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Exploitation of Mining Resources in El Argar Culture: Bronze Age Metallurgy in the Hinterland of the Western Betic Cordillera (Southeastern Iberian Peninsula)

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ABSTRACT

This research addresses the territorial organisation of metallurgical production during the El Argar Bronze Age (2200–1550 cal BC) in the inner areas of El Argar territory through lead-isotope and trace element analyses of geological copper ores, archaeometallurgical remains and copper-based artefacts. Results from 31 mineral and 35 archaeological samples suggest that the exploitation of copper resources in the studied region was significant and had a similar impact than other mining districts of El Argar territory. This, therefore, leads the hierarchical and centralised production model to be questioned. It also appears that the copper ore deposits in the coastal regions that were intensively exploited during the Copper Age were used less intensively in the El Argar period. At that time, copper was mostly procured from ore deposits in the inland areas of El Argar territory: that is, ore deposits within the Alpine orogeny hinterland (inland areas of the Betic Cordillera, from Granada to Baza). Other artefacts were sourced from outside the Alpine geological domain, but still on the fringe of El Argar territory (the foothills of the Sierra Morena-Linares mining district) or even from ore deposits definitely outside El Argar territory itself (the Los Pedroches Variscan region and elsewhere).

1 | Introduction

The role of metals and metallurgy in El Argar society (c. 2200–1550 cal BC) has been widely discussed in the literature since the first archaeological finds of several thousand prehistoric metal objects and metallurgical remains. The Siret brothers already gave metallurgy a leading role in these prehistoric societies in their first publications (Siret and Siret 1890), although they considered it as an exogenous element according to the prevailing historiographical model at the time. Since then, the role of metals in El Argar society has been debated unceasingly (see Aranda Jiménez, Montón-Subías, and Sánchez Romero 2021 for a review). Although very few production remains have been analysed (Mongiatti and Montero-Ruiz 2020; Moreno Onorato et al. 2010; Rovira Llorens et al. 2015), there is general consensus that the increase in metal production with respect to the Copper Age (c. 3000–2200 cal BC) and

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the introduction of new metals such as bronze and silver did not imply a significant change in the technological process of metallurgical production. Metal ore continued to be processed in domestic contexts through crucible metallurgy, without furnace structures, which implied limited production and poor control over temperature and pressure. This metallurgical process was clearly inefficient and produced viscous slag that did not separate well from the metal, a feature that is shared with contemporary cultures elsewhere on the Iberian Peninsula (Rovira-Llorens and Montero Ruiz 2013). Nevertheless, there was some innovation regarding previous technologies and the introduction of annealing allowed greater versatility in the manufacturing process. For example, it multiplied the manufacture of ornaments with respect to the Copper Age, when they were almost absent (Murillo-Barroso and Montero Ruiz 2012). All in all, the few innovations (bronze alloy, silver and annealing) had little impact on improving extractive efficiency and the higher volume of metals produced was mostly the result of an increase in labour rather than the effect of more efficient technological innovations. However, even though no significant technological changes were induced, there was a shift in the social value of metals, which now played a major role in power and gender demarcation in El Argar society.

In recent decades, scientific debate has focused on the territorial organisation of El Argar metal production. When compared to the Copper Age, there are fewer settlements with evidence of smelting (slag and crucibles with adhered slag). This has been interpreted as a greater centralisation of metallurgical production (e.g., Lull et al. 2010). According to this hypothesis, production would had taken place mainly in the Upper Guadalquivir valley at Peñalosa (Jaén) using the mineral resources of the Sierra Morena foothills (the Linares mining district) (Figure 1). Once smelted, the metal (copper and silver) would have been supplied to the principal settlements of the El Argar region that centralised the finishing of the objects in workshops controlled by the social elites. Finally, metal objects would have been distributed from these workshops to other lower-ranking settlements. This search for new mineral sources would, among other factors, be one of the explanations for the expansion of El Argar culture from the Almeria-Murcia coastal lands to inland areas in the Upper Guadalquivir valley. It remains to be explained, however, which settlements would have been the suppliers of the raw material for the territory before this expansion took place around 1950 cal BC (Lull et al. 2009, Figure 2).[1]

Peñalosa unquestionably stands out from the other El Argar archaeological sites due to the magnitude of its metallurgical production. Slag fragments have been found in nearly all the settlement's houses, as well as in a dump on its outskirts. In total, 20.5 kg of slag (16.2 from the dump), 13 kg of ore, 2094 crucible fragments and 8 moulds have been quantified for a

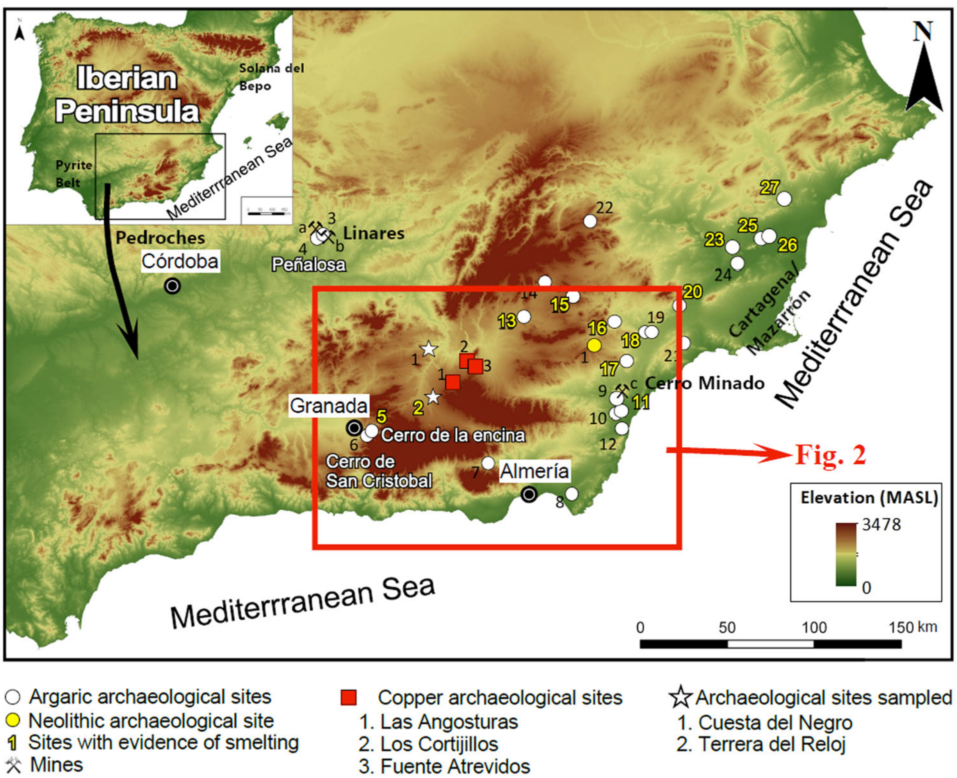

FIGURE 1 | Location of sites mentioned in the text. White dots, El Argar archaeological sites: 1. Cuesta del Negro; **2. Terrera del Reloj** (both of them studied in this paper); 3. Siete Piedras; 4. *Peñalosa*; **5. Cerro de la Encina**; 6. Cerro San Cristóbal; 7. Piedras de Canjayar; 8. *El Barranquete;* 9. *Fuente Álamo*; 10. Lugarico Viejo; **11. *El Argar***; 12. *Gatas;* **13. Cerro de la Virgen**; 14. Puebla de Don Fadrique; **15. Topares; 16. Cerro de las Víboras; 17. *Rincón de Almendricos*; 18. Lorca**; 19. *Murviedro*; **20.*La Bastida***; 21. *Cabezo Negro;* 22. Cerro de las Viñas; **23. Cobatillas;** 24. *Monteagudo*; **25. San Antón; 26. Laderas del Castillo; 27. El Tabayá**. Red dots, Copper Age archaeological sites: 1. Las Angosturas; 2. Los Cortijillos; 3. Fuente Atrevidos. Yellow dot, Neolithic archaeological site: 1. Cerro de Los López. Mines with prehistoric mining evidence mentioned in the text: (a) José Palacio; (b) Polígono; and (c) Cerro Minado. Sites with evidence of smelting are highlighted in bold and sites with LIA analyses by OXALID in italics. Base map designed by A. Uriarte (LabTel, IH-CSIC).

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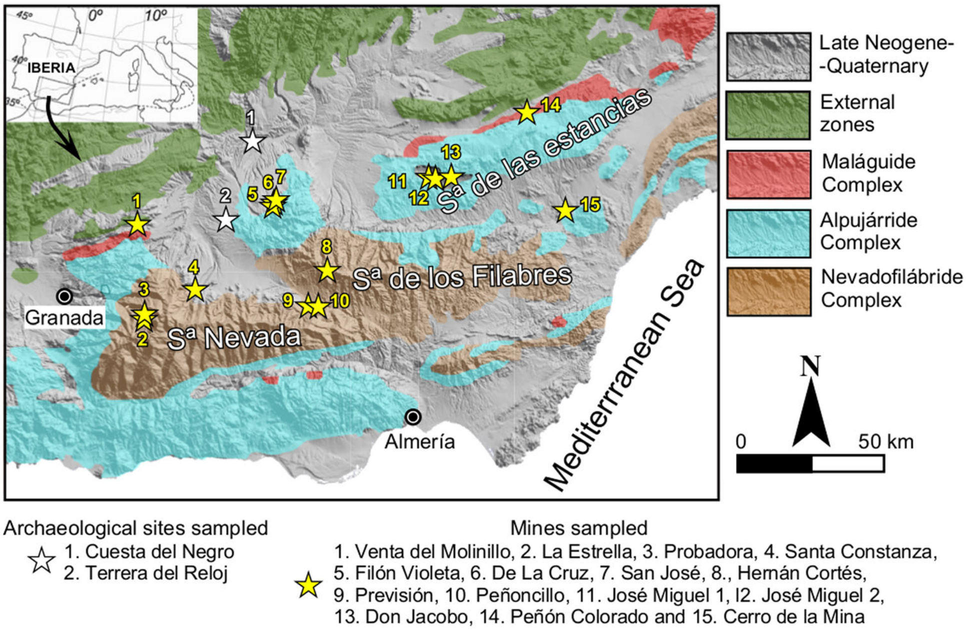

FIGURE 2 | Geological sketch map of the Betic Cordillera showing the mines sampled (yellow): 1. Venta del Molinillo, 2. La Estrella, 3. Probadora, 4. Santa Constanza, 5. Filón Violeta, 6. De La Cruz, 7. San José, 8. Hernán Cortés, 9. Previsión, 10. Peñoncillo, 11. José Miguel 1, l2. José Miguel 2, 13. Don Jacobo, 14. Peñón Colorado and 15. Cerro de la Mina, and archaeological sites with finds analysed in this paper (red): 1. Cuesta del Negro, 2. Terrera del Reloj.

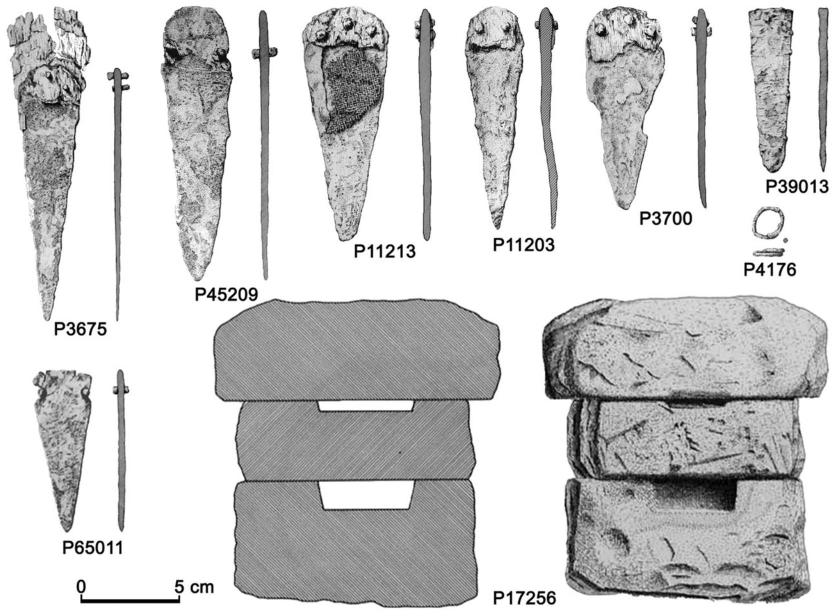

FIGURE 3 | Some of the archaeological objects analysed. Modified from Carrasco Rus 1973.

period of about 400 years, most of them recovered in the excavated area of the dump (3.95 m^3) (Moreno et al. 2017). While these figures could seem low from an industrial metallurgical perspective, they are comparatively high when compared to the few grams or hundreds of grams documented at other prehistoric sites. However, the number of metal objects found at this site is no higher than that in other El Argar settlements, raising the possibility that metal was distributed from Peñalosa to other El Argar settlements or even to other cultural areas outside the El Argar world. However, this hypothesis does not necessarily imply that there was a centralised model of metal production and distribution. The difference between a centralised and a decentralised model lies in the interdependent relationship that emerges when defining production relationships on a territorial scale.

While metallurgy was an important activity in the Sierra Morena mountain range, where the Peñalosa settlement is located, Argarian metallurgical production could also have been organised on a regional basis, with several production sites located in different areas, without this ruling out an exchange of objects on a macro-regional scale (Montero-Ruiz 1994; Murillo-Barroso, Montero-Ruiz, and Aranda Jiménez 2015). In this decentralised

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TABLE 1 | Geological copper ores analysed.

ID	Complex	Age	Min.	Sample	Mine	Municipality
PA25303A	Alpujárride	Triasic (Cover, Baza)	Pb-Fe-Cu	Azurite	Filón Violeta	Oria
PA25292	Alpujárride	Triasic (Cover, Baza)	Pb-Fe-Cu	Azurite	Mina La Cruz	Baza
PA25283B	Alpujárride	Triasic (Cover, Baza)	Pb-Fe-Cu	Malachite	Mina La Cruz	Baza
PA25284	Alpujárride	Triasic (Cover, Baza)	Pb-Fe-Cu	Malachite	Mina La Cruz	Baza
BA-05	Alpujárride	Triasic (Cover, Baza)	Pb-Fe-Cu	Malachite	Mina San José	Baza
PA252311A	Alpujárride	Triasic (Cover, Baza)	Pb-Fe-Cu	Malachite	Mina San José	Baza
PA25297A	Alpujárride	Triasic (Cover, Sierra de las Estancias)	Cu	Malachite	Don Jacobo	Oria
PA25298	Alpujárride	Triasic (Cover, Sierra de las Estancias)	Cu	Malachite	Don Jacobo	Oria
PA25299	Alpujárride	Triasic (Cover, Sierra de las Estancias)	Cu	Malachite	Don Jacobo	Oria
PA25300	Alpujárride	Triasic (Cover, Sierra de las Estancias)	Cu	Malachite	Don Jacobo	Oria
PA25301A	Alpujárride	Triasic (Cover, Sierra de las Estancias)	Cu	Malachite	José Miguel 2	Oria
PA25313	Maláguide	Palaeozoic (Base, hydrothermal, Sierra Arana)	Pb-Cu	Malachite	Venta del Molinillo	Huétor Santillán
PA25315	Maláguide	Palaeozoic (Base, hydrothermal, Sierra Arana)	Pb-Cu	Malachite	Venta del Molinillo	Huétor Santillán
PA25280A	Maláguide	Palaeozoic (Base, hydrothermal, Sierra Arana)	Pb-Cu	Azurite	Venta del Molinillo	Huétor Santillán
PA25281A	Maláguide	Palaeozoic (Base, hydrothermal, Sierra Arana)	Pb-Cu	Azurite	Venta del Molinillo	Huétor Santillán
PA25279A	Maláguide	Palaeozoic (Base, hydrothermal, Sierra Arana)	Pb-Cu	Azurite	Venta del Molinillo	Huétor Santillán
PA25306	Maláguide	Permic-Triasic (Cover, Sedimentary, Vélez-Rubio)	Cu	Malachite	Peñón Colorado	Vélez Rubio
PA25305A	Maláguide	Permic-Triasic (Cover, Sedimentary, Vélez-Rubio)	Cu	Malachite	Peñón Colorado	Vélez Rubio
PA25293	Maláguide	Permic-Triasic (Cover, Sedimentary, Vélez-Rubio)	Cu	Azurite	Cerro de la Mina	Vélez Rubio
PA25294	Maláguide	Permic-Triasic (Cover, Sedimentary, Vélez-Rubio)	Cu	Malachite, Azurite	Cerro de la Mina	Vélez Rubio
PA25296A	Maláguide	Permic-Triasic (Cover, Sedimentary, Vélez-Rubio)	Cu	Malachite, Azurite	Cerro de la Mina	Vélez Rubio
PA25266A	Nevado Filábride	Palaeozoic (Base)	Fe-Cu	Chalcopirite, Malachite	Mina Hernán Cortés	Caniles
PA25270	Nevado Filábride	Palaeozoic (Base)	Cu-Ag-Pb	Chalcopirite, Malachite	Mina La Probadora	Güejar-Sierra
PA25260	Nevado Filábride	Palaeozoic (Base)	Cu	Malachite	Peñoncillo 1	Abla
PA25261A	Nevado Filábride	Palaeozoic (Base)	Cu	Malachite	Peñoncillo 2	Abla
PA25307A	Nevado Filábride	Palaeozoic (Base)	Cu	Malachite	Previsión	Fiñaña
PR-02	Nevado Filábride	Palaeozoic (Base)	Cu	Malachite, Azurite	Previsión	Fiñaña
PA25308A	Nevado Filábride	Palaeozoic (Base)	Cu	Malachite, Azurite	Previsión	Fiñaña
SC-02	Nevado Filábride	Palaeozoic (Base)	Cu	Malachite	Santa Constanza	Jerez del Marquesado
PA25317	Nevado Filábride	Palaeozoic (Base)	Cu	Malachite	Santa Constanza	Jerez del Marquesado
PA25316	Nevado Filábride	Palaeozoic (Base)	Cu	Malachite	Santa Constanza	Jerez del Marquesado

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TABLE 2 | Archaeological objects analysed. AMS dates from Cámara Serrano and Molina González (2009) calibrated using OxCal v4.4.4.

Chronology	Site	ID	Type	Alloy	Context	C14 cal BC (2σ)	Trace elements	LIA	Proposed provenance
Surface	Cuesta del Negro, Purullena	P superficial	Awl	Arsenical Copper	Surface			X	
		P279	Lump	Bronze	Domestic		X	X	
		P60002A	Arrowhead	Bronze	Domestic		X	X	Linares?
		P60002b	Dagger	Copper	Domestic		X	X	Nevado Filábride
El Argar Culture (2200–1550 cal BC)	Cuesta del Negro, Purullena	P13164	Metal Frags.	Bronze	Domestic		X	X	Linares
		P45087	Thread	Bronze	Domestic		X	X	Linares
		P4571	Awl	Arsenical Copper	Domestic		X	X	Linares?
		P9219	Chisel?	Copper	Domestic		X	X	
		P9783	Awl	Arsenical Copper	Domestic		X	x	Maláguide
		P39013	Dagger	Arsenical Copper	Tomb 2	1922–1745 1921–1743	X	X	
		P4176	Ring	Arsenical Copper	Tomb 8	1677–1507		X	
		P11203	Dagger	Arsenical Copper	Tomb 9	1871–1620 1871–1619	X	X	Linares?
		P11213	Dagger	Arsenical Copper	Tomb 10	1919–1700	X	X	Nevado Filábride
		P3676_r	Rivet	Arsenical Copper	Tomb 16	2012–1771	X	X	Maláguide
		P3676_d	Dagger	Arsenical Copper	Tomb 16		X	X	Linares
		P3700_r	Rivet	Arsenical Copper	Tomb 19	2032–1826 1942–1700	X	X	Nevado Filábride
		P3700_d	Dagger	Arsenical Copper	Tomb 19		X	X	
		P45209	Dagger	Arsenical Copper	Tomb 29	1872–1619 1627–1457	X	X	
		P65011	Rivet	Arsenical Copper	Tomb 35		X	X	Linares

(Continues)

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TABLE 2 | (Continued)

Chronology	Site	ID	Type	Alloy	Context	C14 cal BC (2σ)	Trace elements	LIA	Proposed provenance
Post-El Argar	Cuesta del Negro, Purullena	P67006	Awl	Bronze	Domestic		X	X	Linares
		P337	Scrapings from a crucible	Arsenical Copper	Domestic			X	
		P17064	Awl	Arsenical Copper	Domestic		X	X	Nevado Filábride
Surface	Terrera del Reloj, Dehesas de Guadix	DG83015	Blade	Arsenical Copper	Surface			X	
Unknown	Terrera del Reloj, Dehesas de Guadix	DG1024	Copper prill	Arsenical Copper	Unknown			X	
		DG183	Rivet	Arsenical Copper	Unknown			X	
El Argar Culture (2200–1550 cal BC)	Terrera del Reloj, Dehesas de Guadix	DG6295	Awl	Arsenical Copper	Tomb 4			X	Alpujárride
		DG3133	Small Axe	Arsenical Copper	Tomb 6		X	X	Nevado Filábride?
		DG4325	Ring	Arsenical Copper	Tomb 11			X	
		DG4261-1	Ring	Arsenical Copper	Tomb 13			X	Nevado Filábride
		DG4261-2	Ring	Arsenical Copper	Tomb 13			X	Linares
		DG4298	Dagger	Arsenical Copper	Tomb 15		X	X	Linares
Copper Age (3100–2200 cal BC)	Las Angosturas	PA2435A	Copper ore		Domestic			X	Cerro Minado?
	Los Cortijillos	PA4974	Copper ore		Surface			X	Nevado Filábride
	Fuente Atrevidos	PA5025	Copper ore		Surface			X	Cerro Minado?
	Piedras de Canjayar	1935/4/ CANJ/2/1	Dagger 3R		T2			X	

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model, the lower number of sites with evidence of smelting compared to the Copper Age could reflect the different spatial organisation of the settlements. In El Argar villages, buildings were densely packed together and the open spaces typical of earlier Copper Age settlements were replaced by narrow streets. Houses were now rectangular or trapezoidal in shape with internal divisions, in contrast to the circular buildings that distinguished the previous Chalcolithic villages. These changes in urbanism would have forced a polluting activity such as metallurgy, which generated a significant amount of fumes and had some fire risks, to be undertaken away from the habitat areas (sectors that are generally not excavated during archaeological campaigns). This is actually the case of Peñalosa, where the rubbish dump outside the settlement concentrates 80% of the metallurgical evidence. Although in lesser amounts than at Peñalosa, the slag remains reported at another 15 El Argar sites (Figure 1) are a reflection of the necessary existence of other sites with extractive activities in El Argar culture areas (see Murillo-Barroso, Montero-Ruiz, and Aranda Jiménez 2015 and references therein).

In a context in which widespread recycling is not observed and therefore the primary production signatures are not likely to be extensively blurred (e.g., Montero-Ruiz, Murillo-Barroso, and Rovira-Llorens 2020), these two contrasting models (centralised or decentralised) should have produced different lead-isotope data patterns. In a centralised production model, the lead-isotope composition of most El Argar objects should converge with the ore deposits from the eastern Sierra Morena (and the José Palacio and Polígono mines in Linares, see Figure 1), which are known to have been used at Peñalosa (Hunt et al. 2011). On the other hand, in a decentralised model, lead-isotope data would indicate a greater variety of copper ore deposits as potential metal sources (Murillo-Barroso, Montero-Ruiz and Aranda Jiménez 2015).

A pioneering lead-isotope study of 33 copper-based artefacts from the El Argar culture area was carried out in the 1990s as part of the Gatas Project (Stos-Gale 2001; Stos-Gale, Hunt, and Gale 1999). They were mainly from sites on the coastal lands of El Argar territory (Figure 1). In addition, 48 analyses of copper ores from areas around those sites were published by Arribas and Tosdal (1994), Arribas Rosado and Arribas Moreno (1995), Dayton and Dayton (1986), Graeser and Friedrich (1970) and Stos-Gale, Hunt, and Gale (1995, 1999). Despite the limitations of a then-pioneering lead-isotope methodology applied to the provenance of archaeological metals and the limited number of geological samples available for comparison, various provenances ranging from the Sierra Morena to the southwest of the Iberian Peninsula were proposed. It can be noted that the copper and bronze objects analysed by Thermal Ionisation Mass Spectrometry with Multicollector (MC TIMS) and compiled in the OXALID database of the Isotrace Laboratory at the University of Oxford (http://oxalid.arch.ox.ac.uk/) currently include another 14 pieces from the Gatas archaeological site that were not published at the time of that interpretation.

Since that first Gatas Project, the number of analysed silver- and copper-based objects has increased considerably, especially at the inland settlements of the western Betic Cordillera hinterland (Bartelheim et al. 2012; Brandherm et al. 2022; Müller-Kissing 2022; Murillo-Barroso 2013; Murillo-Barroso, Montero-Ruiz, and Aranda Jiménez 2015; Murillo-Barroso et al. 2024). Similarly, the number of lead-isotope analyses of copper and lead–silver ores from the Iberian Peninsula has increased exponentially in the last 20 years (Brandherm et al. 2022; Gauss 2016; Hunt 2003; Hunt, Contreras Cortés, and Arboledas Martínez 2011; Klein et al. 2009; Montero Ruiz and Murillo-Barroso 2010; Murillo-Barroso et al. 2019; Renzi et al. 2016; Rodríguez et al. 2023; Rodríguez Vinceiro et al. 2018; Sáez et al. 2021; Santos Zaldegui et al. 2004; Tornos and Chiaradia 2004). A compilation of these data on minerals and artefacts in the Iberlid database (https://www.ehu.eus/ibercron/iberlid) includes more than 1000 entries. However, despite the increased amount of data, there is still a significant lack of information on potential copper ore deposits that could have been exploited by El Argar society.

In this paper, we present new data from the hinterland of El Argar territory. They contribute to filling in some of these gaps and allow us to depict a more solidly based interpretation of metallurgical production organisation in El Argar society.

2 | Geological Background

This study has focused on the northern fringe of the Sierra Nevada, Sierra Arana, Sierra de Baza, Sierra de Filabres and Sierra de las Estancias mountain ranges in the southeast of the Iberian Peninsula. These are part of the Betic Cordillera that extends for more than 600 km (Figure 2). The Betic Cordillera constitutes the westernmost end of the Mediterranean Alpine Orogen and is composed of two major domains: the External and Internal Zones. The former consists mainly of Mesozoic and Tertiary sedimentary rocks deposited in the southern palaeo-margin of the Variscan Iberian massif. The latter are mainly metamorphic Palaeozoic and Mesozoic rocks distributed in three superposed tectonic complexes: from bottom to top, the Nevado-Filábride, Alpujárride and Maláguide complexes.

2.1 | The Nevado–Filábride Complex

Located at the lowest tectonic position in the Internal Zones, the Nevado–Filábride Complex contains the highest-grade metamorphic rocks in the Betic Cordillera. It crops out in the core of antiforms in the Nevada, Baza, Filabres, Alhamilla, Cabrera, Almagrera and Almenara mountain ranges, as well as in Lomo de Bas (Lorca) and the coastal alignment of Cartagena (Figure 2). The main lithologies are graphite-bearing micaschists and quartzites of Palaeozoic age with marbles and alleged ophiolitic metabasites (Lozano Rodríguez 2019 and references therein) of Jurassic to Cretaceous age.

Copper mineralisation in the Nevado–Filábride Complex appears within the so-called Mulhacén section, in low temperatures, hydrothermal veins and fractures. The main primary ores are siderite, with disseminations of galena, pyrite, chalcopyrite, sphalerite, cinnabar, stibnite, tetrahedrite and arsenopyrite. The gangue is made up of albite, kaolinite, white mica, quartz, chlorite and calcite. Iron oxides, azurite and malachite are common secondary (alteration) minerals (Arana Castillo 1973).

The main deposits are those located on the northern edge of the Sierra Nevada. Stone hammers and prehistoric pottery found at the

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TABLE 3 | Results of trace element compositions of geological ore samples by ICP-MS. See the complete table of results in File S2.

ID	GC	Na (ppm)	Mg (ppm)	Al (ppm)	P (ppm)	K (ppm)	Ca (ppm)	Fe (%)	Mn (ppm)	Co (ppm)	Ni (ppm)
PA25303A	A	40.7	193	404	117	54.1	22,660	0.9	73.6	3.41	5107
PA25292	A	90.2	2039	3746	146	1988	56,700	0.9	370	2134	5783
PA25283B	A	192	407	1308	308	364	70,680	0.3	140	14.4	6888
PA25284	A	118	311	391	313	119	20,450	0.1	158	3.72	131
PA252311A	A	43.0	933	394	41.0	157	40,310	0.1	495	78.3	191
BA-05	A	58.4	919	901	328	267	60,400	0.3	220	2006	9391
PA25297A	A	56.9	18,340	6232	185	1666	13,420	2.2	317	628	114
PA25298	A	71.5	33,490	26,790	334	4418	624	1.3	48.6	2926	480
PA25299	A	62.9	71,550	6424	103	1363	34720	0.8	1016	541	98.4
PA25300	A	10.1	25,110	12,950	300	113	477	0.4	51.4	1522	473
PA25301A	A	14.8	2326	1339	103	217	938	14.2	79.3	573	1532
PA25313	M	70.3	18470	2067	20.8	1042	10,190	9.9	2889	241	678
PA25315	M	64.7	752	6534	104	3213	140	8.0	64.8	70.0	176
PA25280A	M	25.5	3128	2780	70.2	791	1731	1.1	1257	588	443
PA25281A	M	38.6	336	6075	112	1594	2492	12.5	86.9	432	906
PA25279A	M	41.0	39,880	3893	0	853	30,130	2.4	7189	363	79.3
PA25305A	M	352	11,310	62,900	566	20740	513	3.3	197	25.0	66.7
PA25306	M	272	9110	51,590	428	16,580	708	2.2	204	36.7	79.2
PA25293	M	744	722	2466	246	1319	339	0.3	76.2	154	258
PA25294	M	124	862	2905	328	1150	781	0.5	112	233	331
PA25296A	M	55.8	280	3806	197	2870	429	0.7	42.4	201	135
PA25266A	NF	144	11,340	123	0	64.7	392	25.8	5374	60.4	329
PA25270	NF	103	3238	209	40.4	64.9	108	25.2	2212	15.5	18.2
PA25260	NF	29.1	485	829	104	57.3	220	33.8	3222	8.52	112
PA25261A	NF	53.8	428	369	1088	302	365	27.7	366	25.5	219
PA25307A	NF	70.6	522	4118	284	391	6685	3.4	3208	657	131
PA25308A	NF	30.7	104	6051	333	90.3	643	8.4	186	50.0	63.0
PR-02	NF	36.8	194	744	2516	101	6080	9.9	1440	213	86.3
PA25316	NF	359	39,430	15,780	364	2412	46,220	1.2	13,080	344	828
PA25317	NF	93.2	4534	4684	171	255	96,310	0.2	2263	172	468
SC-02	NF	437	7403	16,800	340	1914	34,550	1.2	2413	92.7	228

Abbreviations: A, Alpujárride; GC, geological complex; M, Maláguide; NF, Nevado Filábride.

Santa Constanza mine suggest its exploitation in Late Prehistory. Other mines sampled for this study within this complex are La Estrella, Probadora, Previsión, Peñoncillo and Hernán Cortés.

2.2 | The Alpujárride Complex

The Alpujárride Complex is a succession of tectonic nappes with pre-Mesozoic metapelites at the base that are overlaid by a Triassic sedimentary section, fundamentally formed by carbonated rocks. This complex also includes the largest and best outcrops of the subcontinental lithospheric mantle of the world, the Ronda peridotites.

Two types of copper mineralisation appear within the metasedimentary formations of the Alpujárride Complex: stratiform-stratabound Fe–Pb–Zn–Cu deposits of probable sedimentary origin and subvolcanic deposits with a similar chemistry, but filling faults and/or fractures, probably remobilising the former stratiform mineralisation. Copper minerals are mostly sulphides, while minor copper carbonates appear as secondary mineral associations, together with tyrolite, annabergite, cinnabar, goethite, magnetite, proustite and pyrargyrite.

In the Alpujárride Complex, copper ores have been sampled from Filón Violeta, De La Cruz, San José, Don Jacobo and José Miguel-1 and −2 mines (Figure 2). No evidence of prehistoric exploitation has been found at these mines, although prehistoric occupation since the Neolithic has been documented in these areas. Intensive modern mining may, however, have obliterated that evidence.

2.3 | The Maláguide Complex

The Maláguide Complex is located in the highest part of the Internal Zones. It is barely affected by metamorphism and

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Cu (ppm)	Zn (ppm)	As (ppm)	Sr (ppm)	Ag (ppm)	Sn (ppm)	Sb (ppm)	Ba (ppm)	Pb (ppm)	Bi (ppm)	Th (ppm)	U (ppm)
13.4	552	5206	51.6	297	327	1571	43.7	2116	1007	0	6591
20.1	1345	4020	390	31.5	96.8	474	344	790	0	0.439	2023
11.8	16,520	12,570	2480	84.0	133	3012	9374	107,400	1653	0.116	5496
8.0	49,840	36,290	279	145	58.4	3505	74.3	87,000	1148	0	11.5
11.6	95,370	3138	334	30.3	86.8	73.0	15.6	51,490	0.412	0.113	2365
11.4	9210	87,950	464	34.2	29.8	306	31.9	112,200	1651	0.074	29.2
28.0	2424	60,820	95.8	40.5	27.7	550	22.5	9269	0	0.760	13.7
31.0	766	2091	7975	13.5	32.8	2172	40.5	64.8	0	1815	1035
11.0	2345	37,960	72.2	84.4	32.0	79.0	24.4	858	0	0.733	5058
31.4	1684	3153	4633	0.872	18.0	2254	6581	764	0	0.934	1443
21.6	409	2207	12.0	31.3	29.7	24.2	60.6	49.4	0	0.200	34.8
10.5	2027	3074	6495	42.7	45.5	59.1	13.6	6854	116	0.466	0.269
9.3	105	5754	23.0	94.1	26.5	237	97.6	27,640	5313	0.507	0.549
33.4	1823	346	3242	11.1	22.6	102	17.7	1553	1417	0.396	8464
22.3	3732	4410	16.6	60.3	45.8	56.8	274	206	433	0.300	3246
12.7	14,120	40.1	38.8	104	29.5	132	160	146	2.29	0.149	0.288
9.2	67.3	66.8	17.0	0.657	18.6	0.926	225	35.3	0.696	6674	3952
17.3	87.2	109	14.7	0.697	20.6	0.290	168	32.9	0.698	4651	3763
14.1	1017	468	35.8	0.827	9.12	4185	238	7.61	0	2.04	16.4
16.7	1065	710	35.4	0.840	9372	8695	47.4	12.2	0	1335	9242
11.8	2918	9434	159	134	8716	248	163	4652	0.199	1442	28.4
18.6	60.1	276	2313	91.2	12.6	44.4	2888	10.7	4067	0	0
22.1	54.5	170	1574	39.3	10.6	27.6	2856	5876	0.502	0	0
11.8	44.9	19.9	16.0	4384	12.8	2377	6157	2.52	0	0.109	1683
14.1	37.0	1179	33.1	23.5	55.2	16.6	16.2	23.8	7766	0.170	4424
23.4	56.1	7774	167	10.2	32.7	0.502	18.1	10.7	2538	0.528	76.9
20.3	43.2	13,560	31.0	5948	9799	3746	3907	4092	0.689	1443	6983
14.0	30.7	88,120	111	2581	9049	4338	5.33	7898	0	0.156	61.4
13.8	1335	191	148	2419	25.3	50.2	71.1	36.2	0.762	2976	3922
8.0	1979	26.2	505	0.215	6605	0.603	77.9	2.45	0	0.576	4678
18.3	824	123	78.3	0.469	7489	1688	82.1	6271	0.319	2679	3128

overlays the Alpujárride Complex. Its main outcrops are located in the western part, near the city of Málaga and in the Montes de Málaga mountains, although some outcrops are also located north of Granada and Sierra de las Estancias (Figure 2).

In this complex, copper mineralisation occurs mainly in the Saladilla Formation, which is made up of conglomerates, sandstones, pelites, grey dolomites and Permian-Triassic gypsum. Copper minerals in the Maláguide Complex are mostly sulphides. Minor secondary copper carbonates appear to be linked to the presence of basic volcanism in the case of Sierra Arana. In the Sierra de las Estancias area, copper mineralisation appears to have a sedimentary origin, although recent tectonics have played an important role in its final concentration (Carrasco Cantos et al. 1979).

Copper ores have been sampled at the Venta del Molinillo, Peñón Colorado and Cerro de la Mina mines, the last of these just a few kilometres from the Neolithic settlement of Cerro de Los López (Martínez García, Blanco de la Rubia, and Mellado Sáez 1988). Prehistoric pottery and stone hammers have been found in those last two mines, suggesting their exploitation in that period.

3 | Archaeological Background

Several Argaric sites (2200–1550 cal BC) with metal objects and metallurgical remains have been excavated in the study area. Compositional and, in some cases, also metallographic analyses had already been carried out on metal objects from the Castellón Alto, Cuesta del Negro, Terrera del Reloj, Cerro de la Encina and Cerro de San Cristóbal sites (Aranda Jiménez et al. 2012; Arribas et al. 1989; Bashore 2013; Bashore Acero et al. 2014). More recently, a provenance study was undertaken to ascertain the origin of copper-based objects from the settlements of Cerro de la Encina, Cerro de San Cristóbal and La Puebla de Don Fadrique (Álvarez Peanes 2016; Murillo-Barroso,

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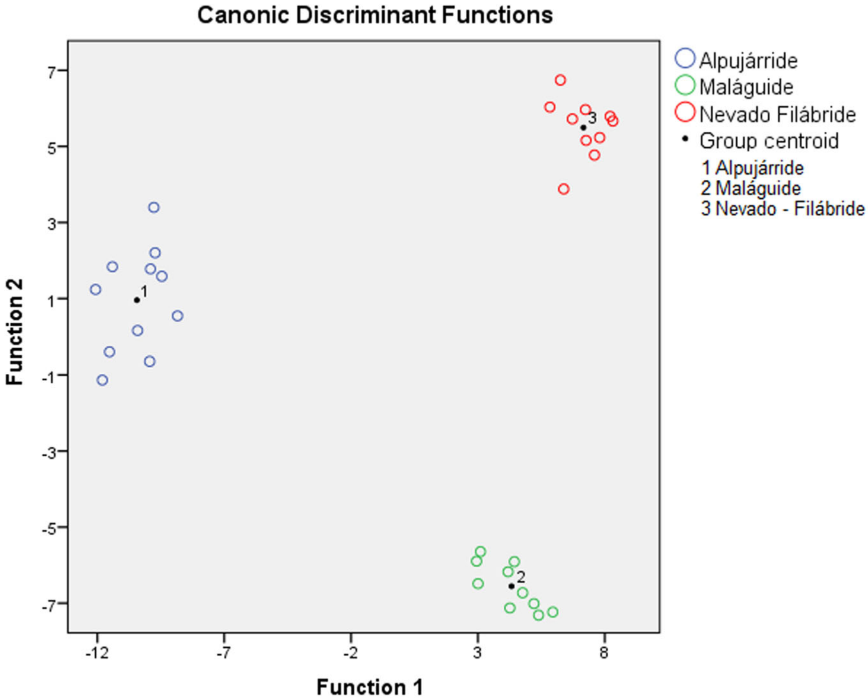

FIGURE 4 | Linear discriminant analysis (LDA) of trace element compositions of copper ore samples from the three geological complexes.

Montero-Ruiz, and Aranda Jiménez 2015). All these sites are located between 10 and 80 km from the aforementioned mining areas discussed in Section 2 (Figure 1).

In this study, we present new trace element and lead-isotope results from the settlements of Cuesta del Negro and Terrera del Reloj (2200–1550 cal BC), which complete the information available for the northern fringe of the Sierra Nevada (Figure 2). Both settlements have the typical El Argar urban planning: built at height, with rectangular houses on terraced areas and burials located inside the houses, mostly in *covachas* (small caves cut into the bedrock) and very occasionally inside *pithoi*. At Cuesta del Negro, 36 tombs were excavated inside the houses and metal objects were found in 21 of them (Figure 3). Several moulds, including a trivalve type, as well as crucible fragments with copper slag adherences are evidence of metallurgical activity in the settlement (Cámara Serrano and Molina González 2009; De La Torre 1973; Jabaloy Sánchez 1978; Molina González and Pareja López 1975). The occupation of Cuesta del Negro occurs mostly during the Early and Middle Bronze Age (c. 1950–1450 cal BC), where all graves are recorded, and absolutes dates are obtained from human remains (Cámara Serrano and Molina González 2009). In this phase, we have most of the metallurgical evidence recorded on the site. In a later period, some houses ascribed to the Late Bronze Age based on decorated *Cogotas* I type pottery (c. 1550–1350 AC) are also documented. At Terrera del Reloj, there were numerous elements that can be linked to metallurgy and mining: at least five stone hammers, copper ore fragments, slag, copper drops and crucible fragments, in addition to the objects found in the graves, such as daggers, awls and ornaments (Arribas et al. 1989). Only two absolute dates have been published from Terrera del Reloj on charcoal (Ambers, Matthews, and Bowman 1987, 1991) but its architecture, graves and material culture evidence an Early and Middle Bronze Age occupation. Most of the metallic objects are actually recovered from El Argar tombs.

4 | Materials and Methods

A set of 31 copper ores from the studied mines (Table 1, File S1) and 31 copper-based objects from the archaeological sites of Cuesta del Negro and Terrera del Reloj as well as few samples from surrounding Copper Age sites (Table 2) were analysed for lead-isotope and trace elemental compositions.[2] Given the degree of corrosion of some of the archaeological objects, only two from Terrera del Reloj and 19 from Cuesta del Negro were suitable for trace element analysis.

For comparison purposes, we also analysed three samples of copper ore collected at the nearby Copper Age archaeological sites of Las Angosturas de Gor, Fuente Atrevidos and Los Cortijillos, as well as a dagger with 3 rivets from Tomb 2 at Piedras de Canjayar (Figure 1, Table 2).

The mineral compositions of the copper ores were determined by X-ray diffraction (XRD) at the CENIM-CSIC using a Bruker AXS model D8 Advance X-ray diffractometer, equipped with a Co tube and Goebel mirror optics to obtain a parallel and monochromatic beam. The working conditions were 40 kV and 30 mA.

Trace element analyses were performed at the Geochronology and Geochemistry—SGIker Facility of the University of the Basque Country UPV/EHU (Spain) using a Quadrupole Inductively Coupled Plasma—Mass Spectrometer (Q-ICP-MS). Details of the methods are published in Murillo-Barroso et al. (2020) and Aragón et al. (2022). The uncertainty of the results corresponds to a 95% confidence level (see analytical errors 2SE in File S2).

Lead-isotope analysis (LIA) was conducted at the Geochronology and Isotope Geochemistry—SGIker facility of the University of the Basque Country (Spain). Aliquots of 0.05 g (mineral samples) and 0.005–0.015 g (archaeological objects) were dissolved with 1 mL of

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7 M HNO3 overnight at 70°C on a heating plate. Lead was purified from the obtained 7 M HNO3 solutions using a miniaturised method adapted from Gale (1996). Lead-isotope ratio measurements were performed by multi-collector inductively-coupled plasma—mass spectrometry (MC-ICP-MS) following the methodology described in Rodríguez et al. (2020). Ore samples were measured in two analytical sessions in June 2017, and measurements of archaeological samples were performed in five analytical sessions in January–February 2016. Analyses of the Certified Reference Material NBS981 during those sessions, with on-line correction for the instrumental mass bias, yielded 206Pb/204Pb = 16.9434 ± 0.0010, 207Pb/204Pb = 15.5009 ± 0.0010, 208Pb/204Pb = 36.7287 ± 0.0026, 207Pb/206Pb = 0.9149 ± 0.0001 and 208Pb/206Pb = 2.1677 ± 0.0001 (average of 13 measurements, with absolute uncertainties twice the standard deviation). Average ratios of NBS981 for this study match the long-term reproducibility obtained at this lab (see File S3).

LIA results will be compared with data from the literature. A first set of analyses was performed by the Oxford Isotrace Laboratory using TIMS (Stos-Gale 2001; STOS-GALE et al. 1995) and are available in OXALID, the Open Access database (http://oxalid.arch.ox.ac.uk/). Since then, the introduction of MC-ICP-MS has considerably increased the reliability (precision and accuracy) of lead-isotope analysis for provenance studies (see Baker et al. 2004; Stos-Gale and Gale 2009; Murillo-Barroso et al. 2019 or Rodríguez et al. 2023 for comparison of the TIMS and MC-ICP-MS techniques). Analyses carried out by MC-ICP-MS at the Geochronology and Isotope Geochemistry SGIker-Facility of the University of the Basque Country UPV/EHU have been published recently. It is necessary to consider the greater analytical error of the analyses performed by TIMS (±0.1%) compared to the recent results obtained with MC-ICP-MS (±0.01%) (see File S3 for further methodological comparison between the results of the Oxford Isotrace Laboratory and the Geochronology and Isotope Geochemistry SGIker-Facility of the University of the Basque Country UPV/EHU).

Object provenances were established following the Stos-Gale and Gale (2009) methodology. Bivariate diagrams were established of LI ratios of all ore data points for each deposit in relation to the data points representing the artefacts. Bivariate diagrams used are mostly $^{206}Pb/^{204}Pb$ vs. $^{207}Pb/^{204}Pb$, $^{206}Pb/^{204}Pb$ vs. $^{208}Pb/^{204}Pb$ and $^{208}Pb/^{204}Pb$ vs. $^{207}Pb/^{204}Pb$ but also $^{207}Pb/^{206}Pb$ vs. $^{208}Pb/^{206}Pb$ and $^{207}Pb/^{206}Pb$ vs. $^{206}Pb/^{204}Pb$. Euclidean distances between the lead-isotope data for artefacts with the whole isotopic database for ores have also been calculated to corroborate concordances established based on bivariate diagrams, especially for artefacts plotting on overlapping areas of two isotopic fields. Any concordance established is evident in all bivariate graphs.

5 | Results

5.1 | Mineralogical Aspects

The mineralogy of the studied ore samples is heterogeneous, especially those from the Nevado Filábride Domain. In general, they are cupro–ferrous mineral associations. Relevant to the chemical composition of ores and metals is the presence of arsenic-bearing minerals in all the ore samples, namely, conichalcite ($CaCuAsO_4[OH]$), clinoclase ($Cu_3AsO_4[OH]_3$), chenevixite (Cu_2-$Fe^{3+}{}_2[AsO_4]_2[OH]_4$) and cornwallite ($Cu_5[AsO_4]_2[OH]_4$). Cerussite ($PbCO_3$) and segnitite ($PbFe_3{}^{3+}AsO_4[AsO_3OH][OH]_6$) have also been identified in ores from the Alpujárride Complex.

5.2 | Trace Element Analyses

Arsenic was detected in all analysed ore samples (Table 3). It reaches the highest contents in ore samples from the Alpujárride Complex (0.2%–8.7% As), with As/Cu ratios above 2/100 in 80% of the samples and above 10/100 As/Cu for 50% of the samples (up to a maximum As/Cu of 77/100). In contrast, only 40% and 30%, respectively, of the samples from the Maláguide Complex and Nevado Filábride Domain have As/Cu ratios above 2/100 and in no case does it exceed 8/100 As/Cu. The highest As/Cu ratio is from sample Pr-02 from the Nevado Filábride Domain (63/100).

The samples from the Alpujárride Complex also show high levels of zinc and lead (≤ 9.5% and ≤ 11.2%, respectively) and significant nickel content (≤ 0.7%).

TABLE 4 | Standardised canonical discriminant function coefficients of copper ore samples analysed.

	Function	
	1	**2**
Na	4.129	4.535
Mg	−1.253	−3.425
Al	6.540	18.948
P	−0.779	0.774
K	8.128	−16.111
Ca	−2.746	−1.658
Sc	−0.736	−4.659
Ti	5.389	−2.544
V	−0.228	−1.763
Cr	1.988	6.523
Fe	3.169	5.844
Mn	7.864	2.795
Co	3.957	−0.122
Ni	−3.992	4.078
Zn	−4.765	4.793
As	−3.155	1.295
Rb	−0.285	−2.162
Sr	−2.575	2.207
Ag	−1.767	−0.587
Sn	9.201	1.254
Sb	−1.555	−2.009
Ba	0.326	−0.004
Pb	8.206	−1.043
Bi	−0.450	−3.156
Th	−13.204	0.226
U	1.892	4.105

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TABLE 5 | Results of trace element compositions of archaeological objects by ICP-MS. A threshold of 1% Sn or As has been established for their classification as Arsenical Copper (As up to 6.26%) or Bronzes (Sn up to 6.16%).

ID	Alloy	Na (ppm)	Mg (ppm)	Al (ppm)	K (ppm)	Ca (ppm)	Ti (ppm)	V (ppm)	Fe (ppm)	Mn (ppm)	Co (ppm)
DG3133	CuAs	< LMD	8.56	4.29	< LMD	< LMD	< LMD	< LMD	7.1	< LMD	0.22
DG4298	CuAs	41.8	5.95	2.07	< LMD	< LMD	1.37	< LMD	14.6	0.24	0.28
P11203	CuAs	179	103	26.6	45.1	97.5	4.49	1.23	41.4	4.75	0.92
P11213	CuAs	67.1	11.3	12.3	< LMD	17.5	1.53	< LMD	35.4	0.37	0.40
P17064	CuAs	138	179	102	109	132	4.79	3.69	191	3.07	0.11
P3676-R	CuAs	149	53.4	25.2	79.7	152	2.84	0.76	40.5	3.35	1.01
P3676-D	CuAs	882	297	724	462	559	29.4	5.26	578	6.07	10.1
P3700-R	CuAs	2579	449	305	1710	2061	9.17	6.59	193	4.66	2.27
P3700-D	CuAs	598	93.7	200	205	390	11.7	0.82	129	1.75	0.66
P39013	CuAs	2001	169	147	1265	1432	10.4	2.23	151	3.41	0.14
P45209	CuAs	31.5	7.12	3.8	< LMD	17.2	1.18	0.36	23.5	1.46	2.02
P4571	CuAs	349	1475	2511	845	1323	44.2	8.42	1952	26.8	1.27
P60002B	Cu	194	36.9	35.7	241	< LMD	< LMD	2.47	97.7	1.95	0.38
P65011	CuAs	3428	204	57.2	2868	2401	7.00	1.82	37.3	5.90	0.19
P9219	Cu	601	1871	2658	1304	1649	37.5	48.7	1898	29.8	3.53
P9783	CuAs	857	796	562	994	1022	16.8	19.9	1616	22.1	0.31
P279	Br	72.9	12.9	17.8	< LMD	< LMD	< LMD	< LMD	7.1	< LMD	4.19
P67006	Br	115	885	2355	675	1188	32.2	10.5	1524	22.4	1.00
P60002A	Br	55	43.6	134	35.5	37.6	1.97	0.21	473	2.73	16.5
P13164	Br	50.8	327	884	249	318	9.92	1.27	228	9.55	68.1
P45087	Br	48.3	13.8	7.74	< LMD	14.9	1.60	< LMD	13.8	0.29	9.94
QCS (Recovery %)		111	104	102	117	109	97	103	102	126	107
Error %		0.01	0.02	0.02	0.25	0.17	0.02	0.01	0.03	0.01	0.01
MDL (ppb)		1.04	0.87	0.39	7.00	6.46	0.52	0.12	1.00	0.03	0.02

Abbreviations: Br, bronze; CuAs, arsenical copper; < LMD, below the limit of detection; LMD, limit of detection; QCS, quality control solution.

The samples from the Nevado Filábride Domain, on the other hand, are the only ones to present significant levels of silver, up to 0.6% (Table 3 and File S2).

In order to better visualise these differences, a Linear Discriminant Analysis (LDA) including all elements that passed the tolerance test was carried out (Figure 4 and Table 4). It reflects 100% of the total variance with two functions and clearly separates the three domains based on their trace element compositions.

The copper-based objects studied are nearly pure copper or arsenical copper (Cu ≥ 93.9%; Table 5). Only five objects may be considered bronze alloys (Sn = 1.2%–6.2%). Arsenic was detected in almost all cases, although generally at low levels (2% on average and up to a maximum of 6.3%). It is significant that no arsenic was documented in any of the bronze objects (0.2% As maximum). In addition to the absence of As, significant differences can also be seen in other elements. Bronzes have higher average values of Ni, Ag or Sb (452, 528 and 131 ppm, respectively, compared to 51, 278 and 71 ppm, respectively, in the arsenical copper) and especially of Pb: four of the five bronze objects exceeded 1450 ppm of Pb, with a maximum value of 3.2% Pb, compared to the ≤ 341 ppm Pb in the arsenical copper objects. In contrast, Bi is the only element found at lower levels in the bronze objects with respect to those of arsenical copper ones (16 and 148 ppm on average, respectively) (Figure 5). These compositional differences suggest the use of different copper ore deposits for the manufacture of bronzes and arsenical coppers.

5.3 | Lead-Isotope Analysis

The isotopic composition of the geological samples does not present an excessive dispersion, with all the samples lying between 18.28 and 18.70 for the isotopic ratio $^{206}Pb/^{204}Pb$, between 15.65 and 15.69 for $^{207}Pb/^{204}Pb$ and between 38.44 and 38.92 for $^{208}Pb/^{204}Pb$ (Table 6). Ore deposits from the three geological groups, the Alpujárride, Maláguide and Nevado Filábride complexes, plot in different fields in bivariate diagrams, although with some overlaps (Figure 6).

The samples from the Alpujárride Complex are the most homogeneous ($^{206}Pb/^{204}Pb = 18.34–18.44$, $^{207}Pb/^{204}Pb = 15.673–15.680$, $^{208}Pb/^{204}Pb = 38.555–38.633$, $^{208}Pb/^{207}Pb = 2.09–2.10$ and $^{208}Pb/^{206}Pb = 0.850–0.854$), with the exception of sample PA25301A, which shows higher values, especially in

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Ni (ppm)	Cu (%)	Zn (ppm)	As (ppm)	Se (ppm)	Ag (ppm)	Sn (ppm)	Sb (ppm)	Pb (ppm)	Bi (ppm)	Th (ppm)	U (ppm)
55.0	99.5	440	13,674	12.5	809	27.2	63.8	46.4	88.1	0.003	0.003
23.0	94.1	381	21,509	8.93	167	< LMD	75.3	228	285	< LMD	0.002
197	96.7	399	11,647	6.55	180	< LMD	< LMD	10.5	82.0	0.006	0.014
7.63	99.7	416	27,345	7.71	94	< LMD	122	< LMD	69.8	0.005	0.003
31.4	99.6	397	13,995	8.27	384	67.0	164	341	97.0	0.030	0.217
6.72	99.1	101	13,152	18.0	77.0	75.3	15.2	11.0	89.1	0.005	0.008
3.81	94.6	83.2	27,740	14.0	55.4	92.7	13.9	17.4	51.5	0.133	0.051
50.5	98.9	115	14,937	22.7	147	244	127	44.2	127	0.046	0.093
104	96.6	79.4	38,794	< LMD	< LMD	54.6	57.7	19.6	2.45	0.031	0.042
1.63	94.2	77.8	62,641	< LMD	101	131	20.7	112	1018	0.022	0.128
235	98.6	383	14,285	33.1	294	206	275	163	199	0.004	0.003
9.90	98.9	359	21,400	12.1	136	217	32.2	29.3	103	0.698	1.19
43.7	97.0	112	7204	10.8	430	772	65.5	18.5	43.0	< LMD	0.006
26.1	95.9	319	10,732	7.23	390	125	33.7	125	37.6	0.010	0.012
10.7	96.3	323	6269	17.2	206	74.0	10.6	34.1	54.8	0.733	2.79
13.4	94.7	364	25,860	7.40	976	60.6	52.4	8.34	27.8	0.122	0.705
430	99.2	409	2719	13.6	1039	11,986	327	1481	39.5	0.005	0.012
9.03	94.9	323	10	3.23	368	14,513	61.2	242	8.7	0.850	2.84
466	96.8	339	1.94	< LMD	446	24,750	47.1	7397	3.4	0.032	0.063
936	98.0	322	1.99	< LMD	102	41,293	89.4	32147	17.9	0.204	0.465
418	93.9	367	3.92	2.62	687	61,612	129	7747	12.8	< LMD	0.002
102	116	103	104	102	109	104	103	97	98	100	91
0.01	0.01	0.01	0.02	0.03	0.50	0.05	0.02	0.05	0.07	0.10	0.12
0.14	61.41	0.67	0.32	1.16	7.79	23.41	0.33	0.66	0.01	0.00	0.00

the $^{206}Pb/^{204}Pb$ ratio: $^{206}Pb/^{204}Pb = 18.65$, $^{207}Pb/^{204}Pb = 15.689$, $^{207}Pb/^{204}Pb = 38.627$, $^{208}Pb/^{207}Pb = 2.07$ and $^{208}Pb/^{206}Pb = 0.840$. As this sample significantly differs from all other samples of the Alpujárride Complex, at the present state of research, we will consider it an outlier and will not be included in the definition of its isotopic field (Figure 6). However, in this paper, fields are defined only by 10 samples and their isotopic fields might enlarge if further samples are measured.

Two subgroups are distinguished within the Maláguide Complex: ore deposits of sedimentary origin ($^{206}Pb/^{204}Pb = 18.47–18.55$, $^{207}Pb/^{204}Pb = 15.679–15.691$ and $^{208}Pb/^{204}Pb = 38.63–38.79$) and hydrothermally formed deposits. The latter have ratios with similar values to those of the Alpujárride Complex ($^{206}Pb/^{204}Pb = 18.28–18.34$, $^{207}Pb/^{204}Pb = 15.657–15.677$, $^{208}Pb/^{204}Pb = 38.44–38.54$, $^{208}Pb/^{207}Pb = 2.100–2.102$ and $^{208}Pb/^{206}Pb = 0.854–0.856$), with the exception of the sample PA25281A at $^{206}Pb/^{204}Pb = 18.63$, $^{207}Pb/^{204}Pb = 15.683$, $^{208}Pb/^{204}Pb = 38.72$, $^{208}Pb/^{207}Pb = 2.077$ and $^{208}Pb/^{206}Pb = 0.841$. Similar to the case of the Alpujárride, this sample differs from all other samples of the hydrothermal Maláguide Complex and is not included in the definition of its isotopic field.

Finally, the samples from the Nevado–Filábride Complex present greater isotopic dispersion ($^{206}Pb/^{204}Pb = 18.42–18.70$, $^{207}Pb/^{204}Pb = 15.662–15.686$ and $^{208}Pb/^{204}Pb = 38.02–38.93$) and partially overlap with the Alpujárride Complex field (Figure 6).

The isotopic compositions of the studied ore deposits located in the hinterland of El Argar territory (in red in Figure 7) differ considerably from those of other El Argar areas, such as the Mulhacén and Alpujárride Complexes in the Vera Basin (Almería) (in blue in Figure 7), the materials corresponding to the tabular cover of the Linares (Jaén) mining district (in green in Figure 7) and the materials from Cartagena/Mazarrón (in black in Figure 7). The detailed geological information of these zones as well as the isotopic results can be found in Arribas and Tosdal (1994), Baron, Rico, and Antolinos Marín (2017), Gauss (2016), Graeser and Friedrich (1970), Klein et al. (2009), Murillo-Barroso et al. (2019), Santos Zalduegui et al. (2004) and STOS-GALE et al. (1995).

Regarding the results for the archaeological object samples, most objects have $^{206}Pb/^{204}Pb$ ratios between 18.26 and 18.68 (Table 7, Figure 8). Two subsamples (rivet and blade) were

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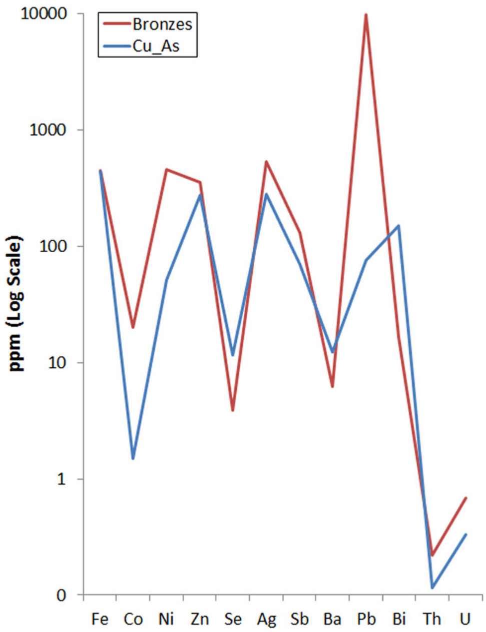

FIGURE 5 | Average of trace elements of arsenical copper and bronze objects analysed in this paper. Note higher levels of Ni, Ag and especially Pb on bronze objects than in arsenical copper ones. Bi is the only element with a higher value on the arsenical copper objects if compared to bronze ones.

analysed from daggers P3676 and P3700; interestingly, for each of the studied daggers, the two subsamples have different isotopic (and elemental) compositions.

6 | Discussion

In this paper, we analysed 31 mineral and 35 archaeological samples, 31 of them from two settlements in the hinterland of El Argar culture and further 4 samples from nearby Copper Age sites (Figure 2). These results should be looked at together with the trace element and lead-isotope compositions of objects from Cerro de la Encina and Cerro de San Cristóbal, which are also located in western Sierra Nevada and were published in 2015 (Murillo-Barroso, Montero-Ruiz, and Aranda Jiménez 2015). However, incomplete information about the copper mines made it impossible to identify the metal provenance for 15 of the 23 objects analysed at that time (Murillo-Barroso, Montero-Ruiz, and Aranda Jiménez 2015). The new data provided in this work, together with the recent data published for the Vera Basin (Murillo-Barroso et al. 2019), allow a better understanding of the territorial organisation of copper resource exploitation in El Argar society and some issues can be highlighted.

6.1 | Metallurgical Production and Metal Selection and Use

In terms of metal use, it is interesting to note that regarding the two daggers for which both the blade and the rivet were analysed, the two elements had different isotope and trace element compositions and therefore do not appear to have been made with the same casting (Figure 8, Table 5). This is a trait that confirms the need for repair due to the wear and tear of the daggers highlighted in previous studies (Montero Ruiz and Murillo-Barroso 2021). Different isotope compositions were also obtained for the two rings found in Tomb 13 at Terrera del Reloj (DG4261-1 and DG4261-2) (Figure 8). In that case, this could indicate that these pieces were not made specifically for the grave goods but were accumulated during the lifetime of the deceased.

More interesting is the fact that bronze objects and production remains (the low-tin bronze lump and the crucible scrapings) from Cuesta del Negro are well grouped in the lead-isotope plots (Figure 8), although the crucible has a post-El Argar chronology and the lump is a surface find (Table 2). No tin was detected in the crucible scrapings, suggesting that the same copper resources may have been used for both bronze and copper metallurgy. The low tin levels of the lump are more consistent with El Argar metallurgy than with later examples. If the bronze lump at Cuesta del Negro is from the El Argar period, it would constitute, together with the crucible recently analysed at El Argar site (Mongiatti and Montero-Ruiz 2020), some of the first evidence of the El Argar bronze production process. In any case, these production remains would evidence the continuity of raw material procurement in the post-El Argar phase (1550–1300 cal BC). These data are important since, among the more than 20 kilos of slag and the crucible fragments documented at Peñalosa, so far, no evidence of bronze production has been documented (Moreno Onorato et al. 2010). Likewise, the local production at Terrera del Reloj may be envisaged from the similarity of isotopic compositions of the copper prill DG1024 and the copper ring found in Burial 11 (DG4325), which are also very close to the production remains of Cuesta del Negro (Figure 8).

Regarding alloys, three sites, Cuesta del Negro, Cerro de la Encina and Cerro de San Cristóbal, have objects in both bronze and arsenical copper, although there are significant differences between them. At Cuesta del Negro, bronzes plot in the area of $^{206}Pb/^{204}Pb = 18.26–18.30$, $^{207}Pb/^{204}Pb = 15.63–15.65$ and $^{208}Pb/^{204}Pb = 38.43–38.50$ (Figure 9). Some copper objects, such as the rivet P65011 or the crucible slag from the same site, plot in this area. This could point to a local production of bronze (as suggested by the bronze lump) or to imported copper and bronze objects from the same origin, although, except for the rivet P65011, almost none of the copper objects from the site coincide isotopically with those made of bronze. We observe greater isotopic differences between copper and bronze at Cerro de la Encina and Cerro de San Cristóbal (Figure 9). Bronzes from Cerro de la Encina have more radiogenic isotopic composition than the copper artefacts from the same sites. Similarly, bronze objects from Cerro de San Cristóbal have higher $^{207}Pb/^{204}Pb$ ratios than arsenical coppers from the same site (Figure 9) (Murillo-Barroso, Montero-Ruiz and Aranda Jiménez 2015). These different tendencies do not seem to be significant enough to reflect two systematically different arsenical copper and bronze productions, although they may reflect different mineral resources. This is especially evident in the case of the bronze ring MO39260, bracelet MO39255 and dagger MO39281 from Tomb 21 at Cerro de la Encina (Ns. 9, 10 and 11 in Figure 9).

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TABLE 6 | Results of lead-isotope analyses of geological samples by MC-ICP-MS.

ID	GC	$^{206}Pb/^{204}Pb$	Uncertainty (2SE)	$^{207}Pb/^{204}Pb$	Uncertainty (2SE)	$^{208}Pb/^{204}Pb$	Uncertainty (2SE)	$^{208}Pb/^{206}Pb$	Uncertainty (2SE)	$^{207}Pb/^{206}Pb$	Uncertainty (2SE)
PA25303A	A	18.3467	0.0007	15.6730	0.0006	38.5573	0.0016	2.10160	0.00003	0.85427	0.00001
PA25292	A	18.4026	0.0007	15.6741	0.0007	38.6071	0.0017	2.09791	0.00004	0.85173	0.00001
PA25283B	A	18.4055	0.0005	15.6760	0.0005	38.6337	0.0015	2.09904	0.00003	0.85170	0.00001
PA25284	A	18.3510	0.0007	15.6751	0.0006	38.5728	0.0017	2.10194	0.00003	0.85418	0.00001
PA252311A	A	18.3670	0.0007	15.6745	0.0007	38.5877	0.0018	2.10092	0.00004	0.85340	0.00001
BA-05	A	18.3575	0.0007	15.6749	0.0007	38.5830	0.0017	2.10176	0.00004	0.85387	0.00001
PA25297A	A	18.3757	0.0008	15.6814	0.0008	38.5631	0.0020	2.09859	0.00004	0.85338	0.00001
PA25298	A	18.4437	0.0007	15.6806	0.0006	38.5556	0.0017	2.09044	0.00003	0.85019	0.00001
PA25299	A	18.3634	0.0008	15.6759	0.0008	38.5552	0.0020	2.09957	0.00004	0.85365	0.00001
PA25300	A	18.3808	0.0007	15.6805	0.0007	38.5631	0.0018	2.09800	0.00003	0.85309	0.00001
PA25301A	A	18.6577	0.0007	15.6891	0.0006	38.6276	0.0016	2.07033	0.00003	0.84090	0.00001
PA25305A	MS	18.5544	0.0007	15.6862	0.0006	38.7929	0.0017	2.09077	0.00003	0.84542	0.00001
PA25293	MS	18.5496	0.0007	15.6864	0.0007	38.7183	0.0018	2.08729	0.00004	0.84564	0.00001
PA25306	MS	18.5121	0.0009	15.6797	0.0008	38.7394	0.0020	2.09266	0.00003	0.84700	0.00001
PA25294	MS	18.4826	0.0007	15.6817	0.0007	38.6357	0.0018	2.09038	0.00003	0.84846	0.00001
PA25296A	MS	18.4710	0.0008	15.6912	0.0007	38.6402	0.0020	2.09194	0.00004	0.84951	0.00001
PA25281A	MH	18.6370	0.0007	15.6837	0.0007	38.7212	0.0018	2.07765	0.00004	0.84153	0.00001
PA25315	MH	18.3468	0.0007	15.6774	0.0006	38.5409	0.0017	2.10069	0.00004	0.85450	0.00001
PA25313	MH	18.3431	0.0006	15.6752	0.0006	38.5344	0.0017	2.10076	0.00004	0.85456	0.00001
PA25280A	MH	18.3323	0.0008	15.6731	0.0008	38.5247	0.0020	2.10147	0.00003	0.85495	0.00001
PA25279A	MH	18.2837	0.0008	15.6575	0.0007	38.4470	0.0019	2.10280	0.00003	0.85636	0.00001
PA25266A	NF	18.6080	0.0007	15.6854	0.0007	38.8818	0.0018	2.08953	0.00003	0.84294	0.00001
PA25270	NF	18.6873	0.0007	15.6700	0.0006	38.9253	0.0017	2.08298	0.00003	0.83854	0.00001
PA25260	NF	18.5964	0.0021	15.6745	0.0018	38.7661	0.0045	2.08460	0.00007	0.84288	0.00002
PA25261A	NF	18.7058	0.0007	15.6863	0.0006	39.0258	0.0017	2.08629	0.00003	0.83858	0.00001
PA25307A	NF	18.3869	0.0007	15.6629	0.0007	38.5310	0.0018	2.09557	0.00004	0.85185	0.00001
PA25308A	NF	18.5348	0.0015	15.6699	0.0012	38.7434	0.0032	2.09030	0.00006	0.84543	0.00002
PR-02	NF	18.4402	0.0007	15.6762	0.0006	38.5992	0.0017	2.09321	0.00003	0.85011	0.00001
PA25316	NF	18.4232	0.0009	15.6781	0.0009	38.6400	0.0023	2.09735	0.00003	0.85100	0.00001
PA25317	NF	18.6332	0.0014	15.6746	0.0012	38.7975	0.0031	2.08217	0.00006	0.84122	0.00002
SC-02	NF	18.6715	0.0009	15.6779	0.0008	38.9391	0.0021	2.08549	0.00004	0.83967	0.00001

Abbreviations: A, Alpujárride; GC, geological complex; MH, Maláguide (Hydrothermal); MS, Maláguide (Sedimentary); NF, Nevado Filábride.

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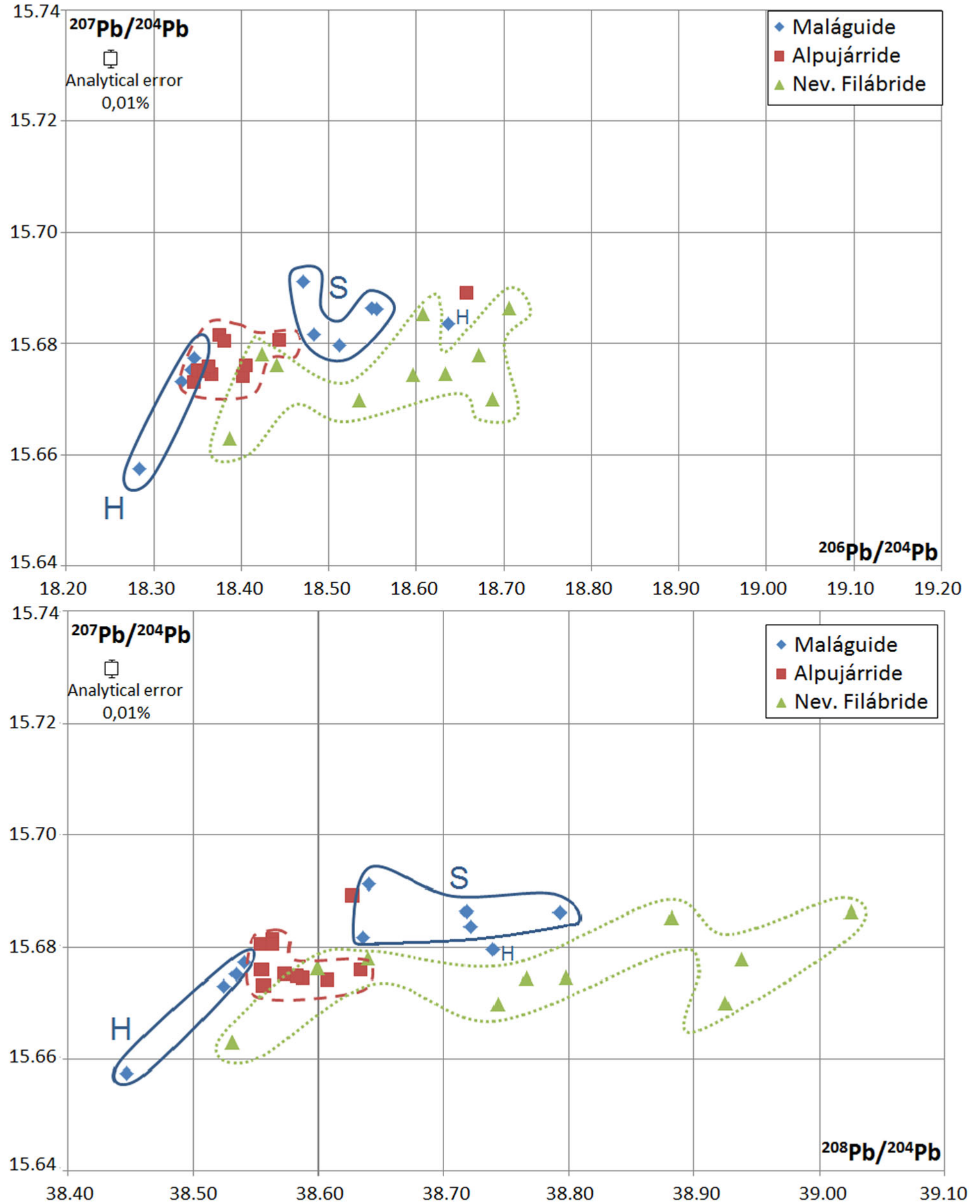

FIGURE 6 | Isotopic ratios of copper ore samples from the Maláguide (of hydrothermal = H and sedimentary = S origins), Alpujárride and Nevado Filábride complexes. Note that the Maláguide outlier is a hydrothermal sample.

6.2 | Local Copper Provenance in the Hinterland of El Argar Culture

The lead-isotope ratios of some of the metal objects of the studied region are compatible with those of neighbouring copper deposits, suggesting local use of the surrounding ore resources (Figure 9). Thus, the awl P9783 and the rivet of dagger P3676 from Cuesta del Negro (Ns. 2 and 3 in Figure 9) plot in the lead-isotope field of sedimentary copper deposits from the Maláguide Complex. Likewise, ring DG4261-1 from Terrera del Reloj (N.4 in Figure 9) and the metal fragments P60002b (N.8), awl P17064 (N.6), dagger P11213 (N.7) and the rivet of dagger P3700 (N.5) from Cuesta del Negro can be associated with copper deposits from the Nevado Filábride Domain. Finally, awl DG6295 from Tomb 4 at Terrera del Reloj (N.1 in Figure 9) can be linked to copper deposits from the Alpujárride Complex. According to Euclidean distances in the isotope space, there is one sample from Previsión mine, in the Nevado Filábride complex, which is geometrically closer (at a Euclidean Distance of 0.0185), although there are also three samples from Don Jacobo mine, in the Alpujárride Complex, at Euclidean Distances of 0.0187, 0.020 and 0.022. Although the isotopic fields have been defined on the basis of 10 geological samples (and thus they can be modified if further samples are analysed), we have preferred to be very strict on the provenance assignation and only propose a possible provenance to the objects that plot inside the isotopic fields defined with the available up-to-date information even if this may entail an underestimation of these resources exploitation. However, doing so, some objects such as, for instance, the small axe DG3133 from Terrera del Reloj (N.12 in Figure 9), have not been associated with the Nevado Filábride Complex, as they fall outside (although close) this isotopic field in some bivariate graphs.

This would mean that at least 30% of the finds analysed from these two archaeological sites are compatible with local copper deposits. The fact that one object has a post-El Argar chronology

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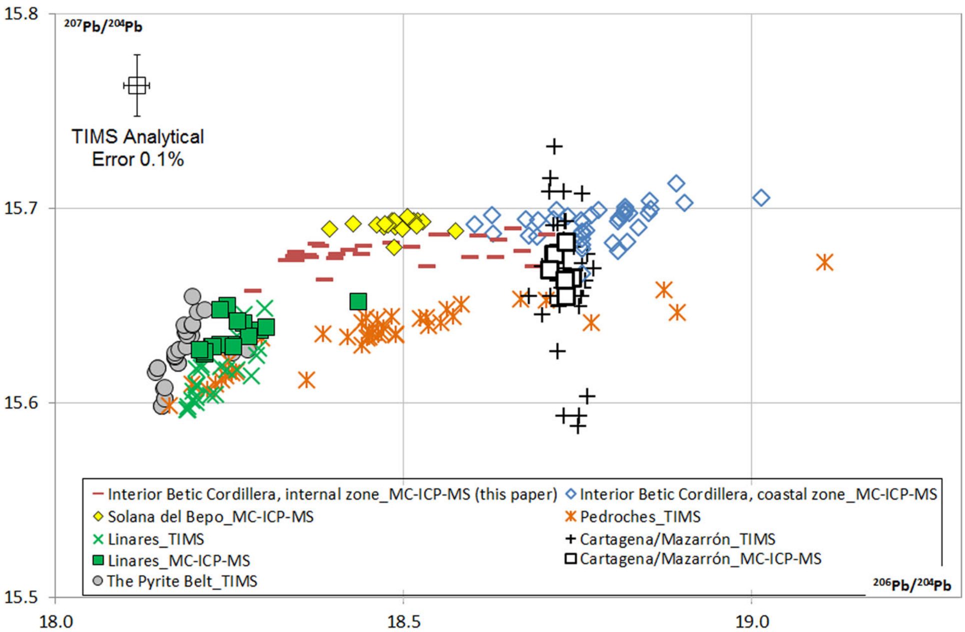

FIGURE 7 | Isotopic ratios of the complexes analysed in this paper (red) compared to other mining districts within El Argar territory, that is, Linares in Jaén province (green), the Vera Basin in Almería province (blue) and the Cartagena/Mazarrón mining district in Murcia province (black) as well as other mining districts outside El Argar territory discussed in the text, mainly Los Pedroches (orange), the Pyrite Belt in the Iberian SW (grey) and Solana del Bepo in the Iberian NE (yellow). Data collected from Arribas and Tosdal (1994), Baron, Rico, and Antolinos Marín (2017), Graeser and Friedrich (1970), Hunt (2003), Klein et al. (2009), Müller et al. (2014), Montero-Ruiz (2017), Murillo-Barroso et al. (2019), Rodríguez et al. (2023), Santos Zalduegui et al. (2004), STOS-GALE et al. (1995) and this study. Analytical Error only applies to TIMS analyses; for MC-ICP-MS data, errors are smaller than symbols.

evidences the continuity in the exploitation of these resources. Local copper mining is confirmed by the isotope composition of the ore found at the Copper-Age settlement of Los Cortijillos, which also plots in the field of the Nevado-Filábride mines and very close to some of the analysed metal objects (Figure 9). However, the remaining two-thirds of the analysed objects (21 out of 30) are not compatible with local copper sources and imply that either copper ores from elsewhere were imported to be smelted in the study area or that metal objects were produced elsewhere and distributed throughout El Argar territory (although, as stated, if more samples are analysed from these mining districts, the extent of their isotopic fields might enlarge, matching some of these unprovenanced objects). It is striking that none of the objects from Cerro de la Encina and Cerro de San Cristóbal can be associated with the mineralisations of this area, the closest geographically to both sites, as the lead-isotopic compositions of objects from these two settlements are less radiogenic than both the analysed ores and the objects from Cuesta del Negro and Terrera del Reloj (Figure 9).

6.3 | Regional Exploitation of Copper Resources

It is clear that local ore deposits were a secondary source for metal production by El Argar societies in the hinterland of the territory. The wide dispersion of objects observed in lead-isotope bivariate diagrams supports a decentralised model of metal production with a heterogeneous supply of raw materials. In the case of Cuesta del Negro, only a few copper objects were produced from local mines. Bronze objects, a bronze lump and the rest of the copper objects from this settlement have less radiogenic lead and plot in a linear array from local mines toward the well-known Linares and Los Pedroches mining districts, on the fringe of El Argar territory (Figure 10).

The contribution of the Linares (Jaén) mining district in copper and silver production is well attested (e.g., Bartelheim et al. 2012; Brandherm et al. 2022; Hunt et al. 2011; Murillo-Barroso, Montero-Ruiz, and Aranda Jiménez 2015). Specifically, four bronze objects from Cuesta del Negro (arrowhead P60002A –N.2 in Figure 10–, post-El Argar awl P67006 –N.3–, thread P45087 –N.4– and dagger P13164 –N.5–) and at least one of the four bronzes from Cerro de San Cristóbal (bracelet OSC7002 –N.6–) can be associated with the pre-Alpine (Variscan) Linares (Jaén) isotope field (Figure 10). The relatively high levels of lead found in some of these pieces (≤ 3.2%) are also compatible with the lead-rich copper minerals found at Peñalosa. It is noteworthy, however, that there is no evidence of bronze production at Peñalosa, despite the abundant metallurgical remains. Therefore, the most likely explanation is that local smelters at Cuesta del Negro acquired ores from the eastern Sierra Morena (Linares) to be smelted on site either in pure form or mixed with some local ores in different proportions. This interpretation may also be applied to copper objects from Cerro de la Encina and bronzes from Cerro de San Cristóbal, as they plot in the same array in lead-isotope diagrams. The fact that the low-tin bronze lump P279 and the copper slag P337 from Cuesta del Negro plot close to the Linares isotopic field and remain in a peripheral zone of the Maláguide Complex in bivariate diagrams opens up the possibility of a double scenario at this site. On the one hand, bronze manufactured with copper from Sierra Morena could have arrived in a metallic state, either as

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TABLE 7 | Results of lead-isotope analyses of archaeological samples by MC-ICP-MS.

ID	Site	Context	Type	$^{206}Pb/^{204}Pb$	Error (2SE)	$^{207}Pb/^{204}Pb$	Error (2SE)	$^{208}Pb/^{204}Pb$	Error (2SE)	$^{208}Pb/^{206}Pb$	Error (2SE)	$^{207}Pb/^{206}Pb$	Error (2SE)	Proposed provenance
DG1024	TR	Domestic	Copper prill	18.3440	0.0007	15.6588	0.0007	38.5260	0.0018	2.1002	0.00004	0.85362	0.00001	
DG183	TR	Unknown	Rivet	18.4726	0.0007	15.6647	0.0007	38.6955	0.0019	2.0947	0.00004	0.84800	0.00001	
DG3133	TR	T6	Small Axe	18.4169	0.0008	15.6564	0.0007	38.5324	0.0020	2.0922	0.00004	0.85011	0.00001	Nevado Filábride?
DG4261-1	TR	T13	Ring	18.3926	0.0007	15.6673	0.0006	38.5850	0.0019	2.0978	0.00005	0.85183	0.00001	Nevado Filábride
DG4261-2	TR	T13	Ring	18.3199	0.0010	15.6497	0.0010	38.4774	0.0026	2.1003	0.00005	0.85424	0.00002	Linares
DG4298	TR	T15	Dagger	18.2625	0.0010	15.6428	0.0010	38.4233	0.0026	2.1039	0.00005	0.85655	0.00002	Linares
DG4325	TR	T11	Ring	18.3501	0.0008	15.6597	0.0008	38.5240	0.0021	2.0994	0.00004	0.85338	0.00001	
DG6295	TR	T4	Awl	18.3841	0.0010	15.6735	0.0009	38.5460	0.0025	2.0967	0.00004	0.85256	0.00001	Alpujárride
DG83015	TR	Surface	Blade	18.4745	0.0007	15.6640	0.0006	38.6719	0.0018	2.0933	0.00004	0.84787	0.00001	
P11203	CN	T9	Dagger	18.3063	0.0008	15.6470	0.0008	38.4084	0.0022	2.0981	0.00005	0.85473	0.00001	Linares?
P11213	CN	T10	Dagger	18.4145	0.0011	15.6647	0.0010	38.5635	0.0027	2.0942	0.00005	0.85067	0.00002	Nevado Filábride
P13164	CN	Domestic	Metal frags	18.2669	0.0007	15.6373	0.0007	38.4301	0.0020	2.1038	0.00004	0.85605	0.00001	Linares
P17064	CN	Domestic	Awl	18.4004	0.0007	15.6610	0.0007	38.5304	0.0020	2.0940	0.00005	0.85112	0.00001	Nevado Filábride
P279	CN	Domestic	Copper lump	18.3063	0.0007	15.6581	0.0007	38.5064	0.0018	2.1035	0.00003	0.85534	0.00001	
P337	CN	Domestic	Crucible scrapings	18.3110	0.0008	15.6558	0.0007	38.5095	0.0020	2.1031	0.00004	0.85499	0.00001	
P3676_d	CN	T16	Dagger	18.2824	0.0008	15.6489	0.0008	38.4347	0.0020	2.1023	0.00005	0.85595	0.00001	Linares
P3676_r	CN	T16	Rivet	18.5458	0.0008	15.6805	0.0008	38.6480	0.0022	2.0839	0.00005	0.84550	0.00001	Maláguide
P3700_d	CN	T19	Dagger	18.6741	0.0022	15.6570	0.0020	38.7280	0.0053	2.0739	0.00009	0.83843	0.00002	
P3700_r	CN	T19	Rivet	18.3910	0.0007	15.6645	0.0007	38.5452	0.0019	2.0959	0.00004	0.85174	0.00001	Nevado Filábride
P39013	CN	T2	Dagger	18.4351	0.0009	15.6621	0.0008	38.6128	0.0024	2.0945	0.00005	0.84958	0.00001	
P4176	CN	T8	Ring	18.3512	0.0006	15.6647	0.0005	38.5381	0.0015	2.1000	0.00003	0.85361	0.00001	
P45087	CN	T31	Thread	18.2806	0.0006	15.6423	0.0006	38.4510	0.0016	2.1034	0.00003	0.85568	0.00001	Linares
P45209	CN	T29	Dagger	18.6831	0.0009	15.6766	0.0008	38.6879	0.0022	2.0707	0.00004	0.83908	0.00001	
P4571	CN	Domestic	Awl	18.2917	0.0015	15.6515	0.0014	38.4162	0.0036	2.1002	0.00008	0.85567	0.00002	Linares?
P60002A	CN	Domestic	Arrowhead	18.2849	0.0008	15.6486	0.0007	38.4714	0.0021	2.1040	0.00004	0.85582	0.00001	Linares
P60002b	CN	Domestic	Fragment	18.4516	0.0007	15.6707	0.0007	38.6069	0.0020	2.0923	0.00005	0.84928	0.00001	Nevado Filábride
P65011	CN	T35	Rivet	18.2733	0.0007	15.6426	0.0007	38.4440	0.0020	2.1038	0.00004	0.85603	0.00001	Linares

(Continues)

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TABLE 7 | (Continued)

ID	Site	Context	Type	$^{206}Pb/^{204}Pb$	Error (2SE)	$^{207}Pb/^{204}Pb$	Error (2SE)	$^{208}Pb/^{204}Pb$	Error (2SE)	$^{208}Pb/^{206}Pb$	Error (2SE)	$^{207}Pb/^{206}Pb$	Error (2SE)	Proposed provenance
P67006	CN	Domestic	Unknown	18.2951	0.0006	15.6487	0.0006	38.4833	0.0018	2.1035	0.00004	0.85535	0.00001	Linares
P9219	CN	Domestic	Chisel?	18.6138	0.0009	15.6796	0.0009	38.7092	0.0024	2.0796	0.00005	0.84236	0.00001	
P9783	CN	Domestic	Metal frags	18.5505	0.0025	15.6862	0.0021	38.7041	0.0052	2.0864	0.00006	0.84560	0.00002	Maláguide
PA4974	LC	Surface	Ore	18.4585		15.6738		38.6869		2.0959		0.84913		Nevado Filábride
PA5025	FAt	Surface	Ore	18.9842		15.7099		38.7146		2.0393		0.82752		Cerro Minado?
PA2435A	LA	Surface	Ore	18.8057		15.7063		38.663		2.0559		0.83519		Cerro Minado?
1935/4/CANJ/2/1	PC	T2	Dagger 3R	18.3860		15.6735		38.5749		2.0980		0.85246		

Abbreviations: CN, Cuesta del Negro; FAt, Fuente Atrevidos; LA, Las Angosturas; LC, Los Cortijillos; PC, Piedras de Canjayar; TR, Terrera del Reloj.

finished objects or ingots (e.g., lump P279) to be cast on site. On the other hand, ore from the Sierra Morena could have been brought to the settlement to be smelted directly on site. In both scenarios, copper from the Maláguide Complex could have been mixed with that from Linares.

Other pieces that can be associated with the Linares deposits are the blade of dagger P3676_d (N.7 in Figure 10), rivet P65011 (N.8), awl P4571 (N.9) and dagger P11203 (N.10) from Cuesta del Negro, although the last two objects plot on the periphery of the Linares isotopic field at $^{208}Pb/^{204}Pb$. To these ring DG4261-2 (N.11) and dagger DG4298 (N.12) from Terrera del Reloj can be added, as well as dagger MO55208 (N.13) and bracelets MO39279 (N.14) and MO39258 (N.21) from Cerro de la Encina, although these plot on its edge in some bivariate graphs and this last object would be outside Linares if only MC-ICP-MS data are used for the definition of the Linares mining district (Figure 7) (Rodríguez et al. 2023). This means that between 18% and 22% of the objects (excluding post-El Argar and surface finds) from the hinterland of El Argar culture would have been manufactured with copper ores from the Linares mining district (Jaén). A similar percentage (20%–25%) of objects originating from Linares is obtained from sites in the coastal lands of Almería, including El Argar and El Oficio (Murillo-Barroso et al. 2024) or Murcia (Brandherm et al. 2022, 138).

We also observe some concordances with other copper deposits in El Argar area (i.e., the deposits of the coastal area, Almería-Cartagena, where some of the main Argarian sites are located) (Figure 10). The bronze bracelet MO39255 from T21 at Cerro de la Encina (N.1 in Figure 10) could be associated with the Cerro Minado copper mine in the Vera basin, although it plots on the border of its isotopic field. Two copper ores found at the Copper-Age settlements of Fuente Atrevidos and Las Angosturas plot over the isotope field of Cerro Minado as well. The copper ores from Fuente Atrevidos and Las Angosturas are pure raw materials and their isotopic composition undoubtedly indicates potential sourcing mine(s) or mining district(s). Another sample, dagger MO39281 from Tomb 21 at Cerro de la Encina (N.22 in Figure 10), is related to the copper deposits in the Segura Valley (Murcia) (Brandherm et al. 2022).

Other metal objects from these settlements also indicate potential trade in ore (or metals) from outside El Argar territory. The halberd from Puebla de Don Fadrique, dagger OSC15014 from Cerro de San Cristóbal (N.15 in Figure 10), bronze awl MO39257 (N.16) and dagger MO39264 (N.17), although the latter contains > 20% Ag that could alter its isotopic signature, as well as bracelet MO39261 (N.18), awl MO55209 (N.19) and dagger MO21292 (N.20) from Cerro de la Encina, all plot within the Los Pedroches isotopic field in the Sierra Morena mountains, outside the area corresponding to El Argar culture.

Further on, the scraper OSC11015 from Cerro de San Cristóbal (N.23 in Figure 10) matches the isotope field of the pre-Alpine (Variscan) Pyrite Belt in southwestern Iberia (beyond the Betic Cordillera) and the bronze ring MO39260 from Cerro de la Encina (N.24) plots in all bivariate diagrams within the isotopic field of the Solana del Bepo mine in northeastern Iberia, a copper deposit exploited since the first half of the second millennium BC (Rafel et al. 2019). Other pieces whose origins

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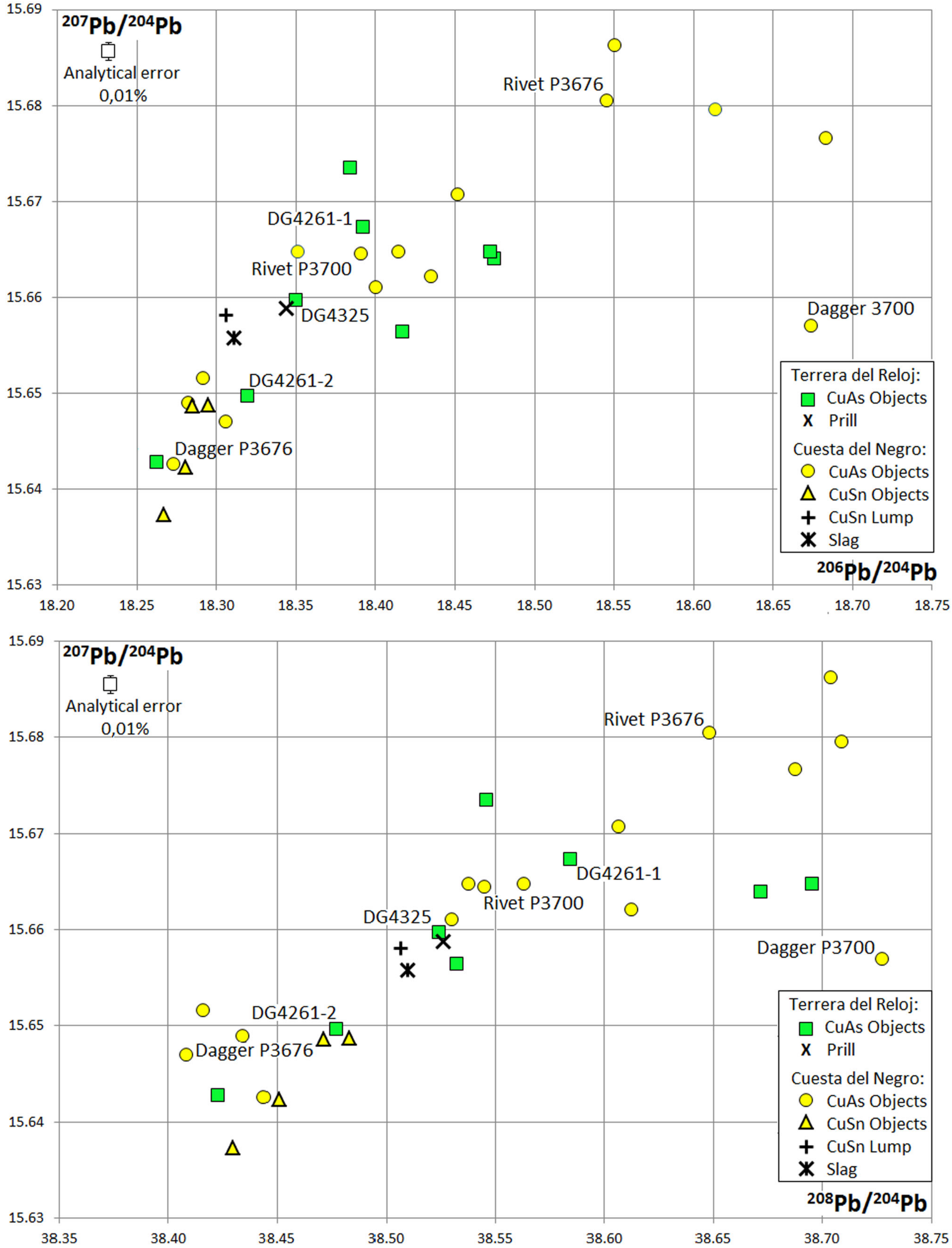

FIGURE 8 | Legend on next page.

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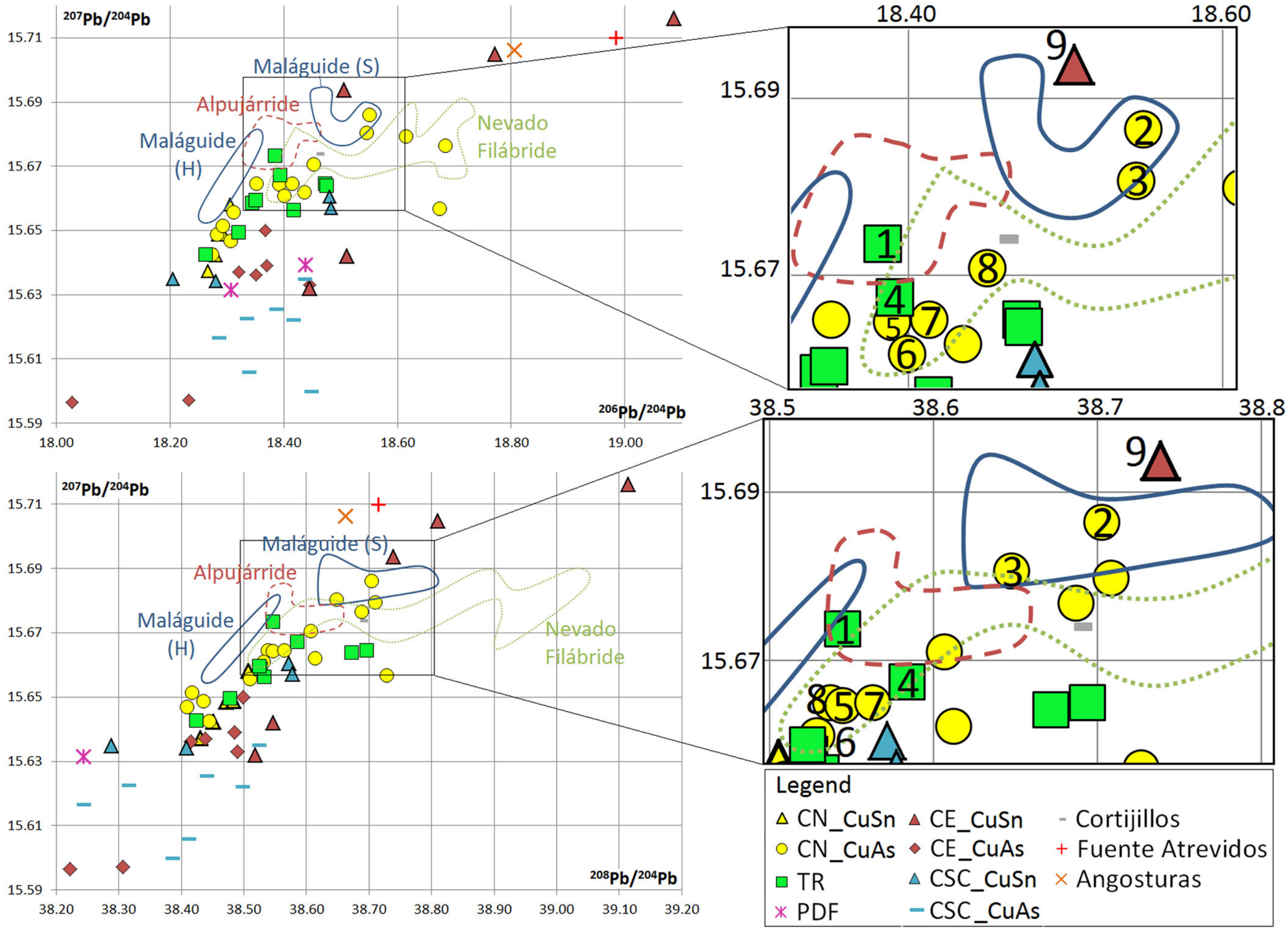

FIGURE 9 | Isotopic ratios of the archaeological objects from all El Argar archaeological sites in the area (CE, Cerro de la Encina; CN, Cuesta del Negro; CSC, Cerro de San Cristóbal; PDF, Puebla de Don Fadrique; TR, Terrera del Reloj) as well as the copper ore samples from the Copper Age settlements of Las Angosturas, Cortijillos and Fuente Atrevidos projected on top of the isotopic field of the mining districts analysed. Analytical errors are smaller than symbols. 1. Awl DG6295 from Tomb 4 at Terrera del Reloj, 2. The metal fragments P9783 from Cuesta del Negro, 3. The rivet of dagger P3676 from Cuesta del Negro, 4. Ring DG4261-1 from Terrera del Reloj, 5. The rivet of dagger P3700 from Cuesta del Negro, 6. Awl P17064 from Cuesta del Negro, 7. Dagger P11213 from Cuesta del Negro, 8. Metal fragment P60002b from Cuesta del Negro, 9. Bronze ring MO39260 from Tomb 21 at Cerro de la Encina, 10. Bronze bracelet MO39255 from Tomb 21 at Cerro de la Encina, 11. Bronze dagger MO39281 from Tomb 21 at Cerro de la Encina and 12. Small axe DG3133 from Terrera del Reloj. Archaeological data compiled from Murillo-Barroso, Montero-Ruiz and Aranda Jiménez (2015), Álvarez Peanes (2016) and this study.

extend beyond the El Argar area are nail OSC13006_71 from Cerro de San Cristóbal (N.25 in Figure 10), for which an origin in the mineralisations of the pre-Variscan (Cadomian) Ossa-Morena was already proposed (Murillo-Barroso, Montero-Ruiz, and Aranda Jiménez 2015). Therefore, this means that 24.5% of the objects found at the archaeological sites in the hinterland of El Argar culture have their origins in mineralisations from outside the El Argar area, mainly from Los Pedroches (Sierra Morena). However, c. 37%–41% of the objects from the hinterland of El Argar culture have yet to be associated with any known mineralisation. Although some of the resources used by El Argar communities may have been completely exhausted and no further evidence might have been left, there are still important *lacunae* of geological characterisation of many of the copper resources known within El Argar territory. Further regional studies like the one presented here will contribute toward filling in this lack of information and in providing a clearer overview of the Bronze Age metallurgical exploitation system.

FIGURE 8 | Isotopic ratios of archaeological objects from Terrera del Reloj (green) and Cuesta del Negro (yellow). A copper prill, a bronze lump and copper slag are plotted in black, as these are not objects but production remains. Note that the rivets of daggers P3700 and P3676 do not plot close to their blades, which could suggest that they were not manufactured with the same metal. The same is true for the two rings from Tomb 13 at Terrera del Reloj (DG4261-1 and DG4261-2), which could indicate that these pieces were not made specifically for the grave goods but were accumulated during the lifetime of the deceased. Production remains (prill, slag and lump) are plotted in a close cluster in all graphs, with a very close match of the copper ring from Burial 11 at Terrera del Reloj (DG4325) and the copper prill from the same site, which could suggest in situ production.

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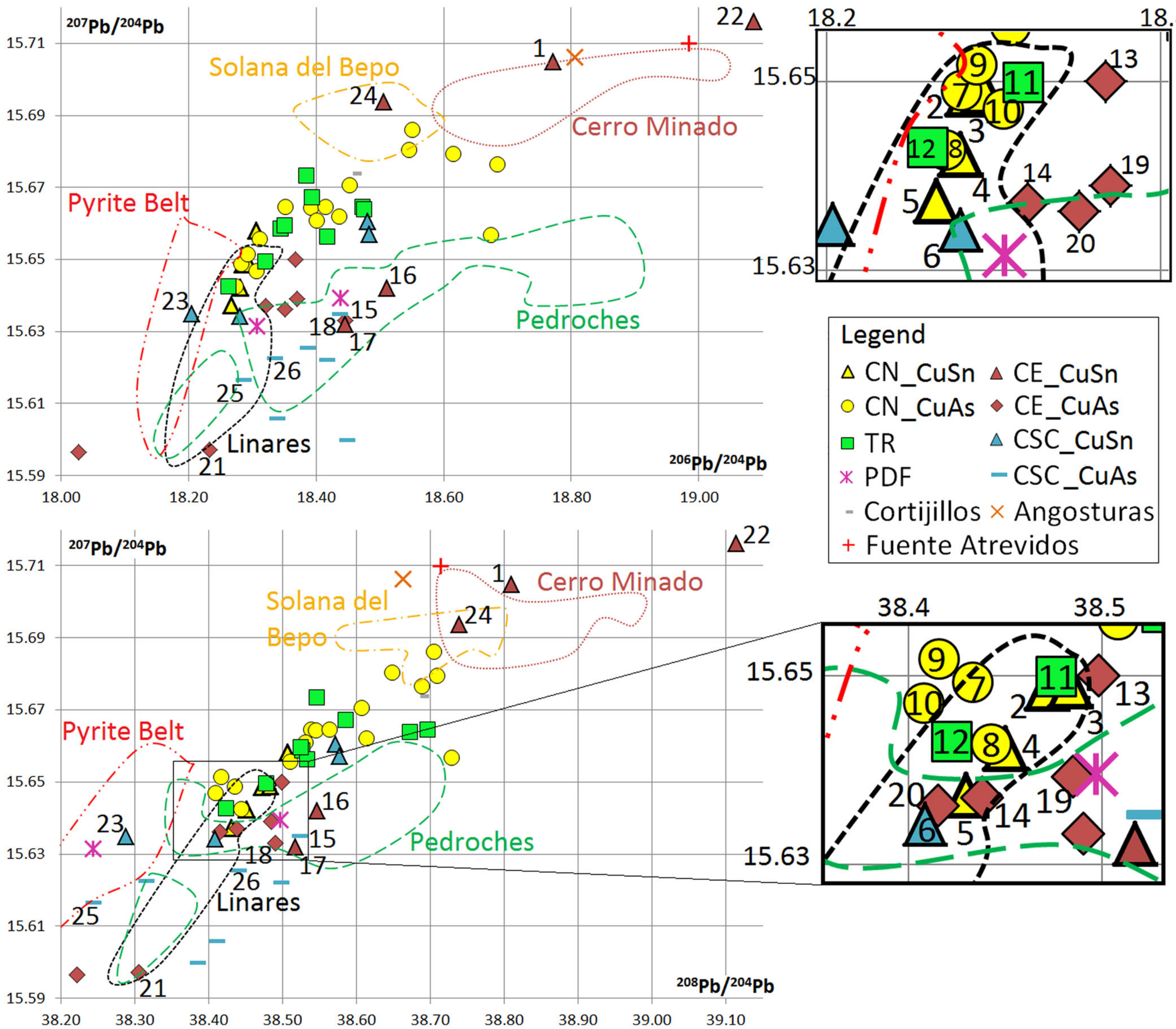

FIGURE 10 | Isotopic ratios of archaeological objects from all El Argar archaeological sites in the area (CE, Cerro de la Encina; CN, Cuesta del Negro; CSC, Cerro de San Cristóbal; PDF, Puebla de Don Fadrique; TR, Terrera del Reloj) and copper ore samples from the Copper Age settlements of Las Angosturas, Cortijillos and Fuente Atrevidos, projected on top of the isotopic field of other mining districts of El Argar territory (Linares in Jaén and Cerro Minado in the Vera Basin), as well as mining districts outside El Argar territory (Solana del Bepo in northeastern Iberia, the Pyrite Belt in southwestern Iberia or Los Pedroches in northern Córdoba province). 1. Bronze bracelet MO39255 from Cerro de la Encina, 2. Arrowhead P60002A, 3. Awl P67006, 4. Thread P45087, 5. Dagger P13164, all from Cuesta del Negro; 6. Bronze bracelet OSC7002 from Cerro de San Cristóbal, 7. Dagger P3676_d, 8. Rivet P65011, 9. Awl P4571, 10. Dagger P11203, all from Cuesta del Negro, 11. Ring DG4261-2, 12. Dagger DG4298, both from Terrera del Reloj, 13. Dagger MO55208, 14. Bracelet MO39279, both from Cerro de la Encina, 15. Dagger OSC15014 from Cerro de San Cristóbal, 16. Bronze awl MO39257, 17. Dagger MO39264, 18. Bracelet MO39261, 19. Awl MO55209, 20. Dagger MO21292, 21. Bracelet MO39258, 22. Dagger MO39281, all from Cerro de la Encina, 23. Scraper OSC11015 from Cerro de San Cristóbal, 24. Bronze ring MO39260 from Cerro de la Encina and 25. Nail OSC13006_71 from Cerro de San Cristóbal. Archaeological data compiled from Murillo-Barroso, Montero-Ruiz and Aranda Jiménez (2015), Álvarez Peanes (2016) and this study. Analytical errors are smaller than symbols. Geological data collected from Hunt (2003), Klein et al. (2009), Montero-Ruiz (2017), Murillo-Barroso et al. (2019), Sáez et al. (2021), Santos Zalduegui et al. (2004) and STOS-GALE et al. (1995).

Beyond these inland areas of El Argar culture, lead-isotope analyses of 64 archaeological objects have been already published, mainly from the coastal strip (Almería-Murcia) (Brandherm et al. 2022; Murillo-Barroso et al. 2024; Stos-Gale, Hunt, and Gale 1999). It is noteworthy that up to 18.7% of the archaeological objects from the Almería-Murcia coastal strip (especially from the Vera basin) can be related to the mineralisations corresponding to the inland area of El Argar culture studied here (Figure 11). This is in contrast to the finds from the archaeological sites closer to these copper deposits, such as Cerro de la Encina and Cerro San Cristóbal.

The dagger from Piedras de Canjayar, Alto Andarax (southeastern Sierra Nevada) plots in all bivariate diagrams with the Alpujárride Complex, which is close to the site, while the dagger from Monteagudo (to the east of the city of Murcia)

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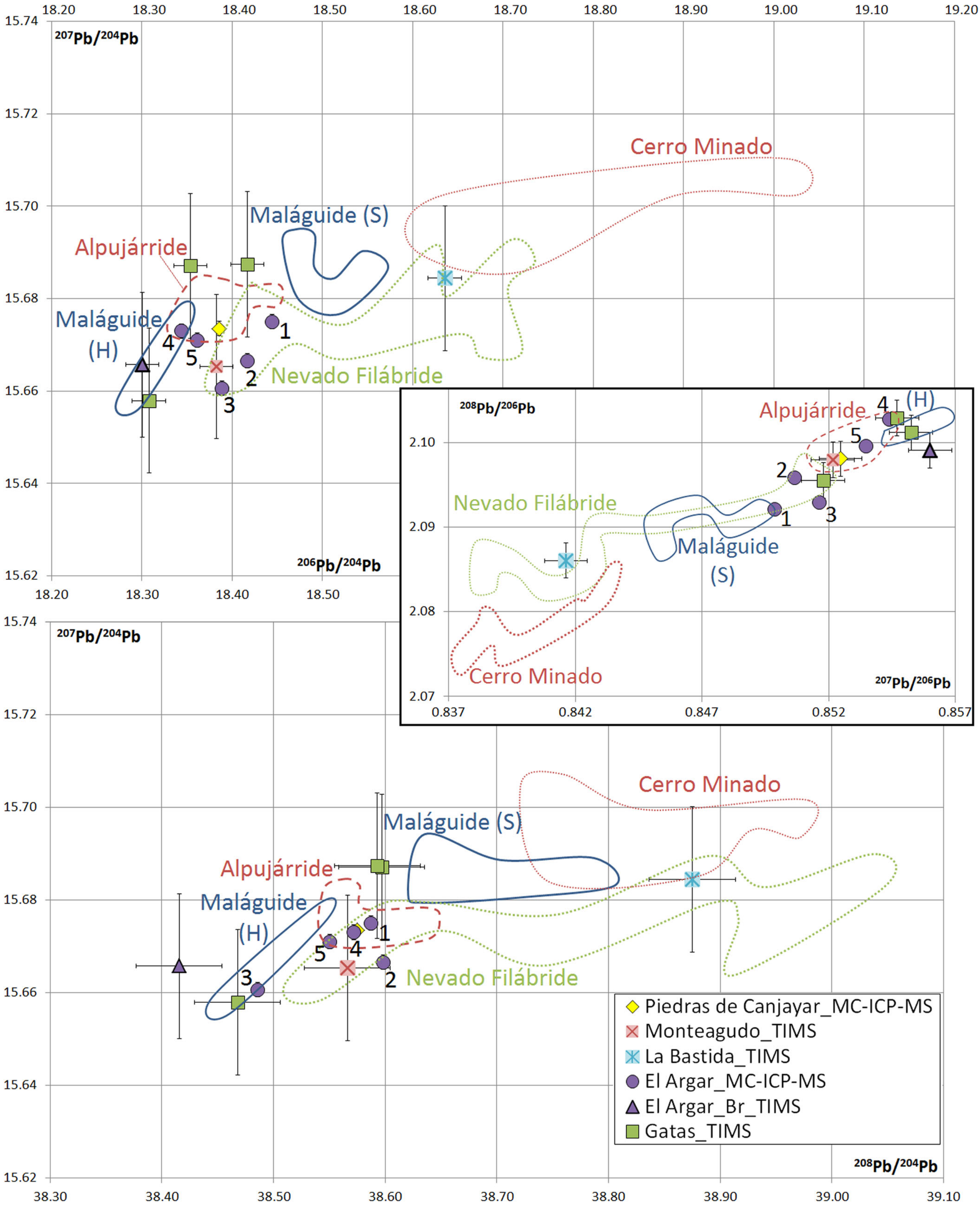

FIGURE 11 | Isotopic ratios of archaeological objects from the coastal regions of El Argar territory (Almería and Murcia provinces) projected on top of the isotopic field of the mining districts analysed in this paper. Note that some objects were analysed in 90 s using TIMS (analytical errors shown) and recent analyses were conducted by MC-ICP-MS (analytical error smaller than symbols). Archaeological data from STOS-GALE et al. (1995) and Murillo-Barroso et al. (2024). 1. Sword from T429 at El Argar, 2. Bracelet from T429 at El Argar, 3. Dagger from T716 at El Argar, 4. Axe from T958 at El Argar and 5. Arrowhead from domestic contexts at El Argar.

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matches the Nevado Filábride Domain, although due to their large analytical error, it could also be associated with the Alpujárride Complex, to which it is better associated in the $^{207}Pb/^{206}Pb–^{208}Pb/^{206}Pb$ ratio. For its part, the dagger from La Bastida (southern Sierra Espuña) plots in an intermediate position between the Nevado Filábride Domain and Cerro Minado (Sierra de Almagro, in the Vera basin) and, given its greater analytical error, both possibilities would be feasible. However, it is better associated with the Nevado Filábride Domain ratio in the $^{208}Pb/^{204}Pb–^{206}Pb/^{204}Pb$ bivariate diagram and especially in $^{207}Pb/^{206}Pb–^{208}Pb/^{206}Pb$ (Figure 11), in which the analytical error of the TIMS analyses is much lower, making a provenance of this sample from the inner area more likely in isotopic terms.

We can also associate five objects from El Argar archaeological site with copper deposits from the inland area of El Argar culture. The sword (N.1 in Figure 11) and bracelet (N.2) from T429 and the dagger from T716 (N.3) match the isotopic field of the Nevado Filábride Domain, although the last one drifts slightly at $^{208}Pb/^{204}Pb$. The axe from T958 (N.4) and an arrowhead from domestic contexts at El Argar (N.5) can be associated with the Alpujárride Complex. Two other objects from Gatas, bracelet T23b and foil G91-2C-M1, plot in all bivariate graphs within the Alpujárride Complex in a peripheral area. However, we cannot rule out this provenance, given the analytical error of these pieces. The bracelet from Tomb 34 at Gatas could also be associated with the Maláguide Complex (Figure 11).

Therefore, the exploitation of the resources from the hinterland area of El Argar culture (the interior of the eastern and central Betic Cordillera mountains) for the supply of copper becomes obvious, both to the settlements surrounding these copper outcrops and to those on the Almería-Cartagena coastline.

7 | Conclusions

This study has expanded the trace element and lead-isotope geochemical data set of copper deposits in El Argar territory with new data from mines in the hinterland of this Bronze-Age culture. The enlarged data set improves our knowledge of the metal exchange networks established both within the El Argar region and with external communities. We have a total of 64 copper-based objects analysed from archaeological sites along the Almería-Cartagena coastline and another 58 from inland sites, making it possible to compare both areas with a similar volume of analysed materials. With regard to the origins, the current scientific information suggests that the exploitation of copper resources from El Argar culture hinterland studied here would have had a comparable relevance to that of the copper resources of the mining district of Linares (Jaén), on the fringe of the El Argar culture domain. Hence, between 17.2% and 24.1% of the archaeological objects from the inland El Argar area would have had a clear origin in the mining district of Linares (Jaén), while 13.8% would have been manufactured with copper ore from the hinterland of El Argar area. In the case of the archaeological sites on the coastal strip of Almería-Murcia, 20.4% of the objects had their origin in Linares (Jaén) and 18.7% in the inland mineralisations studied in this article.

In terms of processing, we know that copper resources from the mining district of Linares (Jaén) were being processed in large quantities at the metallurgical site of Peñalosa (Jaén), while in the settlements of the inland El Argar area, scattered evidence of production is known at the archaeological sites of Cuesta del Negro, Cerro de la Encina, Cerro de la Virgen and Terrera del Reloj (Figure 1).

The representativeness of the copper deposits in the coastal area of Almería-Cartagena is considerably less. Only 6.9% of the archaeological objects from the inland El Argar area would have come from those coastal mining resources, while 8.2% of the archaeological finds on the Almería-Cartagena coast would have been made with ore mined on the coast itself. The data that we have today appear to indicate that the copper deposits intensively exploited during the Copper Age (Delgado-Raack, Escanilla, and Risch 2014; Murillo-Barroso et al. 2020) were later used with less intensity, in favour of the mining resources of other El Argar areas (interior of the Betic Cordillera from Granada to Baza) or very nearby (foothills of the Sierra Morena-Linares mining district), or even from outside El Argar territory itself (such as Los Pedroches and the Iberian Pyrite Belt). 17.2% of the objects from archaeological sites in the inland of the El Argar area would have had an origin from outside the El Argar area (mainly Los Pedroches, Córdoba). However, for 56.3% of the archaeological finds, a specific provenance cannot currently be proposed.

In general, the intensity of metallurgical activity observed in the Copper-Age Almería-Cartagena coastal area seems to have decreased in favour of the inland El Argar areas (from Granada to Jaén). The exploitation of these copper mining resources, especially those of the Linares mining district, was not limited to the supply of metal to the inland of the El Argar area, since the metal from the Linares mining district is also documented in other more distant areas of the Iberian Peninsula, such as Alicante in the Iberian Levant, or the central Meseta (plateau) (Benítez de Lugo Enrich et al. 2022; Galindo San José, Marcos Sánchez, and Montero Ruiz 2018; Montero-Ruiz et al. 2014).

However, this greater metallurgical intensity in the interior of the Sierra Morena, also geared toward trade with the exterior, does not necessarily imply a dependence on the coastal deposits in terms of copper supply, as we observe both the use of various mineralisations and the arrival of copper from other areas outside the El Argar area in similar quantities. These lead-isotope data distributions reflect the decentralisation of the metallurgical activities and processes.

Future advances in the detailed characterisation of the quantity and diversity of mineral resources in the south of the Iberian Peninsula will allow us to better understand the copper exploitation and exchange networks as a resource in El Argar society.

Author Contributions

M. Murillo-Barroso: writing–original draft, funding acquisition, writing–review and editing, conceptualisation, supervision, visualisation. **G. Aranda Jiménez:** writing–review, editing, resources. **J. A. Lozano Rodríguez:** resources, writing–review, editing, visualisation. **A. Lackinger:** writing–review and editing, conceptualisation. **Z. Stos-Gale:** writing–review and editing, resources. **J. Rodríguez:** writing–review, editing, resources,

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validation, formal analysis. **J. I. Gil Ibarguchi:** writing–review, editing and resources. **I. Montero-Ruiz:** conceptualisation, writing–review and editing, resources.

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Data Availability Statement

The data that support the findings of this study are available in the supplementary material of this article.

Endnotes

[1] However, recent radiocarbon dating developments revealed that the area traditionally considered as the core area of the El Argar culture in the Vera basin can be expanded to neighbouring regions. Radiocarbon dates as early as those at sites such as Gatas (Lull et al. 2009, 2011) and Fuente Álamo (Schubart 2012) in the Vera basin can be found in La Bastida and La Almoloya in the Murcia region (Lull et al. 2015), Laderas del Castillo and Tabayá in Alicante (Hernández Pérez, López Padilla, and Jover Maestre 2019, 2021) and Cerro de la Virgen in the Guadix basin (Molina González et al. 2014). This discussion exceeds the scope of this paper, but the expansion of the El Argar society to the inland areas in successive phases should be reconsidered.

[2] The samples of these artefacts were sent by Prof. A. Arribas some decades ago to N. Gale and Z. Stos-Gale for analysis at the Isotrace Laboratory (University of Oxford, UK). These samples were not analysed there and, thanks to this collaboration, have recently been analysed at the University of the Basque Country.

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