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# *Shine on you crazy diamond: Symbolism and social use of fluorite ornaments in Iberia's late prehistory*

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## **Abstract:**

Fluorite ornaments have been recorded in different sites of Europe since Upper Paleolithic. Due to its visual appearance and physical properties, some translucent or transparent mineralogies like fluorite were searched for or casually acquired by late prehistory's human communities. After intensive research on archaeological contexts from the Iberian Peninsula with personal ornaments from 4<sup>th</sup> to 2<sup>nd</sup> millennia BCE, we have recently identified and characterized for the first time an important number of fluorite ornaments, confronting a previous background where little attention was paid. Our work has been carried out in different archaeological collections and museums from the whole Iberian Peninsula by non-destructive techniques (Raman spectroscopy, portable X-ray fluorescence (p-XRF) and X-ray Diffraction (XRD), that revealed the nature of fluorite ornaments and points to its consideration as scarce and highly symbolic items during late prehistory. A total of 36 fluorite beads from 23 sites are here recorded and studied, many of them unpublished or wrong catalogued as other mineralogies. These adornments could have important roles in trade and use among the communities of Iberia from the 4th millennium BCE onwards, because of their scarcity and its recurrent association with important funerary complex and exotic materials. Fluorite ornaments could have been significant and special symbols in the development of new and exclusive raw materials in the context of increasing social complexity and inequality.

**Keywords:** adornments; beads; translucent; fluorite; Iberian Peninsula; late prehistory; Raman spectroscopy; XRD

## **1. Introduction**

Personal adornments (beads, pendants, charms, *etc.*) are determinants to understand symbolic behaviour and identity constructions in prehistoric societies (Bar-Yosef Mayer & Porat 2008; Wright & Garrard 2003), giving key information about the metaphoric codes used



(Tilley 1999) by colour, shape, brightness, and others (Gaydarska & Chapman 2008; Jones & MacGregor 2002).

In the Iberian Peninsula, apart from the long-known social value of amber (for a review see Odriozola *et al.* 2019), transparent or translucent beads are rare, and when recorded they were generally classified as quartz or rock crystal with no further analysis than that of the naked eye. However, besides quartz varieties, there are many more transparent minerals that are not usually considered, for example fluorite, calcite, and many sheet silicates that when thinned to a certain level are capable of transmitting light (Baysal 2017: 6-7). The social value of translucent ornaments made of those minerals would partially stem from its visual and physical properties and its scarcity in the archaeological record (Garrido-Cordero *et al.* 2020a; 2020b).

Fluorite (CaF<sub>2</sub>) has recently received some attention by researchers in different European late prehistory's contexts (Cardoso *et al.* 2012; Garrido-Cordero *et al.* 2020a; 2020b; Goemaere & Philippo 2010; Goemaere *et al.* 2013; Honings *et al.* 2014; Jungels & Goemaere 2007). Fluorite mineral is relatively frequent geologically in the Iberian Peninsula and Western Europe (Anthony *et al.* 2011; Galán & Mirete 1979), but infrequent in archaeological record.

This paper is focused in the role of fluorite adornments as highly scarce and symbolic items, used among the communities of Iberia's late prehistory, a subject on which little research has been previously carried out. The starting point for this study is a remarkable set of unpublished translucent beads that we have characterized and gathered for the first time. The chemical and contextual analysis of these beads inventoried throws new light on the use and social significance of translucent personal adornments during the increase of social complexity from Late Neolithic to Early Bronze Age.

### 1.1. Fluorite ornaments in late prehistory: The broader European context

In Europe, the small-scale exploitation and consumption of fluorite has been documented in Belgium at Upper Paleolithic sites, mostly during the Magdalenian period (Goemaere *et al.* 2013; Jungels & Goemaere 2007). In some cases, a large quantity was used, and the mineral was worked *in situ* (440 g of fluorite at the site of Chaleux) and distributed locally and regionally for the manufacture of adornments (Goemaere *et al.* 2013). No prehistoric fluorite mines have been identified in Europe. In fact, ethnographic data suggest that outcrops of this type of raw material could have been exploited just occasionally and opportunistically by small groups without any complex logistical organization (Brandl & Trnka 2014: 124).

The frequency of its consumption for objects of personal adornment seems to increase during the Neolithic, but is still small compared with other raw materials. In the late 5<sup>th</sup> and early 4<sup>th</sup> millennia BCE, some Belgian sites have yielded both manufactured fluorite fragments and others in the process of being made into beads and pendants (Delye *et al.* 2011; Vermeersch *et al.* 1990). At Thieusies, several worked fluorite fragments in different colours and four finished beads came to a total weight of 158.4 g (Vermeersch *et al.* 1990: 51-52).

From the Late Neolithic to the Copper Age (5<sup>th</sup>-3<sup>rd</sup> millennia BCE) the number of sites where this raw material was used to make adornments increases considerably. Fluorite beads have been found at Neolithic sites in the north of France (Hauzeur & Cauwe 2012: 39), in the Paris Basin (Polloni 2008) and in the south-west, especially between Perpignan and Narbonne (Roscian *et al.* 1992: 233-234). Their distribution coincides with the main outcrops of fluorite in the Central Massif and the Pyrenees, and supports the idea of local exploitations near the outcrops and distribution on a regional scale. In Predynastic Egypt (4<sup>th</sup> millennium BCE), fluorite ornaments are also reported (Nai 2014: 77).

Fluorite continued to be used occasionally for personal adornments in the 2<sup>nd</sup> millennium BCE. Fluorite beads are referred at Bronze Age levels in Mol, Belgium (Warmenbol 2001) or Midea, Greece (Demakopoulou *et al.* 1996).

Unlike other parts of Europe, no personal adornment made from fluorite is known in the Iberian Peninsula before the 4<sup>th</sup> millennium BCE, although many sites with fluorite have not been dated by radiocarbon or the finds are not directly associated with a dated level.

Before this paper, only Cardoso *et al.* (2012) confronted regional identification of five fluorite beads in Portugal. Also, fluorite beads were reported in the Iberian peninsula in the megaliths of Gabrieles 6 (Blanco & Rothenberg 1981: 284), La Velilla (Villalobos 2015: 304), Mámoa 5 do Leandro (Ribeiro & Loureiro 2015) and dolmen of Areita (Gomes *et al.* 1998), while one bead of fluorite was identified for the Bronze Age tomb 111 of Fuente Álamo (Pozo *et al.* 2002).

## 2. Materials and methods

A total of 23 sites in the Iberian Peninsula with a total number of 36 items made of fluorite are reported in this work (Figure 1, Table 1). Archaeological background on the sites studied here can be found at Supplementary Material 1. From this total, 28 fluorite beads from 20 sites were directly analyzed for this paper (Figure 2) by Raman spectroscopy and portable XRF. Also, X-ray diffraction (XRD) has been possible to perform when the museums granted the transport of materials to the laboratory.

Table 1. Inventory of fluorite items in Iberian late prehistory. Period legend: LN, Late Neolithic; C, Copper Age; B, Bronze Age. Mineralogy legend: F: fluorite; C: calcite; Q: quartz; Ame: amethyst; Ca: Calcium; Si: Silicon; PL: Photoluminescence.

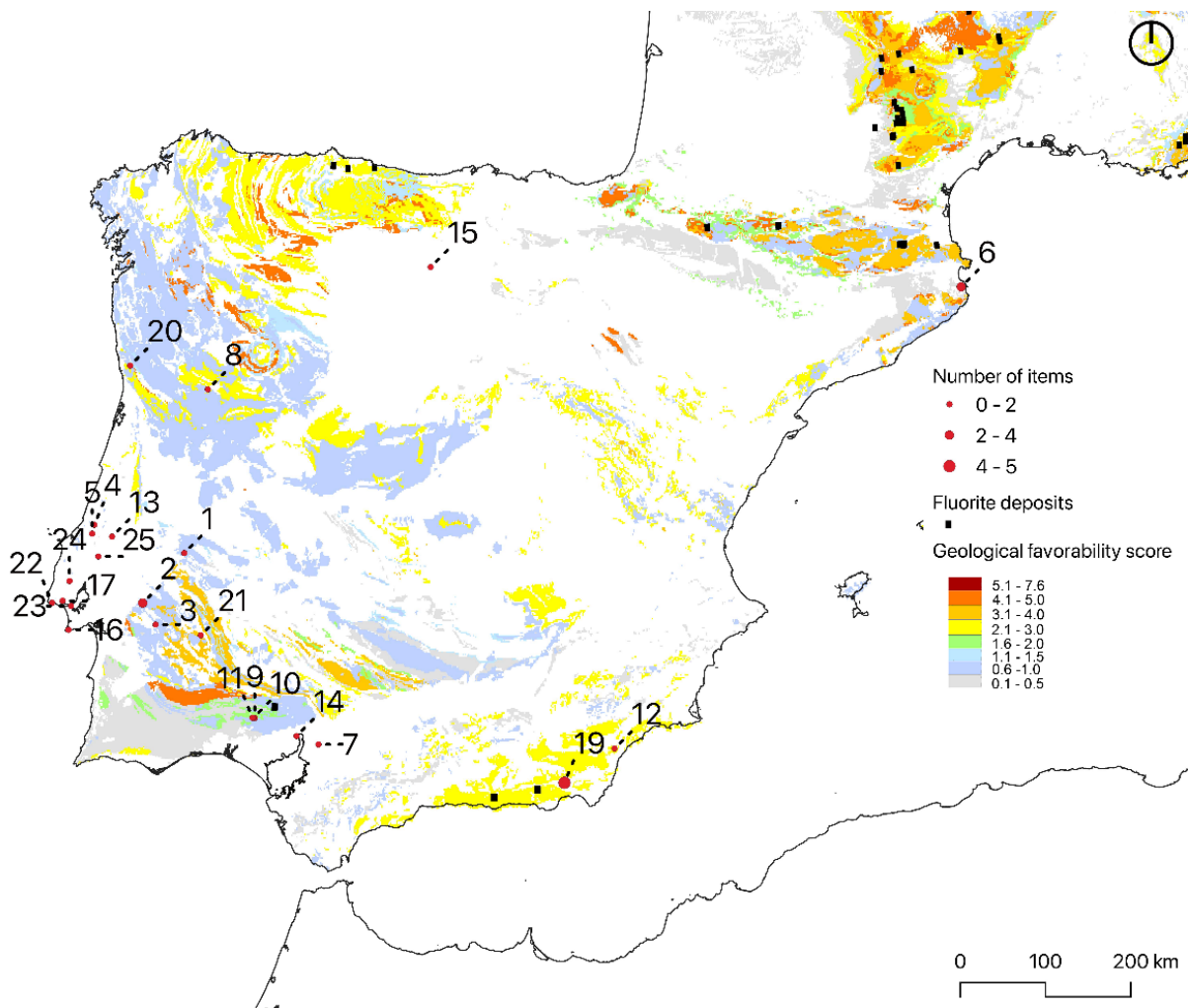
Site	ID	Type	Period	Expected mineralogy	Actual mineralogy	Colour	Weight (g)	Techniques and results		
								p-XRF	Raman	XRD
Casa da Moura	CMR-436	natural cave	LN, C	F	F	transparent smoky	22.6	Ca	F (T <sub>2g</sub> + PL)	-
Poço Velho	CCG-159	natural cave	LN, C	F	-	green	-	Si	-	-
Leceia	LC93-TA-C3	site - fortified	C	F	F	yellow, green	-	Ca	F (PL)	F
Vila Nova de São Pedro	ARQ-VNSP-964-46	site - fortified	C	-	F	transparent smoky	0.7	Ca	-	-
Lapa do Bugio	LB-0011-3	natural cave	LN, C	F	F	green	10.6	Ca	F (PL)	-
Lapa do Bugio	LB-0011-4	natural cave	LN, C	F	F	green	7.2	Ca	F (PL)	-
São Paulo 2	MMA-5743	artificial cave	LN, C	-	F	pale green	26.3	Ca	F (PL)	-
Olival da Pega 1	21815A	megalith	LN, C	-	F	green	6.5	Ca	F (PL)	-
Olival da Pega 1	21815B	megalith	LN, C	-	F	green	2.0	Ca	F (PL)	-
Anta Grande da Comenda da Igreja	2011.54.278	megalith	LN, C	-	F	white	19.8	Ca	F (T <sub>2g</sub> + PL)	-
Anta Grande da Comenda da Igreja	985.51.617	megalith	LN, C	-	F	white	16.0	Ca	F (T <sub>2g</sub> + PL)	-
Anta Grande da Comenda da Igreja	985.51.670	megalith	LN, C	-	F	pale green	3.7	Ca	F (PL)	-
Tituaría	998.16.42	<i>tholos</i>	C	C	F	yellow, green	3.6	Ca	F (PL)	-
Anta dos Penedos	PSM-112	megalith	LN, C	-	F	green	2.6	Ca	F (PL)	-

Site	ID	Type	Period	Expected mineralogy	Actual mineralogy	Colour	Weight (g)	Techniques and results		
								p-XRF	Raman	XRD
de São Miguel										
Gruta da Marmota	MRO/S-25	natural cave	LN	-	F	green	8.0	Ca	F (PL)	-
Gruta da Marmota	MMT-26	natural cave	LN	-	F	green	5.1	Ca	-	F
Cabeço da Ministra	25992-1	natural cave	LN, C	-	F	green	5.6	Ca	-	-
Cabeço da Ministra	25992-2	natural cave	LN, C	-	F	green	5.0	Ca	-	-
Mámoa 5 do Leandro	-	megalith	LN, C	F	-	-	-	-	-	-
Dolmen de Areita	ART-96-60	megalith	LN, C	F	-	pale blue	-	-	-	-
El Pozuelo 1	IG-313/51	megalith	LN, C	Q	F	transparent, yellow	1.7	Ca	F (PL)	-
El Pozuelo 5	-	megalith	LN, C	Q	F	green	2.0	Ca	F (PL)	F
El Pozuelo 7	1848	megalith	LN, C	Q	F	yellow, green	3.2	Ca	F (T <sub>2g</sub> + PL)	-
Los Gabrieles 6	IG-3486/22	megalith	LN, C	F	F	green	21.2	Ca	F (PL)	-
Cueva del Vaquero	-	tholos	C	Q	F	green	90.1	Ca	-	-
“La Emisora”	-	artificial cave?	C	-	F	green	1.2	Ca	-	-
Los Millares 12	-	tholos	C	Ame.	F	violet	-	Ca	F (T <sub>2g</sub> )	-
Los Millares 12	-	tholos	C	Ame.	F	violet	-	Ca	F (T <sub>2g</sub> )	-
Los Millares 12	-	tholos	C	Ame.	F	violet	-	Ca	F (T <sub>2g</sub> )	-
Los Millares 12	-	tholos	C	Ame.	F	violet	-	Ca	F (T <sub>2g</sub> )	-
Los Millares 12	-	tholos	C	Ame.	F	violet	-	-	-	-
La Velilla	-	megalith	LN, C	C	F	green	-	Ca	-	-
Cau de l'Olivar d'en Margall	16477-7	natural cave	C	C	F	translucent green	3.1	Ca	F (PL)	-
Cau de l'Olivar d'en Margall	16477-17	natural cave	C	C	F	translucent white	1.6	Ca	F (PL)	-
Anta Grande do Zambujeiro	ME-3760	megalith	C	-	F	pink	26.5	Ca	F (T <sub>2g</sub> )	F
Fuente Álamo, tomb 111	VI-1	pithos	B	F	F	translucent	-	Ca	F (T <sub>2g</sub> + PL)	-
Fuente Álamo tomb 111	DJ83773	pithos	B	F	F	transparent	0.5	Ca	F (T <sub>2g</sub> + PL)	-

Raman spectroscopy was performed using a portable BWTEK iRaman Plus device. The laser diode operated with a wavelength of 785 nm produces a power of up to 420 mW in the laser port. Filters were not used to reduce the power of the laser. The selected range of the measurement spectrum was between 150 and 3300 cm<sup>-1</sup> with a high efficiency quantum CCD detector. The selected measurement accuracy was 4 cm<sup>-1</sup>. The measurement conditions, as regards the laser power and integration time have varied from one object to another in order to obtain the best possible Raman signal.

Chemical composition was measured by an Oxford Instrument XMET-7500 p-XRF equipped with a Rh tube, a silicon drift detector (SDD), and an automatic 5-position filter changer. Quantification was obtained using the SOILS-LE program based on fundamental parameter (FP) method. This method is the most appropriate when no standardized method is

available or when a large number of elements have to be analyzed (Beckhoff *et al.* 2006: 403).



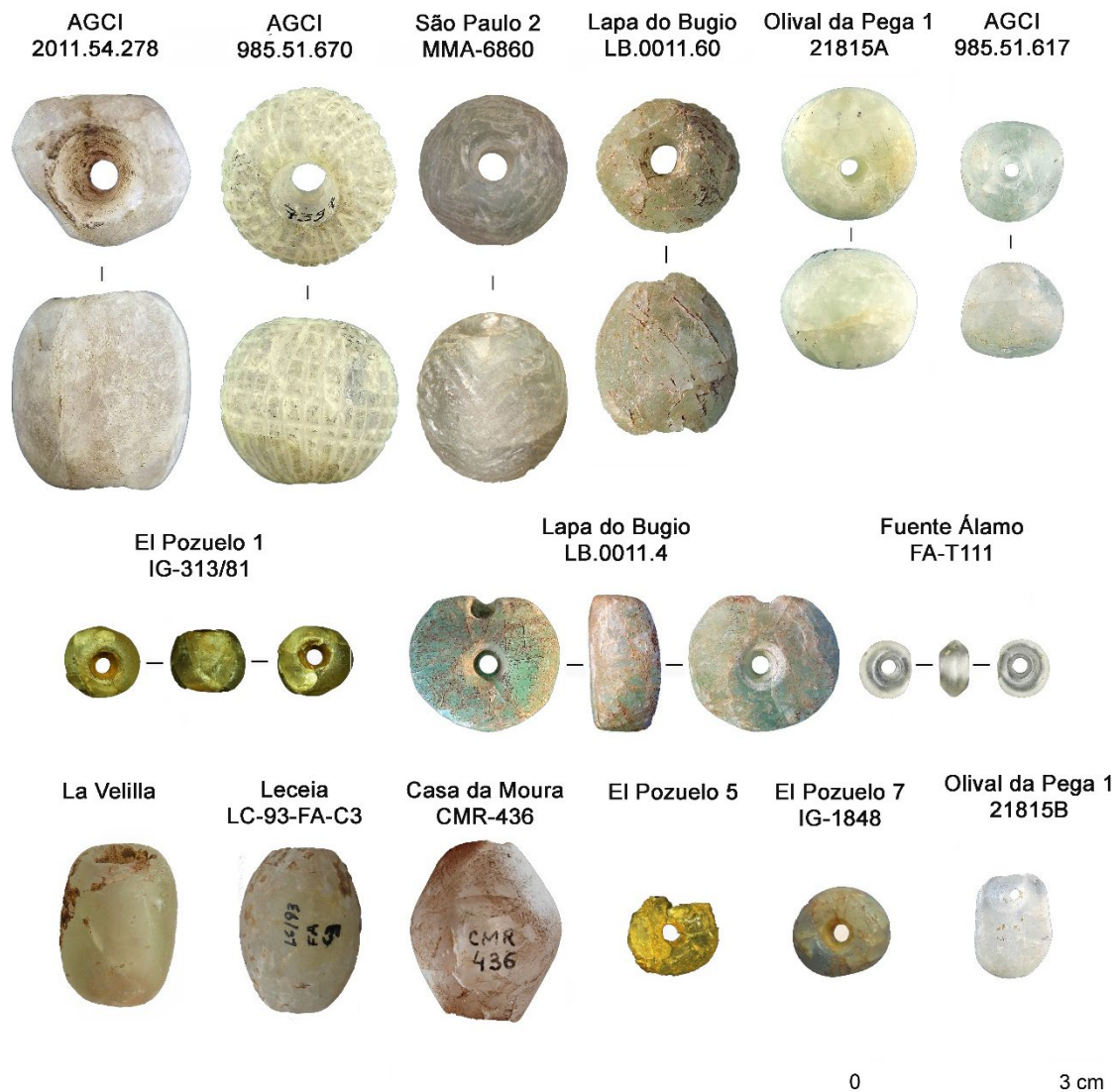
**Figure 1.** Iberian late prehistory sites and materials cited and studied in the text, compared with fluorite geological occurrence in the Iberian Peninsula. 1. Anta Grande da Comenda da Igreja. 2. Anta Grande do Zambujeiro. 3. Anta dos Penedos de São Miguel. 4. Cabeço da Ministra. 5. Casa da Moura. 6. Cau de l'Olivar d'en Margall. 7. Cueva del Vaquero. 8. Dolmen de Areita. 9. El Pozuelo 1. 10. El Pozuelo 5. 11. El Pozuelo 7. 12. Fuente Álamo. 13. Gruta da Marmota. 14. La Emisora (Valencina). 15. La Velilla. 16. Lapa do Bugio. 17. Leceia. 18. Los Gabrieles 6. 19. Mámoa 5 do Leandro. 20. Necrópolis de Los Millares. 21. Olival da Pega 1. 22. Poço Velho. 23. São Paulo 2. 24. Tituarria. 25. Vila Nova de São Pedro.

XRD analysis was performed using a Panalytical X'Pert Pro  $\theta/\theta$  diffractometer equipped with Cu K $\alpha$  source (1.5406 Å) operating at 45 kV and 40 mA. A PixCel detector was used and the data were collected on transmission mode with a 2D detector. Patterns were obtained using a step width of 0.053° 2 $\theta$  between 5° and 70° 2 $\theta$  and a counting time of 155 s per step at ambient temperatures. An incident beam PreFIX module with X-ray mirror for Cu radiation was used to allow non-destructive analysis.

### 3. Results and discussion

Of the 25 Raman analyzed beads only a set formed by Los Millares translucent pale violet beads, El Pozuelo 7 colourless bead and the translucent pale pink beads from Casa da Moura, Anta Grande da Comenda da Igreja, Anta Grande do Zambujeiro and Fuente Álamo tomb 111 (CMR-436, AGCI-2011.154.278, AGCI-985.51.617, ME-3760, FA-T111-DJ83773

and FA-T111-VI-1) records the fluorite diagnostic  $T_{2g}$  Raman band at *c.*  $320\text{ cm}^{-1}$ . Therefore, this set of 11 beads are most likely fluorite (Figure 3).



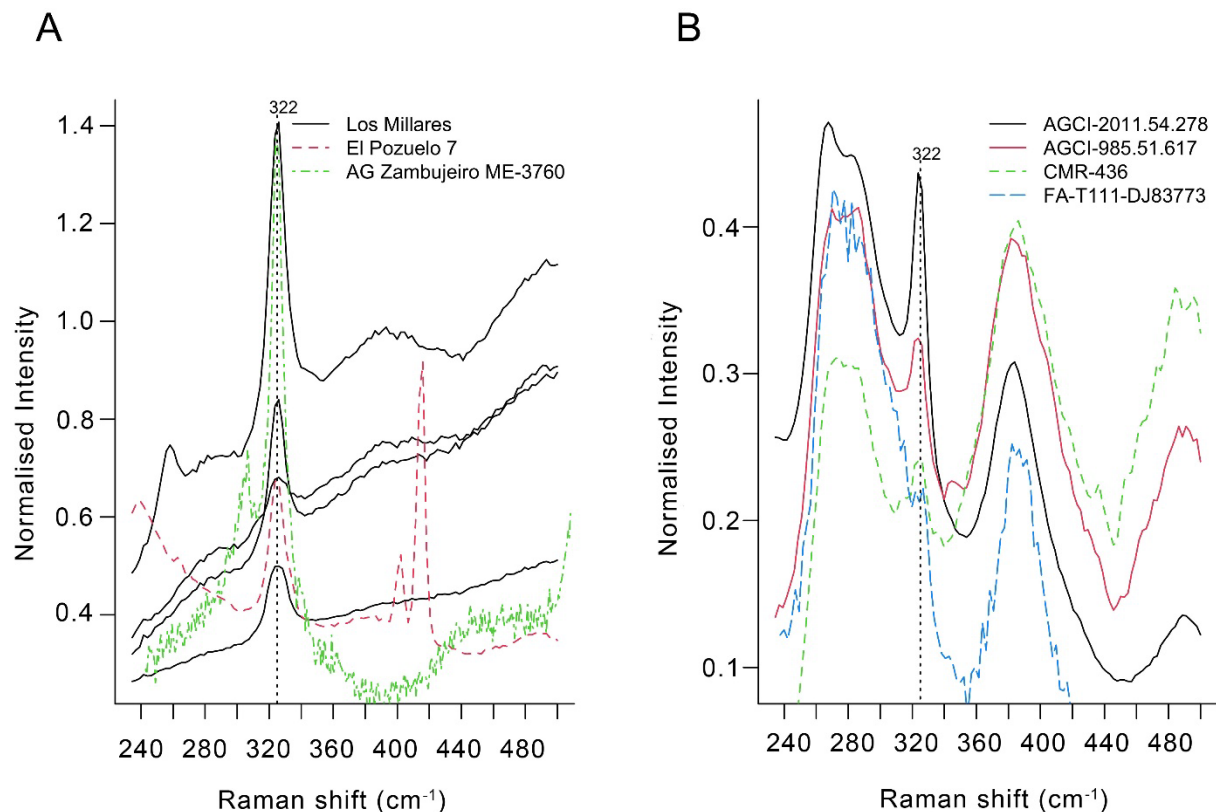
**Figure 2.** A sample of beads analyzed in this paper.

However, the translucent pale pink beads (CMR-436, AGCI-2011.154.278, AGCI-985.51.617, FA-T111-DJ83773 and FA-T111-VI-1) accounts for additional bands to the  $T_{2g}$  Raman active band at lower and higher frequencies that remain to be explained (Figure 3, right). These additional bands to the fluorite diagnostic  $T_{2g}$  Raman band may be caused by 1) the accommodation of Y, Sr, Ba ... and REEs in substitution of Ca (Chen & Stimets 2014; Cherniak *et al.* 2001; Lenz *et al.* 2015; Sverdrup 1968; Tu & Sievers 2002); 2) by radiation-induced defects (Alencar *et al.* 2016), which could also cause colourless fluorites to become coloured (Alencar *et al.* 2016; Schmalzl *et al.* 2003); or 3) as argued recently by Vandenabeele and Edwards (2018: 327-328) could be caused by the interference pattern (ripple) generated by the edge filters in luminescent samples.

Beside these features in the diagnostic region, a set of bands appear in the frequency-shift region between  $1000$  and  $2500\text{ cm}^{-1}$  that are the result of the  $785\text{ nm}$  laser induced fluorescence and photoluminescence (PL) (Figure 4). Fluorite PL pattern is most likely due to the presence of  $\text{REE}^{3+}$  ions, mainly Nd, at  $C_{4v}$  sites (Freeth & Jones 1982; Payne *et al.* 1991). PL bands have been formerly proposed as mineral characteristic (Chen & Stimets 2014),



therefore the match between fluorite reference PL spectra and the here recorded would support the identification of all the analyzed specimens as fluorite. However, no exhaustive database exists for mineral identification by means of the PL spectra and their usability is therefore limited and still under research. Recent studies (Zhuk 2017) have shown that in some cases calcite shows PL spectra very similar to that of fluorite leading to a misclassification of the specimen, and therefore preventing the immediate classification of these beads as fluorite. This is true because Ca substitutions by REE elements in calcite crystal lattice result in a similar laser induced photoluminescence to that of fluorite crystals (Gaft *et al.* 2001).

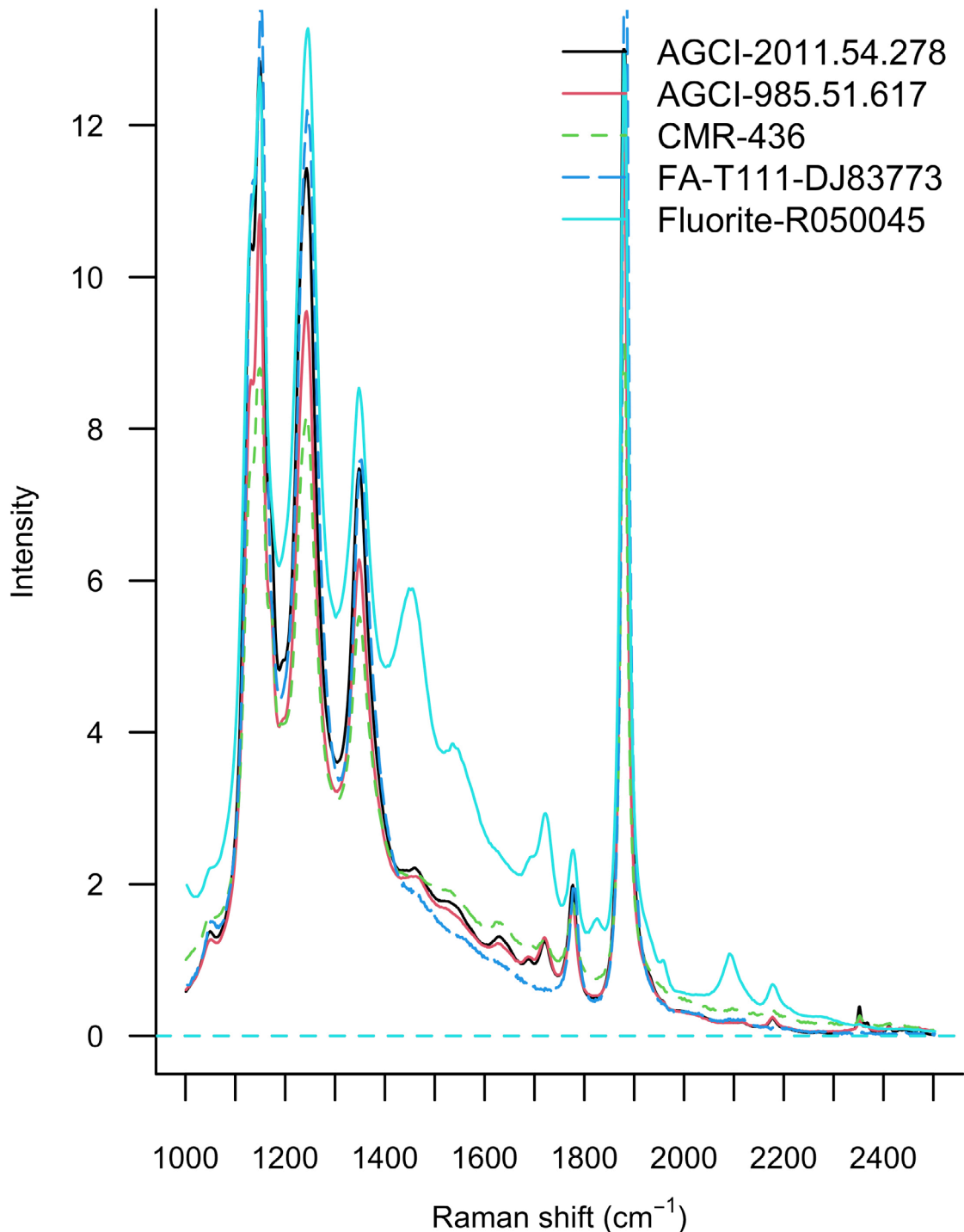


**Figure 3.** Raman spectra in the diagnostic region, **left)** set of samples that shows solely the fluorite diagnostic  $T_{2g}$  Raman band at  $c. 320 \text{ cm}^{-1}$ , **right)** set of samples that shows the fluorite diagnostic  $T_{2g}$  Raman band at  $c. 320 \text{ cm}^{-1}$  and additional bands at lower and higher frequencies.

Except for Los Millares specimens that lack the  $1137 \text{ cm}^{-1}$  band and show differences in position and intensity of the bands in this region, that might be associated with the accommodation of  $\text{REE}^{3+}$  in substitution for Ca (Chen & Stimets 2014; Cherniak *et al.* 2001; Lenz *et al.* 2015; Sverdrup 1968; Tu and Sievers 2002), the spectra of this set of beads (Figure 4) accounts for the typical fluorite PL spectrum (Andò & Garzanti 2014).

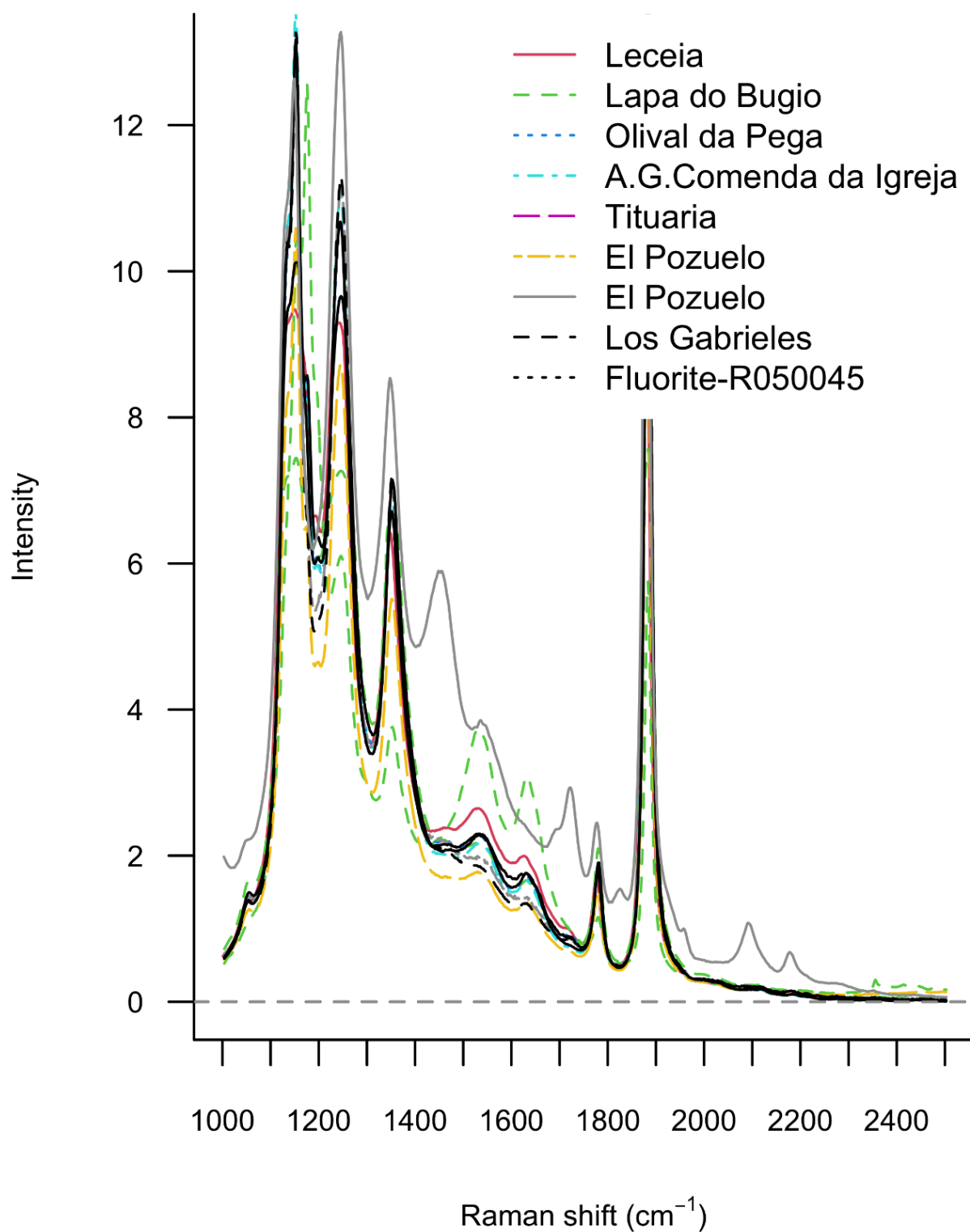
Therefore, based on 1) the presence of the  $T_{2g}$  Raman band, 2) the coincidence of the PL of the specimens with that of fluorite, and 3) the chemical composition of the beads; fluorite is proposed as the main mineral phase for these 11 beads. Even though 15 of the analyzed beads (LB-0011, LB-0012, OP-21815B, OP-21815A, AGCI-985.51.670, TIT-988.16.42, MRO/S-25, SP2-5743, PSM-112, LC93-FA-C3, El Pozuelo 1, El Pozuelo 5, Los Gabrieles 6 and Cau de l'Olivar d'en Margall 16477-7 and 16477-17) lack the  $T_{2g}$  band, their chemical composition and PL spectra (Figure 5) make it impossible for us to exclude fluorite as the main mineral phase of these 15 beads. Calcite is off the table for these 15 specimens because they all lack the calcite Raman spectral features at  $284$ ,  $710$  and  $1085 \text{ cm}^{-1}$  (Chen & Stimets

2014; Sood *et al.* 1981). In addition to p-XRF and Raman spectroscopy, XRD analysis has been performed on a selection of this set of 15 beads and on ME-3760. Leceia LC-93-FA-C3 (see Cardoso *et al.* 2012: 39; Figure 4), Gruta da Marmota MMT-26 and Anta Grande do Zambujeiro ME-3760 matches the standard XRD fluorite pattern (Figure 6, left), while El Pozuelo 5 (Figure 6, right) matches that of fluorite (1-77-2251) and copper selenide [Cu<sub>2</sub>Se]. Although only 4 beads have been submitted to XRD analysis, we might argue that this set of beads are most likely fluorites.



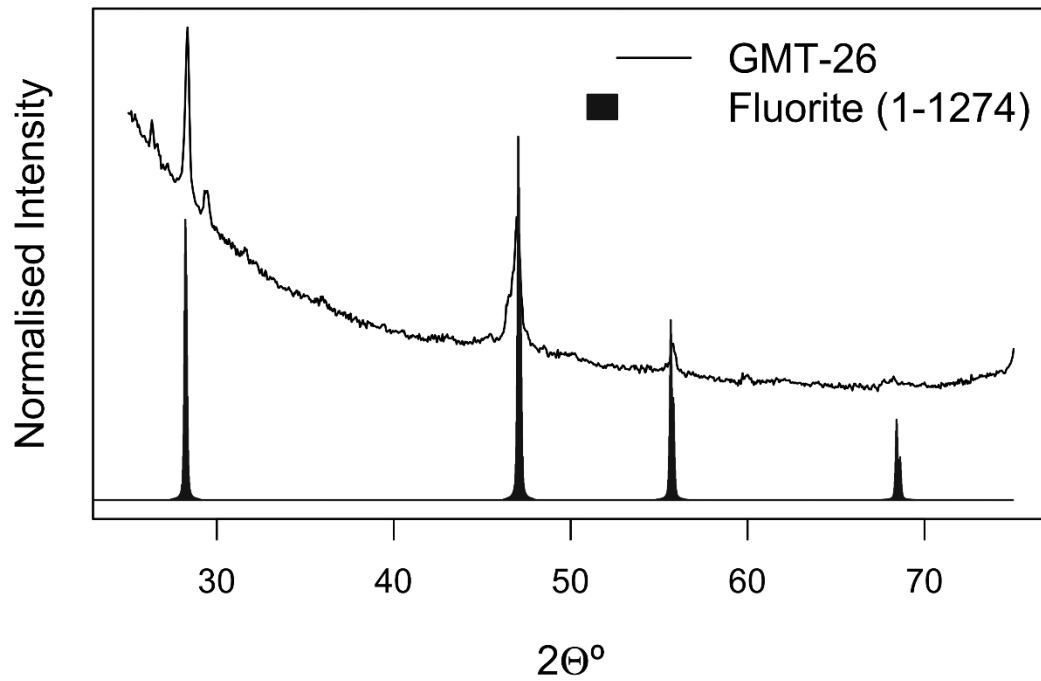
**Figure 4.** Raman spectra in the frequency-shift region between 1000 and 2500 cm<sup>-1</sup> showing the typical fluorite PL spectra.



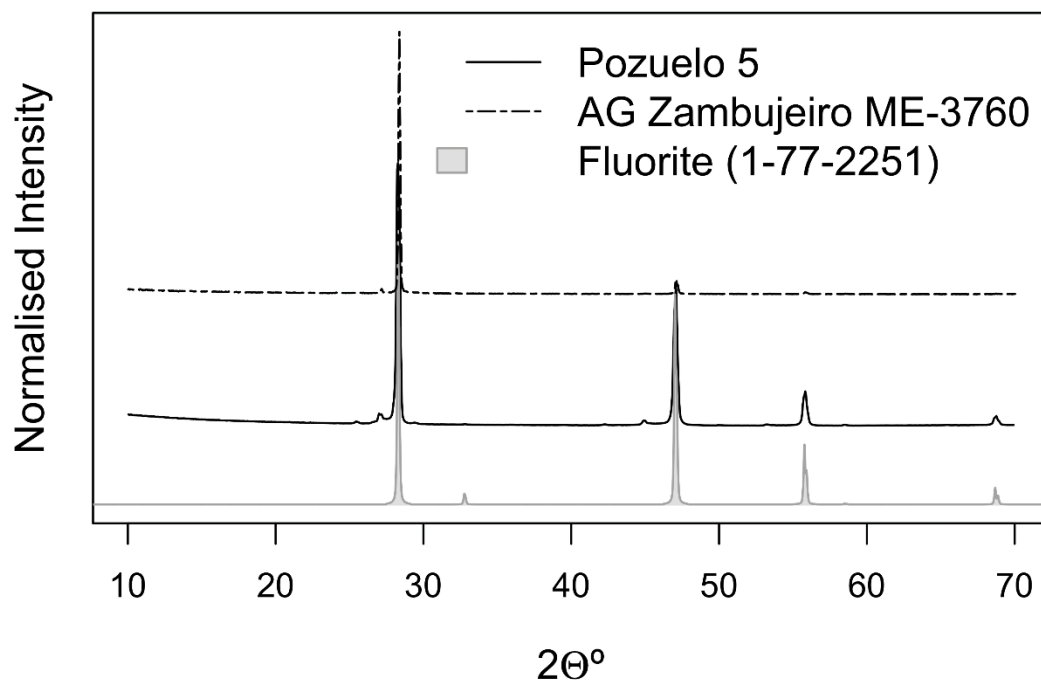


**Figure 5.** Raman spectra in the frequency-shift region between 1000 and 2500 cm<sup>-1</sup> showing the typical fluorite PL spectra lacking the T<sub>2g</sub> Raman band.

A



B



**Figure 6.** XRD pattern of samples MMT-26 from Gruta da Marmota (A) and El Pozuelo 5 and Anta Grande do Zambujeiro (B) compared to fluorite reference card (1-77-2251).

Apart from the aforementioned fluorite beads, ornaments from La Velilla, Vila Nova do São Pedro (ARQ-VNSP-964.064), Cabeço da Ministra (25992-1 and 25992-2), as well as translucent green minerals from Cueva del Vaquero, and La Emisora had a chemical composition and optical properties compatible with that of fluorite (Table 1).

#### **4. Conclusions. Fluorite consumption in Iberia's late prehistory: Perspectives on its social use and symbolism**

The use of fluorite can be circumscribed to the period from the mid-4<sup>th</sup> to the mid-3<sup>rd</sup> millennia BCE, while a few isolated cases attest an interesting symbolic continuity in the 2<sup>nd</sup> millennium in south-east Iberia, in Tomb 111 at Fuente Álamo. So far, only the two fluorite beads from have been documented in that chronology (Pozo *et al.* 2002; Schubart *et al.* 2006).

In the Iberian Peninsula, as in France and Belgium, fluorite was used little, either because of the difficulty in finding large fluorite crystals or because of its consideration amongst other transparent and translucent raw materials. Ornaments made with this mineral are present in various types of contexts during the 4<sup>th</sup>-3<sup>rd</sup> millennia BCE, both funerary and non-funerary. Orthostatic megaliths (16 sites) and natural caves (9 contexts) represent most of the sites where they have been found, as well as in artificial caves (4 sites) and *tholoi* (3 sites). Two habitational contexts, the Chalcolithic sites of Leceia and Vila Nova de São Pedro, also evidence the consumption of fluorite ornaments (Table 1, Supplementary Material 1). It has only been found at two non-mortuary sites; in the third millennium fortified settlements of Leceia and Vila Nova de São Pedro, which are relatively near to each other by the Tagus estuary. At practically all the sites, the fluorite appears in the form of finished adornments and at present no remains of production or objects in the process of being fabricated are known. The only elements of fluorite in mineral form that are known are both from the Province of Seville: the large piece in the *tholos* of Cueva del Vaquero (Alcalá de Guadaíra), and in La Emisora sector in Valencina de la Concepción.

The distribution area of the fluorite mineral is mainly situated on the Atlantic seaboard (Figure 1), where sites possess local or regional access to outcrops, or are distant from sources but positioned on strategic nodes of communication. Its distribution therefore appears to be marked by the Douro and Tagus river basins, with their tributaries, and to a lesser extent by the Guadalquivir estuary and the Guadiana basin (Figure 1). An important concentration is in Évora, particularly at Anta Grande da Comenda da Igreja and Anta Grande do Zambujeiro. Although at many archaeological sites the fluorite may have been obtain in the proximity, the fluorite beads from the area of the Tagus estuary have been interpreted as exotic elements arriving through exchange networks, as the best known fluorite outcrops are several hundreds of kilometers away (Cardoso *et al.* 2012).

The situation around the Tagus estuary, particularly on the Lisbon and Setubal peninsulas, throughout the 3<sup>rd</sup> millennium BCE but especially in the first half, seems to support the idea of groups or centers that received rare raw materials (amber, ivory, variscite and fluorite) and “exclusive” or prestigious objects (*e.g.*, votive elements in limestone and lagomorphic pendants). They may have been the final destination of the route that the finds of fluorite at sites near the Atlantic coast seem to draw. A similar situation can be observed around the Guadalquivir estuary, where specially Valencina de la Concepción-Castilleja de Guzmán site (Seville) but also the megalithic necropolis of El Gandul have yielded a large number of exotic objects, burials and unusual raw materials (see García 2013 for a review on Valencina). However, the proximity of outcrops of fluorite at Morón and El Castillo de las Guardas (Calderón 1910: 409) may support a local provenance for that mineral. This may also be the case of fluorite beads in the megalithic monuments in the western Sierra Morena, in Huelva.

The circulation of large heavy pieces is noteworthy and mostly coincides with the most spectacular items that have been found precisely in prestigious mortuary contexts: the beads from São Paulo 2 (26.3 g), Anta Grande do Zambujeiro (26.5 g), Casa da Moura (22.6 g), and bead 2011.54.278 from Anta Grande da Comenda da Igreja (19.8 g) represent 47.3% of the total analyzed in Portugal (171.8 g). The total weight of the 23 fluorite objects from the Iberian Peninsula that have been weighted is 296.6 g. Spanish fluorite objects amount to 124.8 g, of which 90.1 g corresponds to the fragment of mineral at Cueva del Vaquero and 21.2 g to the fluorite bead in Dolmen 6 at Los Gabrieles.

Fluorite's property of light transmission, its transparency or translucence and the fact that it can be tainted a wide palette of colours that are unusual in nature, like pink, violet or blue, most probably lent them social and symbolic significance and invested them with special meanings and connotations. Indeed, it seems that the pink and violet shades, the more unusual ones, appear in the megaliths where individuals of higher rank were inhumed, for example, Anta Grande da Comenda da Igreja, São Paulo 2, Anta Grande do Zambujeiro and Los Millares 12.

Besides its organoleptic properties, it seems therefore reasonable to suggest that fluorite beads would have had an "added value" based on their exoticism and rarity, as characterization allows to compare its rare proportion of use as ornamental raw material. The idea of this special consideration and rareness of fluorite ornaments is strengthened by its proportion compared with other raw materials used for objects of personal adornment: *exotica* as amber counts to date with 647 items (Odriozola *et al.* 2019), against the 36 fluorite ornaments reported here. As personal belongings, beads may have been items whose use and possession differentiated certain individuals within their communities, materializing social differences by means of their asymmetric display among the living and the dead. In addition to the former, the association of fluorite beads to other non-local raw material items of great value strongly suggests the high-ranking status of the people wearing such items. For example, Tombs 12 and 63 at Los Millares are considered "prestigious" (Chapman 2008), as the grave goods included pottery with symbolic decoration, singular ivory objects and amber beads (5 and 1 respectively) (Chapman 2008; Leisner & Leisner 1943: 25-26; Odriozola *et al.* 2019).

The find of large decorated fluorite beads in Anta Grande da Comenda da Igreja and the artificial cave of São Paulo 2 (Figure 2) indicates not only a formal similarity between the two items (practically perfect spherical shapes) and a possible association with the 4<sup>th</sup>-3<sup>rd</sup> millennia BCE transition, but also their relationship to other prestige items, such as amber beads and lagomorphic pendants (Barros & Espírito-Santo 1997; Leisner & Leisner 1965: 121-122), in these outstanding monuments and their rich grave goods assemblages. Although smaller in size, the fluorite bead from Gruta da Marmota MMT-26 is also decorated with incised vertical lines. Bearing in mind the scarcity of decorated beads in the archaeological record and the singularity of the examples, due to their size and weight, they can be regarded as "special" objects possessing significant symbolism.

As such, it is reasonable to assume that although the raw material was relatively easily available throughout the communities that inhabited Iberia in the Copper Age, only the kin groups, factions or individuals who were buried in monumental tombs were able to afford the added value that allowed the production of such sophisticated objects as decorated fluorite beads. The coincidences between tomb 111 at Fuente Álamo and tomb 454 of El Argar (see Garrido-Cordero 2015), with high status young woman with rich grave goods including translucent beads, point to this consideration for the 2<sup>nd</sup> millennium BCE, where high social differences are displayed. However, it is important to note that, paradoxically, most artifacts studied in this paper, particularly the decorated beads, cannot be ascribed to any particular

individual due to the collective nature of the funerary records or the recording of the items in the late 19<sup>th</sup>- early 20<sup>th</sup> century of our era.

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### Data statement

All the data used here is owned by the authors and was collected by the authors in the general frame of the research projects *Redes y Rocas Raras en la Península Ibérica y el Mediterráneo (VI-II Milenio AC)* (HAR2017-83474-P) and *Nuevas Tecnologías Aplicadas al Estudio de la Movilidad e Intercambio: Cuentas Verdes y Cerámica Decorada con Rellenos Blancos del VI al II Milenio Ane en la Península Ibérica desde una Perspectiva Multidisciplinar* (HAR2012-34620), both funded by the MINECO (Spain). Apart from the available data on this paper (supplementary files 2), ongoing works and a public server on construction will provide open-access data related to the objectives of this research.

### List of supplementary files

Supplementary file 1

“Garrido-Cordero et al.-supplementary file 1 - Contexts of the sites.doc”

Contexts of the sites studied and reported in the text.

Supplementary file 2

“Garrido-Cordero et al.-supplementary file 2 - p-XRF.doc”

p-XRF compositional data of the samples analysed in this research.

### References

- Alencar, I., Ruiz-Fuertes, J., Schwartz, K., Trautmann, C., Bayarjargal, L., Haussühl, E. & Winkler, B. 2016, Irradiation effects in CaF<sub>2</sub> probed by Raman scattering: Irradiation effects in CaF<sub>2</sub> probed by Raman scattering. *Journal of Raman Spectroscopy*, 47: 978-983. doi:10.1002/jrs.4927
- Andò, S. & Garzanti, E. 2014, Raman spectroscopy in heavy-mineral studies. *Geological Society, London, Special Publications*, 386: 395-412. doi:10.1144/SP386.2

- Anthony, J.W., Bideaux, R.A., Bladh, K.W. & Nichols, M.C. 2011, *Handbook of mineralogy*. Mineralogical society of America, Chantilly, USA. Accessed: 12 December 2019. URL: <http://www.handbookofmineralogy.org>
- Bar-Yosef Mayer, O. & Porat, N. 2008, Green stone beads at the dawn of agriculture. *PNAS*, 105(25): 8548-8551. doi:10.1073/pnas.0709931105
- Barros, L. & Espírito-Santo, P. 1997, Gruta artificial de S. Paulo. *Setúbal Arqueológica*, 11-12: 217-220. (in Portuguese) (“The artificial cave of S. Paulo”)
- Baysal, E.L. 2017, Personal ornaments in Neolithic Turkey, the current state of research and interpretation. *Arkeoloji ve Sanat*, 155: 1-22.
- Beckhoff, B., Kanngießer, B., Langhoff, N., Wedell, R. & Wolff, H. 2006, *Handbook of practical X-ray fluorescence analysis*. Springer, 863 p.
- Blanco, A. & Rothemberg, B. 1981, Exploración arqueometalúrgica de Huelva (EAH). Labor, Barcelona, 320 p. (in Spanish) (“Archeometallurgical explorations in Huelva”)
- Brandl, M. & Trnka, G. 2014, Contemporary rock crystal mining in Minas Gerais, Brazil: An ethno-archaeological case study. In: *Lithic raw material resources and procurement in Pre and Protohistoric times* (Bostyn, F. & Giligny, F., Eds.), BAR International Series Vol. 2656, Oxford: p. 121-130.
- Calderón, S. 1910, Los minerales de España. Junta para Ampliación de Estudios e Investigaciones Científicas, Madrid, 977 p. (in Spanish) (“Minerals of Spain”)
- Cardoso, J.L., Domínguez, S. & Martínez, J. 2012, Ocorrência de contas de fluorite no Neolítico Final e no Calcolítico da Estremadura (Portugal). *Estudos Arqueológicos de Oeiras*, 19: 35-42. (in Portuguese) (“Fluorite beads occurrence in Estremadura’s Late Neolithic and Chalcolithic”)
- Chapman, R. 2008, Producing inequalities: Regional sequences in later prehistoric southern Spain. *Journal of World Prehistory*, 21: 195-260. doi:10.1007/s10963-008-9014-y
- Chen, H. & Stimets, R.W. 2014, Fluorescence of trivalent neodymium in various materials excited by a 785 nm laser. *American Mineralogist*, 99: 332-342. doi:10.2138/am.2014.4311
- Cherniak, D., Zhang, X., Wayne, N. & Watson, E. 2001, Sr, Y, and REE diffusion in fluorite. *Chemical Geology*, 181: 99-111. doi:10.1016/S0009-2541(01)00267-4
- Demakopoulou, K., Divari-Valakou, N., Åström, P. & Walberg, G. 1996, Excavations in Midea 1994. *Opuscula Atheniensia*, 21: 13-32.
- Delye, E., Gilson, S.P. & Noiret, P. 2011, L’occupation Michellsberg de Pont-de-Bonne, Rocher du Vieux-Château (Modave, province de Liège, Belgique). Présentation liminaire. *Revue Archéologique de Picardie*, 28: 497-505. (in French) (“The Michellsberg occupation of Pont-de-Bonne, Rocher du Vieux-Château (Modave, Liège province, Belgium”)
- Freeth, C.A. & Jones, G.D. 1982, Zeeman infrared spectra of calcium and strontium fluoride crystals containing cerium and neodymium. *Journal of Physics C: Solid State Physics*, 15: 6833-6849. doi:10.1088/0022-3719/15/33/019
- Gaft, M., Panczer, G., Reisfeld, R. & Uspensky, E. 2001, Laser-induced time-resolved luminescence as a tool for rare-earth element identification in minerals. *Physics and Chemistry of Minerals*, 28: 347-363. doi:10.1007/s002690100163



- Galán, E. & Mirete, S. 1979, Introducción a los minerales de España. Instituto Geológico y Minero de España, Madrid, 420 p. (in Spanish) (“Introduction to the minerals of Spain”)
- García, L. 2013, El asentamiento de la Edad del Cobre de Valencina de la Concepción: estado actual de la investigación, debates y perspectivas. In: *El asentamiento prehistórico de Valencina de la Concepción (Sevilla): Investigación y tutela en el 150 aniversario del descubrimiento de La Pastora* (García, L., Vargas, J.M., Hurtado, V., Ruiz, T. & Cruz-Auñón, R., Eds.), Universidad de Sevilla, Sevilla: p. 21-59. (in Spanish) (“The Copper Age site of Valencina de la Concepción: Current state of the investigation, debates and perspectives”)
- Garrido-Cordero, J.A. 2015, El uso del cuarzo y el cristal de roca en la Prehistoria andaluza. Estado de la cuestión y análisis de un fenómeno cultural. *Revista Atlántica-Mediterránea de Prehistoria y Arqueología Social*, 17(1): 187-200 (in Spanish) (“Use of quartz and rock crystal in Andalusian late prehistory. State of the question and analysis of a cultural phenomenon”)
- Garrido-Cordero, J. A., Odriozola, C. P., Sousa, A. C., Gonçalves, V. S. 2020a, Fluorite and translucent beads in Iberian Late Prehistory. *Materials and Manufacturing Processes*, 35(13): 1424-1430. doi:10.1080/10426914.2020.1753069
- Garrido-Cordero, J. A., Odriozola, C. P., Sousa, A. C., Gonçalves, V. S., Cardoso, J. L. 2020b, Distribution and consumption of fluorite and translucent ornaments in the Iberian Peninsula from 6<sup>th</sup> to 2<sup>nd</sup> millennia BC. *Trabajos de Prehistoria*, 77(2): 273-283. doi:10.3989/tp.2020.12256
- Gaydarska, B. & Chapman, J. 2008, The aesthetics of colour and brilliance – or why were prehistoric persons interested in rocks, minerals, clays and pigments. In: *Geoarchaeology and Archaeomineralogy. Proceedings of the International Conference* (Kostov, R. I.; Gaydarska, B. & Gurova, M., Eds.) St Ivan Rilski, Sofia: p. 63-66.
- Goemaere, E. & Philippo, S. 2010, La fluorite de Seilles. In: *Terres, pierres et feu en vallée mosane. L'exploitation des ressources naturelles minérales de la commune de l'Andene*. Institut Royal de Sciences Naturelles de Belgique: p 491-500. (in French) (“Earth, stones and fire in the Mosane valley. The mineral natural resources exploitation in the Andene commune”)
- Goemaere, E., Honings, J., Jungels, C., Golitko, M., Degryse, P. & Schneider, J. 2013, Discerning geological and geographical sources of Upper Paleolithic fluorites by rare earth elements and Sr-isotopic geochemistry. *Journal of Archaeological Science*, 40: 2892-2901. doi:10.1016/j.jas.2013.02.029
- Gomes, L.F.C., Sobral, P., Perpétuo, J.M.A. & Marrafa, C. 1998, O dólmen de Areita (S. João da Pesqueira, Viseu). In: *Actas do Colóquio “A Pré-História na Beira Interior” (Tondela, 21 a 23 de novembro de 1997)* (Da Cruz, D.J., Coord.), Estudos Pré-históricos vol. VI, Centro de Estudos Pré-históricos da Beira Alta, Porto: p. 33-93. (in Portuguese) (“The dolmen of Areita (S. João da Pesqueira, Viseu)”)
- Hauzeur, A. & Cauwe, N. 2012, La parure au Néolithique. In: *La parure du Cro-Magnon á Clovis* (Cattelain, P., Bozet, N. & Di Stazio, G.V., Dirs.), Guides Archéologiques du Malgré-Tout, Éditions du CEDARC, Bruxelles: p. 36-45. (in French) (“The ornaments in the Neolithic”)

- Honings, J., Goemaere, E., Bostyn, F., Monchablon, C., Leroy-Langelin, E., Degryse, P., Golitko, M., Collet, H. & Jadin, I. 2014, Rare earth elements and Sr-isotopic geochemistry: Tools to determinate the geological and geographical origins of Neolithic fluorites. In: *Proceedings of the 39th International Symposium on Archaeometry, May 28th-June 1st, 2012, Leuven, Belgium* (Scott, R. B., Braekmans, D., Carremans, M. & Degryse, P., Eds.), Centre for Archaeological Sciences, KU Leuven: p. 145.
- Jones, A. & MacGregor, G. 2002, Introduction. Wonderful things: Colour studies in archaeology from Munsell to materiality. In: *Colouring the past. The significance of colour in archaeological research* (Jones, A. & MacGregor, Eds.), Berg, New York: p. 1-22.
- Jungels, C., Goemaere, E. 2007, La fluorite: une matière première inhabituelle en Préhistoire, *Notae Praehistoricae*, 27: 27-39. (in French) (“Fluorite: An unusual raw material in prehistory”)
- Leisner, G., Leisner, V. 1943. *Die Megalithgräber der Iberischen Halbinsel: der Süden*. Römisch-Germanische Forschungen, 17. Madrider Forschungen 1. Walter de Gruyter, Berlin, 619 p. (in German) (“The megalithic tombs of the Iberian Peninsula: The south”)
- Leisner, G., Leisner, V. 1965. *Die Megalithgräber der Iberischen Halbinsel: der Westen*. Madrider Forschungen Band 1/3. Deutsches Archäologisches Institut abteilung Madrid. Walter de Gruyter, Berlin, 471 p. (in German) (“The megalithic tombs of the Iberian Peninsula: The west”)
- Lenz, C., Nasdala, L., Talla, D., Hauzenberger, C., Seitz, R. & Kolitsch, U. 2015, Laser-induced REE<sup>3+</sup> photoluminescence of selected accessory minerals - An “advantageous artefact” in Raman spectroscopy. *Chemical Geology*, 415: 1-16. doi:10.1016/j.chemgeo.2015.09.001
- Nai, X. 2014, *Ancient Egyptian beads*. Chinese Academy of Social Sciences and Springer, Berlin, Heidelberg, 173 p. doi:10.1007/978-3-642-54868-0
- Odrizola, C.P., Sousa, A.C., Mataloto, R., Boaventura, R., Andrade, M., Villalobos, R., Garrido-Cordero, J.A., Rodríguez, E., Martínez-Blanes, J.M., Avilés, M.A., Daura, J., Sanz, M. & Riquelme, J.A. 2019, Amber, beads and social interaction in the Late Prehistory of the Iberian Peninsula: An update. *Archaeological and Anthropological Sciences*, 11(2): 567-595. doi:10.1007/s12520-017-0549-7
- Payne, S.A., Caird, J.A., Chase, L.L., Smith, L.K., Nielsen, N.D. & Krupke, W.F. 1991, Spectroscopy and gain measurements of Nd<sup>3+</sup> in SrF<sub>2</sub> and other fluorite-structure hosts. *Journal of the Optical Society of America B*, 8: 726. doi:10.1364/JOSAB.8.000726
- Polloni, A. 2008, Parures individuelles et sépultures collectives à la fin du Néolithique en Bassin parisien. *Préhistoires Méditerranéennes*, 14: 75-89. (in French) (“Individual ornaments and collective burials at the end of Neolithic in the Paris basin”)
- Pozo, M., Casas, J. & Medina, J.A. 2002, Estudio mineralógico de componentes ornamentales pétreos procedentes de un yacimiento de la Cultura del Argar (Fuente Álamo, Almería). *Boletín Geológico y Minero*, 113(2): 131-142. (in Spanish) (“Mineralogical study of lithic ornaments from an Argar’s culture site (Fuente Álamo, Almería)”)

- Ribeiro, A.T. & Loureiro, L. 2015, O núcleo Megalítico do Taím/Leandro, o caso de estudo das mamoaas 4 e 5 do Leandro, concelho da Maia, Porto, Portugal. In: *Actas do V Congresso do Neolítico Peninsular* (Gonçalves, V., Diniz, M. & Sousa, A.C., Eds.), Monografias 8, UNIARQ (Centro de Arqueologia da Universidade de Lisboa), Lisbon: p. 522-531. (in Portuguese) (“The megalithic nucleus of Taím/Leandro, the case study of dolmens 4 and 5 of Leandro, Maia county, Porto, Portugal”)
- Roscian, S., Claustre, F. & Dietrich, J.E. 1992, Les parures du Midi méditerranéen du Néolithique ancien à l’Age du Bronze: origine et circulation des matières premières. *Gallia Préhistoire*, 34: 209-257. (in French) (“The ornaments of the Mediterranean south from the Early Neolithic to the Bronze Age: Raw material origin and circulation”)
- Schmalzl, K., Strauch, D. & Schober, H. 2003, Lattice-dynamical and ground-state properties of CaF<sub>2</sub> studied by inelastic neutron scattering and density-functional methods. *Physical Review B*, 68: 144301. doi:10.1103/PhysRevB.68.144301
- Schubart, H., Pingel, V., Kunter, M., Liesau vonLettow-Vorbeck, C. & Hägg, I. 2006, Estudios sobre la tumba 111 de Fuente Álamo (Almería). *Spal*, 15: 103-148. (in Spanish) (“Studies on the tomb 111 of Fuente Álamo (Almería)”)
- Sood, A.K., Arora, A.K., Umadevi, V. & Venkataraman, G. 1981, Raman study of temperature dependence of lattice modes in calcite. *Pramana*, 16: 1-16. doi:10.1007/BF02847889
- Sverdrup, T.L. 1968, Yttrifluorite-yttrocerite-cerfluorite in Norwegian pegmatites. *Norsk Geol. Tidsskr*, 48: 245-252.
- Tilley, C. 1999, *Metaphor and material culture*. Blackwell, Oxford, 316 p.
- Tu, J.J. & Sievers, A.J. 2002, Experimental study of Raman-active two-level systems and the boson peak in LaF<sub>3</sub>-doped fluorite mixed crystals. *Physical Review B*, 66: 094206. doi:10.1103/PhysRevB.66.094206
- Vandenabeele, P. & Edwards, H.G. 2018, *Raman spectroscopy in archaeology and art history: Volume 2*. Royal Society of Chemistry, Cambridge, 349 p.
- Vermeersch, P.M., Vynckier, G. & Walter, R. 1990, Thieusies, Ferme de l’Hosté, site Michelsberg. II. Le matériel lithique. *Studia Praehistorica Belgica*, 6: 5-70. (in French) (“Thieusies, Ferme de l’Hosté, a site Michelsberg. II. The lithic assemblage”)
- Villalobos, R. 2015, *Análisis de las transformaciones sociales en la Prehistoria Reciente de la Meseta norte española (milenios VII-III CAL A.C.) a través del empleo de la variscita y otros minerales verdes como artefactos sociotécnicos*. Doctoral thesis at the Department of Prehistory, Archaeology, Social Anthropology and Historiographical Science and Techniques, University of Valladolid, 582 p. (in Spanish) (“Analysis of social transformations in the Spanish northern Meseta’s late prehistory (7th-3rd millennia cal BCE) by the use of variscite and other greenstones as sociotechnical artifacts”)
- Warmenbol, E. 2001, L’Âge du Bronze. *Anthropologica et Praehistorica*, 117: 107-119. (in French) (“The Bronze Age”)
- Wright, K. & Garrard, A. 2003, Social identities and the expansion of beadmaking in Neolithic Western Asia. New evidence from Jordan. *Antiquity*, 77(286): 267-284. doi:10.1017/S0003598X00092267

Zhuk, Y. 2017, *Critical assessment of the mineralogical collections at Uppsala University using Raman spectroscopy*. Bachelor degree thesis presented at the Department of Earth Sciences, Uppsala universitet. Uppsala, 240 p.