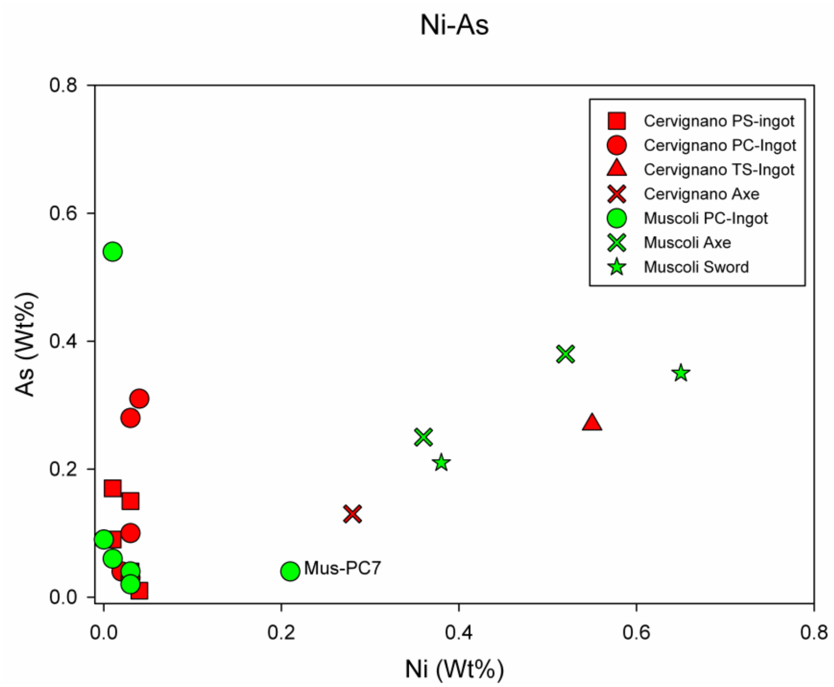

Fig. 7 Scatter plot of As and Ni showing a clear correlation between these elements for the objects and the TS-ingot. Among PC and PS-ingots, Mus-PC-7 is the only sample in which appreciable levels of Ni were detected

5.2 Metallographic observations

Metallographic examinations showed that all ingots possess a dendritic structure, as expected in as-cast items (Fig. 8a). For what concerns the objects, the examination of the microstructure provides information about the degree of working, annealing and homogenisation. The cross-section of the sword Mus-S3, representative of the handle area, exhibited a microstructure consistent with a plastic deformation followed by annealing cycles, since recrystallized and twinned grains were observed (Fig. 8b). Moreover, numerous slip-lines across the α-phase crystals are evident, which testify a strain-hardened state due to plastic deformation in the last step of the refining process. These observations suggest that also the handle of the sword has been subjected to plastic deformation after the casting. The cross-section of Mus-S2 (Fig. 8c) is instead exclusively characterized by large twinned crystals, attesting the complete annealing and the absence of further deformations in this area.

Moreover, the metallographic observations showed that only the outer parts of the blades were finished by hammering, in order to obtain cutting edges. In swords, the manufacturing process commonly involved a heating treatment performed to recover ductility through recrystallization, and a subsequent hammering in order to confer the desired bronze mechanical properties. All these features are in complete accordance with the metallographic studies carried out on swords from Olmo di Nogara (Verona province, Italy) in which the blades show signs of hammering and annealing cycles and a final cold-working, in order to improve the hardening of the alloy (Angelini 2005). In this regards, all the analysed swords have adequate properties for practical use, proving that not only could these swords be used for ceremonial rituals or as status-display, but also they could be efficient in battle.

Differently, the socketed axes were sampled near the handle-hole and their microstructure suggests a weak plastic deformation after casting, probably carried out to remove the slight imperfections that may result from the casting in two-part mould. The presence of distorted dendrites, slip-lines and only few polygonal grains (Fig. 8d) supports this interpretation. The incomplete absorption of δ-phase by the α-phase is further indication that annealing was not fully reached, not surprising, if we consider that the sample was extracted near the handle

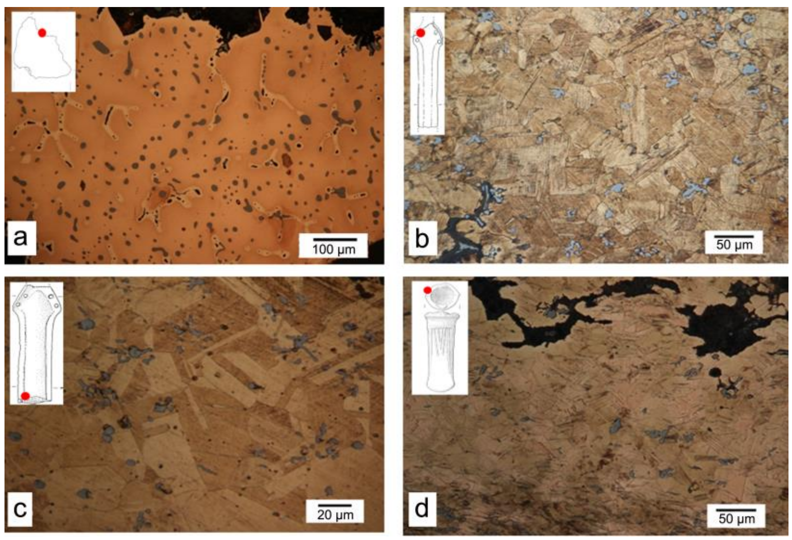

Fig. 8 RL-OM micrographs after etching ($FeCl_3$). – a) Dendritic microstructure of the Cer-PC-56 ingot; b) Bronze sword Mus-S3 sampled on the handle area. Note the slip lines and the twin bands. c) Bronze sword Mus-S2 sampled on the central part of the blade. Note the equiaxial grains with twins, testifying the last annealing process. d) Axe Mus-Ax6, sampled on the handle area. Note the clearly visible residual coring that persists despite the twinned grains

5.3 Provenance question

The Pb-isotope compositions of the selected ingots, swords and axes (Table 3) was compared with those of relevant ore deposits from different published databases (OXALID; BRETTSCAIFE; AAcP; Ling et al. 2014; Nimis et al. 2012; Nimis et al. 2018; Artioli et al. 2016). Because of the significant isotopic overlap between some of the potential ore sources, provenancing of a given object may best be tackled by using a statistical approach. This can be useful to provide, at least, a preliminary discrimination of the possible mining sites. For this purpose, we have first calculated the Euclidean distances in the 3D space, ranking the shortest distances between each measured point and the closest ores in the database (Baxter 2003; Stos-Gale and Stos 2009). Then, we have used a slightly more sophisticated approach that involves the Kernel Density Estimation (KDE; Scaife et al. 1999; Baxter 2000; Hsu et al. 2018), which allows for a visual interpretation of the data density of the reference data in 2D projections. In any case, robust identification of the geographical origin of the copper requires a combination of chemical, isotopic and archaeological information on the possible ore sources that are known to have been exploited in the Bronze Age.

Both the Euclidean test and the visual comparison with Kernel Densities (Fig. 9) suggest an isotopic similarity of the Cervignano and Muscoli samples with the ores from the Eastern Alps (Italy and Austria) and, in one case, Southern Tuscany. Note that, since the minimum number of samples required to perform KDE is 20 (Baxter, 2000), only single points were reported for deposits from the Carnia and Carinthia regions.

All but one of the PC-ingots and objects from Muscoli exhibit a Pb-isotope composition perfectly compatible with one of the major groups of ore deposits from the Eastern Southalpine defined by Nimis et al. (2012; 2018) and Artioli et al. (2016), i.e., the Alto-Adige-Trentino and Veneto (AATV) field (Fig. 9). These deposits are mostly represented by chalcopyrite–sphalerite–galena–pyrite veins and are thus compatible with the compositions of the PC-ingots and objects. Three samples from Cervignano (Cer-PS-77, Cer-PS-82 and Cer-PC-72) appear to cluster together in both Pb–Pb diagrams (Fig. 9), showing a less radiogenic signal identical to that of the Valsugana Volcanogenic Massive Sulphide (VMS) ore deposits (e.g., Calceranica, Vetriolo and Valle Imperina; Nimis et al. 2012; Nimis et al. 2018; Artioli et al. 2016), some of which are known to have been exploited already in the second half of the 3rd millennium BC (Preuschen, 1973; Artioli et al. 2016). Also in these deposits the copper mineral is chalcopyrite, which can be associated with minor amounts of sphalerite and galena. Some Austrian chalcopyrite-rich ores from Carinthia (Koppel and Schroll, 1983; Koppel and Weber, 1997) overlap with the Valsugana VMS field, but to date there is no evidence that these ores were exploited in prehistory.

The other Cervignano ingots (Cer-PC-56, Cer-PC-70, Cer-PC-79 and Cer-PS-62) fall in an intermediate region between the two major Valsugana VMS and Southalpine AATV fields, suggesting two possible interpretations. The mismatch with the established Pb-isotope fields could indicate the existence of yet unexplored mines. However, this area has been extensively surveyed and all geological types of ore deposits occurring in the region have been investigated, therefore the existence of a missing isotopic signal is not the most favourable hypothesis. Alternatively, the linear arrangement of these samples in both the $^{206}Pb/^{204}Pb$ vs $^{208}Pb/^{204}Pb$ and $^{206}Pb/^{204}Pb$ vs $^{207}Pb/^{204}Pb$ diagrams (Fig. 9) could indicate mixing of materials from distinct copper sources (cf. Stos 2009), either during the smelting stage or at a later stage. The latter interpretation was proposed for the slags from the Italian Eastern Alps near Trento (Addis 2013), which also showed an intermediate isotopic compositions between the Valsugana VMS and Southalpine AATV ore deposits, and is the most plausible hypothesis. The compositional data, which indicate that all these ingots are made of pure or almost pure copper, primarily smelted from chalcopyrite associated with sphalerite and galena, are also consistent with this interpretation.

The absence of Sb in all but one of our specimens excludes an involvement of ore deposits from Carnia, which do show some overlap with the AATV field (Fig. 9), but in which tetrahedrite is typically a major, if not dominant, constituent (Artioli et al. 2016). In this regard, it is interesting to consider the composition of the ingot Cer-PS-64, which is rich in Sb (1.9%) and shows an isotopic composition intermediate between the Valsugana VMS and Southalpine AATV fields (Fig. 9). In this case, the high Sb concentration suggests mixing between chalcopyrite from Valsugana (e.g., Calceranica/Vetriolo), having low amounts of impurities, and Sb-rich fahlerz, such as tetrahedrite from Carnia or from minor tetrahedrite-rich occurrences belonging to the AAVT group (Artioli et al. 2016; Fig. 9). From a technological point of view, the addition of Sb could be done intentionally, in order to improve the forgeability of the alloy, but this hypothesis is not supported by the presence in the hoard of objects having the same composition.

Finally, one ingot from Muscoli (Mus-PC7) shows a distinct, highly radiogenic composition, enriched in $^{207}Pb/^{204}Pb$ and, especially, $^{208}Pb/^{204}Pb$. In this case, an effective attribution is difficult, since the copper has a low impurity content (only 0.2% Ni). The Euclidean test would suggest the use of copper from Sothern Tuscany (Fig. 9) to smelt this ingot, although a mixing between copper-ores from Valsugana VMS or Southalpine AATV and highly radiogenic ores, such as those from the Mitterberg region (Pernicka et al., 2016) cannot be excluded.

Concerning the weapons, the Pb-isotope compositions of the investigated axes and swords indicate some of the Southalpine AATV deposits (particularly, those in the Val dei Mocheni, Trentino) as potential copper sources. Minor mixing with copper from Valsugana VMS, as suggested for the Cervignano ingots, cannot be excluded. This tendency is in agreement with predominant position of the Southeastern Alps in the production and distribution of copper around 1500 BC, when mines in the Italian Alps became a main supplier to central and northern Europe (Angelini 2005; Angelini et al. 2015; Artioli et al. 2016; Melheim et al. 2018; Ling *submitted*).

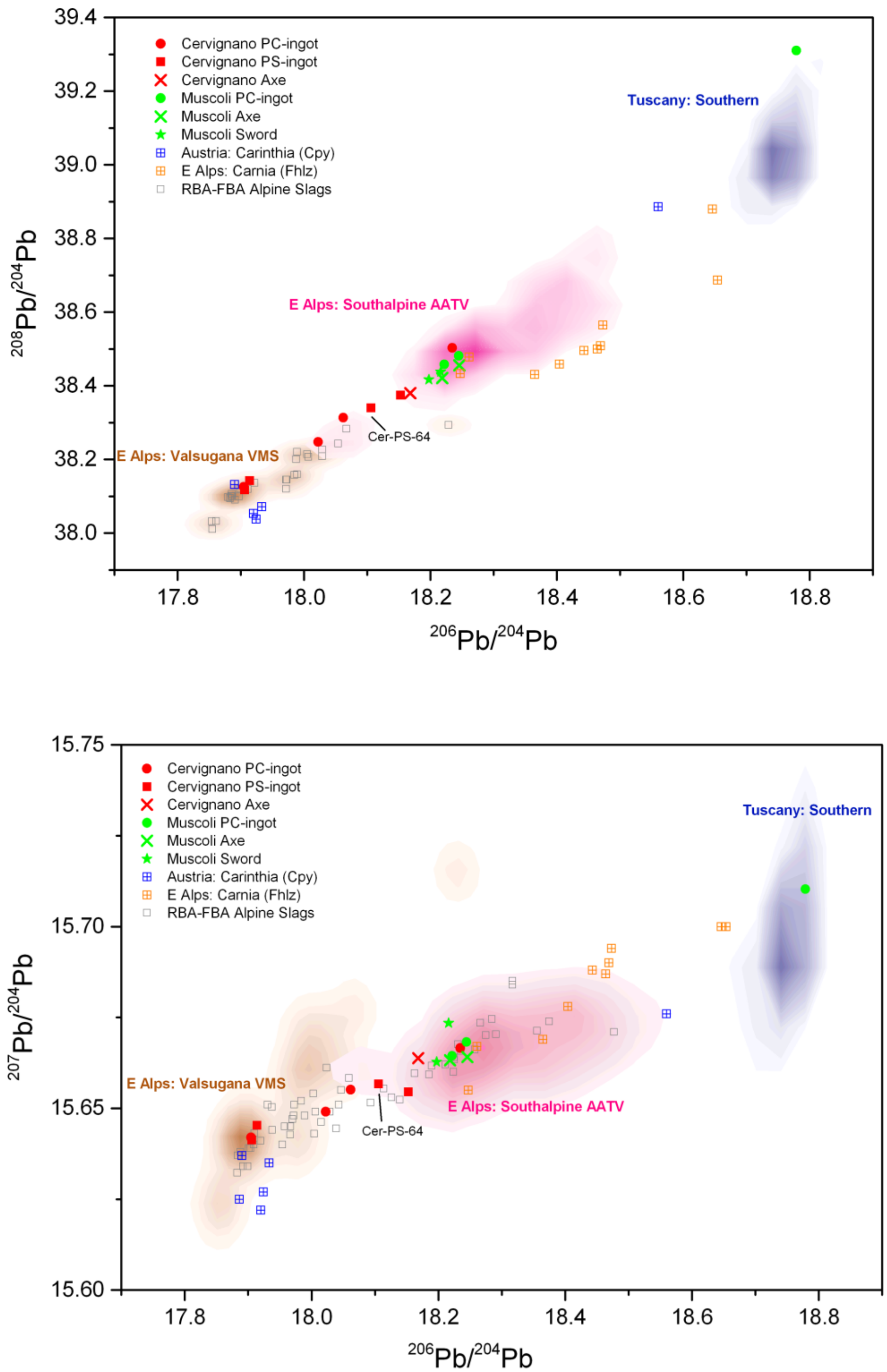

Fig. 9 Lead isotope ratios diagrams of the ingots and the artefacts from the hoards of Cervignano and Muscoli compared with the possible ore-sources. The analytical uncertainty is equivalent or smaller than the size of the symbols

6. Conclusions

LBA semi-worked copper objects (ingots) from the Lower Friuli Plain (Cervignano and Muscoli hoards) show a relatively homogeneous chemical composition, which indicates the employment almost exclusively of chalcopyrite (± sphalerite, galena) as ore charge. This result is compatible with the unavailability of oxide ores during the LBA in Europe and with the necessity of employing sulphide ores in order to answer the ever increasing demand for metal (Rapp 2013). Differences in the amounts of impurities, such as sulphides and segregations, between different types of ingots from the same hoards can be ascribed to the different technological knowledge of ancient smiths. In particular, in the Cervignano hoard, the PS-ingots can be interpreted as being less refined than the PC-ingots, suggesting either the lack of the last refining step or the use of a less evolved process. The presence of a bronze ingot from Cervignano suggests that, within the established copper trade system, alloyed bronze was circulating as ingot already in the RBA.

Other intriguing observations were deduced from the analysis of the weapons and tools. All the analysed weapons are tin bronzes; the swords are made of carefully controlled and homogeneous alloy, whereas variable Sn contents are found in the axes, ensuring anyway a good castability and the workability of the alloy. The low concentrations of Fe and Zn denote a further purification of the metal with respect to the coexisting ingots in the same hoard; moreover, the rare presence of δ-phase and (α+δ)-eutectoid phase suggests well-controlled manufacturing processes characterized by low cooling rate. All the axes and swords underwent extensive plastic deformation followed by annealing cycles. Most of the samples, taken from the blade's edge, were in a strain-hardened state, according to the functional use of the weapons. Only one sample, taken from the central part of the blade of a sword from Muscoli, showed exclusively twinned grains.

Concerning the provenance of ingots, swords and axes, the majority of the samples exhibit lead-isotope and chemical compositions perfectly compatible with the copper ore deposits from the Italian Southeastern Alps (Valsugana VMS and Southalpine AATV groups). Three samples fit precisely the composition of ores from the Calceranica and/or Vetriolo mines (upper Valsugana, Trentino), thus supporting the exploitation of these mines during the MBA-RBA. The compositions of other samples most likely reflect the use of mixed chalcopyrite-rich sources from the same regions. Only two samples differ from the others: a single Sb-rich ingot from Cervignano suggests a possible small flow of metal from Carnia and one other from Muscoli, showing a highly radiogenic lead isotope composition can be interpreted as copper from southern Tuscany or as a mixing using copper-ore from Valsugana VMS or Southalpine AATV and a small quantity of copper from more northerly Alpine sources (Mitterberg). Our results reinforce the hypothesis that, during the RBA and the transition to the FBA, the Friuli-Venezia Giulia area was a leading region for the metal flow, strongly connected with the Alpine area for the metal supply and with some "occasional" contacts with mainland Italy.

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Table 1. Information about the find context, category of items, typology and chronology of all the analysed Bronze Age objects within this study. The materials are presently preserved to the National Archaeological Museums of Aquileia. L= Length, W= Width, H=Height, Ø= Diameter

Sample	Typology	Description	Dimensions (cm)	Weight (g)	Context	Chronology	References
Cer-Ax	Winged-axe	Complete	L 14.2 - W blade 4.4 - H 2.3	208	Cervignano hoard	MBA-RBA	Borgna 2001, 2004; Vitri 1991; Tuniz et al. 1986
Cer-PC-56	Plano-convex ingot	Fragment	L 9.5 - W 6.8- H 2.6	447	Cervignano hoard	RBA	\
Cer-PC-57	Plano-convex ingot	Fragment	Ø 6.8 - H 2.3	336	Cervignano hoard	RBA	\
Cer-PC-66	Plano-convex ingot	Fragment	Ø 5.6 - H 3.2	441	Cervignano hoard	RBA	\
Cer-PC-70	Plano-convex ingot	Fragment	L 6.5 -W 3.5 - H 3.8	315	Cervignano hoard	RBA	\
Cer-PC-72	Plano-convex ingot	Fragment	Ø 4.6 - H 2.9	244	Cervignano hoard	RBA	\
Cer-PC-79	Plano-convex ingot	Fragment	L 5.1 - W 3.5 - H 1.7	109	Cervignano hoard	RBA	Tuniz et al. 1986
Cer-PS-61	Parallel-surface ingot	Fragment	L 7.1 - W 6.5 -H 2.6	556	Cervignano hoard	RBA	\
Cer-PS-62	Parallel-surface ingot	Fragment	L 7.8 -W 5.1 - H 3.4	539	Cervignano hoard	RBA	\
Cer-PS-64	Parallel-surface ingot	Fragment	L 4.6 - W 3.5 - H 2.4	216	Cervignano hoard	RBA	Tuniz et al. 1986
Cer-PS-77	Parallel-surface ingot	Fragment	L 3.6 - W 2.5 - H 1.6	66	Cervignano hoard	RBA	\
Cer-PS-82	Parallel-surface ingot	Fragment	L 2.7 - W 1.9 - H 1.0	26	Cervignano hoard	RBA	\
Cer-TS	Tongue-shaped ingot	Fragment	L 5.3 - W 4.1 - H 0.8	91	Cervignano hoard	RBA	Borgna 2001; Tuniz et al. 1986
Mus-S2	Flange-hilted sword, Cetona-type	Broken	L 15.0 - W blade 2.8	117	Muscoli hoard	RBA	Anelli 1949; Bianco Peroni 1970; Borgna 2001
Mus-S3	Flange-hilted sword, Sacile-type	Broken	L 11.8 - W blade 3.4	189	Muscoli hoard	MBA	Anelli 1949; Bianco Peroni 1970; Vitri 1983; Borgna 2001
Mus-Ax6	Socketed axe, "V" decoration	Complete	L 14.4 - W blade 5.1 Ø handle hole 4.7 x 6.2	547	Muscoli hoard	RBA-FBA	Anelli 1949; Borgna 2001 Carancini 1984; Marchesetti 1903; Vitri 1983
Mus-Ax7	Socketed axe, pseudo-wings decoration	Complete	L 9.2 - W blade 3.3 Ø handle hole 3.5 x 3.2	139	Muscoli hoard	FBA	Anelli 1949; Borgna 2001 Carancini 1984; Marchesetti 1903; Vitri 1983
Mus-PC-1	Plano-convex ingot	Fragment	L 17.7 - W 12.7 - H 7.2	3013	Muscoli hoard	RBA-FBA	Tuniz et al. 1986
Mus-PC-2	Plano-convex ingot	Fragment	L 10.5 - W 5.1 - H 4.3	1453	Muscoli hoard	RBA-FBA	\
Mus-PC-3	Plano-convex ingot	Fragment	L 12.6 - W9.0 - H 5.2	1415	Muscoli hoard	RBA-FBA	\
Mus-PC-6	Plano-convex ingot	Fragment	L 6.8 - W 3.8 - H 2.7	405	Muscoli hoard	RBA-FBA	\
Mus-PC-7	Plano-convex ingot	Fragment	L 7.0 - W 3.6 - H 2.5	256	Muscoli hoard	RBA-FBA	\
Mus-PC-8	Plano-convex ingot	2 Fragments	Fr.A: L 3.1 -W 2.6 - H 0.9; Fr.B: L 3.7 - W 4.1 - H 2.9	Fr.A: 41 Fr.B: 164	Muscoli hoard	RBA-FBA	\

Table 2. SEM-EDS and EPMA chemical analyses (Wt %) of the α-phase metal of all the analysed objects and ingots. EPMA data are calculated as a mean of 5÷7 points analysis. d.l. = detection limit; SD = standard deviation of the measures; * = arithmetic mean between Cu-rich and Cu-poor phases

	SEM-EDS							EPMA													
Sample	**Cu**	**Sn**	**Fe**	**S**	**Pb**	**Sb**	**Ni**	**S**	**Cl**	**Mn**	**Fe**	**Co**	**Ni**	**Cu**	**Zn**	**As**	**Ag**	**Sn**	**Sb**	**Pb**	**Bi**
d.l. (%)								**0.03**	**0.02**	**0.03**	**0.02**	**0.02**	**0.03**	**0.1**	**0.05**	**0.06**	**0.06**	**0.05**	**0.05**	**0.12**	**0.45**
Cer-Ax	89.3	8.9		0.4	0.8		0.5	0.01	0.00	0.00	0.07	0.02	0.28	88.45	0.01	0.13	0.14	10.56	0.00	0.04	0.00
SD	*0.4*	*0.3*		*0.0*	*0.2*		*0.2*	*0.02*	*0.01*	*0.00*	*0.02*	*0.02*	*0.02*	*0.60*	*0.01*	*0.05*	*0.03*	*0.82*	*0.00*	*0.03*	*0.00*
Cer-PS-61	94.4		1.0	1.9	2.6			0.01	0.00	0.01	0.44	0.06	0.03	98.86	0.64	0.04	0.07	0.05	0.15	0.07	0.00
SD	*1.6*		*0.2*	*0.3*	*1.3*			*0.01*	*0.00*	*0.02*	*0.05*	*0.02*	*0.02*	*0.16*	*0.06*	*0.05*	*0.03*	*0.04*	*0.10*	*0.08*	*0.01*
Cer-PS-62	96.5		0.5	1.4	1.5			0.01	0.00	0.00	0.07	0.01	0.01	99.79	0.08	0.17	0.06	0.01	0.00	0.11	0.00
SD	*1.4*		*0.2*	*0.9*	*0.9*			*0.01*	*0.00*	*0.01*	*0.01*	*0.02*	*0.01*	*0.41*	*0.06*	*0.06*	*0.03*	*0.02*	*0.01*	*0.10*	*0.00*
Cer-PS-64	92.4		2.6	1.7	1.3	1.9		0.01	0.00	0.00	2.52	0.07	0.01	96.37	0.38	0.09	0.03	0.18	0.09	0.04	0.02
SD	*1.1*		*0.5*	*0.5*	*0.4*	0.1		*0.01*	*0.00*	*0.00*	*0.21*	*0.02*	*0.01*	*0.88*	*0.06*	*0.06*	*0.03*	*0.11*	*0.09*	*0.09*	*0.04*
Cer-PS-77	95.6		1.9	1.1	1.4			0.01	0.00	0.00	1.73	0.10	0.04	98.22	0.30	0.01	0.05	0.03	0.05	0.06	0.00
SD	*0.5*		*0.2*	*0.1*	*0.8*			*0.01*	*0.01*	*0.01*	*0.23*	*0.02*	*0.02*	*0.89*	*0.03*	*0.01*	*0.02*	*0.03*	*0.05*	*0.06*	*0.00*
Cer-PS-82	93.6		3.5	1.4	1.5			0.01	0.01	0.01	2.22	0.08	0.03	97.76	0.27	0.15	0.10	0.07	0.16	0.01	0.00
SD	*0.6*		*0.3*	*0.5*	*0.8*			*0.01*	*0.01*	*0.01*	*0.45*	*0.03*	*0.03*	*1.03*	*0.06*	*0.06*	*0.05*	*0.04*	*0.05*	*0.03*	*0.00*
Cer-PC-56	95.0		1.4	1.7	1.9			0.01	0.01	0.01	0.76	0.05	0.02	99.43	0.56	0.04	0.06	0.02	0.07	0.09	0.00
SD	*0.4*		*0.1*	*0.5*	*0.5*			*0.01*	*0.01*	*0.01*	*0.05*	*0.01*	*0.02*	*0.54*	*0.06*	*0.04*	*0.04*	*0.03*	*0.04*	*0.07*	*0.00*
Cer-PC-57	94.3		1.6	1.6	2.6			0.01	0.01	0.00	0.81	0.05	0.03	99.41	0.22	0.28	0.08	0.05	0.06	0.05	0.00
SD	*0.2*		*0.2*	*0.2*	*0.2*			*0.01*	*0.01*	*0.00*	*0.10*	*0.01*	*0.03*	*0.34*	*0.03*	*0.20*	*0.05*	*0.05*	*0.05*	*0.05*	*0.00*
Cer-PC-66	95.2		1.5	1.5	1.8			0.01	0.00	0.01	0.90	0.04	0.02	99.34	0.23	0.04	0.04	0.04	0.05	0.00	0.06
SD	*0.6*		*0.1*	*0.2*	*0.9*			*0.01*	*0.00*	*0.01*	*0.05*	*0.02*	*0.02*	*0.56*	*0.05*	*0.02*	*0.05*	*0.02*	*0.03*	*0.00*	*0.13*
Cer-PC-70	98.1			1.9	tr.			0.02	0.01	0.00	0.27	0.07	0.04	99.94	0.04	0.31	0.09	0.09	0.05	0.05	0.00
SD	*0.5*			*0.5*				*0.04*	*0.01*	*0.00*	*0.04*	*0.01*	*0.03*	*0.68*	*0.03*	*0.12*	*0.05*	*0.06*	*0.02*	*0.05*	*0.00*
Cer-PC-72	96.7			1.4	1.9			0.00	0.00	0.01	0.12	0.02	0.03	99.85	0.07	0.03	0.07	0.03	0.14	0.16	0.00
SD	*0.7*			*0.3*	*0.9*			*0.01*	*0.00*	*0.01*	*0.03*	*0.01*	*0.02*	*0.21*	*0.05*	*0.04*	*0.04*	*0.04*	*0.05*	*0.14*	*0.00*
Cer-PC-79	98.2			1.8	tr.			0.01	0.01	0.00	0.37	0.02	0.03	99.16	0.11	0.10	0.08	0.04	0.16	0.07	0.00
SD	*0.1*			*0.1*				*0.01*	*0.01*	*0.00*	*0.03*	*0.01*	*0.02*	*0.29*	*0.03*	*0.04*	*0.04*	*0.01*	*0.07*	*0.08*	*0.00*
Cer-TS	76.3	20.2	1.1	0.5	0.6		1.2	0.00	0.00	0.01	0.79	0.06	0.55	85.88	0.01	0.27	0.02	13.48	0.00	0.05	0.00
SD	*1.7*	*1.6*	*0.1*	*0.2*	*0.6*		*0.0*	*0.00*	*0.01*	*0.01*	*0.08*	*0.01*	*0.04*	*0.86*	*0.01*	*0.09*	*0.03*	*0.59*	*0.00*	*0.07*	*0.00*
Mus-S2	84.6	12.1		0.8	0.7		1.6	0.11	0.01	0.01	0.01	0.04	0.65	86.69	0.02	0.35	0.05	11.97	0.00	0.06	0.00
SD	*0.7*	*0.2*		*0.2*	*0.6*		*0.2*	*0.20*	*0.01*	*0.01*	*0.01*	*0.01*	*0.07*	*0.32*	*0.03*	*0.07*	*0.03*	*0.26*	*0.00*	*0.10*	*0.00*
Mus-S3	85.1	12.1		0.9	0.4		1.3	0.02	0.01	0.01	0.05	0.04	0.38	86.76	0.01	0.21	0.04	12.66	0.00	0.05	0.02
SD	*1.3*	*1.1*		*0.2*	*0.6*		*0.4*	*0.02*	*0.01*	*0.01*	*0.02*	*0.02*	*0.05*	*0.49*	*0.03*	*0.04*	*0.05*	*0.41*	*0.00*	*0.07*	*0.05*
Mus-Ax 6*	90.2	8.2		0.7	0.9		1.2	0.00	0.01	0.01	0.03	0.03	0.36	92.87	0.01	0.25	0.05	7.12	0.09	0.13	0.00
SD	*0.9*	*0.7*		*0.5*	*0.7*		*0.3*	*0.01*	*0.01*	*0.01*	*0.02*	*0.01*	*0.03*	*0.99*	*0.01*	*0.08*	*0.03*	*0.82*	*0.05*	*0.23*	*0.00*
Mus-Ax 7	84.5	12.2		0.4	1.4		1.4	0.01	0.01	0.01	0.08	0.03	0.52	86.60	0.00	0.38	0.03	12.25	0.00	0.08	0.04
SD	*0.3*	*0.3*		*0.3*	*0.6*		*0.3*	*0.01*	*0.01*	*0.01*	*0.02*	*0.02*	*0.03*	*0.43*	*0.00*	*0.05*	*0.02*	*0.49*	*0.00*	*0.13*	*0.10*
Mus-PC-1	96.2			1.5	2.4			0.01	0.01	0.01	0.25	0.02	0.01	99.67	0.04	0.06	0.11	0.06	0.02	0.05	0.00
SD	*0.9*			*0.5*	*1.2*			*0.01*	*0.01*	*0.01*	*0.03*	*0.02*	*0.01*	*0.30*	*0.04*	*0.04*	*0.03*	*0.03*	*0.03*	*0.04*	*0.00*
Mus-PC-2	97.3			1.4	1.3			0.02	0.01	0.01	0.01	0.01	0.01	99.55	0.01	0.54	0.10	0.03	0.03	0.01	0.00
SD	*0.6*			*0.4*	*0.7*			*0.01*	*0.01*	*0.01*	*0.01*	*0.01*	*0.01*	*0.44*	*0.01*	*0.12*	*0.10*	*0.02*	*0.02*	*0.01*	*0.00*
Mus-PC-3	96.9			1.5	1.6			0.02	0.01	0.01	0.01	0.01	0.00	99.96	0.00	0.09	0.31	0.00	0.01	0.04	0.00
SD	*0.6*			*0.2*	*0.5*			*0.02*	*0.01*	*0.01*	*0.01*	*0.01*	*0.01*	*0.76*	*0.00*	*0.05*	*0.13*	*0.01*	*0.01*	*0.05*	*0.00*
Mus-PC-6	95.2		3.0	1.8				0.00	0.01	0.01	2.34	0.01	0.03	98.26	0.01	0.04	0.03	0.02	0.00	0.03	0.00
SD	*0.3*		*0.0*	*1.8*				*0.01*	*0.01*	*0.01*	*0.47*	*0.01*	*0.02*	*0.50*	*0.03*	*0.04*	*0.03*	*0.02*	*0.01*	*0.03*	*0.00*
Mus-PC-7	97.3		1.0	1.7				0.00	0.00	0.01	0.45	0.02	0.21	99.19	0.01	0.04	0.01	0.01	0.00	0.05	0.00
SD	*0.7*		*0.2*	*0.7*				*0.01*	*0.01*	*0.01*	*0.04*	*0.01*	*0.03*	*0.34*	*0.02*	*0.04*	*0.01*	*0.01*	*0.00*	*0.07*	*0.00*
Mus-PC-8	95.9			1.8	2.2			0.01	0.00	0.01	0.16	0.01	0.03	99.29	0.28	0.02	0.06	0.03	0.02	0.09	0.00
SD	*1.0*			*0.1*	*0.9*			*0.01*	*0.00*	*0.01*	*0.03*	*0.01*	*0.02*	*0.41*	*0.05*	*0.03*	*0.05*	*0.02*	*0.02*	*0.06*	*0.00*

Table 3. Samples analysed and Pb isotope values divided by typology. 2SE is analytical uncertainties calculated as twice the individual in-run precision on the samples

	Sample	Type	206/204	±2SE	207/204	±2SE	208/204	±2SE
Bronze ingots	CER-PC-56	Plano-convex	18.022	0.001	15.649	0.001	38.247	0.004
	CER-PC-70	Plano-convex	18.235	0.001	15.667	0.001	38.503	0.003
	CER-PC-72	Plano-convex	17.905	0.001	15.642	0.001	38.126	0.004
	CER-PC-79	Plano-convex	18.062	0.001	15.655	0.002	38.314	0.005
	CER-PS-64	Parallel- surface	18.106	0.001	15.657	0.002	38.340	0.005
	CER-PS-82	Parallel- surface	17.914	0.002	15.645	0.002	38.142	0.005
	CER-PS-62	Parallel- surface	18.153	0.001	15.655	0.002	38.375	0.005
	CER-PS-77	Parallel- surface	17.906	0.002	15.641	0.002	38.117	0.006
	MUS-PC-2	Plano-convex	18.245	0.002	15.668	0.003	38.482	0.008
	MUS-PC-3	Plano-convex	18.222	0.001	15.664	0.001	38.458	0.005
	MUS-PC-7	Plano-convex	18.779	0.006	15.710	0.005	39.310	0.012
Axes	Cer-Ax	Winged-axe	18.168	0.002	15.664	0.002	38.380	0.005
	Mus-Ax6	Socketed axe	18.246	0.002	15.664	0.002	38.455	0.006
	Mus-Ax7	Socketed axe	18.218	0.002	15.663	0.002	38.421	0.005
Swords	MUS-S2	Flange-hilted sword, Cetona-type	18.197	0.001	15.663	0.002	38.417	0.005
	MUS-S3	Flange-hilted sword, Sacile-type	18.216	0.002	15.673	0.002	38.438	0.006