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The Chalcolithic metallurgical tradition of Northeast Iberia and its later influence: new analyses and a synthesis

La tradición metalúrgica calcolítica del nordeste ibérico y su influencia posterior: nuevos análisis y una síntesis

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Abstract: Our knowledge of Chalcolithic copper smelting practices across Iberia is extensive but lacks regional nuance. This prevents an assessment of regional trajectories that may depart from a general 'Iberian technological tradition', including their origins and diachronic evolution. This paper contributes to these questions by analysing 3rd millennium BC copper production residues from various sites in Northeast Iberia: Covas Cartanyà, del Buldó, Josefina d'Escornalbou, de l'Heura, and Balma del Duc. Using pXRF, OM, and SEM-EDS, we characterise the ores used and the metallurgical operations conducted (including smelting of copper carbonates, co-smelting of sulphidic and oxidic ores, and melting), and discuss them in relation to their contexts, contemporaneous materials and available lead isotopes data. We reveal Northeast Iberian metallurgy as a distinct and versatile tradition that selectively adopted elements from Southern Iberia and Southern France traditions, and trace the legacy of these idiosyncratic practices in copper and bronze making over two millennia.

Keywords: Chalcolithic; copper; smelting crucibles; slag; technological tradition.

Resumen: *Aunque nuestro conocimiento sobre la producción de cobre durante el Calcolítico en la península ibérica es amplio, carece de matices regionales. Esto impide entender las trayectorias regionales que podrían apartarse de una 'tradición tecnológica ibérica' generalizada, incluyendo sus orígenes y evolución diacrónica. En este artículo se contribuye a estas cuestiones analizando una serie de residuos de producción de cobre del III milenio a. C. de varios yacimientos del nordeste: Covas Cartanyà, del Buldó, Josefina d'Escornalbou, de l'Heura y Balma del Duc. Utilizando pXRF, MO y MEB-EDS, se han caracterizado los minerales utilizados y las operaciones realizadas (reducción de carbonatos de cobre, co-reducción de minerales de cobre oxídicos y sulfídicos y fusión de metal). Los resultados obtenidos se han contextualizado en relación con sus contextos arqueológicos, materiales contemporáneos y análisis previos de isótopos de plomo. Con todo esto, se defiende que la metalurgia del nordeste ibérico constituye una tradición aparte, caracterizada por su versatilidad, que adoptó selectivamente elementos de las tradiciones del sur peninsular y francés. El artículo concluye presentando cómo las características idiosincráticas calcolíticas influyen en la producción de cobre y bronce en el área a lo largo de más de dos milenios.*

Palabras clave: *Calcolítico; cobre; vasijas de reducción; escoria; tradición tecnológica.*

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1. INTRODUCTION

Iberia has attracted considerable research as one of the independent foci of metallurgical invention of the world, including compositional and microstructural analyses of slag and technical ceramics to understand the nature of copper-based extractive metallurgy (Ruiz-Taboada and Montero-Ruiz, 1999; Montero-Ruiz and Murillo-Barroso, 2017; Montero-Ruiz *et al.*, 2021). Notwithstanding some isolated studies scattered across the Peninsula (*e. g.*, Sesma

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Sesma, 1993; La Duc *et al.*, 2022) and the pan-Iberian approach of the *Proyecto de Arqueometalurgia* (Rovira Llorens and Montero-Ruiz, 2018), scientific studies have largely focused on the Southeast (*e. g.*, Carrilero Millán and Suárez Márquez, 1989; Keesmann *et al.*, 1991; Müller *et al.*, 2004; Escanilla, 2016; Montero-Ruiz and Murillo-Barroso, 2016; Murillo-Barroso *et al.,* 2017, 2020), the Southwest (*e. g.*, Hunt Ortiz, 2003; Sáez *et al.*, 2003; Nocete, 2006; Müller *et al.*, 2007; Müller and Monge Soares, 2008; Nocete *et al.*, 2008), and the Northeast (*e. g.*, Soriano Llopis, 2010; Rafel Fontanals *et al.*, 2017, 2018a; Montes-Landa *et al.*, 2021). Despite the focus on these areas, scarce attention has been paid to defining their specific characteristics (but see the studies by Obón Zuñiga *et al.*, 2020; Murillo-Barroso *et al.*, 2025a). Thus, results have been interpreted following generalising statements that characterise Iberian metallurgy as rudimentary, happening in simple infrastructures using open vessels as crucibles, and involving copper carbonates (Rovira and Montero-Ruiz, 2013). This leads to the impression of the existence of a universal copper-based tradition relevant for the whole Peninsula, and oversimplifies the great variety of socio-economic realities, environments, external influences and ways of living of early Iberian populations, which surely favoured distinct behavioural patterns to produce copper. We therefore need to add regional nuance to start drawing comparisons to other areas of the Peninsula, and to better understand how these early peculiarities affected subsequent developments.

To contribute to this collective task, we focus on Northeast Iberia and present a set of analyses of crucibles, slags and copper ores from Tarragona: Cova Cartanyà (Vilavert), Cova del Buldó (Montblanc), Balma del Duc (Montblanc), Cova Josefina d'Escornalbou (Ruidecanyes) and Cova de l'Heura (Ulldemolins) (Fig. 1). These sites, together with the crucibles from Bauma del Serrat del Pont (Tortellà, Girona; La Bauma henceforth) (Montes-Landa *et al.*, 2021), yield the earliest evidence of copper making in the Northeast so far, dated to the second half of the 3rd millennium BC. Characterising these materials using portable X-Ray Fluorescence (pXRF), Optical Microscopy (OM), and Scanning Electron Microscopy/Energy Dispersive Spectroscopy (SEM-EDS) allowed creating a detailed definition of the early copper-based technological tradition of the Northeast and its peculiarities. Furthermore, we compared our characterisation to previous data available from later Northeastern sites (Minferri (Juneda) and Vilars (Arbeca) both in Lleida, and Mas Castellar (Pontós) and Ullastret in Girona, Fig. 1) spanning from 2100 to 200 BC, a timeframe corresponding to the Early-Middle Bronze Age (EMBA) up until the Iberian Iron Age. These comparisons served to ascertain the weight of tradition –namely, how 3rd millennium BC features were retained for over two millennia.

Fig. 1. Map with the sites, mines, and mining districts mentioned in the text (base layer: Institut Cartogràfic i Geologic de Catalunya under a CC BY 4.0 license; minerals: Martín Cólliga *et al.*, 1999).

2. CHALCOLITHIC-EARLY BRONZE AGE METALLURGY IN THE NORTHEAST

To understand the origins of metallurgy in the Northeast, it is necessary to go back to the Late Neolithic-Chalcolithic transition at the turn of the 3rd millennium BC. At this time, the so-called Veraza groups populated the Northeast and Southern France. These mobile communities practiced agriculture and stockbreeding but did not smelt metal. Veraza people, however, had access to copper and gold items, including beads, ornamental small sheets, flat axes, and tongue daggers (Soriano Llopis, 2010). The earliest copper objects from the Northeast have been considered French imports given the typological parallels with items from the Paris Basin and Languedoc (Mille and Carozza, 2009) and because of their sulphidic inclusions and other characteristic impurities typical of French ores (Bourgarit *et al.*, 2003; Rovira, 2005; Soriano Llopis, 2010; Ambert *et al.*, 2014). Smelting technology only arrived in the Northeast with the Bell Beaker phenomenon in ~2500 BC. Bell Beaker peoples scattered nonuniformly across the Northeast, so they were contemporaneous to Veraza groups and generally shared the same lifestyle (Soriano Llopis, 2010).

Evidence of production residues and crucible sherds at this time spreads across the region (Fig. 1), including sites such as La Bauma, Cova Joan d'Os (Tartareu, Lleida), Cova del Frare (Matadepera, Barcelona), Cova Freda de Montserrat (Collbató, Barcelona), and Vapor Gorina (Sabadell, Barcelona) (synthesis in Martín Cólliga *et al.*, 1999; Soriano Llopis, 2010). Of special importance are the sites with metallurgical production remains at El Priorat county: Cova Josefina d'Escornalbou (Cova Josefina henceforth), Balma del Duc, Cova del Buldó, Cova Cartanyà, Cova de l'Heura, and Cova de Porta-Lloret (Siruana, Tarragona) (synthesis in Rafel Fontanals *et al.*, 2018a; Montes-Landa *et al.*, 2021). Absolute dating is not available for most contexts, but they are broadly ascribed to the late 3rd millennium BC, mostly based on artefact typology.

The finds related to extractive metallurgy include shallow open crucibles, small slag lumps, ore fragments, and casting residues. Previous pXRF analyses have related these materials to copper-based activities (references above). No furnaces have been found. Thus, the metallurgy conducted was likely rudimentary, based on open-crucible technology, and unconcerned with optimum efficiency. The small number of finds also suggests that it was likely conducted sporadically. These characteristics, however, fall within the general generalisations for the rest of Iberian metallurgy.

Burials were conducted in some of the caves where metallurgical debris has been found, but there is no compelling evidence to relate metallurgical activities to the burial contexts, except for Cova del Buldó (see below) (Martín Cólliga *et al.*, 1999; Soriano Llopis, 2010). Moreover, the characterisation of level II.5 of La Bauma and of other slightly later context at Cova de l'Heura as metal manufacturing settings (see below) (Vilaseca Anguera, 1952; Alcalde *et al.*, 1998; Rovira and Ambert, 2002; Rafel *et al.*, 2016; Montes-Landa *et al.*, 2021) rules out the interpretation of the rest of isolated crucibles found within caves as evidence of metallurgy being an esoteric activity linked to cave environments.

Although metallurgy had a generally domestic character, La Bauma stands out for having the largest archaeometallurgical assemblage of the region: 65 crucible sherds from a minimum of 19 vessels. This site has the earliest radiocarbon-dated contexts with extractive metallurgy evidence of the area linked to Bell Beaker pottery (2878-2479 cal BC), and metallurgy was still conducted at La Bauma until the EBA (Early Bronze Age). Previous analyses on seven crucibles revealed copper smelting activities using ores from Solana del Bepo (Ulldemolins, Tarragona), Turquesa (Cornudella del Montsant, Tarragona), Les Ferreres (Rocabruna, Girona), and a fourth unknown source (Fig. 1). Decorated and undecorated repurposed common pots were used for smelting, and these were made using the same clay, which contained both mineral and organic (plant) inclusions (Montes-Landa *et al*., 2021).

The hitherto available evidence from the Northeast also supports that the origins of extractive metallurgy were polymetallic. La Bauma is also the earliest Iberian bronze production site. This is supported by ED-XRF analyses on a Chalcolithic crucible (4.6 wt%Sn), melting residue (7.58 wt%Sn), and arrowhead (7.09 wt%Sn). The latter two finds have a Sn content very close to the perceptual threshold for Sn alloying (~6 wt%Sn) (Kuijpers, 2017; Radivojevic *et al.*, 2018). This opens the debate on the intentionality of this alloy through a potential natural alloying smelting process of copper-tin ores that are locally available (Alcalde *et al.*, 1998). Furthermore, at Cova de l'Heura, a lead bead typologically similar to Southern France specimens but isotopically compatible to the Molar-Bellmunt-Falset (MBF henceforth) mining district was identified (Montero, 2017). This supports that copper, lead and bronze were extracted in the region since early times.

This polymetallic nature differs from the Southern Iberian traditions (where early extractive metallurgy only concerns copper or arsenical copper) and is more consistent with the French tradition. Here, some analyses carried out on daggers also point towards an early adoption of bronze metallurgy (Guilaine and Cattin, 2015), and similar lead beads dated to the Late Neolithic and Chalcolithic are commonly found (Arnal *et al.*, 1979).

Southern Iberia and Southern France have independent, older copper-based metallurgical traditions. For Iberia, the oldest smelting crucible comes from Cerro Virtud (Cuevas del Almanzora, Almería), dated to the 5th millennium BC (Ruiz-Taboada and Montero-Ruiz, 1999). Importantly, no further production residues have been identified in the Peninsula until the beginning of the 3rd millennium BC. This gap, however, has been explained in relation to the social value of metal at this time (Montero-Ruiz *et al.*, 2021). In the case of Southern France, early metallurgical evidence dates back to the late 4th or early 3rd millennium BC at Cabrières (Ambert *et al.*, 2002; Laroche, 2019).

From an engineering point of view, the main differences between the early Southern Iberia and Southern France traditions are (1) the use of oxidic/carbonate *vs* sulphidic and oxidic ores, (2) the use of crucibles *vs* open pits for smelting, and (3) the use of limited *vs* extended annealing, respectively (Ruiz-Taboada and Montero-Ruiz, 1999; Ambert *et al.*, 2002, 2011, 2014; Rovira and Ambert, 2002; Bourgarit *et al.*, 2003; Rovira, 2005; Rovira and Montero-Ruiz, 2013; Montero-Ruiz and Murillo-Barroso, 2016; Murillo-Barroso and Montero-Ruiz, 2017; Rovira and Renzi, 2017; Montero-Ruiz *et al.*, 2021). As extractive technology in the Northeast develops in the late 3rd millennium BC, assessing the technological characteristics of the regional metallurgy can also help to contextualise the transmission of copper-based technology into this area.

3. MATERIALS AND CONTEXTS

3.1. Cova Cartanyà (Vilavert, Tarragona)

Cova Cartanyà is a cave with a small corridor to the right of its entrance, followed by a main chamber and a chasm. Pottery and animal bones were found at the corridor. The main chamber yielded pottery (including Bell Beaker pots), bone objects, faunal remains, stone tools, and sporadic human bones. The limited contextual information available prevents further specifics, but its excavator assigned a chronology between the Late Neolithic-Chalcolithic and the Late Bronze Age based on artefact typology (Vilaseca Anguera, 1926, 1973), and more recent research concurs (Miró, 1995; Martín Cólliga *et al.*, 1999; Rovira and Ambert, 2002). The materials recovered suggest that the cave may have been sporadically inhabited and that, despite the presence of human remains, it was not primarily used for burial.

The crucible analysed, held by the Museu d'Arqueologia Salvador Vilaseca (Reus), was not initially recognised during excavations, but later found during a revision of materials (Martín Cólliga *et al*., 1999). Although it could not be accessed for photographic documentation, Martín Cólliga *et al*. (1999) describe it as a hemispherical vessel, with a diameter of 20 cm. Previous lead isotopes analyses (LIA henceforth) on the crucible residue ruled out the Montsant and the MBF mining districts as metal sources (Fig. 1) (Montero, 2017; Rafel Fontanals *et al.*, 2018a).

3.2. Cova del Buldó (Montblanc, Tarragona)

Cova del Buldó is a cave with two entrances communicated by a corridor. Below the second entrance, a lower level can be accessed. The corridor and one of the lower-level chambers host communal inhumations. Finds from the corridor include at least five adults and three children. Only one burial (burial A) had associated perforated shells and some pottery. In contrast, the lower-level chambers yielded scattered human remains, including adult victims of the Carlist Wars and two infant mandibles attributed to prehistoric times. Pottery sherds were also identified together with a crucible fragment (Fig. 2). No Bell Beaker pots were found. Despite the lack of habitational evidence, it is possible that the cave was used for this purpose too (Vilaseca and Iglésies, 1929; Vilaseca Anguera, 1973; Martín Cólliga *et al.*, 1999; Soriano Llopis, 2010).

The crucible sampled is held at the Museu d'Arqueologia Salvador Vilaseca (Reus). It has a ~18-20 cm diameter. Previous LIA indicated that the copper processed in this crucible might come from Turquesa mine (Fig. 1). However, there was some disagreement regarding specific ratios (Soriano Llopis, 2010; Rafel *et al.*, 2016; Montero, 2017).

3.3. Balma del Duc (Montblanc, Tarragona)

Excavation of Balma del Duc was undertaken following a partial looting. The site is a rock shelter 3 m deep and 23 m wide. It had temporary occupations between the Late Neolithic-Chalcolithic and the Middle Bronze Age,

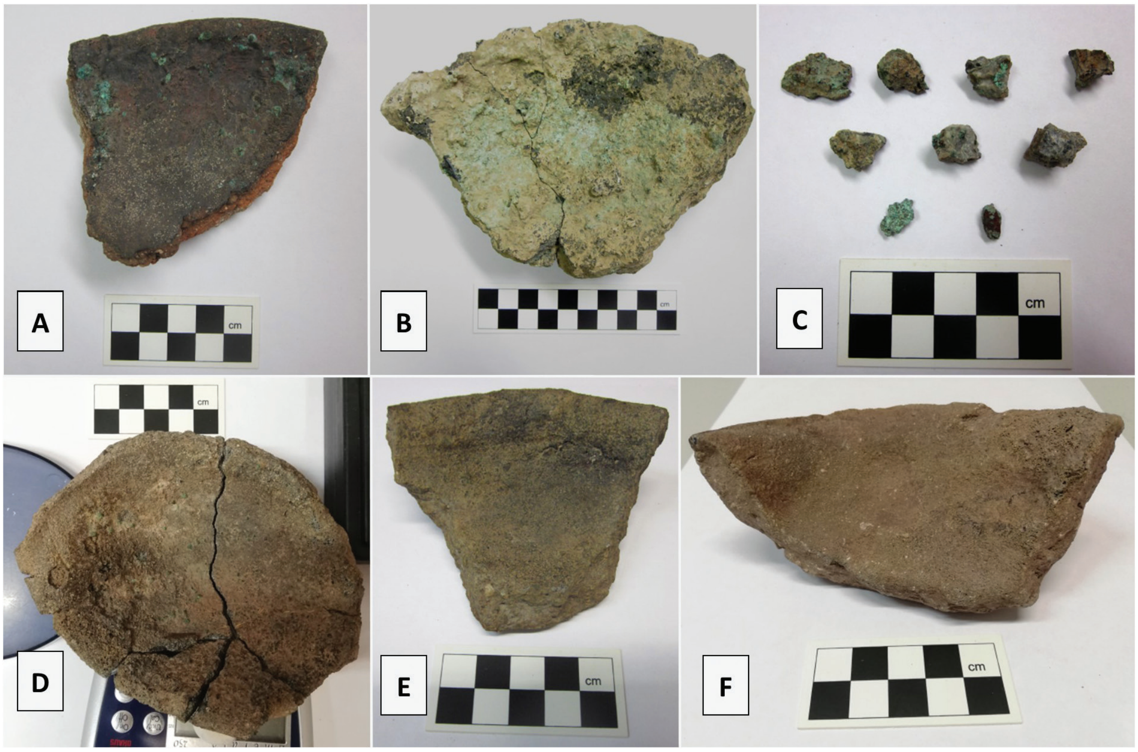

Fig. 2. A) Crucible CB-PA22253 from Cova del Buldó; B) Crucible CH4769 from Cova de l'Heura; C) Mineral remains from Cova de l'Heura; D) Crucible CJ3236 from Cova Josefina; E) Crucible CJ3237 from Cova Josefina; F) Crucible CJ3238 from Cova Josefina.

based on pottery from different periods (including Bell Beaker), and bone and flint tools. Finds dated to the Iberian Iron Age, Roman, and Medieval times were also recorded (Solé, 1982; Estrada Martín and Nadal Lorenzo, 1993). The excavators (Solé, 1982) do not mention the three crucible fragments belonging to two vessels later reported by Soriano Llopis (2010). Thus, they lack contextual information. One vessel is described as very similar to Cova del Buldó specimen, and the possibility that they belong to the same specimen has been suggested (Martín Cólliga *et al.*, 1999; Soriano Llopis, 2010). It was not possible to access these crucibles at the Museu-Arxiu de Montblanc in order to corroborate this.

Rovira and Ambert (2002) dated Balma del Duc crucibles to Chalcolithic times, but their poorly defined chrono-stratigraphic context(s) (Rafel *et al.*, 2016) has led other authors to link them to the EBA instead (Soriano Llopis, 2010). They have 10 cm and 9 cm diameters, respectively (Soriano Llopis, 2010). Previous LIA on one of them matched the copper minerals from Turquesa mine (Fig. 1) (Rafel *et al.*, 2014; Montero-Ruiz, 2018; Rafel Fontanals *et al.*, 2018b).

3.4. Cova Josefina d'Escornalbou (Ruidecanyes, Tarragona)

Cova Josefina, excavated in the 1920s, is a cave with no stratigraphic sequence. It has two chambers communicated by a corridor. The materials that have been recovered are dated from the Late Neolithic-Chalcolithic up to Visigothic times: flint tools, stone beads, perforated shells, and pots of different types, including Bell Beaker material. Three crucible sherds, a flat copper axe, two arrow heads (dated to the Bronze Age), and an awl mould fragment were recovered. Three stone hammers and one flat stone (a polisher?) might be also related to metallurgical work (Serra-Vilaró, 1925; Martín Cólliga *et al.*, 1999; Soriano Llopis, 2010; Rafel Fontanals and Soriano, 2017a). Together with the habitational and metallurgical evidence, remains of at least four individuals were identified at the bottom of the cave with no reported connections with the metallurgical finds (Serra-Vilaró, 1925).

The crucibles are stored at the Museu Diocesà de Tarragona (Fig. 2). CJ3236 has a ~12 cm diameter; CJ3238 and CJ3237 diameters could not be calculated. Although the pXRF analyses on CJ3237 showed enrichments in copper, arsenic, and lead comparable to CJ3238 and CJ3236 (Tab. SF.13 in Supplementary File 1), it was not sampled because it lacks substantial residues. Thus, a sample was taken only from the other two.

3.5. Cova de l'Heura (Ulldemolins, Tarragona)

Cova de l'Heura is a cave that consists of a corridor followed by a sub-rectangular area. In its deeper area, over 60 bodies were uncovered. Moreover, numerous flint tools and production residues point to the existence of a flint knapping station. Stone beads and ceramics were also reported. No Bell Beaker pots were recovered, but other items like the stone wrist-guard found are typical of these communities. A crucible sherd (~34 cm diameter) and other (s)melting residues/ore fragments support metallurgical activity too (Fig. 2). A bronze bead, a lead bead, an arsenical copper awl, and an elongated bronze item were also described.

The first habitational occupation, the flint knapping station, the lead bead, and the awl were dated to the Late Neolithic-Chalcolithic. During the early 3rd millennium BC, the collective burial took place, and the metallurgical activities and the bronze bead were attributed to the beginning of the Bronze Age (Vilaseca Anguera, 1952; Rovira and Ambert, 2002; Soriano Llopis, 2010; Rafel *et al.*, 2016).

LIA on the copper awl were consistent with Turquesa mine. The crucible charge was linked to either Barranc Fondo (Cornudella del Montsant, Tarragona) or Solana del Bepo mines (Fig. 1) although minor elements do not perfectly match. LIA on two mineral fragments (CH4403(1) and CH4403(10)) fit Barranc Fondo mine, although minor elements do not perfectly fit either. Previous pXRF analyses on these minerals indicated iron-rich copper with zinc, silver, tin, antimony and lead, and occasional bismuth impurities, which are not consistent with the known geochemistry of the deposit. The lead bead was linked to the MBF mining district (Fig. 1) (Montero-Ruiz *et al.*, 2012; Rafel *et al.*, 2014, 2016; Montero, 2017; Montero-Ruiz, 2018). These materials are kept at the Museu d'Arqueologia Salvador Vilaseca (Reus). A sample was taken from the crucible, as well as from four production residues/ore fragments, including the two previously sampled for LIA.

3.6. Other Chalcolithic sites not sampled

Although the materials already presented are the bulk of the available late 3rd millennium BC production remains in the Northeast, there are a few other objects not considered in this study that can also be generally ascribed to the same context. Tab. 1 (and Fig. 1 for location) summarises them for reference.

Site	Find(s)	Chronology	Why not sampled	References
Cova Joan d'Os	Amorphous mass (8.17 wt%Sn)	Unknown	Lack of clear dating	Colomines Roca, 1925; Pérez Arrondo and López de Calle Cámara, 1986; Equip Minferri, 1997; Rovira Port, 1998
Cova del Frare	Three crucible sherds	Chalcolithic-EBA	Lack of sufficient attached residue (Martín Cólliga, pers. comm., 2021)	Martín Cólliga, 1980, 2002; Martín *et al.*, 1981; Rovira *et al.*, 1997
Cova Freda de Montserrat	Plano-convex ingot (7.67 wt%Sn)	After LBA?	Lack of clear dating	Colomines Roca, 1925; Martín Cólliga *et al.*, 1999; Martín Cólliga, 2002; Montero-Ruiz *et al.*, 2010; Oms *et al.*, 2019
Vapor Gorina	Small casting residues	Chalcolithic	Access restrictions	Roig Buxó *et al.*, 2008; Montero, 2017
Cova de Porta Lloret	Ingot-like mass (2.85 wt%Sn)	Chalcolithic to Bronze Age	Lack of clear dating	Vilaseca, 1957; Rafel *et al.*, 2008, 2016; Soriano Llopis, 2010; Montero, 2017; Rafel Fontanals and Soriano, 2017b

Tab. 1. Other primary production finds from Northeast Iberia not sampled in this study.

4. METHODS

The materials here studied were initially photographed, weighted and measured. Then, pXRF analyses (Olympus Vanta M-series, 40 kV, 30 seconds acquisition time per beam, Geochem method run at 3 beams) were conducted on their irregular, unprepared surfaces to guide sampling. The resulting data is reported in wt% and rounded to two decimal places. Thus, '0.00' denotes potential presence of elements at the trace level, while 'BDL' refers to an element below the detection limits of the instrument. In any case, pXRF data must be considered qualitatively.

The 10 samples were cut using a Dremel with a diamond-impregnated disk, embedded in epoxy resin and ground and polished down to 3 µm using SiC paper and diamond paste. Optical microscopy was performed using a Keyence VHX-600 with both plane and cross-polarised light. After optical microscopy, the resin blocks were then C-coated to perform SEM-EDS analyses (Zeiss GeminiSEM 300, Ultim Max Silicon Drift EDS detector from Oxford Instruments, 20 kV, 8.5 working distance, 4.5 spot size, 5 process time, 700,000 cpa). A cobalt standard was used to calibrate the beam, and the resulting spectra were processed using Aztec. BSE images were also taken.

SEM-EDS compositions are given in wt%, reporting only concentrations 3σ above the background. Normalised results are provided, with the analytical totals indicated. Non-metallic phases are reported as conventional compounds with the oxygen calculated by stoichiometry, but oxygen in metallic phases is reported as measured. Bulk compositions are the mean of at least three different measurements of different areas at x100 magnification (1065 × 795 µm) comprising matrix, inclusions and porosity. When reporting the composition of the glassy and ceramic matrices, the numbers correspond to the mean of three different areas that avoided inclusions and porosity. In this case, a standardised area of analysis was not used because samples were highly heterogeneous. When calculating mean values, if an element is 'BDL' in one or several measurements, such value is substituted by a 0. The compositions reported for other metallic or mineral phases correspond to a single measurement. Area analyses were preferentially used, except for very small phases.

For samples that preserve both unaltered ceramic and a slag layer (*i.e.* Cova del Buldó, Balma de Duc, Cova Josefina and Cova de l'Heura crucible samples), ratio comparisons between the ceramic and the slag were used to distinguish chemical enrichment in the slag that might simply constitute molten ceramic, from other elements that derived from the charge. Assuming all Al in the slag comes from molten ceramic, the ratios of selected oxides to Al_2O_3 were compared to investigate chemical contributions to slag that might derive from ore or ash, as opposed to ceramic. When, for a given ratio, the difference between slag and ceramic is higher than 1, we interpret this enrichment as related to contributions from ore gangue minerals. More moderate enrichments below this threshold were attributed to charcoal ash. It is acknowledged that the use of these ratios only offers an approximation. When considering the potential impurities linked to copper ores in the slag, if the available evidence does not fully confirm the genetic link of a specific element to an ore, this element was classified as a 'possible impurity'.

A basalt standard from the Columbia River (BCR-2), a tin bronze standard (BCR-691 E) and a leaded bronze standard (BCR-691 D) were used to test the accuracy and precision of the SEM-EDS. As these samples were analysed in the same batch as other samples for a complementary study already published, a report of accuracy and precision can be found in Supplementary Information 10 of Montes-Landa *et al.* (2025).

5. RESULTS

This section discusses the analytical results. Full analytical data can be found in the Supplementary File 1.

5.1. Cova Cartanyà

CC1008 crucible slag is rich in Fe (59.9 wt%FeO in bulk). Its glassy matrix is a Ca-Fe aluminosilicate (15.6 wt%CaO; 26 wt%FeO). Ca enrichment is probably related to charcoal addition, as this element was not part of the ore fragments characterised (see below), but minor gangue contributions cannot be ruled out. Minor S, Cu, Zn, As and Ba detected in the glassy matrix (Tab. 2) were linked to the ore.

While Fe-Si and Mg-Fe-Si crystals occur in the glassy matrix, this is dominated by Fe oxide crystals (magnetite). These usually trap Cu-based prills in an arrangement consistent with Cu ore relics. These prills are metallic Cu (or Cu-Cl derived from corrosion), or Cu sulphides (up to 23.3 wt%S) with As ranging between BDL-9.1 wt%As (Fig. 3A). Some of the sulphides also contain minor Ag, and between 1.2-9.9 wt%Fe. Both Cu and Cu-S prills are roughly equally frequent. This characterisation is consistent with a sulphidic, Fe-rich Cu ore such as chalcopyrite ($CuFeS_2$), which may have contained some oxidic patches, minor As, Ag, Ba, and Zn impurities, and some Mg.

5.2. Cova del Buldó

CB-PA22253 crucible slag is richer in Si, S, Cl, Ca, Mg, Fe, Cu, and Zn than the associated ceramic (Tab. 2). The small Mg enrichment might be derived from charcoal ash contributions as opposed to gangue materials (Fig. 4A). Cl most likely results from post-depositional corrosion. Some Si might have entered the charge as gangue.

	ID	Description	Na_2O	MgO	Al_2O_3	SiO_2	P_2O_5	SO_3	Cl	K_2O	CaO	TiO_2	MnO	FeO	CuO	ZnO	As_2O_3	BaO	Analyt. Total
Cova Cartanyà	CC1088	Slag bulk	BDL	1.6	4.0	19.8	BDL	1.1	1.1	0.5	3.6	BDL	BDL	59.9	8.2	0.3	BDL	BDL	103.3
	CC1988	Slag matrix	0.1	1.5	10.3	40.6	0.5	0.4	BDL	2.9	15.6	0.1	0.2	26.0	0.6	0.5	0.5	0.3	108.3
Cova del Buldó	CB-PA22253	Slag bulk	0.2	2.9	11.2	32.7	BDL	0.3	0.3	1.3	7.2	0.6	0.1	37.9	5.2	0.1	BDL	BDL	100.2
	CB-PA22253	Alt. ceramic bulk	1.3	2.5	24.3	53.2	BDL	BDL	BDL	3.5	1.1	1.2	0.1	10.3	2.4	BDL	BDL	BDL	82.8
	CB-PA22253	Slag matrix	0.1	3.2	12.8	41.6	0.0	BDL	1.8	9.8	0.6	0.2	29.1	0.3	0.4	0.1	BDL	BDL	104.1
	CB-PA22253	Ceramic matrix (alt.)	2.5	0.6	20.5	63.6	BDL	0.1	5.3	3.2	0.1	BDL	2.1	2.0	BDL	2.5	BDL	BDL	106.4
Balma del Duc	BD-PA22251	Slag bulk	0.3	2.2	10.1	33.6	0.0	0.3	0.2	1.1	7.0	0.4	0.1	42.2	2.3	0.0	BDL	BDL	101.7
	BD-PA22251	Ceramic bulk	1.1	1.4	26.1	58.6	BDL	BDL	0.2	2.6	1.2	1.4	BDL	7.3	0.1	BDL	BDL	BDL	75.1
	BD-PA22251	Slag matrix	0.4	0.6	14.5	45.8	BDL	BDL	BDL	1.9	14.3	0.5	BDL	21.4	0.6	BDL	BDL	BDL	101.8
	BD-PA22251	Ceramic matrix	1.0	3.7	31.6	47.5	BDL	BDL	BDL	3.5	0.8	0.7	0.1	11.1	BDL	BDL	BDL	BDL	100.9
Cova Josefina	CJ3236	Slag bulk	1.1	3.4	16.7	46.6	2.6	0.1	0.3	3.8	13.3	0.6	2.2	7.2	1.6	BDL	0.1	0.5	83.4
	CJ3236	Ceramic bulk	0.9	1.2	21.8	59.0	BDL	BDL	0.4	6.0	1.3	0.5	1.0	8.0	BDL	BDL	BDL	BDL	62.9
	CJ3236	Slag matrix	0.9	5.7	17.0	39.6	4.5	BDL	BDL	2.8	16.3	0.6	2.4	7.7	1.6	BDL	0.4	0.4	106.6
	CJ3236	Ceramic matrix	0.4	5.4	25.6	45.5	0.1	BDL	BDL	2.9	1.5	0.6	0.3	17.6	BDL	BDL	BDL	BDL	109.3
	CJ3238	Cer. (inner) bulk	1.0	2.1	19.0	61.8	BDL	BDL	0.2	3.7	3.9	0.9	0.1	7.3	BDL	BDL	BDL	BDL	66.9
	CJ3238	Cer. (outer) bulk	0.9	1.8	22.1	60.8	BDL	0.1	0.5	4.2	3.1	0.8	BDL	5.8	BDL	BDL	BDL	BDL	59.9
	CJ3238	Slag matrix	0.4	4.6	18.5	51.7	BDL	BDL	0.0	2.3	6.7	1.5	0.2	13.1	1.0	BDL	BDL	BDL	106.8
	CJ3238	Ceramic matrix	0.7	5.4	26.8	48.7	0.1	BDL	0.1	3.8	2.6	1.2	0.3	10.4	BDL	BDL	BDL	BDL	101.7
Cova de l'Heura	CH4769	Slag bulk	BDL	1.3	10.4	26.5	BDL	7.0	0.0	1.2	7.6	0.3	0.2	26.2	18.9	0.3	BDL	0.1	99.6
	CH4769	Ceramic bulk	0.1	1.0	23.9	51.1	0.1	0.1	0.5	3.5	10.3	2.1	BDL	7.0	0.4	BDL	BDL	BDL	51.9
	CH4769	Glassy matrix	BDL	2.3	8.0	38.4	0.1	BDL	0.1	3.0	24.4	0.2	0.5	20.5	1.1	0.6	BDL	0.8	102.6
	CH4769	Ceramic matrix	0.2	1.3	34.3	46.3	0.2	BDL	0.3	4.1	8.4	BDL	BDL	4.8	0.1	BDL	BDL	BDL	99.7

Tab. 2. SEM-EDS results of bulk analyses (slag and ceramic), as well as of glassy and ceramic matrices compositions of all the crucibles analysed. BDL = below detection limits.

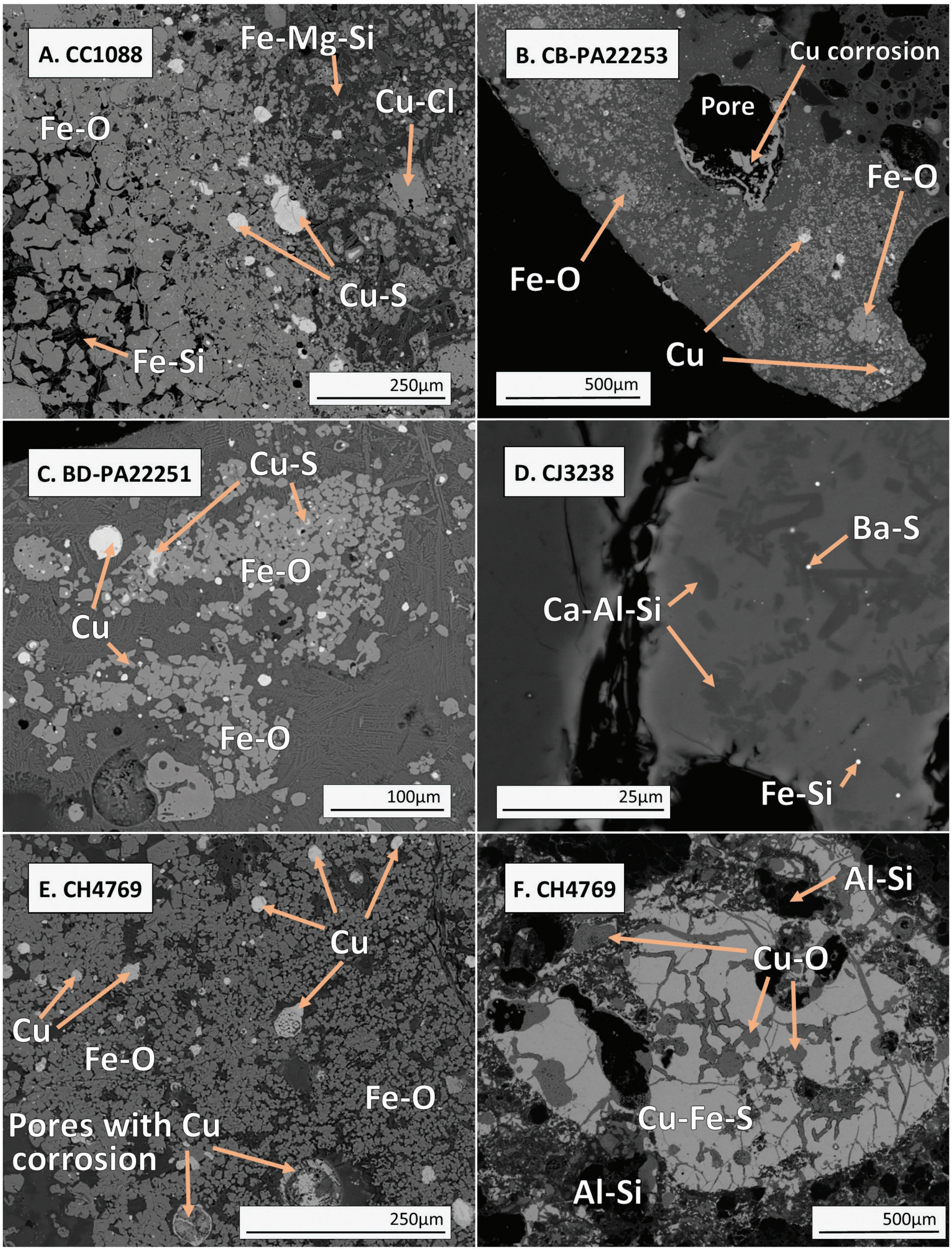

Fig. 3. BSE micrographs. A-C and E) Different clusters of Fe oxide crystals trapping Cu-based prills interpreted as Cu ore relics; D) A glassy patch with Ca-Al-Si crystals, and Ba-S and Fe-Si prills, the latter containing minor Cu; F) Sulphidic Cu ore relic consisting in a Cu-Fe sulphide phase with Cu oxide phases developing in the internal cracks.

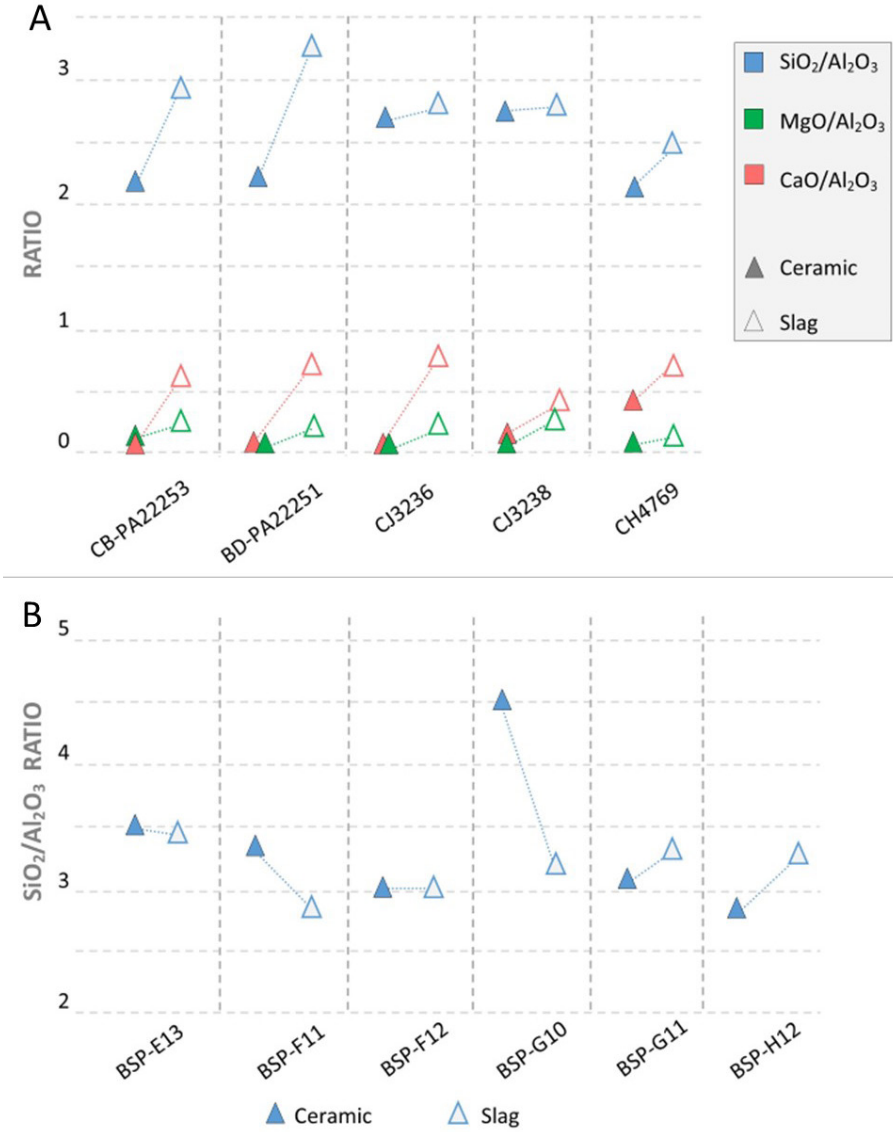

Fig. 4. A) Ratios relative to Al_2O_3 of different elements of the ceramic and slag bulk compositions of Cova del Buldó (CB), Balma del Duc (BD), Cova Josefina (CJ) and Cova de l'Heura (CH); B) SiO_2/Al_2O_3 ratios of the ceramic and slag bulk compositions of the crucibles from La Bauma.

CB-PA22253 glassy matrix is a Ca-Fe aluminosilicate (9.8 wt%CaO; 29.1 wt%FeO) with clusters of Fe oxide crystals (magnetite) trapping Cu prills, which are interpreted as Cu ore relics (Fig. 3B). The prills contain sporadic Ag, Sn, As, and/or Bi, and between 1.6-6.1 wt%Fe. No Cu-S phases were detected. A semi-dissolved Ca-Al-Si mineral was identified in the glassy matrix too, with no parallels in the altered ceramic analysed, so it might be residual gangue contributing to the Ca enrichment in the slag (0.6 CaO/Al_2O_3 and 0.04 CaO/Al_2O_3 for the ceramic and slag respectively, Fig. 4A). Some Fe-Al-Si crystals were also identified, likely resulting from the reaction of Fe gangue with the siliceous matrix. This evidence supports a Cu smelting operation of a Fe-rich Cu-ore bearing some Ca, and with minor Ag, Sn, As, Zn, S and Bi.

The ceramic fabric is non-calcareous (3.2 wt%CaO). No voids consistent with organic (plant) inclusions could be identified in the altered ceramics preserved. The mineral inclusions analysed are summarised in Tab. 3. Their small size (below 400 µm), and their inhomogeneous distribution suggest that they are natural clay inclusions.

	ID	Organic	Quartz	Ti	Fe-Ti	Fe-Ti-Si	Fe-Si	Fe-Al-Si	Fe-Mg-Al-Si	REE-P-Al-Si	Ca-Al-Si	Pyroxene	Zr-Si	K-feldspar	Max. size mineral (mm)	Max. size organic; plant (mm)
C. del Buldó	**CB-PA22253**	*	■		■	■	■			■					0.2	
B. del Duc	**BD-PA22251**	*	■	■									■		0.4	
C. Josefina	**CJ3236**	■	■	■					■				■	■	2	5
	CJ3238		■	■	■	■					■	■	■	■	3	
C. de l'Heura	**CH4769**	■	■	■				■				■	■	■	0.3	1.5

Tab. 3. Inclusions in technical ceramics from Cova del Buldó, Balma del Duc, Cova Josefina and Cova de l'Heura (no comprehensive analysis).
* Very little ceramic material prevented an accurate evaluation of presence/absence.

5.3. Balma del Duc

BD-PA22251 crucible slag, when compared to the ceramic proper, is richer in Mg, Si, S, Ca, Fe, Zn and Cu (Tab. 2). The small Mg increase (0.05 MgO/Al_2O_3 and 0.21 MgO/Al_2O_3 for the ceramic and slag respectively, Fig. 4A) might be consistent with fuel ash as opposed to gangue. However, the larger Ca increase (0.05 CaO/Al_2O_3 and 0.7 CaO/Al_2O_3 for the ceramic and slag respectively) is comparable to that of Cova del Buldó crucible, which was related to Ca-bearing gangue. However, as Ca was not found in the ore relics analysed (see below), it might alternatively represent charcoal ash. The ceramic preserved is non-calcareous (0.8 wt%CaO), so Ca-related ceramic contributions were discarded. Finally, the considerable Si enrichment observed (2.2 SiO_2/Al_2O_3 and 3.3 SiO_2/Al_2O_3 for the ceramic and slag respectively) might derive from siliceous gangue.

BD-PA22251 glassy matrix is a Ca-Fe aluminosilicate (14.3 wt%CaO; 21.4 wt%FeO). Fe-Si-Mg crystals populate the 'devitrified' glassy matrix together with big clusters of Fe oxide crystals (magnetite), Cu prills, Cu sulphides (with up to 27.6 wt%S), and Cu-Fe sulphates that were interpreted as Cu ore relics (Fig. 3C). The Cu-bearing phases sometimes have up to 1.2 wt%As. Similar prills also appear outside the magnetite clusters.

As in Cova Cartanyà, the ore smelted was possibly sulphidic, perhaps combined with Cu carbonates. The charge would also contain Zn and As impurities, and potentially Ca. Moreover, previous pXRF analyses (Soriano Llopis, 2010; Montero, 2017) detected 0.16 wt%Sn, so minor Sn could be present.

The ceramic preserved is considerably altered, preventing the observation of voids consistent with organic (plant) inclusions. Tab. 3 summarises the mineral inclusions found. Their varied shapes, inhomogeneous distribution, and maximum size (200 µm) support their natural presence in the clay.

5.4. Cova Josefina d'Escornalbau (Cova Josefina)

Surface pXRF analyses on CJ3236 and CJ3238 show enrichments in S, Cu, As, Sn, and Pb in the inner altered surface of both specimens. CJ3236 inner surface was also richer in P and Zn, and CJ3238 had more Ba (Tab. SF.13 in Supplementary File 1). This is consistent with SEM-EDS results.

CJ3236 has a thin red inner slag layer (~100-500 µm thick, Fig. 5A) with 1.6 wt%CuO. This layer is richer in Ca, Mg, S, Cu, As, and Ba than the ceramic (Tab. 2). Given its thinness, the Ca and Mg enrichments might be attributed to charcoal ash contributions instead of gangue components, from which more substantial slag might be expected. However, it must be noted that the Ca increase in the slag (0.06 CaO/Al_2O_3 and 0.8 CaO/Al_2O_3 for the ceramic and slag respectively, Fig. 4A) is consistent with that of the samples from Cova del Buldó and Balma del Duc related to Cu smelting operations instead. The Ca-Al-Si crystals analysed (anorthite) might represent the reaction of charcoal with the decomposing ceramic. The Fe-Mg-Al-Si crystals observed are most likely derived from ceramic inclusions (Tab. 3).

CJ3238 only has glassy patches made of altered ceramic. Its inner surface is richer in Ca, Fe, and Cu (1.0 wt%CuO, Tab. 2). Ca enrichment is minor, and likely related to fuel ash, and to Ca-bearing minerals identified in the ceramic paste (Tab. 3; Fig. 4A). A Cu-Fe-Al-Si prill (8.3 wt%Cu), Fe-Si prills with up to 1.5 wt%Cu, a Ba-S

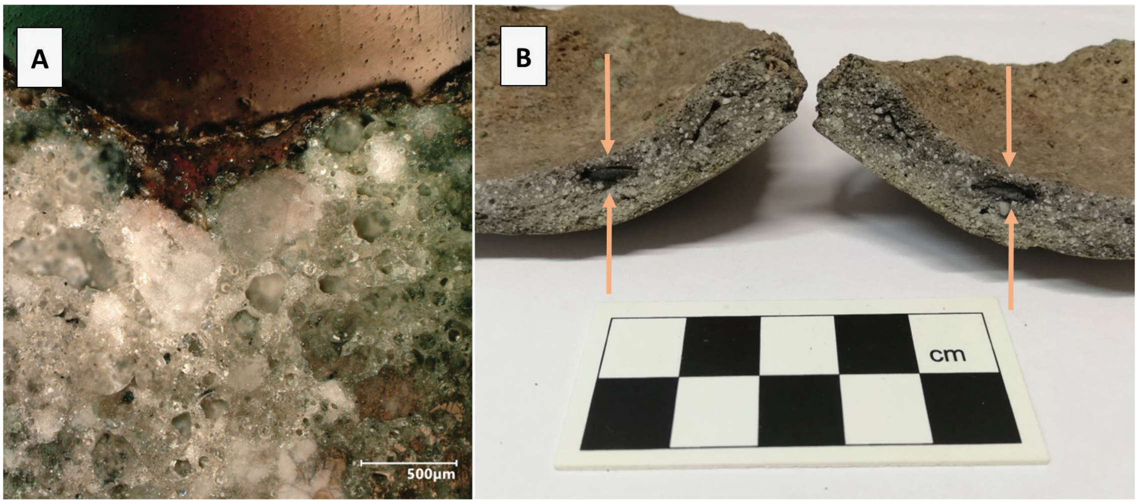

Fig. 5. A) XPL micrograph of CJ3236; slag layer (scale = 500 µm); B) CJ3236 sherds with relics of burnt organic (plant) inclusions (arrows).

prill, and a Ba-S oxidised phase attached to the inner surface of the crucible were also identified (Fig. 3D). Ca-Al-Si, Ca-Fe-Al-Si, and Fe-Al-Si (with minor Cu) crystals were also found. They are consistent with the decomposition of ceramic inclusions that absorbed some of the available Cu (Tab. 3).

The thin slag layers of both crucibles and the scarcity of mineral phases within them might indicate that they were involved in quick operations, perhaps Cu melting. There is no conclusive evidence to support smelting. SEM-EDS and pXRF analyses support the use of Cu containing S, As, Ba, Sn, Zn, and Pb impurities.

SEM-EDS analyses of the clay matrices (Tab. 2) show a broadly similar composition involving non-calcareous clays (below 3 wt%CaO). Similar mineral inclusions of up to 3000 µm (CJ3238) and 2000 µm (CJ3236) were documented. This potentially indicates the use of the same clay for both ceramics. The variability of mineral inclusions, inhomogeneous distribution, and varied sizes in both samples might support natural presence. CJ3236 preserved some organic (plant) inclusions (Fig. 5B) of about 5000 µm. No organic inclusions were observed in CJ3238.

5.5. Cova de l'Heura

Starting with the ore fragments analysed, all of them turned out to be dominated by chalcopyrite at different stages of thermal decomposition. CH4403(1) is a Cu-S mass with variable Fe (4.9-15.5 wt%Fe) (Tab. 4). It consists of Cu sulphide crystals (matte) with up to 26.9 wt%S, surrounded by a Cu oxide or a Cu-Fe-O (23.0 wt%Fe) phase (Fig. 6A). A Bi-Ca-O phase (61.1 wt%Bi, 8.1 wt%Ca) was sporadically observed too. CH4403(1) likely represents the initial thermochemical alteration of a sulphidic Cu mineral such as chalcopyrite. In the absence of SiO_2, the Fe reacted with some of the Cu available. It is possible that some Cu carbonate patches were also dispersed among the dominant hypothesised chalcopyrite ore, facilitating the Fe absorption. This ore also contained minor Bi and Ca.

CH4403(2) has high Fe (25.4 wt%FeO), Si (56.9 wt%SiO_2) and Cu (8.6 wt%CuO), and some S (3.5 wt%SO_3), and Ba (0.3 wt%BaO) in its bulk composition (Tab. 4). Zn (0.2 wt%ZnO) was detected in the glassy matrix (Tab. SF.24 in Supplementary File 1), made of a Cu-Fe silicate with up to 7.2 wt%SO_3. All these elements were linked to the Cu ore used. Within this glassy matrix, big patches of Cu-Fe sulphides (matte) with Ag and Zn impurities were identified. These were sometimes surrounded by Cu-Fe sulphates resulting from their corrosion, and Fe oxide crystals. Ba-S-O prills with minor Sr were also analysed. Moreover, well-delimited areas of a Cu-Fe aluminosilicate phase with 4.7 wt%SO_3 were identified close to these phases, probably resulting from the reaction of the Fe and Cu with the silicate gangue (Fig. 6B). This would be in agreement with the quartz relics seen in other areas. In the inner cracks of the quartz grains, Cu oxide corrosion phases accumulated, together with Fe-Al Si phases and Cu-Fe phases with up to 2.2 wt%SO_3 and occasional Pb (1.0 wt%PbO).

In sum, CH4403(2) is likely the resulting product of a Fe-bearing sulphidic Cu ore thermochemically altered, potentially chalcopyrite. It had silicate gangue, as well as minor Ag, Zn, Ba, Sr, and Pb impurities.

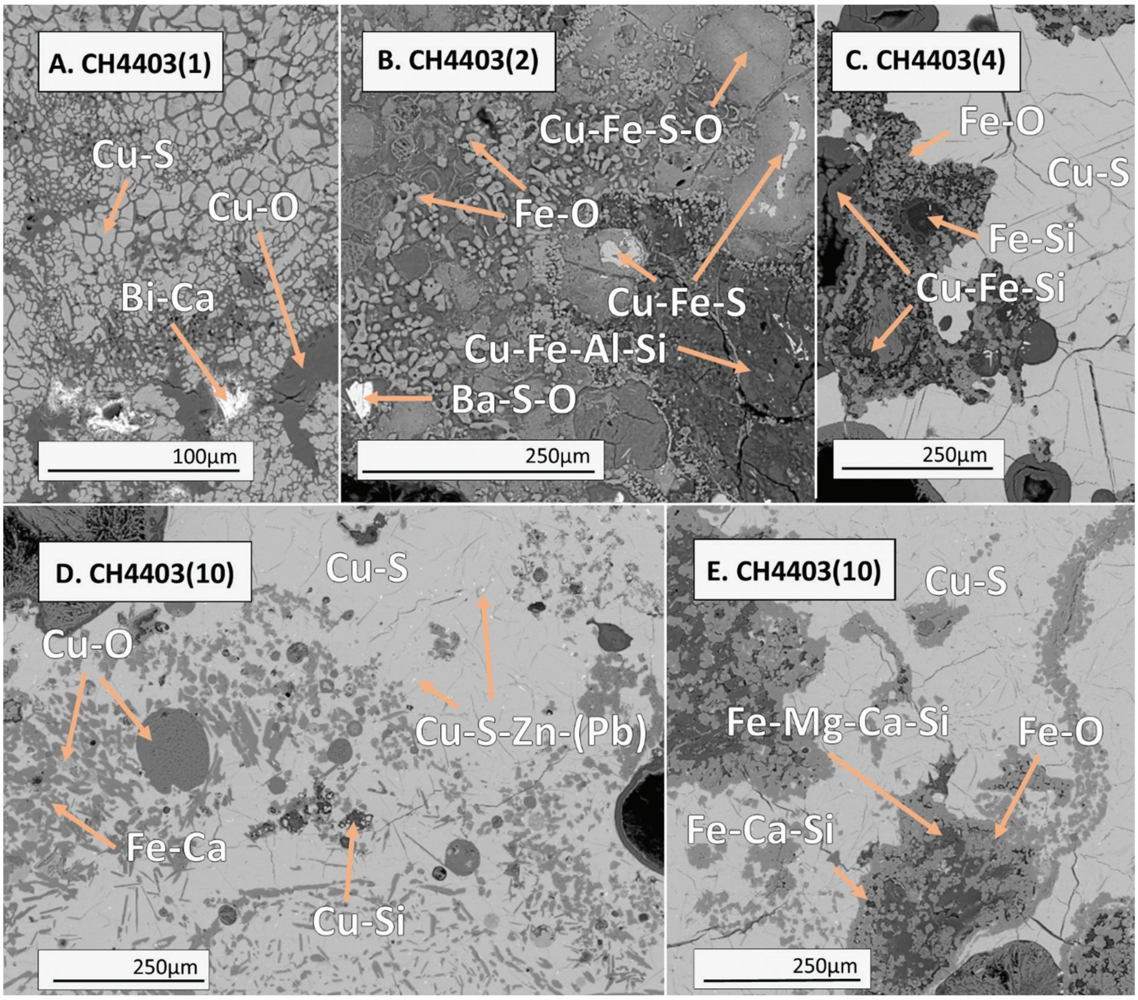

Fig. 6. A-E) BSE images of different ore remains from Cova de l'Heura. The microstructures are described in the main text.

Turning to CH4403(4), this is a Cu-Fe-S silicate (16.8 wt%SO_3; 23.9 wt%FeO; 39.3 wt%CuO) (Tab. 4) made of Cu sulphide patches (matte) with up to 23.4 wt%S and 1.2 wt%Fe. These are surrounded by clusters of Fe oxide crystals related to a Fe-Si phase and a Fe-Cu-Si phase with up to 2.3 wt%SO_3 (Fig. 6C). CH4403(4) was thus also characterised as a sulphidic Cu ore fragment, probably chalcopyrite, subjected to thermochemical alteration. This made the Fe to oxidise and react with the scarce available gangue and surrounding Cu.

ID	MgO	Al_2O_3	SiO_2	P_2O_5	SO_3	Cl	K_2O	CaO	TiO_2	MnO	FeO	CuO	ZnO	BaO	Analyt. Total
CH4403(2)	0.3	4.3	56.9	BDL	3.5	BDL	0.1	0.3	0.2	0.1	25.4	8.6	BDL	0.3	91.0
CH4403(4)*	0.8	1.1	11.9	0.5	16.8	BDL	BDL	5.0	BDL	0.1	23.9	39.3	BDL	0.6	112.2
CH4403(10)	0.3	1.8	BDL	BDL	26.3	BDL	0.0	2.2	BDL	BDL	25.6	43.2	0.5	BDL	129.9
	O	**Al**	**Si**	**S**	**Ti**	**Fe**	**Cu**								**Analyt. Total**
CH4403(1)	22.9	0.0	0.1	14.4	0.2	8.2	54.3								112.2

Tab. 4. SEM-EDS results of bulk compositions of ore fragments from Cova de l'Heura. All reported as oxides, but CH4403(1), which is reported as measured. * Mean of 2 analyses only; BDL = below detection limits.

To conclude with the altered minerals, CH4403(10) has a Cu sulphide matrix (matte, 24.5 wt%S) with up to 9.0 wt%Fe and minor Zn. This is populated by tiny Cu-Zn-S and Cu-Zn-Pb-S prills (the latter containing 1.2 wt%In). Clusters of Fe-Ca crystals associated to Cu oxide prills were observed within the matte matrix (Fig. 6D), as well as clusters of Fe and Fe-Ca-Mg-Si crystals and a Fe-Ca-Si phase with 0.6 wt%BaO (Fig. 6E). Within these clusters, Cu prills with minor Ag, Sn, and Pb were identified. Many were biphasic, with the secondary phase being a Cu-S-Pb alloy (7.8 wt%S; 8.4 wt%Pb). In other areas, Cu-Si phases within the matte matrix were close to Fe-Mg crystals.

CH4403(10) was interpreted as another sulphidic Fe-rich Cu ore fragment, perhaps chalcopyrite, subjected to initial thermochemical transformations. This allowed some of the available Ca, Mg and Si gangue to react with the Fe and Cu. It is important to note the initial formation of metallic Cu with very low S (in bulk) already in the shape of small prills. The mineral processed also had minor Ag, Pb, Zn, Sn, In, and Ba impurities.

While SEM-EDS analyses helped to establish the use of chalcopyrite, pXRF –with lower detection limits– indicates that the impurities of the ore fragments are broadly similar, which might be indicative of a similar source (Tab. SF.21 in Supplementary File 1). According to pXRF, all samples have Si, Fe, Ca, Zn, As, Sn, Ba, and Pb. CH4403(1) also has Ag, Zr and Sr, which should be added to the Bi found in sporadic Bi-Ca-O phases in this sample. The consistent presence of substantial amounts of S in all mineral samples indicates that these were not roasted –the latter usually producing a S-poor, red surface (Rostoker, 1975). Thus, these are likely remnants of a smelting operation in a relatively reducing atmosphere, as seems to be confirmed by their attested processing within crucible CH4769.

CH4769 is the crucible analysed. Its slag layer is enriched in Mg, Si, S, Ca, Fe, Cu, Zn, and Ba when compared to the preserved ceramic. As Mg and Ca enrichment is considerably small (0.04 MgO/Al_2O_3 and 0.12 MgO/Al_2O_3; 0.4 CaO/Al_2O_3 and 0.7 CaO/Al_2O_3 for the ceramic and slag respectively), it is more consistent with charcoal ash rather than gangue (Fig. 4A). Although the Si enrichment is not very high (2.1 SiO_2/Al_2O_3 and 2.5 SiO_2/Al_2O_3 for the ceramic and slag respectively), some siliceous materials could have entered as gangue. The rest of listed elements were interpreted as linked to the Cu ore used.

The glassy matrix is a Fe-Ca aluminosilicate (24.4 w%CaO; 20.5 wt%FeO) with Cu ore relic microstructures. Firstly, clusters of Fe oxide crystals (magnetite) trap numerous Cu-based prills (Fig. 3E). These are mostly pure Cu with up to 0.2 wt%S, and in some occasions, they are Cu sulphides with up to 19.9 wt%S. Importantly, solid-state smelting microstructures have been identified within these clusters, which supports their interpretation as Cu ore relics (Montes-Landa *et al.*, 2024). Based on the low S of the trapped prills, these Fe-bearing Cu ore relics are mostly of oxidic nature. S was only a minor impurity.

Secondly, Cu-Fe sulphide masses (matte) with up to 1.8 wt%Zn were identified too. They present internal Cu oxide masses and cracks also filled up by Cu oxide phases. The Cu oxide masses probably represent the post-depositional corrosion of metallic Cu (comparison in Hauptmann, 2011, fig.19.13). These Cu-Fe sulphide masses are usually surrounded by (Cu)-Fe-Ca-Al-Si crystals with sporadic Ba and Pb impurities probably related to the ore (Fig. 3F). These masses were interpreted as sulphidic Cu ore relics, likely chalcopyrite. It is possible that it was mixed with some Cu carbonate patches that facilitated the development of metallic Cu inside the matte matrix.

The use of sulphidic ores is also confirmed by independent Cu sulphide prills observed under OM and analysed by SEM-EDS. This evidence links CH4769 to the chalcopyrite ore fragments already analysed. Moreover, pXRF on the inner surface of this crucible also detected over 0.02 wt% of Sn and Ag which fits the impurities in the altered chalcopyrite samples above. Although As was not detected in the crucible slag, its volatility might explain its absence in this sample.

Finally, semi-dissolved quartz and pyroxenes were detected in the slag, but these are consistent with other ceramic mineral inclusions (Tab. 3). Their maximum size (250 µm) and their inhomogeneous distribution point to their natural presence in the clay. Elongated voids compatible with burnt organic (plant) inclusions of up to 1500 µm were observed. Interestingly, this crucible presents a calcareous fabric (8.4 wt%CaO in ceramic matrix, and 10.3 wt%CaO in the bulk ceramic composition).

The identification of both oxidic and sulphidic copper minerals potentially links this crucible to a co-smelting operation to produce copper, whereby copper is reduced by direct oxide-sulphide interaction without an intermediate matte stage. This can happen incidentally (through the selection of naturally occurring rocks that exhibit distinct colours and contain both minerals) or by mixing separate ores (also distinguishable by colour) in the same crucible. Whatever the case, the extraction of copper out of this co-smelting operation likely happened in a single step, as proposed by Rostoker *et al.* (1989).

6. DISCUSSION

In this section, the new data will be discussed together with previously analysed Chalcolithic metallurgical ceramics from La Bauma (Montes-Landa *et al.*, 2021), to identify the main traits of the early copper-based technological tradition of the Northeast. Thereafter, such characteristics will be tracked in time in later assemblages (Montes-Landa *et al.*, 2025) dated between 2100-200 BC (Minferri, Vilars, Mas Castellar and Ullastret, Fig. 1).

6.1. The Chalcolithic technological tradition

6.1.1. Technical ceramics

The diameters of the crucibles studied range between 9 and 34 cm, which are comparable to other common pots. Their ceramic pastes combine poorly sorted mineral inclusions and some burnt-out organic (plant) materials (Tab. 3). Mineral inclusions are of varied size and have a low melting point (*e. g.*, Fe minerals and feldspars), suggesting that no specific treatment of the clay or independent addition of temper was conducted. All these features are consistent with previously analysed materials from La Bauma (Montes-Landa *et al.*, 2021).

The clay used was non-calcareous (below 3.5 wt%CaO) except for CH4769 (8.4 wt%CaO), which is slightly richer in alkalis (total sum 14 wt%) and Al (34.2 wt%Al_2O_3) (Tab. 2). In general, when the alkali and the alkali earth oxides are considered against the Al_2O_3 of these fabrics, results suggest vessels with a high refractoriness, especially given the considerably high Al presence (Fig. 7) (Freestone, 1989; Martinón-Torres and Rehren, 2014). Nevertheless, no evidence was found that this type of clay was deliberately selected for metallurgical purposes.

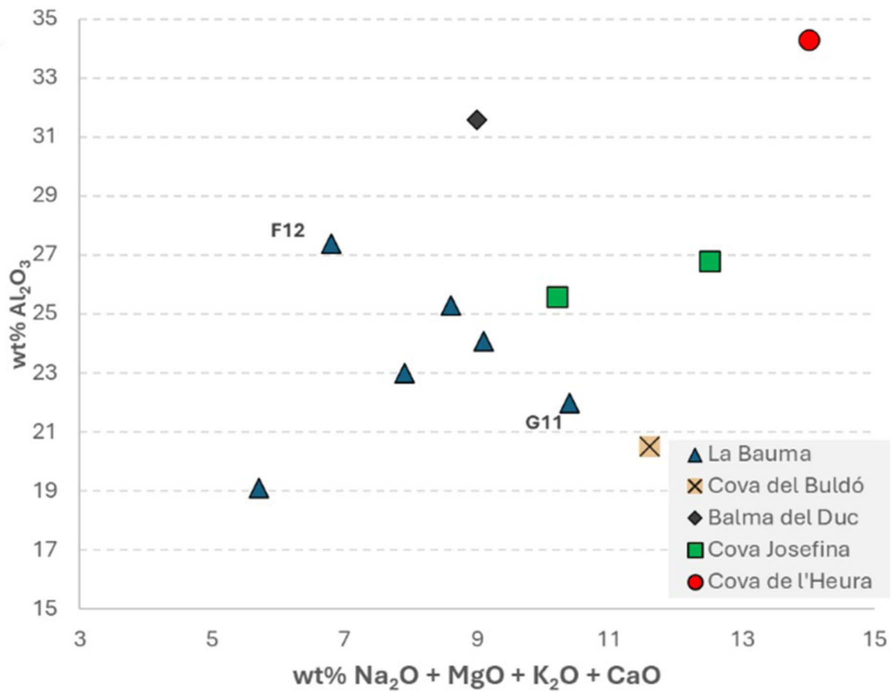

Fig. 7. Al_2O_3 and alkalis content of the Chalcolithic-EBA crucible fabrics. G11 and F12 are the decorated crucibles from La Bauma.

To assess if Chalcolithic crucibles were purpose-made ceramics or reused common pots, it can be said that in the two case studies where crucibles and common pots were analysed petrographically (La Bauma and Cova del Frare), the results showed no significant differences between fabrics (Clop García, 2000; Montes-Landa *et al.*, 2021). Coarse tempering of crucibles was however reported for La Bauma (Clop García, 2000), which might denote intentional selection to improve toughness and thermal shock resistance (Martinón-Torres and Rehren, 2014). The other site with more than one crucible analysed (n = 2) is Cova Josefina. Ceramic paste analyses suggest a similar clay matrix for both (Tab. 2). The mineral inclusions and organic (plant) inclusions are also consistent between specimens (Tab. 3). Because both crucibles –and the third one not sampled– are typologically different but have parallels in other common pots, it is likely that these were repurposed vessels too.

The presence of organic (plant) inclusions in many of the analysed crucibles is significant (Tab. 3) because these are not common in Southern Iberian crucibles (Hook *et al.*, 1991; Murillo-Barroso *et al.*, 2017; Pino Curbelo *et al.*, 2019; Mongiatti and Montero-Ruiz, 2020) but can be found in the Chalcolithic crucibles of La Capitelle du Broum (Cabrières, France) (Ambert *et al.*, 2011). Thus, this points to a distinct feature of the Northeast Iberian tradition. However, it is uncertain if the addition of organic matter occurred intentionally, especially if considering that these were likely repurposed pots.

Interestingly, the previously studied EMBA crucible assemblage from Minferri (2100-1650 BC) (Montes-Landa, 2023; Montes-Landa *et al.*, 2025), which is slightly later than these Chalcolithic crucibles, presents more abundant organic (plant) inclusions than the early ceramics analysed here. While the scarcity of organic (plant) inclusions detected in the early crucibles might be affected by the thermal alteration, in the case of Minferri it seems clear that they were deliberately added. If this was the case, they were likely added to both technical and common ceramics, as most of the crucibles analysed from Minferri seem repurposed common pots too. Therefore, it is unclear if the beneficial properties of organic tempering metallurgical vessels (*i.e.* keeping the heat, preventing crucible collapse) were consciously recognised (Martinón-Torres and Rehren, 2014). Furthermore, beyond the Northeast, organic (plant) tempering also appears in crucibles of the Bell Beaker horizon at Lugar Viejo III (Zaragoza), and in other EBA crucibles from the upper Ebro basin: El Abejar I (Navarra), Siete Cabezos, and Moncín (both in Zaragoza), among others (Fig. 1) (Harrison *et al.*, 1990; Sesma Sesma, 1993; Harrison, 1994; La Duc *et al.*, 2022). By the MBA it has been reported in sites in Southern Iberia such as Peñalosa (Jaén, 1750-1550 cal BC) (Moreno Onorato *et al.*, 2010). There, organic (plant) tempering has been associated to crucible manufacture, and it is absent in other common pots studied (Vico Triguero, 2021). This could mean that this feature spread from France into Northeast Iberia, and from there into other parts of Iberia during the Bronze Age.

6.1.2. Slag and ore fragments

Four copper smelting residues, four chalcopyrite fragments, and two potential copper melting operations were identified. Tab. 5 summarises the impurities of the ores/metal used, and the available LIA on production residues (Rafel *et al.*, 2016; Montero, 2017; Montero-Ruiz, 2018).

The iron-rich smelting sample from Cova del Buldó and the sulphidic ore smelted at Balma del Duc were linked to Turquesa mine. Chalcopyrite has been found there, together with malachite, azurite, libethenite ($Cu_2(PO_4)(OH)$), goethite ($FeO(OH)$), covellite (CuS), small amounts of quartz, and crandallite ($CaAl_3(PO_4)(PO_3OH)(OH)_6$) (Montero-Ruiz, 2018). These minerals broadly fit the technological characterisation proposed.

Cova Cartanyà and Cova de l'Heura samples also involved operations with chalcopyrite. LIA on Cova Cartanyà residue does not fit the Montsant or the MBF outcrops. The ore fragments from Cova de l'Heura were consistent with Barranc Fondo mine, although minor elements do not perfectly match. Chalcopyrite has been identified at Barranc Fondo mine (Montero, 2017), so this provenance remains plausible. Equally, LIA on Cova de l'Heura crucible slag fits either Barranc Fondo or Solana del Bepo mines, as both isotopic fields overlap. Importantly, at Solana del Bepo, malachite, azurite, iron oxides, and quartzes were reported together (Rafel *et al.*, 2019), which perfectly fit the microstructure of the oxidic ore observed at Cova de l'Heura crucible slag. Moreover, sulphidic minerals (which are also present in this crucible slag) have been reported both at Solana del Bepo and Barranc Fondo (Montero, 2017). Thus, the identified co-smelting operation might have occurred incidentally (if naturally occurring oxidic and sulphidic patches coexisting at these mines were used) or alternatively, by consciously mixing oxidic and sulphidic minerals. The first option seems more plausible.

Finally, the two Cova Josefina samples might be consistent with a melting operation involving copper with minor sulphur, arsenic, tin lead, and sporadic zinc and barium. No LIA was conducted.

To complete the characterisation of Chalcolithic samples, two previously analysed samples from La Bauma were consistent with smelting Ca-poor ores, and another four samples associated to smelting of Ca-rich ores. LIA linked these materials to Les Ferreres mining district, Turquesa and Solana del Bepo mines, and to a fourth unknown source that is likely within the region because it follows the trend observed for all the metallurgical remains analysed so far in the area (Montes-Landa *et al.*, 2021). A potential fifth copper ore used at La Bauma could be the polymetallic copper-tin ore reported by Alcalde *et al.* (1998), which could be located within Les Ferreres mining district too.

ID	Characterisation	LIA	Mg	Si	P	S	Ca	Fe	Zn	As	Sr	Ag	In	Sn	Ba	Pb	Bi
CC-1008	Smelting	Not from MBF and Montsant															
CB-PA22253		Turquesa															
BD-PA22251		Turquesa												x			
CH4403(1)	Chalcopyrite fragments – smelting	Barranc Fondo															
CH4403(2)																	
CH4403(4)																	
CH4403(10)		Barranc Fondo															
CH4769	Co-smelting of Cu ores	Barranc Fondo/Solana del Bepo															
CJ3238	Melting?																
CJ3236																	

Tab. 5. Characterisation of Chalcolithic-EBA samples, impurities (qualitative) interpreted as related to the copper/copper ore used (SEM-EDS and pXRF data) and potential provenance. Light grey = potential impurity; x = according to other published pXRF analyses.

These results illustrate that both local minerals (Les Ferreres mine) and outcrops ~200 km away (Solana del Bepo and Turquesa mines) were exploited contemporaneously at La Bauma. This contrasts with the rest of sites analysed here, which are only ~25 km away from the matching mineral outcrops (Fig. 1). This difference in the procurement strategy might be linked to the setting and ways of living of these communities, with La Bauma people linked to cattle herding and seasonal mobility to the upper lands (Alcalde *et al.*, 1998) and the other sites to areas with better access to agricultural lands and abundant mineral resources.

The use of a minimum of four ore sources at La Bauma within a single, small setting suggests a complex and versatile resource acquisition strategy. This is in accordance with the highly mobile Chalcolithic-EBA communities, and their lack of a centralised production organisation, although similar versatile strategies have been identified in other more centralised and sedentary settlements in Iberia (*e. g.*, Murillo-Barroso *et al.*, 2017). This use of different ores since the early stages of metallurgy shows a remarkable awareness of regional resources and the development of an opportunistic network of connections with other regional groups that might have supplied some of these minerals. This is especially possible for the mineral sources located at the Montsant mining area (Fig. 1). Other nearby sources such as those at Les Ferreres mining district could have been directly exploited by those at La Bauma. This flexible and adaptive approach to resource acquisition would have ensured access to enough raw materials.

All in all, these analyses attest to the spread of smelting technology across the Northeast during the late 3rd millennium BC. The processing of oxidic ores attested at La Bauma is coherent with the Southern Iberian technological tradition. However, the processing of sulphidic ores in a co-smelting with oxidic ones at Cova de l'Heura and probably also at Cova Cartanyà and Balma del Duc is typical of the technological tradition of Southern France. Smelting sulphidic copper ores is usually complex, as it requires several steps: roasting, smelting, and several matte purification operations (Rostoker, 1975). However, this is an unlikely scenario for the generally rudimentary metallurgy of these early moments. Rostoker *et al.* (1989) already demonstrated that it is possible to produce metallic copper in one-step by co-smelting mixed copper sulphide and oxidic ores. Instances of this process have been documented archaeologically, in many cases occurring incidentally (*e. g.*, Hauptmann *et al.*, 2003). Within Iberia, excluding instances in which oxidic copper ores with sulphidic impurities have been characterised (*e. g.*, Siret and Siret, 1890; San Nicolás del Toro, 1988; Müller *et al.*, 2004; Escanilla, 2016; Murillo-Barroso *et al.*, 2017, 2025a), only sporadic parallels of sulphidic ores use exist at other Chalcolithic sites such as La Ceñuela (Murcia), Cueva del Cañaveralejo (Córdoba) and Valencina de la Concepción (Seville). These examples have been interpreted as unintentional, isolated exceptions dictated by the local geology (Rovira, 2002; Nocete *et al.*, 2008; La Duc *et al.*, 2022). Against these scattered parallels, the three contemporaneous instances of sulphidic ore use in Northeast Iberia here identified cannot be seen as isolated cases, especially considering the nearby Southern France tradition. Analyses of slag from early Southern France Chalcolithic metallurgy indicate that co-smelting of sulphidic and oxidic ores was facilitated by the local mineralogy, which combined both types of minerals (Carozza *et al.*, 1997; Bourgarit and Mille, 2001, 2005; Prange and Ambert, 2005; Burger *et al.*, 2011). Thus, the practice of co-smelting at both sides of the Pyrenees reinforces the hypothesis about a transmission of the metallurgical know-how from Southern France. This hypothesis was previously supported by (1) the close chronologies between metallurgical finds in both regions, and (2) the general absence of early crucibles and slag in the regions separating Southern and Northeast Iberia (Soriano Llopis, 2010).

Furthermore, we should remember that the early exploitation of polymetallic ores for bronze metallurgy at La Bauma (Alcalde *et al.*, 1998; Montes-Landa *et al.*, 2021), might also have parallels in Early Bronze items in France (Guilaine and Cattin, 2015), and that the lead bead found at Cova de l'Heura, the only Chalcolithic lead object from Iberia, morphologically looks like contemporary French specimens, even though LIA point to local production (Rafel *et al.*, 2016). This, added to the new evidence on sulphidic ores processing, supports that the adopted features from Southern France include smelting of different ores (copper, copper-tin and lead) and the recognition of the advantageous use of mixed ore sources (sulphidic-oxidic and copper-tin).

It is important to note that oxidic copper ores are widely available across Northeast Iberia, including in mines where sulphidic minerals are present (Martín Cólliga *et al.*, 1999). In fact, the oxidic ore from Cova del Buldó and the sulphidic ore from Balma del Duc might have come from the same mine (Turquesa). Therefore, the apparently generalised use of sulphidic ores (attested in three of the sites sampled) to produce copper, as opposed to preferentially selecting oxidic minerals, should be discussed.

It could be argued that sulphidic minerals leave more slag behind, and therefore, their use is more archaeologically visible. However, as co-smelting of copper ores was frequently conducted in Southern France, it is possible that during the process of technological transmission, co-smelting was introduced as a suitable process. The people of Northeast Iberia could have learnt to recognise both oxidic and sulphidic ores as good-enough to produce copper since early times. As a result, metallurgists might not have had a strong preference for oxidic minerals. Instead, they adapted the recipe used (direct smelting or co-smelting) to the type of ores available.

Copper carbonates (green or blue) and chalcopyrite (usually known for its brass-yellow colour but often greenish grey/black when weathered, see Haldar (2020) and fig.1.9 in Haldar (2017)) are easily distinguishable by eye. Accordingly, a naturally mixed ore would exhibit patches of mixed material, thereby enabling, to a certain extent, the recognition that two different substances were being introduced together into the crucible. This organic learning process may have played a role in the early introduction of co-smelting (copper ore + cassiterite) to produce bronze in the area (Montes-Landa *et al.*, 2025). At this point, however, oxidic ores needed to be preferentially selected.

Whatever the case, not every feature of the Chalcolithic metallurgical tradition of Northeast Iberia can be linked to Southern France. The use of smelting crucibles in the Northeast links the area to the rest of the Peninsula too, where from Late Neolithic (Cerro Virtud) and Chalcolithic times (Rovira and Ambert, 2002; Montero-Ruiz and Murillo-Barroso, 2016), crucible smelting technology was the favoured way of extracting metal. Although some pyrometallurgical infrastructures have been reported at Valencina de la Concepción and Cabezo Juré (Huelva) (Sáez *et al.*, 2003; Nocete *et al.*, 2008) these have been questioned by Rovira Llorens (2016) and Gauss (2013). This contrasts with the origins of metallurgy in Southern France, where the initial steps of metallurgical development at Cabrières in the early 3rd millennium BC (Roque-Fenetres, La Capitelle de Broum) or the more recent site of Le Planet (2800-2500 BC) favoured pit smelting (Rovira and Ambert, 2002; Bourgarit and Mille, 2005; Costa *et al*., 2021). The introduction of crucible smelting technology in Southern France only occurred in the second half of the 3rd millennium BC (Carozza *et al.*, 1997; Mille and Bourgarit, 1998; Rovira and Ambert, 2002; Labaune, 2010; Gandois *et al.*, 2020).

Thus, the Northeast emerges as a bridge between both traditions during Chalcolithic times combining features developed in both areas to adapt them to their necessities and possibilities. This shows a high permeability to innovations and a critical selection of them.

Furthermore, the apparent dominance of smelting evidence over recycling in the Northeast might suggest good knowledge of the available resources that favoured active production. This behavioural pattern was sustainable because of the wide availability of copper ore deposits in the Northeast (Fig. 1) (Martín Cólliga *et al.*, 1999). Thus, the two melting operations identified in Cova Josefina samples might represent the remelting stage of the metallic copper prills obtained from a previous smelting operation, a feature typical of early Iberian metallurgy (Rovira, 2005; Rovira and Montero-Ruiz, 2013; Murillo-Barroso and Montero-Ruiz, 2017; Rovira and Renzi, 2017). The other crucible from Cova Josefina and the three 'clean' crucible specimens found at Cova del Frare might also indicate that they were involved in similar melting or sporadic recycling activities.

To conclude with this section, it is necessary to point out that only at Cova de l'Heura and La Bauma more than three samples were analysed. However, although several small ore fragments/production residues were recovered from Cova de l'Heura, only one crucible sherd was found. Given the small nature of the ore fragments, it is possible that they represent one or a few operations, as opposed to the minimum of 19 crucibles/operations from La Bauma. Thus, Cova de l'Heura assemblage can be compared to the rest of Northeast Iberian sites, where one to three crucibles have been recovered. This would also explain why at Cova de l'Heura just one or perhaps two ore sources were identified, as opposed to the at least four (but possibly five) ores used at La Bauma (see above).

The uniqueness of La Bauma should also be related to the peculiar context it represents. At the other Northeast Iberian sites, production residues are linked to temporary occupations, and only at Cova del Buldó, to a funerary context. Most of these sites were excavated in the 1920s with rudimentary recording strategies and lack clear stratigraphy. La Bauma excavations were conducted in the early 1990s, and the site was stratified, probably reflecting a more frequent use of space. Thus, smelting operations at La Bauma were developed in different phases, including habitational and production contexts. In the latter, the greater amount of crucible sherds recovered indicates a larger production. Given the similarity in the processing of ores in metallurgical and domestic settings, it can be concluded that the brief change in the function of the site only implied an increase in metal production to meet a time-specific higher demand. This was also true for the rest of sites analysed.

Hence, despite the peculiarities of La Bauma, metal smelting was predominantly undertaken within the domestic sphere and adapted to the generally low social demand. If demand increased, the number of metallurgical operations was multiplied, still using the same rudimentary infrastructures and without explicit attempts at improving the efficiency of their known set-up. In this convenient conservatism, Northeast Iberia shares what would remain a defining characteristic of Iberian metallurgy until the Iron Age (Gómez Ramos, 1996).

6.2. The lasting legacy of the Chalcolithic tradition

It is important to understand how the characteristics of the Northeast Iberian technological tradition affect copper-based production in subsequent periods.

Going back to the EMBA (2100-1650 BC), a previous study of Minferri and Cantorella (Maldà, Lleida) assemblages had revealed the arrival of a range of metallurgical innovations to the Northeast at this time, presumably transmitted from Polada groups in Italy, through Southern France. These are (1) the know-how necessary for alloying bronze, (2) a specific type of purpose-made crucibles with a thicker base and pouring spout, and (3) stone moulds, of which early examples in the Northeast date to 1900-1800 BC (Soriano and Amorós Gurrera, 2014; Soriano and Escanilla, 2016; Peres *et al.*, 2024). Remarkably, the connections between the Northeast and Southern France during this time are also supported by the documented Baltic amber trade that, although started back in Neolithic times, remained active up until this period in the Northeast but absent in other Iberian areas (Murillo-Barroso *et al.*, 2023, 2025b).

Although both purpose-made crucibles and repurposed common pots were used for metallurgical activities at Minferri (2100-1650 BC), purpose-made crucibles disappear from the archaeological record in the Northeast after this time. Arguably, this type of specialised technological ceramics did not provide behaviourally significant improvements in this context. This is supported by the analyses carried out on one purpose-made crucible from Minferri that, when compared to other repurposed fabrics, show that both types of vessels have non-refractory calcareous pastes of comparable qualities (Montes-Landa, 2023; Montes-Landa *et al.*, 2025).

The rejection of purpose-made crucibles and the adoption of bronze technology and stone moulds in the Northeast would be consistent with the already discussed selective adoption of features from Southern Iberia and Southern France technological traditions during Chalcolithic-EBA times. Moreover, it illustrates a key technological characteristic of the regional tradition, which will influence technological choices later on: a commitment to incurring only in low investments during metallurgical practice.

The evolution of copper smelting and bronze alloying technologies documented in Northeast Iberia through the analyses of Minferri, Vilars, Mas Castellar and Ullastret production residues suggests that the technological tradition of this area remained broadly unchanged between the 3rd millennium and at least the 3rd century BC (Montes-Landa, 2023; Montes-Landa *et al.*, 2025). With the exception of bronze technology introduction and stone moulds (Peres *et al.*, 2024), no further remarkable innovations were adopted, including proper furnaces. Metallurgy continued to take place as small-scale operations in crucibles heated from the inside within very simple open structures. Reducing conditions were unstable, resulting in what we could consider inefficient operations that were, however, good enough for the standards of the time. This is demonstrated by the apparent continuation of this way of doing even within the workshop context studied at Ullastret dated to the 450-350 BC (Montes-Landa, 2023; Montes-Landa *et al.*, 2025). This characterisation is consistent with prehistoric Iberian metallurgy in general, which connects this area with the rest of the Peninsula despite the peculiarities already highlighted.

The Chalcolithic-EBA data on metal provenance should also be discussed in relation to later finds. The LIA results available so far are consistent with the use of Les Ferreres (La Bauma), Linda Mariquita (Ullastret), Solana del Bepo (La Bauma, Ullastret), Barranc Fondo (Cova de l'Heura), Turquesa (Cova del Buldó, Balma del Duc, La Bauma?, Ullastret?) and Linares (Minferri) mines (Rafel *et al.*, 2016; Montero, 2017; Montero-Ruiz, 2018; Montes-Landa, 2023; Montes-Landa *et al.*, 2025). For Solana del Bepo case, whose minerals have been found both at La Bauma and Ullastret smelting slag, this would imply transport of minerals from El Priorat to Girona from Chalcolithic (La Bauma) up until Iberian Iron Age times (Ullastret), perhaps using very similar routes. This is particularly noteworthy when one considers that metal trade had been established by the Iron Age. Moreover, an unknown source of copper identified in the production residues from Mas Castellar (dated to the 550-500 BC) is consistent with EMBA metallic items from Cova de Porta Lloret, Rocallaura, and Covas del Cingle Blanc (Arbolí, Tarragona) (Montes-Landa *et al.*, 2025). This also supports a continuity in the use of the same metal sources in the region.

Furthermore, the versatile procurement pattern observed at La Bauma, which combined five sources located between 25 and 200 km away is important, as well as the possible exploitation of two mineral sources at Cova de l'Heura. As already mentioned, these cases exemplify a highly adaptive and versatile strategy to secure metal procurement. This permeated in the later technological tradition in two ways. Firstly, a multi-source exploitation strategy was maintained at sites such as Vilars and Mas Castellar during the Iron Age, Ullastret during Iberian Iron Age times, and possibly Minferri during the EMBA. As in previous times, such strategy allowed sustaining bronze demand. Secondly, the adoption of a flexible pattern of choice towards making technological choices (as seen in the combination of several metal sources within a site, but also in the combination of direct copper smelting and co-smelting of oxidic and sulphidic ores) influenced bronze-making strategies too. Thus, at Minferri, Mas Castellar and Ullastret, different bronze alloying techniques coexisted. Such variability and flexibility in the selection of techniques has been interpreted as a resilience mechanism to sustain demand (Montes-Landa *et al.*, 2025). The adaptive character of the Chalcolithic Northeast Iberian metallurgy remains at the core of such strategy two millennia later.

Also, we should note the matte nodules found at Vilars and Ullastret sites, dated to 450-375 BC and 250-200 BC respectively (Montes-Landa, 2023; Montes-Landa *et al.*, 2025). While these are rare specimens in the context of

Iron Age/Iberian times bronze metallurgy, which is dominated by oxidic minerals, they denote the sporadic handling of sulphidic ores that could have relied on Chalcolithic know-how.

Finally, the contextualisation of the metallurgical operations allows a reassessment of the location of the *chaîne opératoire* in the landscape over time. So far, mainly based on Minferri assemblage and the presence of purpose-made vessels there, it had been contended that since the EMBA the *chaîne opératoire* was broken down into the landscape, with copper smelting being conducted by the mines, and only metal melting occurring on habitation sites (Soriano Llopis, 2010). However, recent results prove that copper smelting and metal alloying were conducted within the settlement (Montes-Landa, 2023; Montes-Landa *et al.*, 2025), thus continuing the Chalcolithic tradition of smelting in domestic spaces, even if the nature of the settlement type had changed. A separation of copper smelting from melting or alloying in the landscape is not observed until the Iron Age at Mas Castellar, but this statement can be misleading: although metallic copper for bronze cementation operations was produced elsewhere, copper ores were still co-smelted with cassiterite to produce bronze on site too. Moreover, at the contemporaneous site of Vilars, copper smelting was still conducted on site within domestic contexts, and this keeps happening later on at Ullastret too. All in all, this data reinforces the weight of the Chalcolithic technological tradition two millennia after its development.

7. CONCLUSIONS

This research has produced new data on the development of smelting technology in Northeast Iberia during the late 3rd millennium BC. Prior to this study, the Chalcolithic crucibles could not be confidently related to smelting operations as opposed to melting ones, due to the absence of microstructural and compositional data. While previous analyses from La Bauma had uncovered the earliest evidence of copper smelting in the region (2878-2479 cal BC) (Montes-Landa *et al.*, 2021), these new analyses allow us to trace the spread of this technology across the area.

The analyses also identified the blended influences of Southern Iberia and Southern France traditions in the early stages of development of metallurgy in the Northeast. While (co-)smelting know-how (to produce copper and lead), use of co-smelting of sulphidic and oxidic ores and possibly organic tempering were adopted from Southern France, crucible-based technology and use of oxidic ores were transferred from Southern Iberia. The combination of characteristic features from both traditions illustrates the ability of Northeast Iberia people to select the relevant innovations that better fit their social and environmental possibilities. This dynamism and critical evaluation of innovations will remain a defining feature of the local technological tradition, as demonstrated by the selective adoption of a new range of metallurgical innovations brought during the EMBA. Thus, Northeast Iberian metallurgy was simple but versatile, allowing for resilience –hence resulting in millennia-long sustainability, which should not be regarded as inflexible conservatism. These contextualised observations challenge technologically deterministic narratives of passive adoption and technological somnambulism (Pfaffenberger, 1988).

Of special interest is the documentation of the use of sulphidic ores at Cova de l'Heura, Balma del Duc, and Cova Cartanyà during Chalcolithic-EBA times. These finds should be added to other scattered evidence across Iberia (Rovira, 2002; Müller *et al.*, 2004; Nocete *et al.*, 2008; La Duc *et al.*, 2022). The multiple instances of sulphidic ore use in Northeast Iberia reinforces a more common use than previously expected, and it likely relates to influences from the French technological tradition. Moreover, matte nodules from the Iron Age and Iberian Iron Age found at Vilars and Ullastret can be consistent with the extended sporadic handling of sulphidic ores for copper production through co-smelting in the Northeast.

The consistent appearance of organic inclusions in Chalcolithic technical ceramics is also a remarkable feature found in Northeast Iberia crucibles. Although we lack conclusive evidence as to whether this was a deliberate practice to enhance the technical properties of the vessels (Martinón-Torres and Rehren, 2014), the use of organic inclusions was consolidated during the EMBA, as demonstrated by Minferri samples (Montes-Landa, 2023; Montes-Landa *et al.*, 2025).

In general, the copper smelting technology in Northeast Iberia remains generally unchanged for at least two millennia. It was characterised by the use of very simple, rudimentary, open structures where small open crucibles were heated from above, and it was conducted in domestic spaces. These crucibles were, in most cases, repurposed vessels with high refractoriness. Even if some purpose-made crucibles appear during the EMBA, these are still manufactured using the same clays. The multiplication of the number of operations allowed maintaining the demand changes while using efficient-enough set ups that did not seek for maximum copper recovery.

This technological tradition was also versatile, which allowed sustaining the necessary demand under the operating conditions. This feature was visible both in the general use of copper mineral sources from different locations at a single site from the Chalcolithic to the Iberian Iron Age, as well as in the combination of several techniques for a given end (*i.e.*, producing copper through co-smelting of oxidic and sulphidic copper ores or by directly smelting oxidic ores; and producing bronze by co-smelting, cementation or recycling). In terms of mineral sources, the outcrops of the Montsant mining districts seem to be continuously exploited during millennia, as well as the unknown likely regional source identified at Mas Castellar whose isotopic signature matches a range of EMBA sites. This supports the passing on of this information over generations.

All this evidence uncovered at Northeast Iberia highlights the importance of studying peripheral areas of metallurgical practice to account for the role of such places in the definition of broader technological traditions and in the development of metallurgy in Iberia and beyond. We believe that further studies of this kind in other areas of Iberia will be fundamental to rebuilding a more accurate narrative on the development of early metallurgy in the western Mediterranean and its subsequent evolution.

SUPPLEMENTARY FILES

The following supplementary material is available on this journal's website: SF1. Full analytical data.

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DECLARATION OF COMPETING INTERESTS

The authors declare that they have not known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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AUTHORSHIP CONTRIBUTION STATEMENT

Julia Montes-Landa: conceptualisation, data curation, formal analysis, funding acquisition, investigation, methodology, project administration, resources, validation, visualisation, writing – original draft, writing – review & editing.

Ignacio Montero-Ruiz*:* data curation, resources, validation, writing – review & editing.

Marcos Martinón-Torres: conceptualisation, resources, funding acquisition, validation, supervision, writing – review & editing.

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