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The variscite « necklace » of the Salpêtre cave, at Pompignan (Gard, France)

A remarkable set of 17 variscite beads was discovered in the 1970s in the Salpêtre cave in Pompignan (Gard, France). Although known and presented to the public in various recent exhibitions, this discovery has never been the subject of a detailed publication. This article aims to fill this gap: it provides the results of the provenance analyses (PIXE). It documents the typology and illustrates the different beads of this original deposit, unique in the south of France. Finally, this discovery is placed in its chronocultural context.

- Blue rock beads called "callaïs" (variscite, turquoise) were socially valued objects during the Neolithic period. They were traded over long distances.

- The necklace from the Salpêtre cave in Pompignan (Gard, France) is the largest known batch of variscite beads in southern France.

- Thanks to the PIXE multi-element analysis, the comparison between rock samples from the main Western European deposits and the archaeological pieces allows to know the origin of the Neolithic blue beads. The 17 beads from Pompignan were analysed.

- The article demonstrates the relationship between a mining deposit (Gavà, Spain) and the destination of the finished products, located several hundred kilometres away (Pompignan, France).

- In general, Neolithic variscite beads are derived from burials and the Pompignan deposit is a unique case for this period. This notion of depositing batches of precious objects will become recurrent in the Chalcolithic and Early Bronze Age.

## The variscite « necklace » of the Salpêtre cave, at Pompignan (Gard, France)

Jean Guilaine, Guirec Querré, † Serge Cours, Jacques Coularou, Hélène Vergély, Jean Vaquer, Muriel Gandelin

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#### 8 Abstract :.

A 17 variscite beads set, buried in the Salpêtre cave in Pompignan (Gard, France) constitutes a
unique Middle Neolithic set of ornaments in the southern France. PIXE (Particle Induced X-ray
Emission) chemical analysis of these coins and reference samples from possible sources indicates a
mineral origin from the Gava mines, near Barcelona (Spain). This discovery reveals an exchange system
of the materials or finished objects on both sides of the Pyrenees mountains during the first half of the
4th millennium BC.

15

16 Keywords : Neolithic of western Europe, Middle Neolithic (Chasséenne culture), ornaments, variscite,

17 provenance, chemical analysis, PIXE (Particle-Induced X-ray Emission)

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20 Adornments, in prehistoric populations, is not just ornaments but also an «identity language», functioning as a sexual, social, or ethnical marker. In Neolithic times, in the Near East as in Europe, 21 22 green stones were largely used in jewellery making. As such, in Israel, from the recent Natufian to the pre-pottery Neolithic B (PPNB), between -11000 and -7000 BC, several minerals bearing this hue 23 (apatite, fluorapatite, chrysocolla, malachite, turquoise, amazonite, serpentinite) were used to make 24 beads. The source of the raw materials was often distant from the user location of the productions. And, 25 the choice of a green hue has sometimes been interpreted as a symbol of vegetation, of growing plants, 26 of fertility (Bar Yosef Mayer and Porat 2008). In Cyprus, picrolite, the local green mineral, was largely 27 exploited in jewellery production from the PPNA (pre-pottery Neolithic A) until the Chalcolithic 28 29 (Guilaine, 2011; Guilaine and Rigaud, 2021; Peltenburg, 1998). During the Neolithic of western Europe, another green rock, the variscite, was used in the fabrication of necklaces during almost the entire period, 30 from the 6<sup>th</sup> to the 3<sup>rd</sup> millennium BCE. This mineral is a kind of hydrated aluminium phosphate, also 31 32 called variscite, the formula for which is AlPO<sub>4</sub>  $\cdot$  2 H<sub>2</sub>O. Its deposits are essentially located within the 33 Iberian Peninsula, but also in France and Sardinia (Fernandez et al. 2019). 34 In Brittany, the presence of beads made of variscite is attested in Neolithic contexts - often funerary 35 ones – since the 19th century. Such ornamental objects were discovered in the great Carnaccean tumuli 36 (Tumiac, Saint-Michel, Mané er Hroeck) as well as several Armorican dolmens (Kercado). Right away,

the question of the identification and origin of the material was raised. As early as 1864, mineralogist

- 38 A. Damour studied a few specimens of these light green minerals and identified them as made of a
- 39 hydrated aluminide phosphate (Damour 1864). Faced with the quality of such productions, A. Damour,

40 followed by A. Lacroix believed in a distant origin of the raw material, and this hypothesis of an exotic,

41 distant rock prevailed for a long time (Veiga Ferreira, 1950).

In southern France, E. Cartailhac was one of the first to be interested in these ornaments, some of which 42 came from the dolmens of Aveyron and of Lozère, where he first started working. In 1876, he 43 44 participated to research on the Castellet hypogeum (Fontvieille, Bouches-du-Rhône), from which an ensemble of 114 beads was extracted. In 1881 he published in the journal he edited, Matériaux, E. 45 Piette's article mentioning the « necklace » made of 93 beads issued from one of the compartments of 46 47 the Halliade monuments at Bartrès (High-Pyreneans; Piette 1881, Mohen 1978). This led him to give an 48 overview of these adornments, the presence of which he identified in Brittany, Spain, Portugal (in the 49 hypogea of Palmela) and in the Midi (Cartailhac 1889). In parallel, research on the north-eastern region of the Iberian Peninsula revealed the existence of numerous beads of the same material within the earth 50 51 or cists of that region. A. Muñoz, in her thesis on the « Sepulcros de fosa » of Catalonia, provided a synthesis of this issue and brought some doubt to the hypothesis of a distant origin (Muñoz 1965). In 52 53 the Midi, a few specimens were brought to the attention of two researchers of the mineralogy department of the Commissariat à l'Énergie Atomique (F. Chantret, A. Guillemaut) and revealed that the term callais 54 55 could regroup variscite, turquoise, or malachite ornaments (Chantret et al. 1970). Variscite ornaments were essentially recognised within the funerary context in individual inhumations (within a dug-out 56 57 grave or a stone box) or in collective tombs (megalithic structures, hypogea, sepulchral caves).

A new and determining impulsion in approaches to the topic came from the recognition, starting from 58 the 70s onwards, of the variscite mines of Gavà, near Barcelona in Spain (Domínguez-Bella 2004, Bosch 59 Argilagos 2019, Molist and Oliva 2019, Moro et al. 1992 and 1995). Following this, renewed attention 60 was given to the geological formations of the Zamora region, where several deposits had been identified 61 62 (Fabregas Valcarce and Rodriguez-Rellan 2019). Then came the identification of the Pico Centeno 63 deposit (Encinasola, Huelva, Spain) in western Andalusia, (Odriozola et al. 2010, Dominguez-Bella and 64 Ramos-Muñoz 2019). Uncovering the existence of these deposits was combined with diverse analyses 65 programs in order to establish precise baseline referentials characterising each of these three sources: Gavà, the Zamora deposit, and Encinasola. In turn, this meant various scenarios for the circulation of 66 variscite and turquoise during the Neolithic (between 5500 and 2000 BC) at the scale of western Europe 67 could be proposed (Querré et al. 2019). 68

The productions from these various deposits may have similarities and differences. Thus the round or barrel beads are ubiquitous. The large pendants with polar perforation from Breton sites come from Encinasola in the fifth millennium and then, from 4000 BC, from Palazuelo de las Cuevas. The Portuguese cylindrical beads come from deposits in the Zamora region. The large biconical beads with axial perforation come mainly from Gava. The small irregular beads from Final Neolithic collective burials in the south of France do not have, due to lack of analysis, a clear provenance (Querré et al. 2019). 76 A remarkable variscite bead ensemble was discovered in the 70s in the Salpêtre cave at Pompignan

- 77 (Gard, France). Whilst known and presented to the public in various recent exhibits, this discovery had
- 78 never been published in detail. This article aims to fill this gap in knowledge and communicate the
- results of the analyses concerning this original ensemble, unique for the south of France.
- 80

#### 81 1. The site

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The Salpêtre cave is located on the commune of Pompignan (Gard, France), 30 km north of the town of
Montpellier (fig. 1). It opens on the northern flank of the Hortus causse, at the foot of the Taillade cliff,
at an average altitude of 380 m (fig. 2 and 3).

86 This cave was subjected to various investigations over a century. The oldest was by A. Jeanjean who

- 87 mentioned its interest in a synthesis on the prehistory of the Basses-Cévennes region (Jeanjean 1871).
- 88 Around 1960, Jacques Audibert, research associate at the CNRS, undertook a little work in the cavity
- 89 (Audibert 1962). These two operations meant middle Neolithic levels (Chasséen) were identified, as
- 90 well as some final Neolithic (Ferrières and Fontbouïsse cultural groups). In the 70s, S. Cours re-sieved
- some of the waste heaps and prospected other areas of the cave. This is when the necklace described in
- 92 this article was discovered. Following this find, S. Cours and J. Coularou dug a deep test pit 10 m from
- 93 the cave entrance, 3 m deep. At that level, they found hardened sediment. They recognised the reworked
- 94 aspect of the Neolithic levels but also identified the existence of palaeolithic levels still in place (Azilian,
- 95 Magdalenian, Mousterian). The Azilian artefacts were described in M. Barbaza's doctoral thesis
- 96 (Barbaza 1981). It is possible that some Neolithic levels still in place are present in the deepest part of
- 97 the cavity, under the spoil heaps from older excavations, which would allow <sup>14</sup>C dating, none of which
- 98 was done at this time.
- 99 In 1972, M. Escalon de Fonton dug a new test pit, looking for the previously recognised Magdalenian
- 100 level. The investigation remained fruitless, but a few years later in 1978-1979, L. Meignen pursued this
- 101 research axis and confirmed the presence of Mousterian occupations.
- 102 The Salpêtre cave is a long corridor following a north-south cleft; it is 80 m long and 2 m large and
- reaches a height of 5 m. Towards the deepest end of the cavity, a lateral diverticulum opens into a smallroom (fig. 4 and 5).
- 105 The variscite bead cache was found 41.5 m from the cave entrance, where the corridor narrows, and the
- walls are only 0.5 m apart. This narrowing is even more marked by the fact that a natural step existsthere, forming a threshold of sorts (fig. 6).
- 108 The beads were discovered grouped within a small space. Perhaps they were originally linked by a thread
- 109 that has now disappeared. In the immediate vicinity, a large ceramic sherd and a pecten shell were found.
- 110 The sherd (9 x 8 cm) has a curved profile and belongs to a vessel with a thick profile (1 cm). It is
- identified as a fragment of the ventral part of a spherical container, the diameter of which is estimated
- to have been around 26 cm. This element bears an engraved decor made of three horizontal strips: a

- 113 central one with a checkered motif, and two surrounding ones (superior and inferior) presenting a motif
- 114 made of four lines arranged as rafters. This resembles spherical to cylindrical collared containers typical
- of the classical *Chasséen*, the widest part of which was often decorated with geometrical patterned strips.
- 116 The mollusc shell is a scallop (a Saint-Jacques shell: *Pecten jacobeus*), rather large (width: 13 cm). We
- 117 cannot exclude that the beads were originally placed within the bivalve.
- 118

### 119 **2. Description of the Beads**

- 120
- 121 The excavated lot includes 17 elements which could originally constitute a necklace (fig. 9)<sup>1</sup>.

122 All the beads, of various sizes, bear witness to a final fabrication state necessitating an important 123 technical investment. Measures were carried out on the bead ensemble: maximum diameter (median point of the maximum width of the transversal section), minimal diameter (median point of the 124 125 minimum width of the transversal section), diameter of the perforation, maximum and minimum 126 thickness (or length), and maximal and minimum diameter of the bead's extremities. These last 127 measures correspond to the term « méplat » used by F. Herbaut and G. Querré (Herbaut, Querré 2004). The maximum diameter of the beads is between 7.2 and 25.5 mm, for a thickness between 5.9 and 128 129 40.1 mm. The maximum diameter of the perforations is comprised between 2.1 and 4.3 mm. 130 Dimensions, weights and color are resumed in table 1 and in table 2.

- 131 Discussing the typological aspect of these beads is difficult, because variscite ornament productions do not correspond to fabrication standards observed on other mineral matter, and we are confronted 132 to a lack of current research (in France) on this aspect of variscite productions (Viel et al. 2019). Looking 133 134 to the reference work on ornaments of the French Midi region (Barge 1982), the vast majority of the 135 beads of the Salpêtre cave ensemble can be qualified as irregular. It is true that the typological 136 classification of these objects is problematic since they often present asymmetries in their longitudinal 137 and transversal sections. In publications, they are generally described as olive-shaped, or flattened 138 beads...
- 139 In order to better define the production criteria of the workshops, which seems to evolve over time, 140 typological classification work could help us qualify these productions, such as the one published by H. 141 Beck in 1926 « Classification and Nomenclature of Beads and Pendants » (Beck 2006). This typological 142 example allows for the description and classification of beads whilst considering their shape from the 143 section and the front. Synthetically speaking, the Salpêtre ensemble presents, in most cases, an 144 elliptical transversal section (more or less regular) combined with a convex cylindrical longitudinal 145 section. Several beads present a transversal section associated with a convex, « barrel-shaped » or 146 cylindrical longitudinal section, with both extremities being convex (n°7 and 8). One bead reveals a 147 quadrangular transversal section (n°5) and another can be qualified as a conic trunk (n°17).
- 148 The polishing of the beads is advanced, giving an intense shine visible on both the bead's extremities 149 and their longitudinal sections. This aspect is also visible on the extremities' surfaces presenting great 150 irregularities such as bead n°13, or on the section of bead n°3. Only one bead (n°2) presents unpolished

<sup>&</sup>lt;sup>1</sup> These artefacts are kept at the Centre de Documentation Archéologique du Gard (CDAG), 396 avenue Joliot Curie, 30900 Nîmes. Their analysis took place within a PCR (Collective Research Program) titled « Milieu et Peuplement en Languedoc Occidental du Néolithique au Bronze ancien » (dir. M. Gandelin)

151 irregularities that can be interpreted as impacts (3 in total), which must have occurred after the bead's

fabrication and bears witness to probable shocks. Besides, traces of red coloured matter (ochres ?; not
analysed) are visible inside these asperities. The polishing is so advanced that it is difficult to discern

traces of the manufacturing techniques used before that stage of fabrication.

Perforations are all cylindrical, and their regularity implies the use of a system to hold the pieces in place, and the use of flint drills mounted on a rotating axis to pierce the beads. A boring step allowed for the regularity of the perforations. These technical observations correspond to the research made on variscite's chaine opératoire in Catalonia (Molist Montana, Oliva Poveda 2019). The morphology of the beads found in the Salpêtre cave is close to those found in the Gavà workshop and find many comparisons with the beads discovered in grave 83 of the Mine (Beta 155.686 : 5220±110 BP ; 4326 – 3793 cal BC (95.4%) ; Bosch Argilagos 2019).

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# 163 3. Chemical and provenance analyses of the callais bead necklace from the Salpêtre cave164 (Pompignan, Gard).

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The 17 callaïs beads probably composing a necklace (fig. 10 and 11) found during the Salpêtre cave excavation at Pompignan were subjected to an elemental chemical analysis in order to confirm their mineralogic identity (variscite) and to determine their provenance. This determination was established using the chemometric model elaborated based on the main European sources of variscite (Querré et al, 2008; 2012; 2014 ; and forthcoming) as well as PIXE analyses of the Pompignan beads.

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#### 172 **3.1. Methodology**

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The analyses were carried out using the PIXE method (*Particle-Induced X-ray Emission*) using the
AGLAE (Elementary Analysis Accelerator of the *Grand Louvre*) set up from the C2RMF (The research
and restoration centre of *Musées de France*) of the Louvre Palace (Paris).

177 The PIXE method was applied to the callais according to a protocol established by one of the authors (G.Q.). This method of elemental analysis is non-destructive and fast. It is based on the fluorescence of 178 179 X-rays characteristic of the material. The elemental composition of archaeological pieces is compared with a reference sample, representative of the 5 largest European variscite deposits, likely to have been 180 181 exploited during the Neolithic. The concentrations of major elements allow us to recognize the mineralogical nature of the veins; the spectrum of minor elements and/or traces shows intrinsic 182 variations, specific to each deposit. The contents of iron, vanadium, chromium, arsenic, silicon, calcium, 183 zirconium, uranium, their absolute variations and concentration rates are particularly relevant to 184 discriminate potential geological sources. This model can be used to determine the provenance of 185 archaeological variscite pieces (Querré et al. 2019). 186

Protons of 3MeV were extracted in the shape of a beam a few micrometres in diameter. The bead surface
to be analysed was placed a few millimetres from the protons exit window. Four SDD detectors gather

the X-Rays created: one for the low energies corresponding to major elements, and three for high energies corresponding to trace elements (fig. 12 a and b). In order to have a mean analysis of the bead's material, the beam scanned the surface over one millimetre square. The acquisition time for the analysis of each bead was six minutes on average. The high and low energy spectrums went through GUPIX and TRAUPIX software analysis, in order to obtain elementary concentrations as well as the detection thresholds for all 24 chemical elements of interest.

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#### 196 **3.2.** Chemical analysis of the beads

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The concentration of the 24 chemical elements for each of the 17 beads can be found in table 3 and table 4. Three chemical elements are present in concentrations higher than 10 % (100.000 ppm): aluminium, silicon and phosphorus. Copper concentrations are low, 65 ppm on average. The beads are therefore indeed variscite, albeit a very impure form considering the high silicon proportions present within. After these three major elements, iron and copper are the most represented, with element concentrations reaching around 2 %. All other elements are found in minor or trace concentrations.

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#### 205 **3.3. Provenance**

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The chemometric model elaborated to determine the variscite's provenance was applied to the beadsfound in the Salpêtre cave.

The concentrations of the discriminating elements were compared to those of the five western European deposits where the beads might come from : the four Spanish deposits, the Gavà deposit (Barcelona) including the Can Tintorer mines (fig. 13, GA, and table 4), the Encinasola deposit and the mines of Pico Centeno in the Huelva region (fig. 13, EN, and table 4), El Bostal in the Zamora region (fig. 13, EB, and table 4), Palazuelo de las Cuevas (fig. 13, PL, and table 4), and the only known French deposit : Pannecé, in the Loire-Atlantique region (fig. 13, PA, and table 4).

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The relative proportions of the three most discriminating elements (V, Cr and Fe) are, on average, the following: V\*=7.5 - Cr\*=7.8 - Fe\*=84.7. These proportions do not correspond to those of the Pannecé deposits, for which vanadium represents 50 % and within which chrome and iron reach 25% of the total each (see figure 13 as well). The average Cr/V ratio is  $1.39 \pm 1.05$  which is not compatible with the Palazuelo analyses (Cr/V = 5.08 ±6.48), nor with those of El Bostal (Cr/V=0.35 ±0.04).

- 221 The U vs Zr diagram (fig. 13B and table 4) allows us to individualise the variscites from El Bostal,
- which has strong uranium concentrations (U =  $306 \pm 267$  ppm) and no zirconium (Zr =  $6 \pm 4$  ppm). In

the case of the Salpêtre beads, the reverse phenomenon is observed, with low uranium concentration

- $(U=9 \pm 5ppm)$ , often inferior to the detection threshold (LDD = 19 ppm), and average zirconium
- 225 presence ( $Zr = 22 \pm 23$  ppm).

- 226 Concerning diagram (Cr+V) vs As (fig. 13C), the concentrations observed in the Pompignan variscite
- beads (Cr+V =  $2926 \pm 1003$  ppm ; As =  $802 \pm 418$  ppm) are close to those of two deposits : Encinasola
- 228 (Cr+V=4198  $\pm$ 1429 ppm ; As = 610  $\pm$ 802 ppm) and Gavà (Cr+V = 809  $\pm$ 397 ppm ; As = 302  $\pm$ 90 ppm)
- but not those of three other sources : (Cr+V =  $2926 \pm 1003$  ppm ; As =  $802 \pm 418$  ppm) (Cr+V = 2926
- 230  $\pm 1003 \text{ ppm}$ ; As = 802  $\pm 418 \text{ ppm}$ ) (Cr+V = 2926  $\pm 1003 \text{ ppm}$ ; As = 802  $\pm 418 \text{ ppm}$ ).
- Silicon concentrations are extremely high in the Salpêtre beads (Si =  $17.6 \pm 6 \%$ ) and can be qualified as
- elevated as far as calcium is concerned (Ca =  $1.8 \pm 1.2$  %). This tends to indicate that the origins of the
- 233 beads is the Gavà deposit, which is the only one to present elevated concentrations of both silicon and
- 234 calcium (Si =  $11.2 \pm 7.4$  %; Ca =  $6.1\pm 8.0$  %) (fig. 13 C and table 4).
- 235 Together, all these geochemical analyses based on the chemometric model bring only one answer to the
- 236 question of the origins of the Salpêtre cave the Gavà deposit, in Spain. The high iron concentrations
- in contrast to the vanadium, the Cr/V ratio, as well as the high proportions of silicon and calcium are the
- 238 most discriminating factors in this case.

This result is coherent with the identification of the site from which the beads originate as *Chasséen*, and thus belonging to the Mediterranean sphere, within which most variscite ornaments come from the Can Tintorer mines (Gavà, Spain). The morphology of the large biconical beads from Pompignan is the same as those deposited in the graves in the Barcelona area.

- 243
- **4.** A cache of ornaments from the southern Pyrenees
- 245

246 The analyses of the Salpêtre cave ensemble (Pompignan, Gard, France) bring precious information on 247 the circulation of ornaments outside their fabrication area in a region - southern France - where these 248 analyses are still rare. Indeed, this ensemble is composed of various typological elements, including 249 large, olive-shaped beads perfectly comparable to the productions found on the southern flank of the 250 eastern Pyrenees, and dated to the evolved Neolithic middle. The meridional cultural context is then that 251 of the Chasséen. The mineralogical composition of the Pompignan pieces demonstrate it is made of variscite coming from the Gavà deposits. Beyond the synchronism proposed between the Sepulcros de 252 fosa and the meridional Chasséen, which has long been argued for (Guilaine and Muñoz 1964), these 253 254 interactions are now better understood. As such, polished alpine stone axes and Bedoulian flint a attested 255 within the Catalan funerary artefacts, whilst variscite ornaments are themselves found within Chasséen funerary deposits of the Toulouse region (Saint-Michel du Touch, Villeneuve-Tolosane) and as far as 256 257 the Rhône (Pompignan), or even the Lyon region (Tassin ; Vaquer 2019).

The Pompignan ensemble also presents an interest in that it is a deposit of rare pieces placed in a cache, made within a cave, rather far from the cave's entrance. This deposit was hidden, perhaps during a ritual act. We must therefore raise the possibility that as early as the middle Neolithic rare pieces were hidden in such caches. This process continued during the Neolithic through the burial of various productions: flint blades (cf. Sainte-Cécile-les-Vignes deposit, Vaucluse, France), copper ores and flat axes (cf. 263 Centeilles deposit, Siran, Hérault, France), or copper ornaments (cf. Miguro cave, Corconne, Gard, 264 France). This process was particularly developed during the Bronze Age (Gasco, Lachenal, Piningre 2018). This hiding of precious objects could signal disparities in the social order and connotate the 265 266 particular status of their owner(s). The example of the Salpêtre cave constitute, in the Midi de la France, a unique example. We should also mention the existence of two deposits in western France « au contact 267 des ouvrages symboliques que sont la stèle en grès de la Table des Marchands et l'enceinte de pierres 268 269 dressées sur l'îlot d'Er Lannic » ("in contact with the symbolic works of the Table des Marchands stelae 270 and the raised stone enclosure of the Er Lannic islet") (Cassen et al. 2019).

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#### 272 5. Chrono-cultural framework: The Early and Middle Neolithic

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274 The chrono-cultural insertion of variscite meridional ornaments used to be subject to debate. J. Arnal, 275 taking as basis the closed ensembles represented by the Catalan tombs (Sepulcros de fosa culture, Spain), 276 systematically attributed them to the middle Neolithic and made it a cornerstone to date the corridor 277 dolmens of eastern Languedoc (Arnal 1963). In opposition, J. Audibert, witnessing the presence of callais beads within meridional collective tombs, dated them as probably Chalcolithic (Audibert 1962). 278 Progressively, further research meant a long-term chronology could be established for variscite 279 280 ornaments (Barge 1982, Roscian et al. 1992), now spanning over two millennia (c. 4500-2200 BC). It is however not impossible that this chronology could be stretched further. Indeed, we are not aware of 281 beads pertaining to the *Cardial-Epicardial* cycle in the Midi ( $\approx$  5500-4500 BC) whilst on the southern 282 Pyrenean side such ornaments are attested in a sporadic manner during the Cardial (Chaves cave, 283 Aragon), and more often during the Epicardial (Cova de les Lladres, in western Vallès, with a deposit 284 285 of 139 pendants), and also in the Molinot (during the third quarter of the 5<sup>th</sup> millennium, during which 286 the first exploitations of the Gavà mines are attested: Bosch 2019, Molist and Oliva 2019). To the North 287 of the Pyrenees, the first known manifestations are attested within the Juberrian context (Llo oppidum, 288 Pyrénées-Orientales, France, around 4500 BC) and become more obvious within the Montbolo-*Chasséen* context during the last centuries of the 5<sup>th</sup> millennium (grave 33 of Crès, Hérault; tomb 59 of 289 Caramany, Pyrénées-Orientales ; silo 19 of Cugnaux, Haute-Garonne, France ; Vaquer 2019). If the 290 291 Pompignan deposit is contemporaneous with the *Chasséen* occupation of the cave, of which a typical 292 decorated sherd was found near the beads, we cannot exclude a date from the end of the 5th millennium. It is during the optimum functioning of the Gavà mines, linked to the peak of the Sepulcros de fosa 293 294 culture (Vallesian/Solsonian, Spain), that the Catalan variscite importations towards the Midi region reach their peak, during the middle - recent Chasséen, between 4000 and 3500 BCE. 295 296 Several sites from the Roussillon, Aude, and Toulouse region, and up to the Rhone corridor (Saint-Paul-

Several sites from the Roussillon, Aude, and Toulouse region, and up to the Rhone corridor (Saint-Paul Trois-Châteaux, Drôme, France) have yielded beads of various forms, including several « heavy »
 pieces, olive-shaped, of a kind well-known in Catalonia (tomb 1 of Villeneuve-Tolosane, the "royal"
 tomb A 185 from Saint-Michel-du-Touch, Toulouse, Haute-Garonne, France) (Méroc and Simonnet

300 1979). Some of the Pompignan pieces show similarities within the same ambiance. Within cave IV from

- 301 las Claousos at Auriac (Aude, France), used as a collective tomb and for which a  ${}^{14}C$  date yielded a
- 302 recent *Chasséen* attribution, six cylindrical or barrel-shaped variscite beads could come from Encinasola
- 303 rather than Gavà (Guilaine et al. 2019). Could it be that this reflects a competition between the two
- 304 deposits (Can Tintorer and Pico Centeno) at a time when Gavà's production was slowing down?
- 305

#### **6. Continuities in Final Neolithic**

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308 We are less informed on the origin of the small meridional variscite beads, which remain abundant in 309 the Midi de la France during the entire final Neolithic, between 3500 and 2200 BCE. Their typology 310 isolates them from the previously evoked examples. Large, olive-shaped or thick beads have more or 311 less disappeared except in rare cases (Fées de Lunas cave, Hérault, France). The only remaining beads 312 are small discoid models, with irregular outlines, not always well calibrated. The ensemble excavated 313 from the Cabrials collective tombs, near Béziers (Hérault, France), with a flattened shape, is dated to 314 the final Neolithic 1 (Tchéremissinoff 2012). Most of the meridional beads issued from collective tombs (dolmens, natural caves, hypogees) with a long-term use, sometimes over a millennium. They can be 315 numerous: minimum 130 in the Clot-de-l'Oste dolmen at Bouisse (Aude, France; Guilaine et al. 2015). 316 317 Two beads from the René-Carrié cave at Termes (Aude, France) are composed of materials attributing their origin to Gavà. The exploitation of the Catalan deposits could therefore have been pursued until 318 319 after 3500 BC, in a less intense manner.

The end of the use of variscite stone could be placed towards the end of the Chalcolithic with the diffusion of the Bell Beaker phenomenon. An ensemble of 23 beads accompanied the ancient Bell Beaker deposit found within the ditch surrounding the Mourral site in Trèbes (Aude, France; Vaquer 1998). This is a regional association, since the callais is not a general marker of the European Bell Beaker. This example shows that the south of France tradition of using this material could still exist whilst the maritime Bell Beaker phenomenon pursued its supraregional expansion.

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#### 327 Conclusion

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The bead ensemble from the Salpêtre cave of Pompignan originates from the Gavà mines, near 329 Barcelona, some 370 km as the crow flies (this distance being a general indication, since the transfer 330 331 implied either crossing the eastern Pyrenees, or their circumvolution by navigating the Gulf of Lion). Beyond the burial of this ensemble, which is reminiscent of a cache, perhaps during a ritual act, this 332 333 discovery confirms the existing relationship between two Neolithic cultures separated by the physical barrier of the Pyrenees. To the south, the Sepulcros de fosa, which exploited the mines, fabricated and 334 335 diffused the ornaments; and to the North, the meridional Chasséen who, from the Toulouse region to the Alps, received these jewels which were appreciated for their "exotic" provenance, relative as it was. 336

- 337 While slighty offset within the Cevenol mountains, the Pompignan ensemble is located relatively close
- to a geographic axis the Bas-Languedoc. Through this axis, Rhone corridor material circulated (blond
- flint from the Vaucluse, alpine eclogite), which travelled opposite ways to the variscite and crossed the
- 340 Pyrenees to furnish cores, flint blades, and polished axes for populations of Catalonia. The Pompignan
- 341 deposit is therefore at the heart of a reciprocity system demonstrating the strong complementarity in
- 342 exchanges between Neolithic cultures of the first half of the 4th millennium BCE.
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#### 344 Références bibliographiques

- Arnal, J., 1963. Les Dolmens du Département de L'Hérault, Presses Universitaires de France. ed,
  Préhistoire. Paris.
- Audibert, J., 1962. La civilisation chalcolithique du Languedoc oriental, Bordighera. ed, monographies
   préhistoriques et archéologiques, 4. Institut International d'Études Ligures, Montpellier.
- Barbaza, M., 1981. Recherches sur l'Epipaléolithique en Languedoc et Catalogne (Thèse de 3ème
  cycle). EHESS, Toulouse.
- Barge, H., 1982. Les Parures du Néolithique ancien au début de l'âge des métaux en Languedoc, CNRS.
  ed. Laboratoire d'anthropologie et de Préhistoire des pays de la Méditerranée occidentale, Marseille.
- Bar-Yosef Mayer, D., Porat, N., 2008. Green stone beads at the dawn of agriculture. Proceedings of the
  National Academy of Sciences of the United States of America. 105, 8548–51.
  https://doi.org/10.1073/pnas.0709931105
- Beck, H.C., 2006. Classification and Nomenclature of Beads and Pendants. BEADS : Journal of the
  Society of Bead Researchers, 1e ed., 1926, 18, pp. 1–76.
- 359 Bosch Argilagos, J., 2019. Gavà (Barcelone), mine et atelier au Néolithique, in: Querré, G., Cassen, S.,
- 360 Vigier, E. (Eds.), La parure en callaïs du Néolithique européen. Presented at the Colloque International,
- 361 Callaïs, Carnac 2015, Archaeopress Archaeology, Oxford, pp. 43–58.
- 362 Cartailhac, É., 1889. La France préhistorique : d'après les sépultures et les monuments, Félix Alcan. ed.
  363 Paris.
- Cassen, S., Boujot, C., Charvet, A., Grimaud, V., Le Maux, N., Le Pennec, C., Querré, G., Vigier, E.,
  Obeltz, C., Prodéo, F., Villes, A., 2019. La parure en callaïs (variscite et turquoise), au Néolithique, dans
  la moitié nord de la France. Corpus et contextes, in: Querré, G., Cassen, S., Vigier, E. (Eds.), La parure
  en callaïs du Néolithique européen. Presented at the Colloque International, Callaïs, Carnac 2015,
  Archaeopress Archaeology, Oxford, pp. 203–279.
- Chantret, F., Guilaine, J., Guillemaut, A., 1970. Analyses de quelques perles méridionales en « callais ».
  Bulletin de la Société préhistorique française. 67, pp. 216–219. https://doi.org/10.3406/bspf.1970.4258
- 371 Damour, A.-A., 1864. Sur la Callaïs : nouveau phosphate d'alumine hydraté recueilli dans un tombeau
  372 celtique du Morbihan. Comptes rendus l'Académie des Sciences, pp. 936–940.
- Domínguez-Bella, S. (2004). Variscite, a prestige mineral in the Neolithic-Aeneolithic Europe. Raw
   material sources and possible distribution routes. Slovak Geological Magazine, 10, 1-2. pp. 147-152.

- 375 Domínguez-Bella, R., Ramos-Muñoz, J., 2019. Pico Centeno prehiostotic variscite muines (Encinasola,
- Huelva, SW Spain), in: Querré, G., Cassen, S., Vigier, E. (Eds.), La parure en callaïs du Néolithique
  européen. Presented at the Colloque International, Callaïs, Carnac 2015, Archaeopress Archaeology,
- **378** Oxford, pp. 71–83.
- 379 Fàbregas Valcarce, R., Rodríguez-Rellán, C., 2019. Palazuelo de las Cuevas and the exploitation of
- variscite in the North- West of Iberia, in: Querré, G., Cassen, S., Vigier, E. (Eds.), La parure en callaïs
  du Néolithique européen. Presented at the Colloque International, Callaïs, Carnac 2015, Archaeopress
- 382 Archaeology, Oxford, pp. 59–70.
- Fernandez, A., Moro, C., Dabard, M.-P., 2019. Les minéralisations de variscite du Sud-Ouest de
  l'Europe : caractéristiques minéralogiques, géochimiques et genèse, in: Querré, G., Cassen, S., Vigier,
  E. (Eds.), La parure en callaïs du Néolithique européen. Presented at the Colloque International, Callaïs,
- 386 Carnac 2015, Archaeopress Archaeology, Oxford, pp. 9–23.
- Jeanjean, A., 1871. L'Homme et les animaux dans les cavernes des Basses-Cévennes. Mémoires de
  l'Académie du Gard, Nîmes, pp. 139–228.
- Gascó, J., Lachenal, T., Piningre, J.-F., 2018. Un monde de dépôts métalliques, in: Guilaine, J., Garcia,
  D. (Eds.), La Protohistoire de La France. Hermann, Paris, pp. 339–353.
- Guilaine, J., 2011. Parures de pierre et objets symboliques, in: Shillourokambos. Un Établissement
  Néolithique Pré-Céramique à Chypre. Les Fouilles Du Secteur 1. Errance/Ecole Française d'Athènes,
  Paris, pp. 789–806.
- Guilaine, J., Muñoz, A.-M., 1964. La civilisation catalane des "Sepulcros de fosa" et les sépultures
  néolithiques du Sud de la France. Revue d'Études Ligures XXX, pp.5–30.
- Guilaine, J., Rigaud, S., 2021. Parures de pierre et objets symboliques, in: Shillourokambos : un
  Établissement Néolithique Pré-Céramique à Chypre. Les fouilles du secteur 3. CNRS, Paris, pp. 435–
  471.
- 399 Guilaine, J., Vaquer, J., Gratuze, B., Querré, G., 2015. Les perles en variscite de la grotte de Las
- Claousos IV et de la grotte René-Carrié, in: Guilaine, J., Vaquer, J., Zammit, J. (Eds.), Grottes
  sépulcrales préhistoriques des Hautes-Corbières. Toulouse, pp. 89–104.
- Herbaut, F., Querré, G., 2004. La parure néolithique en variscite dans le sud de l'Armorique. Bulletin
  de la Société préhistorique française, 101, pp.497–520. https://doi.org/10.3406/bspf.2004.13029
- 404 Méroc, L., 1962. Le village et la sépulture chasséens de Villeneuve-Tolosane (Haute-Garonne, France).
  405 Zephyrus, XIII, pp. 94–96.
- 406 Méroc, L., Simonnet, G., 1979. Les sépultures chasséennes de Saint-Michel-du-Touch à Toulouse
  407 (Haute-Garonne). Bulletin de la Société préhistorique française, Études et Travaux 76, 10-12, pp. 379–
  408 407. https://doi.org/10.3406/bspf.1979.5163
- Mohen, J.-P., 1978. Tumulus des Pyrénées françaises, in: Actes Du 2e Col.loqui Internacional
  d'Arqueología de Puigcerdà. Presented at the Els pobles pre-romans del Pirineu, Institut d'Estudis
  ceretans, Puigcerdà, pp. 97–108.
- Molist Montana, M., Oliva Poveda, M., 2019. La parure en variscite au Néolithique dans la partie est de
  l'Espagne, in: Querré, G., Cassen, S., Vigier, E. (Eds.), La parure en callaïs du Néolithique européen.

- 414 Presented at the Colloque International, Callaïs, Carnac 2015, Archaeopress Archaeology, Oxford, pp.
  415 351–364.
- 416 Moro Benito, MC; Montero Gómez, JM; Gil Agero, M.; Cembranos Pérez, ML; Pérez del Villar
- 417 Guillén, L.; Fernández Fernández, A. ; Hernández, E. (1992) Aluminofosfatitas silúricas de la península
- 418 Ibérica: síntesis preliminar. Boletín de la Sociedad Española de Mineralogía, Vol. 15, nº. 1, pp. 227-
- 419 232. ISSN 0210-6558.
- 420 Moro Benito, MC ; Pérez del Villar Guillén, L.; Fernández Fernández, A. ; Cembranos Pérez, ML ; Gil
- 421 Cid, MD (1995) Las Mineralizaciones estratiformes de variscita (Aluminofosfatitas) silúricas de los
  422 Sinformes de Alcañices (Zamora) y Terena (Huelva): (España). Boletín Geológico y Minero, Vol. 106,
  423 nº 3, pp. 33-49. ISSN 0366-0176.
- 424 Muñoz, A.-M., 1965. La cultura neolítica catalana de los «Sepulcros de Fosa». Universidad de
  425 Barcelona, Instituto de Arqueología y Prehistoria, Barcelona.
- 426 Odriozola, C.P., Linares-Catela, J.A., Hurtado-Pérez, V., 2010. Variscite source and source analysis:
- 427 testing assumptions at Pico Centeno (Encinasola, Spain). Journal of Archaeological Science. 37, pp.
- 428 3146–3157. https://doi.org/10.1016/j.jas.2010.07.016
- 429 Peltenburg, E., 1998. Excavations at Kissonerga-Mosphilia 1979-1992, Studies In Mediterranean
  430 Archaeology. LXX:2, Paul Åströms Förlag, Jonsered.
- 431 Piette, É., 1881. Note sur les tumulus de Bartrès et d'Ossun, Durand. ed, Matériaux pour l'Histoire de
  432 l'Homme. Durand, Toulouse.
- 433 Querré, G., Calligaro, T., Cassen, S., Dabard, M.-P., Domínguez-Bella, S., 2019. Provenance des parures
- 434 en variscite du Néolithique européen : élaboration d'un modèle chimiométrique, in: Querré, G., Cassen,
- 435 S., Vigier, E. (Eds.), La parure en callaïs du Néolithique européen. Presented at the Colloque
- 436International, Callaïs, Carnac 2015, Archaeopress Archaeology, Oxford, pp. 105–128.
- 437 Querré, G., Calligaro, T., Domínguez-Bella, S., Cassen, S., 2014. PIXE analyses over a long period:
- The case of Neolithic variscite jewels from Western Europe (5th–3th millennium BC). Nuclear
  Instruments and Methods in Physics Research Section B : Beam Interactions with Materials and Atoms,
- The 13th International Conference on Particle Induced X-ray Emission (PIXE 2013) 318, part A, pp.
- 440 The 15th International Conference on Fattere induced X-ray Emission (Fixe 2015) 516, part F
- 441 149–156. https://doi.org/10.1016/j.nimb.2013.07.033
- 442 Querré, G., Cassen, S., Domínguez-Bella, S., 2012. La variscite ibérique. Exploitation, diffusion au
- 443 cours du Néolithique, in : Marchand, G., Querré, G. (Eds.), Roches et sociétés de la Préhistoire. Entre
  444 massifs cristallins et bassins sédimentaires. Presses universitaires de Rennes.
- 445 Querré, G., Herbault, F., Calligaro, T., 2008. Transport of Neolithic variscites demonstrated by PIXE
  446 analysis. X- Ray Spectrom. 37, pp. 116–120. https://doi.org/10.1002/xrs.1031
- 447 Roscian, S., Claustre, F., Dietrich, J.E., 1992. Les parures du Midi méditerranéen du Néolithique ancien
- 448 à l'Age du Bronze : origine et circulation des matières premières. Gallia Préhistoire 34, pp. 209–257.
- 449 https://doi.org/10.3406/galip.1992.2303
- Tchérémissinoff, Y.T., 2012. La sépulture collective mégalithique de Cabrials (Béziers, Hérault). Une
  petite allée sépulcrale enterrée du début du Néolithique final. Préhistoires Méditerranéennes 147.

Vaquer, J., 2019. Les éléments de parure en callaïs dans le midi de la france au Néolithique et au
Chalcolithique, in: Querré, G., Cassen, S., Vigier, E. (Eds.), La parure en callaïs du Néolithique
européen. Presented at the Colloque International, Callaïs, Carnac 2015, Archaeopress Archaeology,
Oxford, pp. 333–349.

Vaquer, J., 1998. Le Mourral, Trèbes (Aude) A fortified languedocian late neolithic site reoccupied by
bell beakers, in: Benz, M., Van Willigen, S. (Eds.), Some New Approaches to The Bell Beaker
Phenomenon, Lost Paradise...? Presented at the Proceedings of the 2" Meeting of the "Association
Archéologie et Gobelets," Oxford Archéologie, Feldberg (Germany), pp. 15–21.

Viel, L., Bonnardin, S., Bailly, M., Margarit, X., 2019. Perles supposées en variscite du sud-est de la
France (Arles-Fontvielle, IV<sup>e</sup> mill. av. J.-C.), in: Querré, G., Cassen, S., Vigier, E. (Eds.), La parure en
callaïs du Néolithique européen. Presented at the Colloque International, Callaïs, Carnac 2015,
Archaeopress Archaeology, Oxford, pp. 365–378.

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- 485486 Illustrations
- 488 Fig. 1 Geographic location of the Salpêtre cave (Pompignan, Gard)
- 490 Fig. 2 Topographic location of the cavity (drawing by J. Coularou)
- 492 Fig. 3 Cave entrance (photograph by A. Aigoin)
- 494 Fig. 4 Plan and stratigraphy of the cave. Localisation of the bead ensemble (drawing by N. Boinet)
- 496 Fig. 5 Localisation of the discovery near the base of the cave wall (photograph by A. Aigoin)

498 Fig. 6 - Close up: localisation of the beads near a decorated sherd and a pecten shell (drawing by
 499 J. Coularou)

Fig. 7 – Sherd from the ventral part of a decorated Chasséen globulous recipient discovered near the
 variscite beads (drawing by J. Coularou)

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- 504 Fig. 8 Saint-Jacques shell (*Pecten jacobeus*) discovered near the beads (drawing by J. Coularou)
- 506 Fig. 9 Necklace restitution picture of the 17 variscite beads deposit (photograph by H. Vergély)
- 508 Fig. 10 Numbering of the 17 beads analysed according to the PIXE method (photograph by 509 H. Vergély).
- 511 Fig. 11 Details of the 17 variscite elements composing the ensemble (photograph by H. Vergély).
- Fig. 12 Variscite beads of the Salpêtre cave in Pompignan being analysed through the PIXE method,
  in front of the beam extract from the AGLAE particle accelerator (C2RMF, Louvre Palace, Paris)
  (realisation by G. Querré).
- Fig. 13 Analyses of the variscite beads of the Salpêtre cave in Pompignan (number of objets analysed:
  17; number of analysis : 17). In the diagrams : V-Cr-Fe; U-Zr; (Cr + V) As; Ca-Si in comparison to
  the referential for the 5 principal variscite sources in western Europe: Gavà (Barcelona, Spain; GA),
  Encinasola (Huelva, Spain; EN), El Bostal (Zamora, Spain; EB), Palazuelo (Zamora, Spain; PL),
  Pannecé (Loire-Atlantique, France; PA) (realisation by G. Querré).
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- Table 2 Color of the 17 variscite éléments of the Salpêtre cave deposits (Pompignan, Gard) (realisation
   H. Vergély).
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Table 3 – Chemical elementary composition obtained through the PIXE method and of each of the
Salpêtre cave (Pompignan, Gard). Analyses are expressed as ppm (part per million). In the table, for
each element, we also find the average concentrations, their standard deviation, minimum and maximum
concentrations as well as the detection limits (LDD) during the bead's analyses in July 2016 (realisation
by G. Querré).

- Table 4 Elemental analysis obtained by PIXE for each of the variscite deposits, in order to develop the
  chemometric model. Concentrations expressed in parts per million (ppm) and in elemental form. V\*,
  Cr\* and Fe\* are the relative percentages of vanadium, chromium and iron contents normalized to the
  sum V+Cr+Fe.
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## 541 *Formatting of funding sources*

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laboratory (UMR5608).

<sup>Table 1 – Measures and weights of the 17 variscite éléments of the Salpêtre cave deposits (Pompignan,
Gard) (realisation H. Vergély).</sup> 

## The variscite « necklace » of the Salpêtre cave, at Pompignan (Gard, France)

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#### 8 Abstract :.

A 17 variscite beads set, buried in the Salpêtre cave in Pompignan (Gard, France) constitutes a
unique Middle Neolithic set of ornaments in the southern France. PIXE (Particle Induced X-ray
Emission) chemical analysis of these coins and reference samples from possible sources indicates a
mineral origin from the Gava mines, near Barcelona (Spain). This discovery reveals an exchange
system of the materials or finished objects on both sides of the Pyrenees mountains during the first
half of the 4th millennium BC.

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16 Keywords : Neolithic of western Europe, Middle Neolithic (Chasséenne culture), ornaments, variscite,

17 provenance, chemical analysis, PIXE (Particle-Induced X-ray Emission)

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20 Adornments, in prehistoric populations, is not just ornaments but also an «identity language», functioning as a sexual, social, or ethnical marker. In Neolithic times, in the Near East as in Europe, 21 22 green stones were largely used in jewellery making. As such, in Israel, from the recent Natufian to the pre-pottery Neolithic B (PPNB), between -11000 and -7000 BC, several minerals bearing this hue 23 (apatite, fluorapatite, chrysocolla, malachite, turquoise, amazonite, serpentinite) were used to make 24 beads. The source of the raw materials was often distant from the user location of the productions. 25 And, the choice of a green hue has sometimes been interpreted as a symbol of vegetation, of growing 26 plants, of fertility (Bar Yosef Mayer and Porat 2008). In Cyprus, picrolite, the local green mineral, was 27 largely exploited in jewellery production from the PPNA (pre-pottery Neolithic A) until the 28 Chalcolithic (Guilaine, 2011; Guilaine and Rigaud, 2021; Peltenburg, 1998). During the Neolithic of 29 western Europe, another green rock, the variscite, was used in the fabrication of necklaces during 30 almost the entire period, from the 6<sup>th</sup> to the 3<sup>rd</sup> millennium BCE. This mineral is a kind of hydrated 31 32 aluminium phosphate, also called variscite, the formula for which is  $AIPO_4 \cdot 2 H_2O$ . Its deposits are 33 essentially located within the Iberian Peninsula, but also in France and Sardinia (Fernandez et 34 al. 2019).

In Brittany, the presence of beads made of variscite is attested in Neolithic contexts – often funerary ones – since the 19<sup>th</sup> century. Such ornamental objects were discovered in the great Carnaccean tumuli (Tumiac, Saint-Michel, Mané er Hroeck) as well as several Armorican dolmens (Kercado). Right away, the question of the identification and origin of the material was raised. As early as 1864, mineralogist A. Damour studied a few specimens of these light green minerals and identified them as 40 made of a hydrated aluminide phosphate (Damour 1864). Faced with the quality of such productions,
41 A. Damour, followed by A. Lacroix believed in a distant origin of the raw material, and this
42 hypothesis of an exotic, distant rock prevailed for a long time (Veiga Ferreira, 1950).

In southern France, E. Cartailhac was one of the first to be interested in these ornaments, some of 43 44 which came from the dolmens of Aveyron and of Lozère, where he first started working. In 1876, he participated to research on the Castellet hypogeum (Fontvieille, Bouches-du-Rhône), from which an 45 ensemble of 114 beads was extracted. In 1881 he published in the journal he edited, Matériaux, E. 46 47 Piette's article mentioning the « necklace » made of 93 beads issued from one of the compartments of 48 the Halliade monuments at Bartrès (High-Pyreneans; Piette 1881, Mohen 1978). This led him to give 49 an overview of these adornments, the presence of which he identified in Brittany, Spain, Portugal (in 50 the hypogea of Palmela) and in the Midi (Cartailhac 1889). In parallel, research on the north-eastern 51 region of the Iberian Peninsula revealed the existence of numerous beads of the same material within the earth or cists of that region. A. Muñoz, in her thesis on the « Sepulcros de fosa » of Catalonia, 52 53 provided a synthesis of this issue and brought some doubt to the hypothesis of a distant origin (Muñoz 54 1965). In the Midi, a few specimens were brought to the attention of two researchers of the mineralogy 55 department of the Commissariat à l'Énergie Atomique (F. Chantret, A. Guillemaut) and revealed that the term callais could regroup variscite, turquoise, or malachite ornaments (Chantret et al. 1970). 56 57 Variscite ornaments were essentially recognised within the funerary context in individual inhumations (within a dug-out grave or a stone box) or in collective tombs (megalithic structures, hypogea, 58 sepulchral caves). 59

60 A new and determining impulsion in approaches to the topic came from the recognition, starting from the 70s onwards, of the variscite mines of Gavà, near Barcelona in Spain (Domínguez-Bella 2004, 61 62 Bosch Argilagos 2019, Molist and Oliva 2019, Moro et al. 1992 and 1995). Following this, renewed 63 attention was given to the geological formations of the Zamora region, where several deposits had 64 been identified (Fabregas Valcarce and Rodriguez-Rellan 2019). Then came the identification of the 65 Pico Centeno deposit (Encinasola, Huelva, Spain) in western Andalusia, (Odriozola et al. 2010, Dominguez-Bella and Ramos-Muñoz 2019). Uncovering the existence of these deposits was combined 66 with diverse analyses programs in order to establish precise baseline referentials characterising each of 67 68 these three sources: Gavà, the Zamora deposit, and Encinasola. In turn, this meant various scenarios 69 for the circulation of variscite and turquoise during the Neolithic (between 5500 and 2000 BC) at the scale of western Europe could be proposed (Querré et al. 2019). 70

71 The productions from these various deposits may have similarities and differences. Thus the round or 72 barrel beads are ubiquitous. The large pendants with polar perforation from Breton sites come from 73 Encinasola in the fifth millennium and then, from 4000 BC, from Palazuelo de las Cuevas. The 74 Portuguese cylindrical beads come from deposits in the Zamora region. The large biconical beads with 75 axial perforation come mainly from Gava. The small irregular beads from Final Neolithic collective **76** burials in the south of France do not have, due to lack of analysis, a clear provenance (Querré et al.

**77 2019**).

A remarkable variscite bead ensemble was discovered in the 70s in the Salpêtre cave at Pompignan (Gard, France). Whilst known and presented to the public in various recent exhibits, this discovery had never been published in detail. This article aims to fill this gap in knowledge and communicate the results of the analyses concerning this original ensemble, unique for the south of France.

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- 83 **1. The site**
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The Salpêtre cave is located on the commune of Pompignan (Gard, France), 30 km north of the town of Montpellier (fig. 1). It opens on the northern flank of the Hortus causse, at the foot of the Taillade cliff, at an average altitude of 380 m (fig. 2 and 3).

88 This cave was subjected to various investigations over a century. The oldest was by A. Jeanjean who 89 mentioned its interest in a synthesis on the prehistory of the Basses-Cévennes region (Jeanjean 1871). 90 Around 1960, Jacques Audibert, research associate at the CNRS, undertook a little work in the cavity 91 (Audibert 1962). These two operations meant middle Neolithic levels (Chasséen) were identified, as 92 well as some final Neolithic (Ferrières and Fontbouïsse cultural groups). In the 70s, S. Cours re-sieved 93 some of the waste heaps and prospected other areas of the cave. This is when the necklace described in 94 this article was discovered. Following this find, S. Cours and J. Coularou dug a deep test pit 10 m from the cave entrance, 3 m deep. At that level, they found hardened sediment. They recognised the 95 96 reworked aspect of the Neolithic levels but also identified the existence of palaeolithic levels still in place (Azilian, Magdalenian, Mousterian). The Azilian artefacts were described in M. Barbaza's 97 98 doctoral thesis (Barbaza 1981). It is possible that some Neolithic levels still in place are present in the 99 deepest part of the cavity, under the spoil heaps from older excavations, which would allow <sup>14</sup>C dating,

- 100 none of which was done at this time.
- 101 In 1972, M. Escalon de Fonton dug a new test pit, looking for the previously recognised Magdalenian
- 102 level. The investigation remained fruitless, but a few years later in 1978-1979, L. Meignen pursued

this research axis and confirmed the presence of Mousterian occupations.

The Salpêtre cave is a long corridor following a north-south cleft; it is 80 m long and 2 m large and reaches a height of 5 m. Towards the deepest end of the cavity, a lateral diverticulum opens into a small room (fig. 4 and 5).

107 The variscite bead cache was found 41.5 m from the cave entrance, where the corridor narrows, and

the walls are only 0.5 m apart. This narrowing is even more marked by the fact that a natural stepexists there, forming a threshold of sorts (fig. 6).

110 The beads were discovered grouped within a small space. Perhaps they were originally linked by a

111 thread that has now disappeared. In the immediate vicinity, a large ceramic sherd and a pecten shell

112 were found. The sherd (9 x 8 cm) has a curved profile and belongs to a vessel with a thick profile (1

- 113 cm). It is identified as a fragment of the ventral part of a spherical container, the diameter of which is
- estimated to have been around 26 cm. This element bears an engraved decor made of three horizontal
- strips: a central one with a checkered motif, and two surrounding ones (superior and inferior)
- 116 presenting a motif made of four lines arranged as rafters. This resembles spherical to cylindrical
- 117 collared containers typical of the classical *Chasséen*, the widest part of which was often decorated
- 118 with geometrical patterned strips.
- 119 The mollusc shell is a scallop (a Saint-Jacques shell: *Pecten jacobeus*), rather large (width: 13 cm).
- 120 We cannot exclude that the beads were originally placed within the bivalve.
- 121

#### 122 **2. Description of the Beads**

123

124 The excavated lot includes 17 elements which could originally constitute a necklace (fig. 9)<sup>1</sup>.

125 All the beads, of various sizes, bear witness to a final fabrication state necessitating an important 126 technical investment. Measures were carried out on the bead ensemble: maximum diameter 127 (median point of the maximum width of the transversal section), minimal diameter (median point of 128 the minimum width of the transversal section), diameter of the perforation, maximum and minimum 129 thickness (or length), and maximal and minimum diameter of the bead's extremities. These last 130 measures correspond to the term « méplat » used by F. Herbaut and G. Querré (Herbaut, Querré 131 2004). The maximum diameter of the beads is between 7.2 and 25.5 mm, for a thickness between 5.9 and 40.1 mm. The maximum diameter of the perforations is comprised between 2.1 and 4.3 mm. 132 133 Dimensions, weights and color are resumed in table 1 and in table 2.

Discussing the typological aspect of these beads is difficult, because variscite ornament productions 134 do not correspond to fabrication standards observed on other mineral matter, and we are 135 136 confronted to a lack of current research (in France) on this aspect of variscite productions (Viel et al. 2019). Looking to the reference work on ornaments of the French Midi region (Barge 1982), the vast 137 138 majority of the beads of the Salpêtre cave ensemble can be qualified as irregular. It is true that the 139 typological classification of these objects is problematic since they often present asymmetries in their 140 longitudinal and transversal sections. In publications, they are generally described as olive-shaped, or 141 flattened beads...

142 In order to better define the production criteria of the workshops, which seems to evolve over time, 143 typological classification work could help us qualify these productions, such as the one published by 144 H. Beck in 1926 « Classification and Nomenclature of Beads and Pendants » (Beck 2006). This 145 typological example allows for the description and classification of beads whilst considering their shape from the section and the front. Synthetically speaking, the Salpêtre ensemble presents, in 146 147 most cases, an elliptical transversal section (more or less regular) combined with a convex cylindrical 148 longitudinal section. Several beads present a transversal section associated with a convex, « barrel-149 shaped » or cylindrical longitudinal section, with both extremities being convex (n°7 and 8). One bead 150 reveals a quadrangular transversal section (n°5) and another can be qualified as a conic trunk (n°17).

<sup>&</sup>lt;sup>1</sup> These artefacts are kept at the Centre de Documentation Archéologique du Gard (CDAG), 396 avenue Joliot Curie, 30900 Nîmes. Their analysis took place within a PCR (Collective Research Program) titled « Milieu et Peuplement en Languedoc Occidental du Néolithique au Bronze ancien » (dir. M. Gandelin)

The polishing of the beads is advanced, giving an intense shine visible on both the bead's extremities and their longitudinal sections. This aspect is also visible on the extremities' surfaces presenting great irregularities such as bead n°13, or on the section of bead n°3. Only one bead (n°2) presents unpolished irregularities that can be interpreted as impacts (3 in total), which must have occurred after the bead's fabrication and bears witness to probable shocks. Besides, traces of red coloured matter (ochres ?; not analysed) are visible inside these asperities. The polishing is so advanced that it is difficult to discern traces of the manufacturing techniques used before that stage of fabrication.

Perforations are all cylindrical, and their regularity implies the use of a system to hold the pieces in place, and the use of flint drills mounted on a rotating axis to pierce the beads. A boring step allowed for the regularity of the perforations. These technical observations correspond to the research made on variscite's chaine opératoire in Catalonia (Molist Montana, Oliva Poveda 2019). The morphology of the beads found in the Salpêtre cave is close to those found in the Gavà workshop and find many comparisons with the beads discovered in grave 83 of the Mine (Beta 155.686 : 5220±110 BP ; 4326 – 3793 cal BC (95.4%) ; Bosch Argilagos 2019).

165

166 3. Chemical and provenance analyses of the callais bead necklace from the Salpêtre cave167 (Pompignan, Gard).

168

The 17 callaïs beads probably composing a necklace (fig. 10 and 11) found during the Salpêtre cave excavation at Pompignan were subjected to an elemental chemical analysis in order to confirm their mineralogic identity (variscite) and to determine their provenance. This determination was established using the chemometric model elaborated based on the main European sources of variscite (Querré et al, 2008; 2012; 2014; and forthcoming) as well as PIXE analyses of the Pompignan beads.

174

#### 175 **3.1. Methodology**

176

The analyses were carried out using the PIXE method (*Particle-Induced X-ray Emission*) using the
AGLAE (Elementary Analysis Accelerator of the *Grand Louvre*) set up from the C2RMF (The

179 research and restoration centre of *Musées de France*) of the Louvre Palace (Paris).

180 The PIXE method was applied to the callais according to a protocol established by one of the authors (G.Q.). This method of elemental analysis is non-destructive and fast. It is based on the fluorescence 181 of X-rays characteristic of the material. The elemental composition of archaeological pieces is 182 compared with a reference sample, representative of the 5 largest European variscite deposits, likely to 183 have been exploited during the Neolithic. The concentrations of major elements allow us to recognize 184 185 the mineralogical nature of the veins; the spectrum of minor elements and/or traces shows intrinsic 186 variations, specific to each deposit. The contents of iron, vanadium, chromium, arsenic, silicon, calcium, zirconium, uranium, their absolute variations and concentration rates are particularly relevant 187

to discriminate potential geological sources. This model can be used to determine the provenance of
archaeological variscite pieces (Querré et al. 2019).

Protons of 3MeV were extracted in the shape of a beam a few micrometres in diameter. The bead 190 191 surface to be analysed was placed a few millimetres from the protons exit window. Four SDD 192 detectors gather the X-Rays created: one for the low energies corresponding to major elements, and three for high energies corresponding to trace elements (fig. 12 a and b). In order to have a mean 193 analysis of the bead's material, the beam scanned the surface over one millimetre square. The 194 195 acquisition time for the analysis of each bead was six minutes on average. The high and low energy 196 spectrums went through GUPIX and TRAUPIX software analysis, in order to obtain elementary 197 concentrations as well as the detection thresholds for all 24 chemical elements of interest.

198

#### 199 **3.2.** Chemical analysis of the beads

200

The concentration of the 24 chemical elements for each of the 17 beads can be found in table 3 and table 4. Three chemical elements are present in concentrations higher than 10 % (100.000 ppm): aluminium, silicon and phosphorus. Copper concentrations are low, 65 ppm on average. The beads are therefore indeed variscite, albeit a very impure form considering the high silicon proportions present within. After these three major elements, iron and copper are the most represented, with element concentrations reaching around 2 %. All other elements are found in minor or trace concentrations.

207

#### 208 **3.3. Provenance**

209

The chemometric model elaborated to determine the variscite's provenance was applied to the beadsfound in the Salpêtre cave.

The concentrations of the discriminating elements were compared to those of the five western European deposits where the beads might come from : the four Spanish deposits, the Gavà deposit (Barcelona) including the Can Tintorer mines (fig. 13, GA, and table 4), the Encinasola deposit and the mines of Pico Centeno in the Huelva region (fig. 13, EN, and table 4), El Bostal in the Zamora region (fig. 13, EB, and table 4), Palazuelo de las Cuevas (fig. 13, PL, and table 4), and the only

known French deposit : Pannecé, in the Loire-Atlantique region (fig. 13, PA, and table 4).

- 218
- 219 The relative proportions of the three most discriminating elements (V, Cr and Fe) are, on average, the
- 220 following: V\*=7.5 Cr\*=7.8 Fe\*=84.7. These proportions do not correspond to those of the Pannecé
- deposits, for which vanadium represents 50 % and within which chrome and iron reach 25% of the
- total each (see figure 13 as well). The average Cr/V ratio is  $1.39 \pm 1.05$  which is not compatible with
- the Palazuelo analyses ( $Cr/V = 5.08 \pm 6.48$ ), nor with those of El Bostal ( $Cr/V = 0.35 \pm 0.04$ ).

- 224 The U vs Zr diagram (fig. 13B and table 4) allows us to individualise the variscites from El Bostal,
- which has strong uranium concentrations (U =  $306 \pm 267$  ppm) and no zirconium (Zr =  $6 \pm 4$  ppm). In
- the case of the Salpêtre beads, the reverse phenomenon is observed, with low uranium concentration
- 227 (U=9  $\pm$ 5ppm), often inferior to the detection threshold (LDD = 19 ppm), and average zirconium
- 228 presence ( $Zr = 22 \pm 23$  ppm).
- 229 Concerning diagram (Cr+V) vs As (fig. 13C), the concentrations observed in the Pompignan variscite
- beads (Cr+V =  $2926 \pm 1003$  ppm; As =  $802 \pm 418$  ppm) are close to those of two deposits : Encinasola
- 231 (Cr+V=4198  $\pm 1429$  ppm ; As = 610  $\pm 802$  ppm) and Gavà (Cr+V = 809  $\pm 397$  ppm; As = 302
- $\pm 90$  ppm) but not those of three other sources : (Cr+V = 2926 \pm 1003 ppm ; As = 802 \pm 418 ppm)
- 233  $(Cr+V = 2926 \pm 1003 \text{ ppm}; \text{As} = 802 \pm 418 \text{ ppm}) (Cr+V = 2926 \pm 1003 \text{ ppm}; \text{As} = 802 \pm 418 \text{ ppm}).$
- Silicon concentrations are extremely high in the Salpêtre beads (Si =  $17.6 \pm 6\%$ ) and can be qualified
- as elevated as far as calcium is concerned (Ca =  $1.8 \pm 1.2$  %). This tends to indicate that the origins of
- the beads is the Gavà deposit, which is the only one to present elevated concentrations of both silicon
- and calcium (Si =  $11.2 \pm 7.4$  %; Ca =  $6.1 \pm 8.0$  %) (fig. 13 C and table 4).
- Together, all these geochemical analyses based on the chemometric model bring only one answer to the question of the origins of the Salpêtre cave – the Gavà deposit, in Spain. The high iron concentrations in contrast to the vanadium, the Cr/V ratio, as well as the high proportions of silicon and calcium are the most discriminating factors in this case.
- 242 This result is coherent with the identification of the site from which the beads originate as *Chasséen*,
- and thus belonging to the Mediterranean sphere, within which most variscite ornaments come from the
- 244 Can Tintorer mines (Gavà, Spain). The morphology of the large biconical beads from Pompignan is
- 245 the same as those deposited in the graves in the Barcelona area.
- 246

#### 247 4. A cache of ornaments from the southern Pyrenees

248

249 The analyses of the Salpêtre cave ensemble (Pompignan, Gard, France) bring precious information on the circulation of ornaments outside their fabrication area in a region - southern France - where these 250 analyses are still rare. Indeed, this ensemble is composed of various typological elements, including 251 252 large, olive-shaped beads perfectly comparable to the productions found on the southern flank of the eastern Pyrenees, and dated to the evolved Neolithic middle. The meridional cultural context is then 253 that of the Chasséen. The mineralogical composition of the Pompignan pieces demonstrate it is made 254 255 of variscite coming from the Gavà deposits. Beyond the synchronism proposed between the Sepulcros de fosa and the meridional Chasséen, which has long been argued for (Guilaine and Muñoz 1964), 256 these interactions are now better understood. As such, polished alpine stone axes and Bedoulian flint a 257 258 attested within the Catalan funerary artefacts, whilst variscite ornaments are themselves found within 259 Chasséen funerary deposits of the Toulouse region (Saint-Michel du Touch, Villeneuve-Tolosane) and 260 as far as the Rhône (Pompignan), or even the Lyon region (Tassin ; Vaquer 2019).

The Pompignan ensemble also presents an interest in that it is a deposit of rare pieces placed in a 261 262 cache, made within a cave, rather far from the cave's entrance. This deposit was hidden, perhaps 263 during a ritual act. We must therefore raise the possibility that as early as the middle Neolithic rare 264 pieces were hidden in such caches. This process continued during the Neolithic through the burial of various productions: flint blades (cf. Sainte-Cécile-les-Vignes deposit, Vaucluse, France), copper ores 265 and flat axes (cf. Centeilles deposit, Siran, Hérault, France), or copper ornaments (cf. Miguro cave, 266 Corconne, Gard, France). This process was particularly developed during the Bronze Age (Gasco, 267 268 Lachenal, Piningre 2018). This hiding of precious objects could signal disparities in the social order 269 and connotate the particular status of their owner(s). The example of the Salpêtre cave constitute, in 270 the Midi de la France, a unique example. We should also mention the existence of two deposits in 271 western France « au contact des ouvrages symboliques que sont la stèle en grès de la Table des 272 Marchands et l'enceinte de pierres dressées sur l'îlot d'Er Lannic » ("in contact with the symbolic 273 works of the Table des Marchands stelae and the raised stone enclosure of the Er Lannic islet") 274 (Cassen et al. 2019).

275

#### 276 5. Chrono-cultural framework: The Early and Middle Neolithic

277

278 The chrono-cultural insertion of variscite meridional ornaments used to be subject to debate. J. Arnal, taking as basis the closed ensembles represented by the Catalan tombs (Sepulcros de fosa culture, 279 Spain), systematically attributed them to the middle Neolithic and made it a cornerstone to date the 280 281 corridor dolmens of eastern Languedoc (Arnal 1963). In opposition, J. Audibert, witnessing the presence of callais beads within meridional collective tombs, dated them as probably Chalcolithic 282 283 (Audibert 1962). Progressively, further research meant a long-term chronology could be established 284 for variscite ornaments (Barge 1982, Roscian et al. 1992), now spanning over two millennia (c. 4500-285 2200 BC). It is however not impossible that this chronology could be stretched further. Indeed, we are 286 not aware of beads pertaining to the Cardial-Epicardial cycle in the Midi (≈ 5500-4500 BC) whilst on 287 the southern Pyrenean side such ornaments are attested in a sporadic manner during the Cardial 288 (Chaves cave, Aragon), and more often during the Epicardial (Cova de les Lladres, in western Vallès, with a deposit of 139 pendants), and also in the Molinot (during the third quarter of the 5<sup>th</sup> millennium, 289 during which the first exploitations of the Gavà mines are attested: Bosch 2019, Molist and Oliva 290 2019). To the North of the Pyrenees, the first known manifestations are attested within the Juberrian 291 context (Llo oppidum, Pyrénées-Orientales, France, around 4500 BC) and become more obvious 292 within the Montbolo-Chasséen context during the last centuries of the 5th millennium (grave 33 of 293 294 Crès, Hérault; tomb 59 of Caramany, Pyrénées-Orientales; silo 19 of Cugnaux, Haute-Garonne, France ; Vaquer 2019). If the Pompignan deposit is contemporaneous with the *Chasséen* occupation of 295 296 the cave, of which a typical decorated sherd was found near the beads, we cannot exclude a date from 297 the end of the 5th millennium. It is during the optimum functioning of the Gavà mines, linked to the

298 peak of the Sepulcros de fosa culture (Vallesian/Solsonian, Spain), that the Catalan variscite 299 importations towards the Midi region reach their peak, during the middle - recent *Chasséen*, between 300 4000 and 3500 BCE.

Several sites from the Roussillon, Aude, and Toulouse region, and up to the Rhone corridor (Saint-301 Paul-Trois-Châteaux, Drôme, France) have yielded beads of various forms, including several 302 « heavy » pieces, olive-shaped, of a kind well-known in Catalonia (tomb 1 of Villeneuve-Tolosane, 303 the "royal" tomb A 185 from Saint-Michel-du-Touch, Toulouse, Haute-Garonne, France) (Méroc and 304 305 Simonnet 1979). Some of the Pompignan pieces show similarities within the same ambiance. Within 306 cave IV from las Claousos at Auriac (Aude, France), used as a collective tomb and for which a <sup>14</sup>C 307 date yielded a recent Chasséen attribution, six cylindrical or barrel-shaped variscite beads could come 308 from Encinasola rather than Gavà (Guilaine et al. 2019). Could it be that this reflects a competition between the two deposits (Can Tintorer and Pico Centeno) at a time when Gavà's production was 309 310 slowing down?

311

#### 312 6. Continuities in Final Neolithic

313

We are less informed on the origin of the small meridional variscite beads, which remain abundant in 314 the *Midi de la France* during the entire final Neolithic, between 3500 and 2200 BCE. Their typology 315 isolates them from the previously evoked examples. Large, olive-shaped or thick beads have more or 316 less disappeared except in rare cases (Fées de Lunas cave, Hérault, France). The only remaining beads 317 are small discoid models, with irregular outlines, not always well calibrated. The ensemble excavated 318 from the Cabrials collective tombs, near Béziers (Hérault, France), with a flattened shape, is dated to 319 320 the final Neolithic 1 (Tchéremissinoff 2012). Most of the meridional beads issued from collective 321 tombs (dolmens, natural caves, hypogees) with a long-term use, sometimes over a millennium. They 322 can be numerous: minimum 130 in the Clot-de-l'Oste dolmen at Bouisse (Aude, France; Guilaine et 323 al. 2015). Two beads from the René-Carrié cave at Termes (Aude, France) are composed of materials attributing their origin to Gavà. The exploitation of the Catalan deposits could therefore have been 324 pursued until after 3500 BC, in a less intense manner. 325

326 The end of the use of variscite stone could be placed towards the end of the Chalcolithic with the 327 diffusion of the Bell Beaker phenomenon. An ensemble of 23 beads accompanied the ancient Bell Beaker deposit found within the ditch surrounding the Mourral site in Trèbes (Aude, France; Vaquer 328 329 1998). This is a regional association, since the callais is not a general marker of the European Bell Beaker. This example shows that the south of France tradition of using this material could still exist 330 331 whilst the maritime Bell Beaker phenomenon pursued its supraregional expansion. 332

#### 333 Conclusion

334

The bead ensemble from the Salpêtre cave of Pompignan originates from the Gavà mines, near Barcelona, some 370 km as the crow flies (this distance being a general indication, since the transfer

- implied either crossing the eastern Pyrenees, or their circumvolution by navigating the Gulf of Lion).
- Beyond the burial of this ensemble, which is reminiscent of a cache, perhaps during a ritual act, this discovery confirms the existing relationship between two Neolithic cultures separated by the physical
- barrier of the Pyrenees. To the south, the *Sepulcros de fosa*, which exploited the mines, fabricated and
- diffused the ornaments; and to the North, the meridional *Chasséen* who, from the Toulouse region to
- 342 the Alps, received these jewels which were appreciated for their "exotic" provenance, relative as it
- 343 was. While slighty offset within the Cevenol mountains, the Pompignan ensemble is located relatively
- 344 close to a geographic axis the Bas-Languedoc. Through this axis, Rhone corridor material circulated
- 345 (blond flint from the Vaucluse, alpine eclogite), which travelled opposite ways to the variscite and
- crossed the Pyrenees to furnish cores, flint blades, and polished axes for populations of Catalonia. The
- 347 Pompignan deposit is therefore at the heart of a reciprocity system demonstrating the strong
- 348 complementarity in exchanges between Neolithic cultures of the first half of the 4th millennium BCE.
- 349

351

#### 350 **Références bibliographiques**

- Arnal, J., 1963. Les Dolmens du Département de L'Hérault, Presses Universitaires de France. ed,
  Préhistoire. Paris.
- Audibert, J., 1962. La civilisation chalcolithique du Languedoc oriental, Bordighera. ed, monographies
   préhistoriques et archéologiques, 4. Institut International d'Études Ligures, Montpellier.
- Barbaza, M., 1981. Recherches sur l'Epipaléolithique en Languedoc et Catalogne (Thèse de 3ème
  cycle). EHESS, Toulouse.
- Barge, H., 1982. Les Parures du Néolithique ancien au début de l'âge des métaux en Languedoc,
  CNRS. ed. Laboratoire d'anthropologie et de Préhistoire des pays de la Méditerranée occidentale,
  Marseille.
- Bar-Yosef Mayer, D., Porat, N., 2008. Green stone beads at the dawn of agriculture. Proceedings of
  the National Academy of Sciences of the United States of America. 105, 8548–51.
  https://doi.org/10.1073/pnas.0709931105
- Beck, H.C., 2006. Classification and Nomenclature of Beads and Pendants. BEADS : Journal of the
  Society of Bead Researchers, 1e ed., 1926, 18, pp. 1–76.
- Bosch Argilagos, J., 2019. Gavà (Barcelone), mine et atelier au Néolithique, in: Querré, G., Cassen,
  S., Vigier, E. (Eds.), La parure en callaïs du Néolithique européen. Presented at the Colloque
  International, Callaïs, Carnac 2015, Archaeopress Archaeology, Oxford, pp. 43–58.
- 369 Cartailhac, É., 1889. La France préhistorique : d'après les sépultures et les monuments, Félix Alcan.
  370 ed. Paris.
- Cassen, S., Boujot, C., Charvet, A., Grimaud, V., Le Maux, N., Le Pennec, C., Querré, G., Vigier, E.,
  Obeltz, C., Prodéo, F., Villes, A., 2019. La parure en callaïs (variscite et turquoise), au Néolithique,

dans la moitié nord de la France. Corpus et contextes, in: Querré, G., Cassen, S., Vigier, E. (Eds.), La
parure en callaïs du Néolithique européen. Presented at the Colloque International, Callaïs, Carnac
2015, Archaeopress Archaeology, Oxford, pp. 203–279.

Chantret, F., Guilaine, J., Guillemaut, A., 1970. Analyses de quelques perles méridionales en
« callais ». Bulletin de la Société préhistorique française. 67, pp. 216–219.
https://doi.org/10.3406/bspf.1970.4258

- Damour, A.-A., 1864. Sur la Callaïs : nouveau phosphate d'alumine hydraté recueilli dans un tombeau
  celtique du Morbihan. Comptes rendus l'Académie des Sciences, pp. 936–940.
- 381 Domínguez-Bella, S. (2004). Variscite, a prestige mineral in the Neolithic-Aeneolithic Europe. Raw
   382 material sources and possible distribution routes. Slovak Geological Magazine, 10, 1-2. pp. 147-152.
- 383 Domínguez-Bella, R., Ramos-Muñoz, J., 2019. Pico Centeno prehiostotic variscite muines
  384 (Encinasola, Huelva, SW Spain), in: Querré, G., Cassen, S., Vigier, E. (Eds.), La parure en callaïs du
  385 Néolithique européen. Presented at the Colloque International, Callaïs, Carnac 2015, Archaeopress
  386 Archaeology, Oxford, pp. 71–83.
- 387 Fàbregas Valcarce, R., Rodríguez-Rellán, C., 2019. Palazuelo de las Cuevas and the exploitation of
- variscite in the North- West of Iberia, in: Querré, G., Cassen, S., Vigier, E. (Eds.), La parure en callaïs
  du Néolithique européen. Presented at the Colloque International, Callaïs, Carnac 2015, Archaeopress
- 390 Archaeology, Oxford, pp. 59–70.
- 391 Fernandez, A., Moro, C., Dabard, M.-P., 2019. Les minéralisations de variscite du Sud-Ouest de
- l'Europe : caractéristiques minéralogiques, géochimiques et genèse, in: Querré, G., Cassen, S., Vigier,
  E. (Eds.), La parure en callaïs du Néolithique européen. Presented at the Colloque International,
  Callaïs, Carnac 2015, Archaeopress Archaeology, Oxford, pp. 9–23.
- Jeanjean, A., 1871. L'Homme et les animaux dans les cavernes des Basses-Cévennes. Mémoires de
  l'Académie du Gard, Nîmes, pp. 139–228.
- Gascó, J., Lachenal, T., Piningre, J.-F., 2018. Un monde de dépôts métalliques, in: Guilaine, J.,
  Garcia, D. (Eds.), La Protohistoire de La France. Hermann, Paris, pp. 339–353.
- Guilaine, J., 2011. Parures de pierre et objets symboliques, in: Shillourokambos. Un Établissement
  Néolithique Pré-Céramique à Chypre. Les Fouilles Du Secteur 1. Errance/Ecole Française d'Athènes,
  Paris, pp. 789–806.
- Guilaine, J., Muñoz, A.-M., 1964. La civilisation catalane des "Sepulcros de fosa" et les sépultures
  néolithiques du Sud de la France. Revue d'Études Ligures XXX, pp.5–30.
- Guilaine, J., Rigaud, S., 2021. Parures de pierre et objets symboliques, in: Shillourokambos : un
  Établissement Néolithique Pré-Céramique à Chypre. Les fouilles du secteur 3. CNRS, Paris, pp. 435–
  406 471.
- Guilaine, J., Vaquer, J., Gratuze, B., Querré, G., 2015. Les perles en variscite de la grotte de Las
  Claousos IV et de la grotte René-Carrié, in: Guilaine, J., Vaquer, J., Zammit, J. (Eds.), Grottes
  sépulcrales préhistoriques des Hautes-Corbières. Toulouse, pp. 89–104.
- Herbaut, F., Querré, G., 2004. La parure néolithique en variscite dans le sud de l'Armorique. Bulletin
  de la Société préhistorique française, 101, pp.497–520. https://doi.org/10.3406/bspf.2004.13029

- 412 Méroc, L., 1962. Le village et la sépulture chasséens de Villeneuve-Tolosane (Haute-Garonne,
  413 France). Zephyrus, XIII, pp. 94–96.
- 414 Méroc, L., Simonnet, G., 1979. Les sépultures chasséennes de Saint-Michel-du-Touch à Toulouse
- (Haute-Garonne). Bulletin de la Société préhistorique française, Études et Travaux 76, 10-12, pp. 379–
  407. https://doi.org/10.3406/bspf.1979.5163
- Mohen, J.-P., 1978. Tumulus des Pyrénées françaises, in: Actes Du 2e Col.loqui Internacional
  d'Arqueología de Puigcerdà. Presented at the Els pobles pre-romans del Pirineu, Institut d'Estudis
  ceretans, Puigcerdà, pp. 97–108.
- Molist Montana, M., Oliva Poveda, M., 2019. La parure en variscite au Néolithique dans la partie est
  de l'Espagne, in: Querré, G., Cassen, S., Vigier, E. (Eds.), La parure en callaïs du Néolithique
  européen. Presented at the Colloque International, Callaïs, Carnac 2015, Archaeopress Archaeology,
  Oxford, pp. 351–364.
- 424 Moro Benito, MC; Montero Gómez, JM; Gil Agero, M.; Cembranos Pérez, ML; Pérez del Villar
  425 Guillén, L.; Fernández Fernández, A.; Hernández, E. (1992) Aluminofosfatitas silúricas de la
- 426 península Ibérica: síntesis preliminar. Boletín de la Sociedad Española de Mineralogía, Vol. 15, nº. 1,
- **427** pp. 227-232. ISSN 0210-6558.
- 428 Moro Benito, MC; Pérez del Villar Guillén, L.; Fernández Fernández, A.; Cembranos Pérez, ML;
- Gil Cid, MD (1995) Las Mineralizaciones estratiformes de variscita (Aluminofosfatitas) silúricas de
  los Sinformes de Alcañices (Zamora) y Terena (Huelva): (España). Boletín Geológico y Minero, Vol.
  106, nº 3, pp. 33-49. ISSN 0366-0176.
- 432 Muñoz, A.-M., 1965. La cultura neolítica catalana de los «Sepulcros de Fosa». Universidad de
  433 Barcelona, Instituto de Arqueología y Prehistoria, Barcelona.
- Odriozola, C.P., Linares-Catela, J.A., Hurtado-Pérez, V., 2010. Variscite source and source analysis:
  testing assumptions at Pico Centeno (Encinasola, Spain). Journal of Archaeological Science. 37, pp.
  3146–3157. https://doi.org/10.1016/j.jas.2010.07.016
- 437 Peltenburg, E., 1998. Excavations at Kissonerga-Mosphilia 1979-1992, Studies In Mediterranean
  438 Archaeology. LXX:2, Paul Åströms Förlag, Jonsered.
- 439 Piette, É., 1881. Note sur les tumulus de Bartrès et d'Ossun, Durand. ed, Matériaux pour l'Histoire de
  440 l'Homme. Durand, Toulouse.
- 441 Querré, G., Calligaro, T., Cassen, S., Dabard, M.-P., Domínguez-Bella, S., 2019. Provenance des
  442 parures en variscite du Néolithique européen : élaboration d'un modèle chimiométrique, in: Querré,
  443 G., Cassen, S., Vigier, E. (Eds.), La parure en callaïs du Néolithique européen. Presented at the
- 444 Colloque International, Callaïs, Carnac 2015, Archaeopress Archaeology, Oxford, pp. 105–128.
- 445 Querré, G., Calligaro, T., Domínguez-Bella, S., Cassen, S., 2014. PIXE analyses over a long period:
- 446 The case of Neolithic variscite jewels from Western Europe (5th-3th millennium BC). Nuclear
- 447 Instruments and Methods in Physics Research Section B : Beam Interactions with Materials and
- 448 Atoms, The 13th International Conference on Particle Induced X-ray Emission (PIXE 2013) 318, part
- 449 A, pp. 149–156. https://doi.org/10.1016/j.nimb.2013.07.033

- 450 Querré, G., Cassen, S., Domínguez-Bella, S., 2012. La variscite ibérique. Exploitation, diffusion au
- 451 cours du Néolithique, in : Marchand, G., Querré, G. (Eds.), Roches et sociétés de la Préhistoire. Entre
- 452 massifs cristallins et bassins sédimentaires. Presses universitaires de Rennes.

453 Querré, G., Herbault, F., Calligaro, T., 2008. Transport of Neolithic variscites demonstrated by PIXE
454 analysis. X- Ray Spectrom. 37, pp. 116–120. https://doi.org/10.1002/xrs.1031

- 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, pp.
  209–257. https://doi.org/10.3406/galip.1992.2303
- Tchérémissinoff, Y.T., 2012. La sépulture collective mégalithique de Cabrials (Béziers, Hérault). Une
  petite allée sépulcrale enterrée du début du Néolithique final. Préhistoires Méditerranéennes 147.
- Vaquer, J., 2019. Les éléments de parure en callaïs dans le midi de la france au Néolithique et au
  Chalcolithique, in: Querré, G., Cassen, S., Vigier, E. (Eds.), La parure en callaïs du Néolithique
  européen. Presented at the Colloque International, Callaïs, Carnac 2015, Archaeopress Archaeology,
  Oxford, pp. 333–349.
- Vaquer, J., 1998. Le Mourral, Trèbes (Aude) A fortified languedocian late neolithic site reoccupied by
  bell beakers, in: Benz, M., Van Willigen, S. (Eds.), Some New Approaches to The Bell Beaker
  Phenomenon, Lost Paradise...? Presented at the Proceedings of the 2" Meeting of the "Association
  Archéologie et Gobelets," Oxford Archéologie, Feldberg (Germany), pp. 15–21.
- Viel, L., Bonnardin, S., Bailly, M., Margarit, X., 2019. Perles supposées en variscite du sud-est de la
  France (Arles-Fontvielle, IV<sup>e</sup> mill. av. J.-C.), in: Querré, G., Cassen, S., Vigier, E. (Eds.), La parure en
  callaïs du Néolithique européen. Presented at the Colloque International, Callaïs, Carnac 2015,
  Archaeopress Archaeology, Oxford, pp. 365–378.
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- 494 Illustrations
- 495

- Fig. 1 Geographic location of the Salpêtre cave (Pompignan, Gard) Fig. 2 – Topographic location of the cavity (drawing by J. Coularou) Fig. 3 – Cave entrance (photograph by A. Aigoin) Fig. 4 – Plan and stratigraphy of the cave. Localisation of the bead ensemble (drawing by N. Boinet) Fig. 5 – Localisation of the discovery near the base of the cave wall (photograph by A. Aigoin) Fig. 6 – Close up: localisation of the beads near a decorated sherd and a pecten shell (drawing by J. Coularou) Fig. 7 – Sherd from the ventral part of a decorated Chasséen globulous recipient discovered near the variscite beads (drawing by J. Coularou) Fig. 8 – Saint-Jacques shell (Pecten jacobeus) discovered near the beads (drawing by J. Coularou) Fig. 9 – Necklace restitution picture of the 17 variscite beads deposit (photograph by H. Vergély) Fig. 10 – Numbering of the 17 beads analysed according to the PIXE method (photograph by H. Vergély). Fig. 11 – Details of the 17 variscite elements composing the ensemble (photograph by H. Vergély). Fig. 12 – Variscite beads of the Salpêtre cave in Pompignan being analysed through the PIXE method, in front of the beam extract from the AGLAE particle accelerator (C2RMF, Louvre Palace, Paris) (realisation by G. Querré). Fig. 13 – Analyses of the variscite beads of the Salpêtre cave in Pompignan (number of objets analysed: 17; number of analysis : 17). In the diagrams : V-Cr-Fe; U-Zr; (Cr + V) As; Ca-Si in comparison to the referential for the 5 principal variscite sources in western Europe: Gavà (Barcelona, Spain ; GA), Encinasola (Huelva, Spain ; EN), El Bostal (Zamora, Spain ; EB), Palazuelo (Zamora, Spain; PL), Pannecé (Loire-Atlantique, France; PA) (realisation by G. Querré). Table 1 - Measures and weights of the 17 variscite éléments of the Salpêtre cave deposits (Pompignan, Gard) (realisation H. Vergély). Table 2 - Color of the 17 variscite éléments of the Salpêtre cave deposits (Pompignan, Gard) (realisation H. Vergély). Table 3 - Chemical elementary composition obtained through the PIXE method and of each of the Salpêtre cave (Pompignan, Gard). Analyses are expressed as ppm (part per million). In the table, for each element, we also find the average concentrations, their standard deviation, minimum and maximum concentrations as well as the detection limits (LDD) during the bead's analyses in July 2016 (realisation by G. Querré). Table 4 – Elemental analysis obtained by PIXE for each of the variscite deposits, in order to develop the chemometric model. Concentrations expressed in parts per million (ppm) and in elemental form. V\*, Cr\* and Fe\* are the relative percentages of vanadium, chromium and iron contents normalized to the sum V+Cr+Fe.
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Planimetric view











Necklace restitution Low-resolution



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Beads	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17
maximum diameter	8.56	11.55	11.74	12.5	13.51	15.41	16.43	20.48	25.15	15.02	<b>1</b> 4.17	11.48	9.35	13.7	9.14	1.47	7.21
minimum diameter	6.43	10.33	10.4	12.4	7.88	13.17	12.53	11.79	15.88	12.83	11.77	9.7	8.48	12.8	9.12	6.57	6.88
maximum perforation	2.22	2.75	2.72	3.14	3.25	4.01	4.18	3.82	5.01	3.57	3.82	4.35	3.05	2.56	2.89	2.56	2.88
minimum perforation	2.12	2.61	3.17	2.92	2.8	2.95	3.44	3.59	4.34	3.02	3.18	2.92	2.55	2.4	2.39	2.43	2.61
maximum thickness	8.45	12.22	5.95	13.92	13.04	16.64	21.41	28.76	40.1	20.85	22.88	20.94	17.44	8.89	10.43	8.42	6.1
minimum thickness	7.69	11.3	5.12	13.05	12.29	12.95	<b>2</b> 0.3	27.31	38.1	18.89	18.42	17.7	10.8	7.42	9.45	7.95	5.8
méplat-1 L	6.84	8.27	9.96	9.05	10.22	11.4	10.78	11.45	17.81	11.72	11.44	9.7	7.96	6.95	8.01	5.8	/
méplat-1 l	5.5	7.69	9.22	8.44	7.42	10.92	10.1	9.61	12.75	10.38	8.66	9.64	7.15	5.95	6.1	5.12	/
méplat-2 L	6.57	8.76	10.23	8.44	10.57	11.89	11.39	13.5	16.47	12.97	11.78	11.48	7.54	7.01	7.56	5.56	/
méplat-2 l	5.1	7.93	9.75	7.45	7.72	10.14	9.98	8.85	12.75	11.61	11.26	9.7	/	5.9	6.6	4.78	/
weight in gr.	0.66	2.03	1.16	2.9	2.23	4.99	6.09	9.74	23.81	5.89	5.53	3.36	8.7	1.36	1.02	0.54	0.42
max diameter/méplat 1	1.25	1.40	1.18	1.38	1.32	1.35	1.52	1.79	1.41	1.28	1.24	1.18	1.17	1.97	1.14	1.29	/
max diameter/méplat 2	1.30	1.32	1.15	1.48	1.28	1.30	1.44	1.52	1.53	1.16	1.20	1.00	1.24	1.95	1.21	1.34	/
diam/perf	3.86	4.20	4.32	3.98	4.16	3.84	3.93	5.36	5.02	4.21	3.71	2.64	3.07	5.35	3.16	2.92	2.50
diam/thickness	1.01	0.95	1.97	0.90	1.04	0.93	0.77	0.71	0.63	0.72	0.62	0.55	0.54	1.54	0.88	0.89	1.18

n° beads	color
1	white alteration, green with brownish to black fissures
2	pale green, white alteration
3	white alteration, pale green
4	white alteration, green with black fissures
5	pale green with fine black veinlets
6	white alteration, palegreen with black veinlets
7	white alteration, palegreen with numerous black veinlets
8	white alteration, palegreen with numerous black veinlets
9	pale green, brownish yellow
10	range of pale green with black veinlets, white alteration
11	pale green with numerous black veinlets, white alteration
12	white alteration, green with black fissures
13	blue-green to pale green with broad black vein
14	range of pale green, brownish yellow
15	blue-green to pale green with black veinlets
16	pale green with fine black veinlets
17	pale green with fine black veinlets

Name	Na	Mg	Al	Si	Р	к	Ca	Ti	v	Cr	Mn	Fe	Со	Ni	Cu	Zn	Ga	As	Rb	Sr	Υ	Zr	Nb U	Cr/V	٧*	Cr*	Fe*	Cr+V
bead n°1	4907	6833	91650	239585	49442	37622	49058	1237	1002	3552	44	23234	0	49	231	240	50	294	62	508	17	40	5 14	3.54	3.6	12.8	83.6	4554
bead n°2	5529	1784	139117	144166	146878	15942	14868	1075	733	2421	21	18927	0	6	57	53	262	1599	14	133	2	12	0 11	3.3	3.3	11	85.7	3154
bead n°3	6638	3494	67660	301054	11081	36238	36612	1218	928	2208	40	50860	0	32	270	216	124	931	65	372	4	23	0 7	2.38	1.7	4.1	94.2	3136
bead n°4	3797	2029	102817	239669	83357	23965	19200	214	660	366	15	20281	0	2	86	45	34	796	21	163	3	40	0 7	0.55	3.1	1.7	95.2	1026
bead n°5	2540	1280	103287	248037	81168	19603	20377	514	2480	692	19	14683	0	1	26	44	67	348	18	183	2	47	2 4	0.28	13.9	3.9	82.2	3172
bead n°6	4427	2329	155420	44743	210720	7020	12562	653	1363	1763	25	51879	0	6	320	73	202	1228	1	36	2	59	5 13	1.29	2.5	3.2	94.3	3126
bead n°7	1670	2008	155758	114805	172458	9641	12142	598	1211	480	7	14549	0	3	9	25	30	1614	7	66	5	4	0 8	0.4	7.5	3	89.6	1691
bead n°8	3046	1492	141149	146038	115070	28244	20022	481	1207	744	2	11492	1	6	92	33	43	205	8	96	5	2	1 4	0.62	9	5.5	85.5	1951
bead n°9	2311	1486	116936	190784	122418	19780	10862	1506	1582	1977	15	23781	0	0	157	16	291	800	9	102	2	5	1 0	1.25	5.8	7.2	87	3559
bead n°10	4424	1338	126016	186931	130648	17872	9518	115	1660	872	15	7365	5	2	24	23	23	1280	12	99	15	0	06	0.53	16.8	8.8	74.4	2532
bead n°11	3753	2007	128787	179294	131446	15068	12858	601	1101	2925	19	10470	3	4	30	32	221	674	13	82	4	10	0 12	2.66	7.6	20.2	72.2	4026
bead n°12	4415	2374	106373	201140	101804	20165	15965	445	800	1178	26	47123	0	4	560	84	64	773	16	111	0	22	0 17	1.47	1.6	2.4	96	1978
bead n°13	2328	1455	142144	138116	162112	14235	3958	516	2901	1296	14	18290	0	5	142	23	52	716	3	12	4	2	2 12	0.45	12.9	5.8	81.3	4197
bead n°14	3224	3024	120878	184156	108418	19563	34597	464	683	1405	17	10759	9	13	61	70	89	464	12	12089	7	3	0 18	2.06	5.3	10.9	83.7	2088
bead n°15	7171	5318	113660	193483	117774	25200	16495	959	1750	2519	42	7159	4	14	66	74	91	703	16	39	8	21	0 13	1.44	15.3	22	62.6	4269
bead n°16	3863	1528	152024	119025	176346	11682	5994	379	1566	739	11	11737	4	1	13	21	50	499	5	116	2	1	0 0	0.47	11.2	5.3	83.6	2305
bead n°17	3562	1903	147390	115079	175144	10273	7639	861	1601	1380	20	23116	0	6	101	25	62	710	6	140	4	78	0 10	0.86	6.1	5.3	88.6	2981
Average	3977	2452	124180	175653	123311	19536	17807	696	1366	1560	21	21512	2	9	132	65	103	802	17	844	5	22	1 9	1.39	7.5	7.8	84.7	2926
standard deviation	1494	1510	24726	62025	49965	8660	11925	386	618	921	12	14585	3	13	144	65	86	418	18	2900	5	23	2 5	1.05	4.9	5.9	8.8	1003
min	1670	1280	67660	44743	11081	7020	3958	115	660	366	2	7159	0	0	9	16	23	205	1	12	0	0	0 0	0.28	1.6	1.7	62.6	1026
max	7171	6833	155758	301054	210720	37622	49058	1506	2901	3552	44	51879	9	49	560	240	291	1614	65	12089	17	78	5 18	3.54	16.8	22	96	4554
LDD	184	127	86	108	88	46	81	23	17	13	12	4	92	1	1	1	1	1	12	4	3	15	4 19					

depo sits	can Tintorer, Barcelona (Catalonia, Spain)						nasola, H	uelva (And	alusia, Spair	ı)	El Bos	stal, Zamor	a (Castilla	y Leon, Spa	in)	Palaz	uelo, Zamo	ora (Castill	a y Leon, Sp	ain)	Pannecé, Loire-Atlantique (Pays de Loire, France)						
	Min	Max	Mean	standard deviation	σ%	Min	Max	Mean	standard deviation	σ%	Min	Max	Mean	standard deviation	σ%	Min	Max	Mean	standard deviation	σ%	Min	Max	Mean	standard deviation	σ%		
Na	315	12680	2835	2591	91	0	4302	867	783	90	0	2336	721	597	83	0	5285	1266	1072	85	65	9503	2771	2207	80		
Mg	851	13434	4197	3156	75	0	1811	329	477	145	0	2020	880	597	68	0	2891	604	677	112	0	3485	826	899	109		
AI	17713	207192	140062	41732	30	103031	221506	203260	17109	8	179919	215783	201001	7058	4	37377	217013	185243	43748	24	191379	218757	202580	7651	4		
Si	14567	300262	112706	74521	66	0	37564	4900	7895	161	0	13361	5885	4421	75	0	211451	163448	37102	227	0	10728	2342	3297	141		
Р	29621	239250	157836	63047	40	206093	266367	245676	10696	4	242177	261510	254794	6055	2	138422	258780	237810	22335	9	245949	262549	257847	5090	2		
к	167	7637	2980	2035	68	0	5388	671	842	125	78	1800	672	432	64	0	81919	1769	9052	512	69	2524	1170	689	59		
Ca	4187	345476	61374	80461	131	0	3666	439	515	117	0	6872	641	1343	210	38	19787	846	2183	258	286	4756	1614	1149	71		
Ti	0	15127	2699	3571	132	34	3000	573	600	105	0	812	191	175	91	159	2908	1211	628	52	30	334	134	74	55		
v	1	888	365	243	67	581	5287	2507	921	37	1729	3594	2763	464	17	35	2773	1106	609	55	2594	4333	3249	565	17		
Cr	1	720	443	189	43	125	3860	1691	894	53	550	1281	968	179	18	1207	8980	3734	1527	41	1163	1791	1449	188	13		
Mn	0	184	35	49	142	0	44	6	8	132	0	114	16	24	154	0	174	20	26	129	0	19	6	6	100		
Fe	7306	24426	13079	4600	35	2274	201149	21386	30558	143	3964	26082	8190	5074	62	1130	298779	35907	71654	200	1185	2090	1586	249	16		
Со	0	81	26	22	84	0	55	9	12	130	0	80	23	18	82	0	227	19	41	208	0	18	7	5	78		
Ni	0	586	71	122	172	0	189	11	32	279	0	14	4	4	99	0	70	4	9	220	0	11	3	3	92		
Cu	11	393	98	108	110	0	3197	416	819	197	64	3510	560	1027	183	8	2768	427	871	204	2	112	27	25	92		
Zn	11	163	45	43	96	1	98	21	23	111	217	593	399	88	22	1	138	26	27	107	8	117	22	23	106		
Ga	12	259	45	62	138	0	362	71	79	111	9	73	30	20	65	4	420	137	91	66	92	223	163	23	14		
As	127	432	302	90	30	45	4783	610	802	131	39	210	62	35	57	26	702	138	126	91	3	16	9	3	31		
Rb	0	51	7	11	145	0	201	8	24	299	0	12	4	4	99	0	712	14	80	576	0	4	1	1	126		
Sr	96	34525	4847	10011	207	0	322	20	49	245	0	21	4	6	145	0	2590	55	290	531	0	74	19	22	119		
Y	0	151	19	35	179	0	128	13	25	200	6	55	23	10	43	0	133	7	18	270	0	7	1	2	256		
Zr	0	127	36	26	73	0	256	50	52	104	0	16	6	4	78	0	240	48	46	95	0	10	1	2	235		
Nb	0	42	5	9	201	0	14	2	3	179	0	24	3	6	179	0	35	2	6	252	0	6	1	2	174		
U	0	205	35	53	153	0	152	17	33	193	37	1468	306	267	87	0	262	13	35	268	0	12	4	3	82		
٧*	0	7	3	2	70	1	41	18	11	62	8	37	26	8	31	0	21	8	6	75	44	55	52	3	5		
Cr*	0	6	3	2	50	0	38	13	11	88	3	12	9	3	29	1	59	25	15	62	22	25	23	1	3		
Fe*	88	100	93	4	4	31	98	70	20	29	51	89	65	10	16	31	99	66	20	30	22	31	25	2	10		
Cr/V	0.01	126	6.49	24.48	377	0.06	1.77	0.74	0.41	55.37	0.23	0.41	0.35	0.04	11.52	0.94	56.29	5.08	6.48	127.37	0.4	0.57	0.45	0.04	8.66		
Cr+V	67	1464	809	397	49	1293	9147	4198	4875	34	2315	4875	3731	618	17	1570	10366	4841	1732	36	3848	6116	4698	736	16		