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1 TITLE

2 Lead provenance for medieval decorated tile glazes from Brittany and Anjou (13th-14th c.)

3

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103 **DECLARATION OF INTEREST**

104 Declarations of interest: none.

105

106 **ABSTRACT**

107 Medieval pavements composed of lead-glazed tiles decorated with a variety of techniques

108 continue to inspire questions about the organisation of glaze manufacture, and the supply and

109 origin of lead materials. The tiles analysed in this study are from Suscinio I (a 13th-century

110 pavement) and Suscinio II (a 14th-century pavement), at the Château of Suscinio in Brittany, and

111 also from the 14th-century pavement at the fortified manor house in Brain-sur-Allonnes, Anjou.

112 Lead isotope analysis (LIA) was used to examine samples from 44 lead-glazed tiles, 29 of which

113 are transparent, while 15 are tin-opacified (an exogenous technique in these regions during this

114 period). Five out of the six LIA groups thus identified favour site-specific supply networks, while

115 results for the remaining group, LIA5, indicate a multi-site supply network. After combining LIA

116 results with archaeological and historical data, the most likely provenance for the lead materials

117 in LIA5 is Derbyshire, in the British Isles. Both the importation of ready-to-use glazing mixtures

118 and the use of lead from neighbouring mines can now therefore be discounted as plausible

119 hypotheses for the production of the tin-opacified lead-glaze tiles analysed in this study.

120

121 HIGHLIGHTS

- 122 - Two lead supply strategies: site-specific and multi-site
- 123 - Imported lead supply strategy for both transparent and opaque glazes
- 124 - Derbyshire is the most plausible lead source for the multi-site supply network
- 125 - Tin-opacified glazing mixtures were locally produced

126

127

128 KEYWORDS

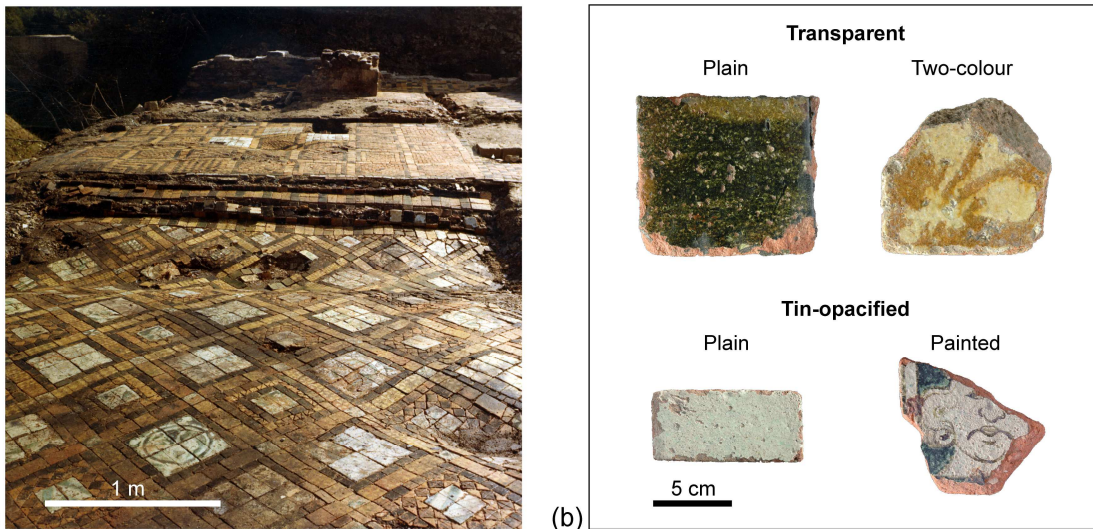
129 Middle Ages; tin-glazed earthenware; high lead glaze; lead isotope analysis (LIA); decorated tile
130 technology; Suscinio (Brittany, France); Brain-sur-Allonnes (Anjou, France)

131

132 1. INTRODUCTION

133 Decorated tile pavements in religious and secular Gothic buildings throughout north-western
134 Europe reflected the power of ecclesiastical and lay princes (Gauthier, 1984; Crépin-Leblond and
135 Rosen, 2000; Rosen and Crépin-Leblond, 2000). These prestigious floor coverings were almost
136 invariably composed of transparent lead-glazed tiles, decorated using various techniques, all of
137 which seem to have been developed locally. In contrast, tin-opacified lead-glaze tiles, also
138 described as tin-glazed tiles (Norton, 1984b), and termed *faïence* or *carreaux stannifères* in
139 French (Norton, 2000), are extremely rare (Bon, 2000; Chapelot, 2000; Démians d'Archimbaud
140 et al., 2000; Jugie, 2000). This technique, originating in the Ummayyad and Abassid Caliphates of
141 Egypt and Western Asia in the 8th to 9th century, spread gradually to the western Mediterranean
142 in the following centuries (Norton, 1984a, 1984b, 1984c; Matin et al., 2018; Pradell and Molera,
143 2020).

144 A pavement with both types of glaze suggests the simultaneous presence of indigenous and
145 exogenous technical knowledge and cultural traditions. One such pavement in present-day
146 France, Suscinio II (Figure 1), was discovered still in place in the extramural chapel at the
147 Château of Suscinio, a residence of the dukes of Brittany from the 13th to the 15th century
148 (André, 1986a, 1986b, 2003). By combining historical and archaeological data with ceramic
149 provenance and technological studies (Métreau et al., 2017), it has been established that: (1) the
150 Suscinio II tiles were probably not imported as finished products, but rather produced nearby
151 with locally available clayey raw materials; (2) tin-opacified lead glazes do not necessarily
152 indicate local experimentation, but could suggest technical knowledge from further afield.
153



154 (a) The medieval pavement of Suscinio II, excavated in 1975 (© G. Chapuy, in Métreau et al., 2015). (b)
155 The four types of decoration on the lead-glazed tiles (© L. Métreau, adapted from Métreau et al., 2017).
156
157

158 In medieval times, lead was used for many purposes (Benoit, 1985; Téreygeol, 2009). Galena
159 (PbS)—also known by potters as *alquifoux* (Picon et al., 1995)—and litharge (PbO) were two of
160 the lead compounds frequently used to lower the silica melting point in glazing mixtures (Abel,
161 1975; Tite et al., 1998). The lead present in archaeomaterials can be characterised by lead isotope

162 analysis (LIA), a technique that has been used since the 1960s (Grögler et al., 1966; Brill and
163 Wampler, 1967; Gale and Stos-Gale, 2000). Studies using LIA on glazes have generally focused
164 on technological choices and specific distribution networks, as well as on lead sources (Mason et
165 al., 1992; Wolf et al., 2003; Stos-Gale, 2004; Iñáñez et al., 2010; Walton and Tite, 2010; Shen et
166 al., 2019; Fornacelli et al., 2020; Medeghini et al., 2020; Chang et al., 2021).

167 In this study, 13 transparent and 13 tin-opacified lead-glaze tiles from Suscinio II, a 14th-century
168 pavement, are investigated through LIA. The results should provide key information about lead
169 sources and supply strategies, in particular the question of local, regional, or international supply
170 networks, and the hypothesis of imported ready-to-use glazing mixtures to produce the tin-
171 opacified glazes, possibly from regions around the western Mediterranean, in particular the
172 Iberian Peninsula. Although Suscinio II is the focal point of this study, further temporal context is
173 provided by Suscinio I, a 13th-century pavement. Further spatial context is provided by the 14th-
174 century pavement of Brain-sur-Allonnes (Anjou). Exploring the origin, initial production, and use
175 of tin-glazing in Brittany and Anjou will contribute to a better understanding of the long-standing
176 tradition of the French tin-glazed earthenware technique and its place within the history of
177 decorated tiles.

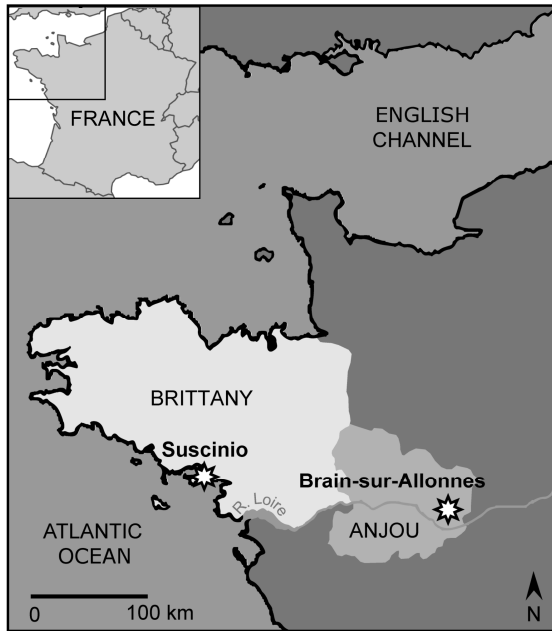
178

179 2. MATERIALS AND METHODS

180 2.1 Decorated tile pavements

181 The tiles are from three different pavements at two archaeological sites (Figure 2): Suscinio I and
182 Suscinio II are pavements from the Château of Suscinio in Brittany; the third pavement is from
183 the fortified manor house at Brain-sur-Allonnes in Anjou. To complement the existing historical
184 and archaeological data for these three pavements, the results of three previous archaeometric
185 studies are compiled in Appendix A.

186



187

188 Figure 2. Map of the study area. Stars indicate pavement sites (L. Métreau, adapted from André and Le Penneç,
189 2003).

190

191 2.1.1 *Suscinio I*

192 The remaining traces of the Suscinio I pavement include approximately 1,500 tiles and tile
193 fragments, with many different geometric forms (André et al., 2016). Stylistic evidence indicates
194 that the pavement may have been laid between 1260 and 1270. Two decorative techniques are
195 identified: plain transparent lead-glazed tiles (ca. 67%), and two-colour transparent lead-glazed
196 tiles (ca. 33%).

197 A lead compound (39–62 wt% PbO) was one of the main components of the glazing mixture,
198 together with silica and clay (Métreau et al., 2012b). Detailed examination of the clay body has
199 identified characteristics similar to those of medieval Breton ceramics, while Brittany and Anjou
200 are the most probable sources for the different types of non-calcareous clayey raw materials
201 (Appendix A).

202 The Suscinio I pavement may well have been similar in style to pavements found in and around
203 the Anjou region (André et al., 2016). It was probably discarded between 1320 and 1330, and
204 replaced by Suscinio II (André, 2003).

205

206 *2.1.2 Suscinio II*

207 The tiles from Suscinio I would have been more difficult to produce and fit together than the
208 simple, standardised, proportionate geometric shapes used for the Suscinio II pavement,
209 composed of approximately 30,000 tiles. Suscinio II may have been purpose-built or redeployed
210 from another site. Its *terminus post quem* (1330) was identified by coins found in the clay bed
211 under the pavement (André, 2003; André et al., 2016).

212 Most of the lead-glazed tiles are transparent (plain, ca. 62%; two-colour, ca. 36%), but some are
213 tin-opacified (plain, ca. 0.5%; painted, ca. 1.5%). The main components of the glazing mixtures
214 were silica and a lead compound (transparent = 50–68 wt% PbO; tin-opacified = 35–
215 45 wt% PbO). Clay was added to the transparent glazing mixture, whereas tin oxide was used to
216 produce the opaque glaze (Métreau et al., 2017). Some of the transparent lead-glazed tiles show
217 signs of tin-oxide contamination, suggesting that both glazing processes took place in the same
218 workshop. Technical flaws on some of the tin-opacified lead-glaze tiles may reflect a poorly
219 controlled firing process. As with Suscinio I, detailed examination of the clay body has identified
220 characteristics similar to those of medieval Breton ceramics. The area around the Château of
221 Suscinio is the most likely source for the non-calcareous clayey raw materials (Appendix A).
222 The tiles from Suscinio II are stylistically similar to decorated tiles found in northern France
223 (e.g. at the Abbey of St Bertin, St Omer), at a distance of over 600 km (André, 2003).

224

225 *2.1.3 Brain-sur-Allonnes*

226 The lead-glazed tiles (approximately 14,000 tiles and tile fragments) found at the fortified manor
227 house at Brain-sur-Allonnes have simple, standardised, proportionate geometric shapes (Norton
228 and Clair, 1980; Clair and Lecompte, 1986). They can be divided into three types: a majority of
229 plain tiles (ca. 97%), and some two-colour tiles (ca. 2.5%), both with transparent lead glaze, and
230 a small number of painted tiles, with tin-opacified lead glaze (ca. 0.5%).

231 The main components of the glazing mixtures were silica and a lead compound (transparent =
232 51–65 wt% PbO; tin-opacified = 44–47 wt% PbO). As with Suscinio II, clay was added to the
233 transparent glazing mixture, whereas tin oxide was used to produce an opaque glaze (Métreau et
234 al., 2012a). The poor quality of the tin-opacified tiles probably indicates problems with the firing
235 process. The area around Brain-sur-Allonnes is the most likely source for the non-calcareous
236 clayey raw materials (Appendix A).

237 Between 1360 and 1370, the manor house was destroyed by fire, thus providing a *terminus ante*
238 *quem* for the production of the decorated tiles. Archaeomagnetic dating of their clay body
239 indicates that they were produced at the end of the 13th century, but the fire may have affected
240 the signal (Goulpeau, 1988). Technical and stylistic studies, however, suggest that they were
241 probably from a single workshop that produced tiles for Anjou and Touraine during the first half
242 of the 14th century (Norton and Clair, 1980; Clair and Lecompte, 1986).

243

244 **2.2. Sample collection and LIA parameters**

245 The number of tiles available for sampling was limited by the destructive nature of the process.
246 Only tiles with no apparent weathering were selected for analysis. As 15 tin-opacified tiles were
247 available, plain and two-colour transparent lead-glazed tiles were collected from each site in
248 similar proportions (Table 1). These tiles were then transported to the Centre of Physics Applied
249 to Archaeology, which is part of the Institute of Research into Archaeomaterials (IRAMAT-

250 CRP2A, CNRS, Bordeaux Montaigne University), and archaeometric studies were used to
 251 identify zones where the impact of weathering was negligible (Métreau et al. 2012a, 2012b,
 252 2017).
 253

DECOR	PAVEMENT			Total
	Suscínio I	Suscínio II	Brain-sur-Allonnes	
Transparent				
PLAIN	3	6	5	14
TWO-COLOUR	3	7	5	15
Tin-opacified				
PLAIN	-	3	-	3
PAINTED	-	10	2	12
Total	6	26	12	44

254
 255 Table 1. Distribution of high lead glaze samples.

256
 257 Before sampling, the glaze surface was cleaned with acetone and rinsed with distilled water. As it
 258 proved impossible to chip small fragments from the surface with a sharp scalpel, a 2 mm-
 259 diameter conical diamond tip (RADIOSPARES) connected to a multifunctional tool with
 260 adjustable rotation speed (DREMEL, 300 model) was used to collect powder samples (2–29 mg).
 261 The tip was cleaned for five minutes in an ultrasonic bath between samples to avoid
 262 contamination. As the glazes are rich in lead (35–68 wt% PbO), a 1 mg sample proved sufficient
 263 (Wolf et al., 2003). These powder samples were digested in 100 µl concentrated hydrofluoric acid
 264 at the Archéologie, Terre, Histoire et Sociétés Laboratory (ARTEHIS, CNRS, MC, University of
 265 Burgundy). The residue after evaporation at 105 °C was dissolved in 50 µl concentrated nitric
 266 acid to convert fluoride into nitrate, and evaporated again at 105 °C. Lead isotope ratios were
 267 then measured using a Nu InstrumentTM MC-ICP-MS and an Apex (TM) desolvating nebulizer at
 268 the Laboratory of Isotope Geology (University of Bern). For mass spectrometric analysis, the
 269 residue was dissolved in 0.5 M nitric acid, diluted to obtain a signal of ca. 20 pA for ²⁰⁸Pb, and

270 spiked with Tl to correct for instrumental mass fractionation. Mercury interference on mass 204
 271 was corrected by measuring ^{202}Hg . Glaze sample measurements were interspersed with
 272 measurements of international standard reference material, NIST SRM 981. The measured Pb
 273 isotopic composition of the reference material (Table 2) overlaps with values reported by
 274 Rehkämper and Mezger (2000). Measured values are therefore presented without bias correction.
 275

Reference	$^{208}\text{Pb}/^{204}\text{Pb}$	$^{207}\text{Pb}/^{204}\text{Pb}$	$^{206}\text{Pb}/^{204}\text{Pb}$	$^{208}\text{Pb}/^{206}\text{Pb}$	$^{207}\text{Pb}/^{206}\text{Pb}$
Galer and Abouchami, 1998	36.7219 ± 44	15.4963 ± 16	16.9405 ± 15	2.16771 ± 10	0.914750 ± 35
Hirata, 1996	36.6800 ± 210	[15.4856]	16.9311 ± 90	2.16636 ± 82	0.914623 ± 37
Rehkämper and Halliday, 1998	36.6969 ± 128	15.4912 ± 51	16.9364 ± 55	2.16677 ± 14	0.914685 ± 49
Rehkämper and Mezger, 2000	36.7000 ± 23	15.4900 ± 17	16.9366 ± 29	2.16691 ± 29	0.91459 ± 13
Thirlwall, 2000	36.7228 ± 80	15.4956 ± 26	16.9409 ± 22	2.16770 ± 21	0.91469 ± 7
Todt et al., 1996	36.7006 ± 34	15.4891 ± 9	16.9356 ± 7	2.16701 ± 13	0.914585 ± 4
White et al., 2000	36.6825 ± 78	15.4899 ± 39	16.9467 ± 76	2.1646 ± 8	[0.9140]
This study (n=11)	36.7084 ± 28	15.4929 ± 9	16.9395 ± 11	2.16704 ± 8	0.91461 ± 4

276
 277 Table 2. Values of NIST SRM 981 from the literature, compared with values obtained in this study. Analytical error
 278 (2 standard errors of the mean) refers to the least significant digits. Results in square brackets were calculated from
 279 the data given in the original publication.
 280

281 2.3 Lead isotope analysis (LIA) interpretation

282 The lead isotope results were then compared with an ore reference database, containing 138
 283 datasets and data points (Appendix B), to eliminate the least plausible candidates and to highlight
 284 potential sources. The ore reference database was compiled for this study from the available
 285 geological and archaeological literature, and from the Oxford archaeological lead isotope
 286 database (OXALID), which is freely available online (Stos-Gale and Gale, 2009). It includes lead
 287 isotope ratios obtained on a variety of minerals, with lead ore as the priority.
 288 Comparing LIA results for ores and archaeological samples will generally eliminate incompatible
 289 sources, but unequivocal identification of provenance is rarely possible (Cattin et al., 2010). The
 290 complexity of geological history may result in vast isotopic fields, with great diversity within the

291 same ore body. By contrast, ore bodies many hundreds of kilometres apart may have similar
292 isotopic compositions, making further discrimination impossible. In addition, ore reference data
293 may be limited in number, even for major mining regions. Old mining works are not always
294 dated; mines may not all have been identified; medieval mines may even have been destroyed
295 (Téreygeol, 2009). Lead ores from different sources may have been mixed together, and lead
296 metal was frequently reused over time (Boni et al., 2000). Mixing of sources can only occur
297 during glaze preparation, because the lead in finished glazes is non-recyclable (Mason et al.,
298 1992).

299 Despite these drawbacks, LIA is considered an effective method when used in conjunction with
300 historical records and archaeological evidence (Wolf et al. 2003; Stos-Gale and Gale, 2009;
301 Hauptmann, 2020). This combined approach provides a reliable way to discriminate between
302 potential sources, and thus validate provenance hypotheses.

303

304 3. RESULTS AND DISCUSSION

305 After a brief presentation of LIA results, the remaining sections will present archaeological and
306 historical arguments regarding glaze manufacture and lead supply strategies, the potential sources
307 proposed in this study, and the provenance of the lead used in the tin-opacified glazes.

308

309 **3.1 Lead isotope data**

310 Table 3 presents the range of values for lead isotope ratios, organised by type of glaze and by
311 decorative technique. Based on these results, six groups can be proposed (LIA1 to LIA6), leaving
312 four outliers. Although LIA2 and LIA3 lie close together, they are treated as separate groups
313 because of the slight difference in isotopic values between plain and two-colour transparent lead-

314 glazed tiles. No such logical subdivision is apparent for the data points in LIA5, despite the broad

315 range of isotope ratios in this group.

316

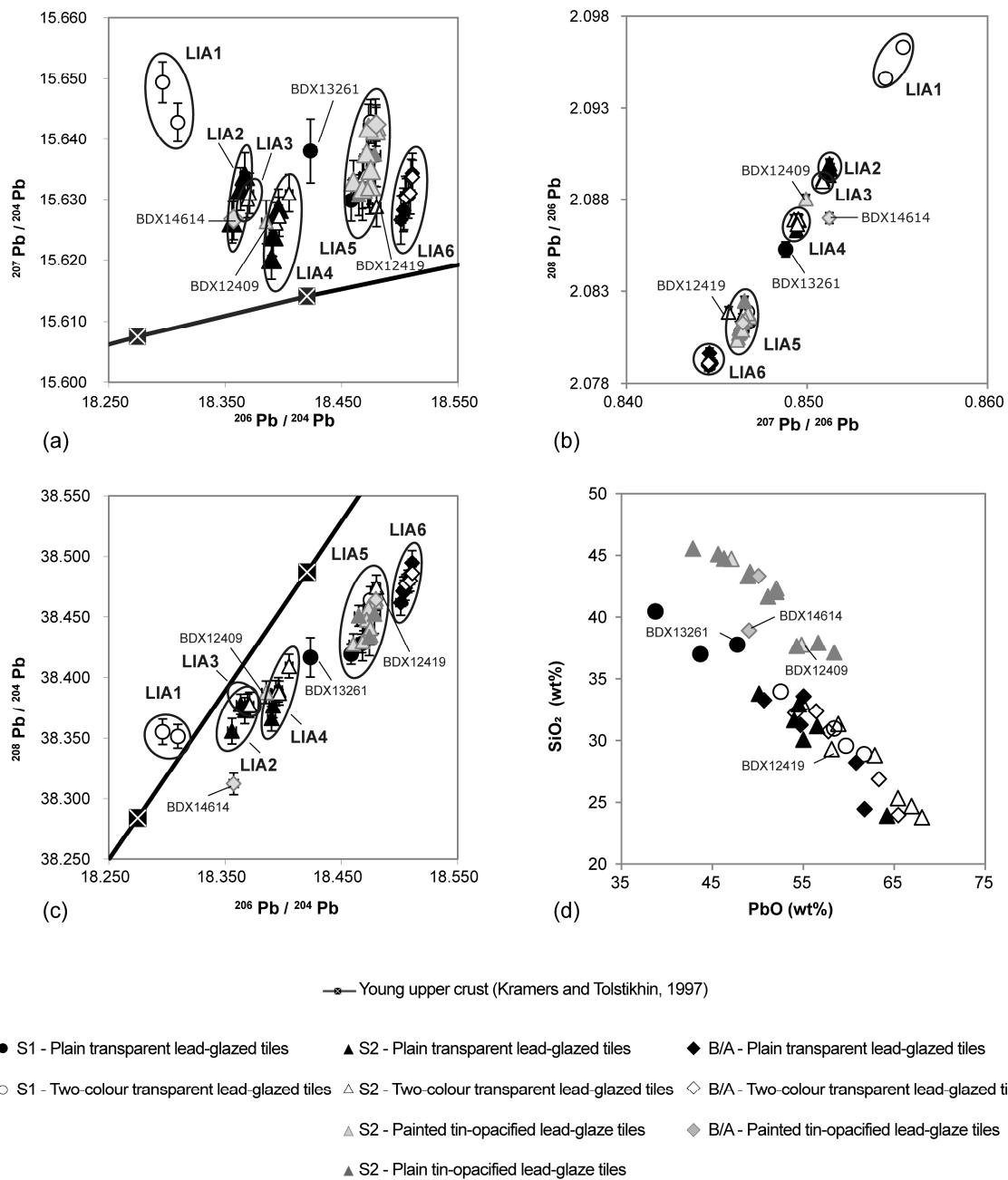
Sample ID and Decor	Body type	PbO (wt%)	LIA group	²⁰⁸ Pb/ ²⁰⁴ Pb	²⁰⁷ Pb/ ²⁰⁴ Pb	²⁰⁶ Pb/ ²⁰⁴ Pb	²⁰⁸ Pb/ ²⁰⁶ Pb	²⁰⁷ Pb/ ²⁰⁶ Pb
SUSCINIO I								
Transparent								
PLAIN								
BDX13257	NC1	43.7 ± 1.1	LIA5	38.428 ± 0.008	15.631 ± 0.003	18.468 ± 0.003	2.08089 ± 0.00026	0.84640 ± 0.00016
BDX13260	NC2	38.7 ± 1.5	LIA5	38.419 ± 0.009	15.630 ± 0.003	18.458 ± 0.004	2.08134 ± 0.00027	0.84677 ± 0.00017
BDX13261	NC3	47.7 ± 1.6	outlier	38.417 ± 0.016	15.638 ± 0.005	18.423 ± 0.005	2.08528 ± 0.00041	0.84882 ± 0.00008
TWO-COLOUR								
BDX13255	NC2	52.5 ± 0.9	LIA5	38.464 ± 0.011	15.643 ± 0.004	18.475 ± 0.004	2.08189 ± 0.00029	0.84669 ± 0.00015
BDX13256	NC2	58.4 ± 1.2	LIA1	38.351 ± 0.010	15.643 ± 0.003	18.309 ± 0.003	2.09462 ± 0.00029	0.85434 ± 0.00015
BDX13258	NC2	61.7 ± 0.7	LIA1	38.355 ± 0.010	15.649 ± 0.003	18.296 ± 0.003	2.09632 ± 0.00030	0.85532 ± 0.00015
SUSCINIO II								
Transparent								
PLAIN								
BDX12420	NC3	54.0 ± 2.1	LIA4	38.377 ± 0.010	15.624 ± 0.003	18.391 ± 0.003	2.08685 ± 0.00029	0.84955 ± 0.00015
BDX12421	NC3	55.0 ± 1.4	LIA2	38.356 ± 0.011	15.626 ± 0.003	18.356 ± 0.004	2.08964 ± 0.00031	0.85130 ± 0.00015
BDX12422	NC3	64.2 ± 0.2	LIA4	38.390 ± 0.010	15.628 ± 0.003	18.396 ± 0.003	2.08687 ± 0.00029	0.84956 ± 0.00015
BDX12423	NC3	54.5 ± 1.1	LIA2	38.378 ± 0.008	15.632 ± 0.004	18.363 ± 0.004	2.08994 ± 0.00028	0.85124 ± 0.00017
BDX12424	NC3	56.5 ± 0.7	LIA2	38.372 ± 0.011	15.634 ± 0.004	18.367 ± 0.004	2.08932 ± 0.00030	0.85120 ± 0.00015
BDX12425	NC3	50.1 ± 0.9	LIA4	38.366 ± 0.010	15.620 ± 0.003	18.390 ± 0.003	2.08631 ± 0.00030	0.84942 ± 0.00015
TWO-COLOUR								
BDX12412	NC3	58.8 ± 2.7	LIA4	38.409 ± 0.010	15.631 ± 0.003	18.405 ± 0.003	2.08688 ± 0.00028	0.84928 ± 0.00015
BDX12413	NC3	62.9 ± 0.8	LIA3	38.377 ± 0.010	15.630 ± 0.003	18.369 ± 0.003	2.08912 ± 0.00029	0.85088 ± 0.00015
BDX12415	NC3	54.8 ± 2.2	LIA5	38.449 ± 0.010	15.634 ± 0.003	18.469 ± 0.004	2.08180 ± 0.00029	0.84650 ± 0.00015
BDX12416	NC3	68.0 ± 0.4	LIA4	38.387 ± 0.010	15.626 ± 0.003	18.394 ± 0.003	2.08691 ± 0.00029	0.84951 ± 0.00015
BDX12417	NC3	66.9 ± 0.4	LIA3	38.377 ± 0.011	15.631 ± 0.003	18.371 ± 0.003	2.08899 ± 0.00030	0.85085 ± 0.00015
BDX12418	NC3	65.4 ± 0.8	LIA4	38.387 ± 0.010	15.627 ± 0.003	18.396 ± 0.003	2.08663 ± 0.00028	0.84946 ± 0.00015
BDX12419	NC3	58.1 ± 0.4	outlier	38.474 ± 0.010	15.629 ± 0.003	18.480 ± 0.003	2.08188 ± 0.00029	0.84566 ± 0.00015
Tin-opacified								
PLAIN								
BDX12402	NC3	43.7 ± 1.1	LIA5	38.451 ± 0.009	15.631 ± 0.003	18.465 ± 0.004	2.08248 ± 0.00025	0.84654 ± 0.00016
BDX12408	NC3	40.1 ± 3.1	LIA5	38.434 ± 0.016	15.633 ± 0.005	18.474 ± 0.005	2.08047 ± 0.00041	0.84618 ± 0.00008
BDX12410	NC3	35.8 ± 2.6	LIA5	38.452 ± 0.016	15.637 ± 0.005	18.479 ± 0.005	2.08098 ± 0.00040	0.84624 ± 0.00008
PAINTED								
BDX12398	NC3	36.9 ± 1.6	LIA5	38.465 ± 0.009	15.642 ± 0.003	18.480 ± 0.004	2.08132 ± 0.00030	0.84641 ± 0.00017
BDX12399	NC3	42.0 ± 0.7	LIA5	38.436 ± 0.009	15.632 ± 0.003	18.473 ± 0.003	2.08071 ± 0.00028	0.84621 ± 0.00016
BDX12400	NC3	39.1 ± 2.3	LIA5	38.463 ± 0.016	15.641 ± 0.005	18.479 ± 0.005	2.08139 ± 0.00040	0.84643 ± 0.00008
BDX12401	NC3	39.7 ± 2.2	LIA5	38.428 ± 0.008	15.633 ± 0.003	18.460 ± 0.004	2.08163 ± 0.00027	0.84685 ± 0.00017
BDX12403	NC3	38.1 ± 1.4	LIA5	38.457 ± 0.010	15.642 ± 0.004	18.473 ± 0.004	2.08181 ± 0.00024	0.84673 ± 0.00016
BDX12404	NC3	43.8 ± 1.6	LIA5	38.435 ± 0.008	15.632 ± 0.003	18.475 ± 0.003	2.08035 ± 0.00027	0.84612 ± 0.00017
BDX12405	NC3	35.4 ± 2.5	LIA5	38.448 ± 0.007	15.638 ± 0.003	18.472 ± 0.003	2.08145 ± 0.00025	0.84657 ± 0.00016
BDX12406	NC3	36.2 ± 1.2	LIA5	38.441 ± 0.008	15.635 ± 0.003	18.475 ± 0.004	2.08080 ± 0.00024	0.84630 ± 0.00016
BDX12407	NC3	45.1 ± 2.2	LIA5	38.430 ± 0.016	15.632 ± 0.005	18.468 ± 0.005	2.08089 ± 0.00040	0.84643 ± 0.00008

BDX12409	NC3	40.8 ± 1.8	outlier	38.388 ± 0.009	15.626 ± 0.004	18.385 ± 0.004	2.08803 ± 0.00026	0.84994 ± 0.00016
BRAIN-SUR-ALLONNES								
Transparent								
PLAIN								
BDX14616	NC4	61.7 ± 0.8	LIA6	38.474 ± 0.010	15.631 ± 0.003	18.505 ± 0.003	2.07909 ± 0.00029	0.84469 ± 0.00015
BDX14617	NC4	50.7 ± 1.5	LIA6	38.471 ± 0.010	15.628 ± 0.003	18.504 ± 0.003	2.07909 ± 0.00031	0.84459 ± 0.00015
BDX14618	NC4	54.7 ± 1.4	LIA6	38.471 ± 0.012	15.628 ± 0.004	18.503 ± 0.003	2.07917 ± 0.00035	0.84465 ± 0.00016
BDX14619	NC4	55.0 ± 2.0	LIA6	38.495 ± 0.010	15.634 ± 0.003	18.511 ± 0.003	2.07963 ± 0.00029	0.84460 ± 0.00015
BDX14620	NC4	60.8 ± 0.6	LIA6	38.462 ± 0.010	15.627 ± 0.004	18.501 ± 0.004	2.07902 ± 0.00029	0.84465 ± 0.00017
TWO-COLOUR								
BDX14621	NC4	63.3 ± 1.3	LIA6	38.486 ± 0.008	15.634 ± 0.003	18.510 ± 0.003	2.07923 ± 0.00027	0.84463 ± 0.00016
BDX14622	NC4	65.4 ± 0.8	LIA6	38.479 ± 0.009	15.630 ± 0.004	18.507 ± 0.004	2.07896 ± 0.00029	0.84454 ± 0.00017
BDX14623	NC4	56.4 ± 1.5	LIA6	38.477 ± 0.008	15.630 ± 0.003	18.505 ± 0.004	2.07911 ± 0.00028	0.84464 ± 0.00016
BDX14624	NC4	54.1 ± 0.4	LIA6	38.480 ± 0.009	15.631 ± 0.003	18.509 ± 0.004	2.07902 ± 0.00024	0.84454 ± 0.00016
BDX14625	NC4	57.8 ± 0.5	LIA6	38.486 ± 0.007	15.634 ± 0.003	18.511 ± 0.003	2.07910 ± 0.00024	0.84454 ± 0.00016
Tin-opacified								
PAINTED								
BDX14614	NC4	45.2 ± 1.3	outlier	38.312 ± 0.009	15.627 ± 0.003	18.357 ± 0.004	2.08697 ± 0.00029	0.85123 ± 0.00017
BDX14615	NC4	45.9 ± 1.2	LIA5	38.464 ± 0.007	15.642 ± 0.003	18.480 ± 0.003	2.08123 ± 0.00025	0.84644 ± 0.00016

318

319 Table 3. Lead content determined by SEM-EDS in previous studies (Métreau et al. 2012a, 2012b, 2017) and lead isotope ratios for the 44 glaze samples.
 320 Analytical error for lead isotope ratios is shown as 2 standard errors of the mean.

321 Figure 3 displays the lead isotopic composition of the samples and their elemental composition:
322 $^{207}\text{Pb}/^{204}\text{Pb}$ vs. $^{206}\text{Pb}/^{204}\text{Pb}$ (Figure 3a), $^{208}\text{Pb}/^{206}\text{Pb}$ vs. $^{207}\text{Pb}/^{206}\text{Pb}$ (Figure 3b), $^{208}\text{Pb}/^{204}\text{Pb}$ vs.
323 $^{206}\text{Pb}/^{204}\text{Pb}$ (Figure 3c), and SiO_2 vs. PbO wt% content (Figure 3d). When compared with the lead
324 evolution model of Kramers and Tolstikhin (1997), all the data points plot above the line for the
325 young upper crust (Figure 3a), indicating that the ore deposit had incorporated some lead deriving
326 from older crust. In Figure 3c, as LIA1 (BDX13256 and BDX13258) contains lead enriched in
327 ^{208}Pb derived from ^{232}Th , this group now lies to the left of the line representing the young upper
328 crust.
329



330

331

332 Figure 3. Lead isotope ratios and elemental composition of glaze samples from S1–Suscinio I, S2–Suscinio II, and
 333 B/A–Brain-sur-Allonnes: (a) $^{207}\text{Pb}/^{204}\text{Pb}$ vs. $^{206}\text{Pb}/^{204}\text{Pb}$; (b) $^{208}\text{Pb}/^{206}\text{Pb}$ vs. $^{207}\text{Pb}/^{206}\text{Pb}$; (c) $^{208}\text{Pb}/^{204}\text{Pb}$ vs. $^{206}\text{Pb}/^{204}\text{Pb}$;
 334 (d) SiO_2 vs. PbO wt% concentration. For the tin-opacified lead glazes, the tin oxide content was subtracted from the
 335 global composition, which was then recast to 100%. The error may be smaller than the symbols.
 336

337 **3.2 Organisation of glaze manufacture and lead supply strategies**

338 The six groups identified by LIA may reflect separate production phases, during each of which
 339 the glazing mixture was prepared with lead from a specific source, for a limited number of tiles
 340 (Table 4).

341

LIA GROUPS AND PAVEMENTS												
DECOR	LIA1	LIA2	LIA3	LIA4	LIA5			LIA6	Outliers			Total
	S1	S2	S2	S2	S1	S2	B/A	B/A	S1	S2	B/A	
Transparent												
PLAIN		3		3	2			5	1			14
TWO-COLOUR	2		2	3	1	1		5		1		15
Tin-opacified												
PLAIN						3						3
PAINTED						9	1			1	1	12
Total	2	3	2	6	17			10	4			44

342

343 Table 4. Decorative techniques and LIA groups for each pavement: S1–Suscínio I, S2–Suscínio II, and B/A–Brain-
 344 sur-Allonnes.
 345

346 Five groups are site-specific, with transparent lead-glazed tiles only: LIA1 (Suscínio I), LIA2,
 347 LIA3, and LIA4 (Suscínio II), and LIA6 (Brain-sur-Allonnes), suggesting that several lead
 348 supply networks could be implemented for the same technique. The results for LIA5, however,
 349 suggest that a single source could have been used for different decorative techniques. Archival
 350 documents indicate that several procurement strategies were used for glaze production,
 351 depending on the market availability of lead, or the opening of a nearby mine as work progressed
 352 (Bon, 1992, pp. 61–93, 120), and on material provided by sponsors (Berthier and Flouzat, 2009),
 353 or even by royal patronage (Madeline, 2009).

354 Almost all the tin-opacified lead-glaze tiles are found in LIA5, apart from two outliers
 355 (BDX12409 from Suscínio II, and BDX14614 from Brain-sur-Allonnes). This group also
 356 includes four transparent tiles, one from Suscínio II, and three from the earlier pavement,
 357 Suscínio I. The tin-oxide contamination of two transparent glaze samples from Suscínio II,

358 BDX12415 (LIA5) and BDX12419 (the closest outlier to LIA5), suggests that these two-colour
 359 tiles must therefore have been produced in the same workshop as the tin-opacified lead-glaze tiles
 360 (Métreau et al., 2017). Medieval pottery workshops producing a variety of wares are known to
 361 have existed in the late 13th century, in the faubourg Sainte-Barbe, Marseille, France, for
 362 example (Thiriot, 1995).

363

364 3.3 Potential sources of lead materials

365 Comparison of the results for the LIA groups with the ore reference database (Appendix B)
 366 shows that, for each of the six groups, around three-quarters of the potential sources of lead
 367 materials are incompatible. The remaining quarter, whether datasets or data points, can be
 368 classified as either possible or plausible sources for the lead used in the glazes (Table 5).

369

LIA GROUPS AND PAVEMENTS						
	LIA1	LIA2	LIA3	LIA4	LIA5	LIA6
	S1	S2	S2	S2	S1+S2+B/A	B/A
INCOMPATIBLE	99	98	98	93	100	107
COMPATIBLE	37	38	38	41	36	29
Possible	29	30	30	24	26	23
Plausible	8	8	8	17	8	6
Good match					2	

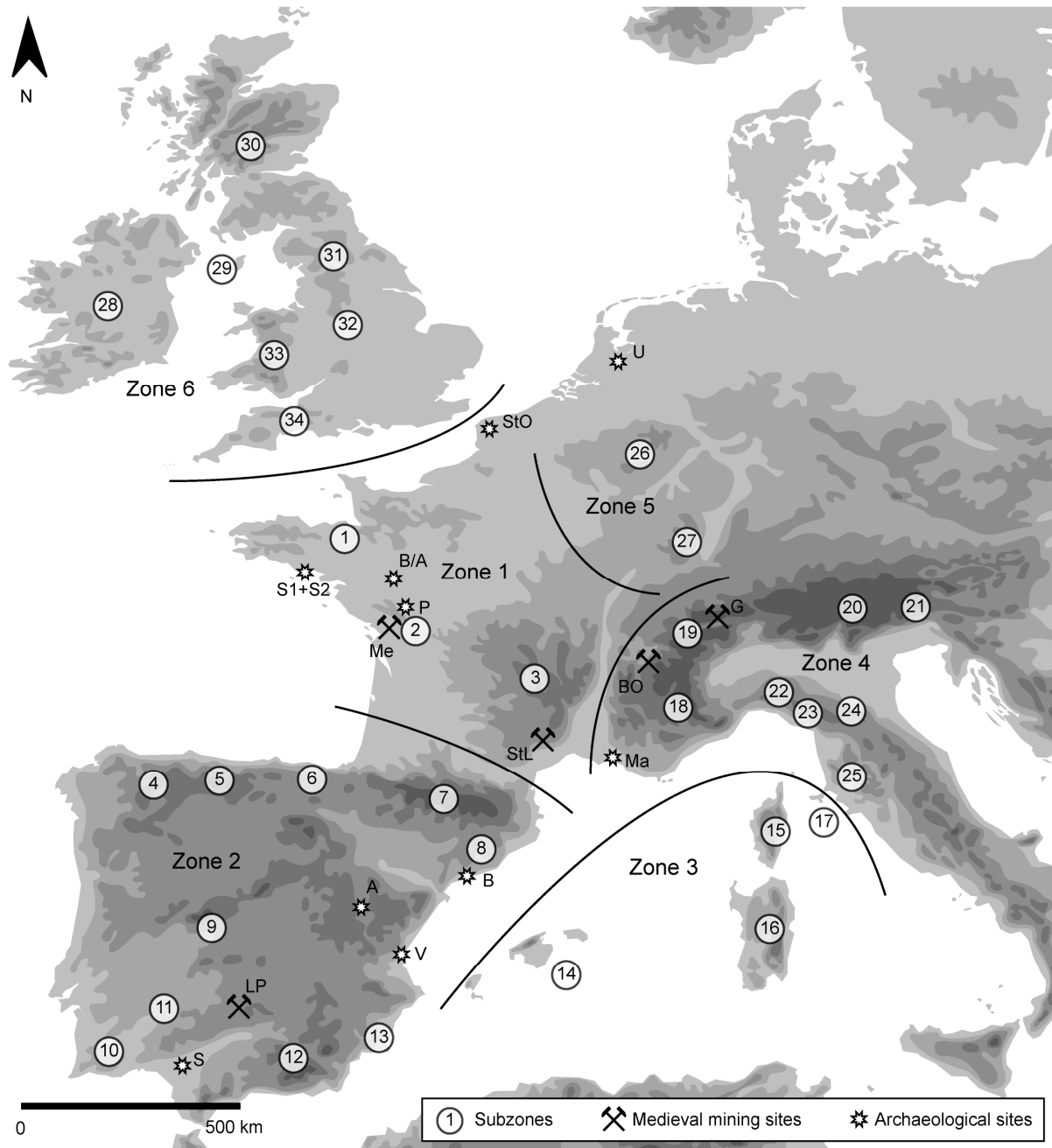
370

371 Table 5. Degree of compatibility between the database (Appendix B), LIA groups, and pavements.

372

373 Possible sources present some drawbacks, because the dataset contains outliers, consists of a
 374 small number of data points, or covers a broad isotopic field. Plausible sources are more likely
 375 candidates, with a narrow isotopic field. Good matches potentially indicate the most likely
 376 sources. To evaluate potentially compatible sources for the lead in the glazes, relevant
 377 archaeological and historical arguments must be explored. For this purpose, the ore reference

378 database (Appendix B) was organised into six major geographic zones, further subdivided into
 379 34 subzones (Figure 4).



380
 381 Figure 4. Map of the ore reference database (Appendix B); **circle**, subzone: **Zone 1:** **1**-Armorican Massif, **2**-Seuil du
 382 Poitou, **3**-Massif Central; **Zone 2:** **4**-West Asturian Leonese, **5**-Cantabrian, **6**-Cantabrian-Basque, **7**-Pyrenees, **8**-
 383 Catalanian Coastal Range, **9**-Central Iberian, **10**-South Portuguese, **11**-Ossa Morena, **12**-Betic Cordillera, **13**-
 384 Southeast Volcanic Province; **Zone 3:** **14**-Balearic Islands, **15**-Corsica, **16**-Sardinia, **17**-Elba; **Zone 4:** **18**-Western
 385 Alps, **19**-Central Alps, **20**-Southern Alps, **21**-Carnic Alps, **22**-Ligurian Apennines, **23**-Apuane Alps, **24**-Modenese
 386 Apennines, **25**-Southern Tuscany; **Zone 5:** **26**-Western Rhenish Massif **27**-Vosges; **Zone 6:** **28**-Ireland, **29**-Isle of

387 Man, **30**-Scotland; **31**-Northern Pennines, **32**-Southern Pennines, **33**-The West, **34**-South West England; **pickaxes**,
 388 medieval mining site: **BO**-Brandes-en-Oisans, **G**-Goppenstein, **LP**-Los Pedroches, **M**-Melle, **StL**-Saint-Laurent-le-
 389 Minier; **star**, archaeological site: **S1+S2**-Suscinio, **B/A**-Brain-sur-Allonnes, **A**-Albarracín, **B**-Barcelona, **Ma**-
 390 Marseille, **P**-Poitiers, **StO**-St Omer, **S**-Seville, **U**-Utrecht, **V**-Valencia.
 391

392 The pavement sites are all located in Zone 1, which will be examined in greater depth in
 393 Section 3.4. Zone 2, discussed in Section 3.5, represents a viable provenance hypothesis for the
 394 tin-opacified glazes (LIA5), as this Islamic tradition spread to the southern and eastern parts of
 395 the Iberian Peninsula during the medieval period (Caiger-Smith, 1985). No datasets in Zone 3 are
 396 plausible sources of lead for the glazes in any of the LIA groups. In Zone 4, although some data
 397 points for Goppenstein (in Subzone 19) are compatible sources of lead for the glazes in LIA5,
 398 and even LIA1 and LIA6, there is no archaeological or historical evidence to confirm the
 399 exploitation of this medieval lead mine before 1474 (Guénette-Beck, 2005; 2019). Two data
 400 points in Col du Lautaret (Subzone 18), about 30 km from the medieval mining district of
 401 Brandes-en-Oisans (Bailly-Maître and Bruno-Dupraz, 1994), are compatible sources of lead for
 402 the glazes in LIA6. In Zone 5, only four datasets in the Western Rhenish Massif (Subzone 26) are
 403 plausible sources of lead for the glazes in LIA4 (Appendix B). Zone 6, presented in detail in
 404 Section 3.6, provides the most promising results, with plausible sources of lead for the glazes in
 405 all six LIA groups, and two good matches for LIA5 (Table 6). The discussion of potential sources
 406 for the lead in the glazes will now focus on Zones 1, 2, and 6, illustrated by figures plotting the
 407 LIA groups against the relevant lead isotope datasets (see also the maps in Appendix B).
 408

Sources	LIA GROUPS AND PAVEMENTS					
	LIA1 S1	LIA2 S2	LIA3 S2	LIA4 S2	LIA5 S1+S2+B/A	LIA6 B/A
PLAUSIBLE						
Zone 1	•	•	•	•	•	
Zone 2				•	•	•
Zone 4	•				•	•
Zone 5				•		
Zone 6	•	•	•	•	•	•

GOOD MATCH

Zone 6

SUBZONE 32. DERBYSHIRE

SUBZONE 34. AVON

•

•

409

410 Table 6. Zone compatibility between lead sources, LIA groups, and pavements. No datasets in Zone 3 are plausible
411 sources.

412

413 **3.4 The local or regional source hypothesis (Zone 1)**

414 Archival documents (1384–1386) attest that lead for some of the tin-opacified lead-glaze tiles
415 was mined at a distance of 45 km from the Palace of Poitiers, for the Duke de Berry (Bon, 1992).

416 The local provenance hypothesis for Suscinio I and II would therefore be the Armorican Massif
417 (Subzone 1), where mines for various metals have been exploited since the Bronze Age.

418 Medieval mining activity nevertheless remains difficult to evaluate (Domergue et al., 2006;

419 Le Carlier de Veslud and Jouanet-Aldous, 2015; Le Carlier de Veslud et al., 2017).

420 Figure 5 presents detailed results for Subzone 1, divided into three categories. Although nine

421 plausible matches are identified (Appendix B), the error margin is greater than the match, making

422 these sources appear less likely. Another potential drawback is that there is often only one data

423 point for each ore body in the Armorican Massif, so that comparison between LIA groups and the

424 database is less robust.

425 The only plausible neighbouring source (less than 50 km from Suscinio) is Plumelin, a data point

426 that matches LIA4. The lead isotopic data for LIA2, LIA3, and LIA4 (Suscinio II) are in the same

427 range as the ore bodies south-west of Pontivy, at a distance of about 75 km, while La Villeneuve,

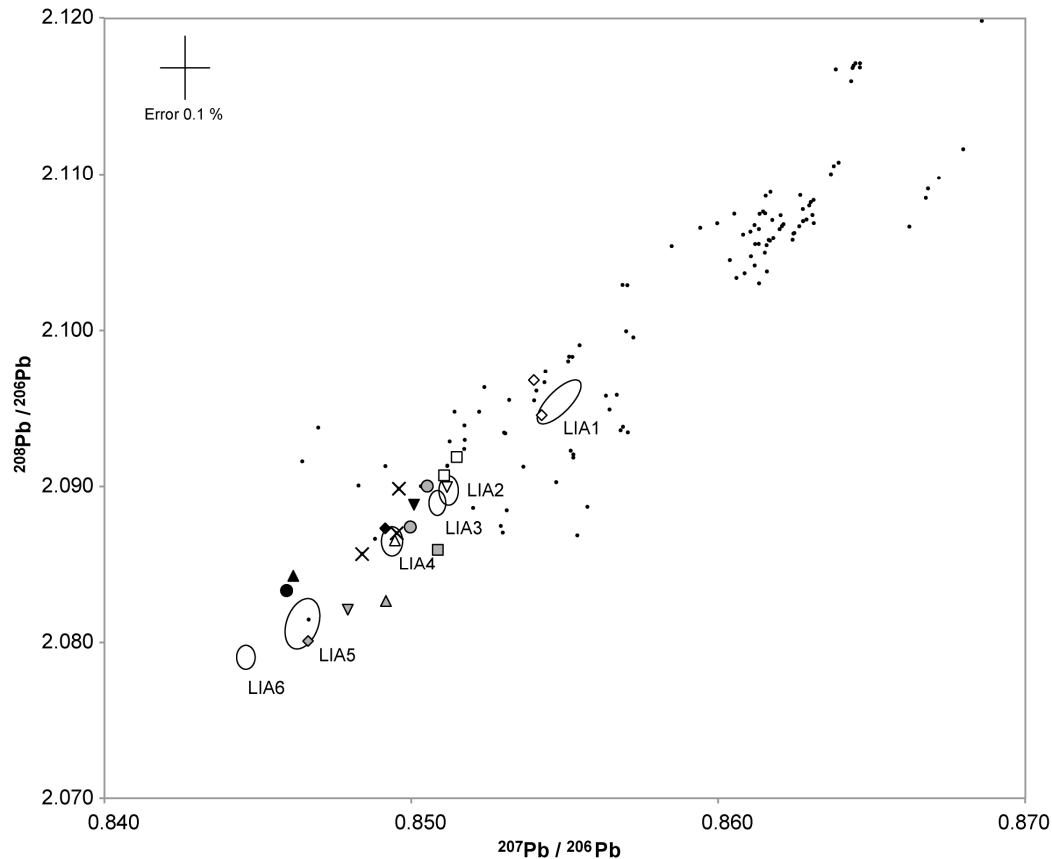
428 about 80 km from Suscinio, is the only dataset compatible with LIA5. Huelgoat, a lead mine

429 about 120 km north-west of Suscinio, which may have been exploited earlier than 1540 (Coativy,

430 2006), is a plausible match for LIA1 (Suscinio I), but with only one data point. Plélauff, a much

431 earlier mine about 100 km away, is a plausible match for LIA2 (Suscino II), but again with only
 432 one data point.

433



Ore bodies in the Armorican Massif

< 50 km from Suscinio

- ▲ Crossac
- Le Sem
- ▼ Plumelin LIA3
- ◆ St Modé (Baud)

SW of Pontivy

- ◇ La Villeneuve LIA5
- △ Le Vouédec
- Locrio
- Quistiave LIA2, LIA3, LIA4
- ▽ St Fiacre

Others

- ◇ Huelgoat LIA1
- △ Kerhuo LIA4
- × La Telhaie LIA4
- ▽ Plélauff LIA2
- Pontpéan
- Non-cited ore bodies

434

435 Figure 5. Plot of LIA groups (black ellipses) and ore data from the Armorican Massif (Appendix B).
 436

437 At a distance of 145 km from Brain-sur-Allonnes, Melle (in Subzone 2) presents a possible
 438 match, but this mine was only exploited from the 6th to the 10th century (Téreygeol et al., 2010).

439 Eleven of the transparent lead-glaze samples from Suscino II (LIA2, LIA3, and LIA4) present a

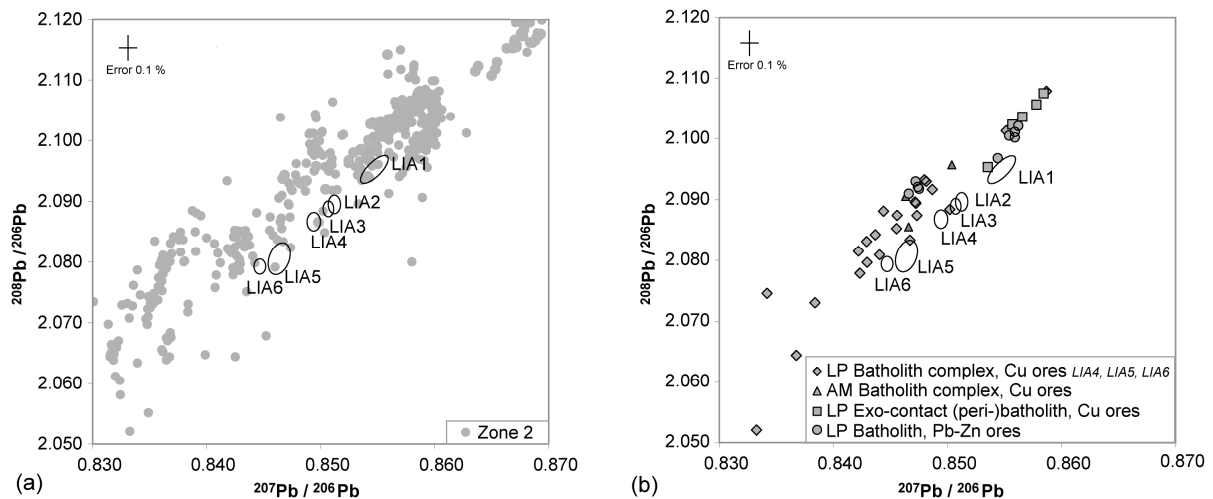
440 plausible match with Saint-Laurent-le-Minier (in Subzone 3), much further south in the Massif
441 Central (Appendix B).

442

443 3.5 The imported glazing mixture hypothesis (Zone 2)

444 A fundamental hypothesis at the beginning of this study was the importation from the Iberian
445 Peninsula of ready-to-use glazing mixtures for the tin-opacified lead glazes. Comparison with the
446 database shows that LIA5 coincides with a sparsely populated area in the isotopic field of Zone 2
447 (Appendix B). The Los Pedroches Batholith complex (in Subzone 9) is the only plausible source
448 (Figure 6), but most of the compatible lead isotope data come from copper ores (Klein et al.,
449 2009). Furthermore, Grañeda Miñión (2008) indicates that mining activity at Los Pedroches,
450 which flourished during the Caliphal period, had already collapsed during the early 12th century.

451



452

453 Figure 6 (a). Data for Zone 2, showing LIA groups; (b) Subzone 9, data from Los Pedroches, showing LIA groups
454 (Appendix B).

455

456 Norton (1984b) put forward the idea that a tiler from a workshop in the Garonne valley might
457 have imported the tin-glazed technique after working at the Abbey of Santas Creus near

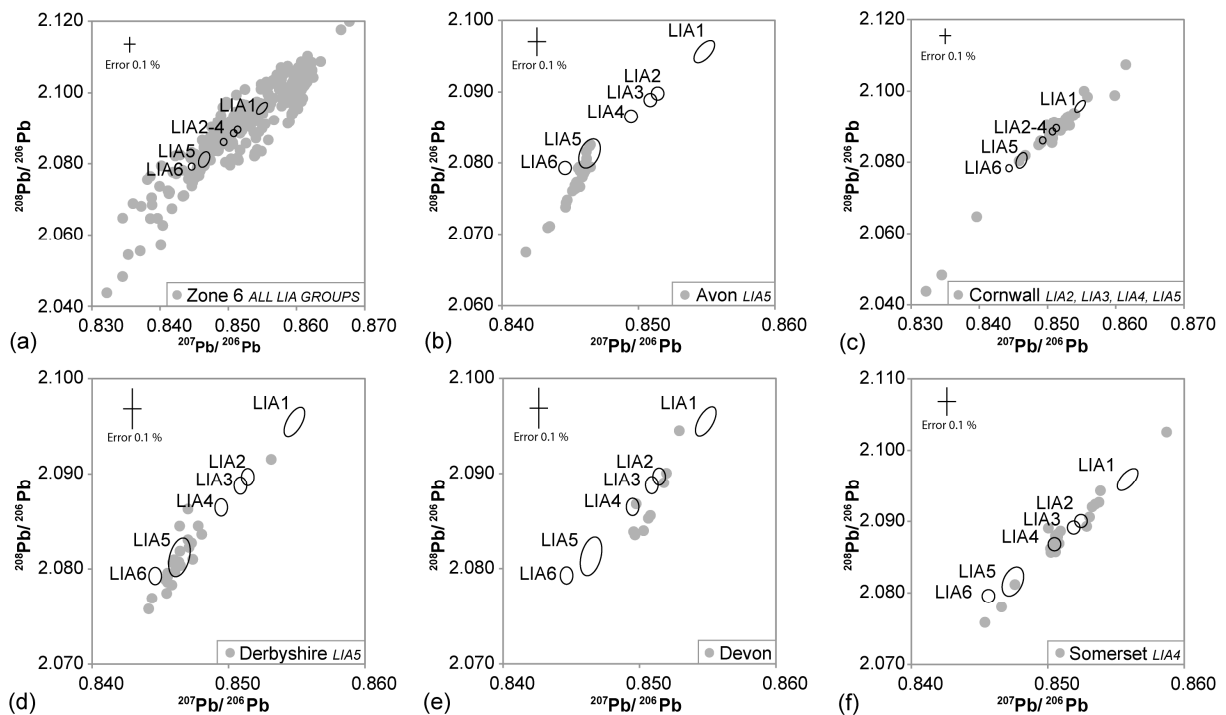
458 Barcelona. Although tin-opacified lead glazes were probably used to decorate ceramics in
459 Catalonia from the 14th century (Iñáñez, 2007), no tin-opacified lead-glaze tiles are described at
460 the Abbey of Santas Creus (Norton, 1984b, fig. 2, p. 137). The isotopic field of the Catalanian
461 Coastal Range (Subzone 8), extended by one uncharacteristic data point, is not coherent with
462 LIA5, nor any of the other LIA groups. Workshops using the tin-glazed earthenware technique,
463 mainly for the production of ceramics, are known to have existed from the 12th century, further
464 south, near Valencia (Manises and Paterna) and Seville (Iñáñez et al., 2008). However, no
465 plausible match for any of the LIA groups exists with the isotopic fields of the Betic Cordillera
466 (Subzone 12) or the South-East Volcanic Province (Subzone 13). The LIA study of four 11th-
467 century tin-opacified lead-glaze ceramics from Albarracín (Marzo et al., 2009) produced results
468 similar to those for LIA5, but no specific Iberian provenance could be identified.

469

470 **3.6 The most plausible hypothesis: lead of British origin (Zone 6)**

471 Comparison between LIA groups and the database identifies Zone 6 as the most plausible
472 provenance for all the lead glazes sampled (Appendix B). The Isle of Man (Subzone 29) is a
473 plausible source for LIA1. In Subzone 31, there are two plausible sources, Cumbria for LIA1 to
474 LIA4, and Durham for LIA4. In Subzone 32, Cheshire is a plausible source for LIA4, LIA5, and
475 LIA6. Subzone 33 is also a plausible source: the Shropshire Hills only for LIA1, Clwyd for all
476 groups except LIA6, and Gwyned for all six groups. In Subzone 34, the plausible sources are
477 Cornwall for LIA2 to LIA5, and Somerset for LIA4 (Figure 7).

478



479 (a) (b) (c) (d) (e) (f)

480 Figure 7. LIA groups (black ellipses) plotted against ore data from Zone 6 (Appendix B).

481

482 Results for Zone 6 also indicate two good matches, Avon and Derbyshire, as potential

483 provenance for LIA5 (Appendix B, Figure 7). A British origin for the lead in the tin-opacified

484 glazes challenges two previously explored hypotheses: the opening of mines in neighbouring

485 regions (Section 3.4; Figure 5), and the importation of ready-to-use glazing mixtures from the

486 Iberian Peninsula (Section 3.5; Figure 6). At the turn of the 14th century, Derbyshire was the

487 main lead-producing area in England and Wales, exporting lead to Northern Europe, mainly via

488 Hanseatic trade routes (Blanchard, 2005), and thus appears to be a more historically plausible

489 solution than Avon. Both Suscinio, on the coast of Brittany, and Brain-sur-Allonnes, in the Loire

490 Valley, had easy access to Atlantic trade routes. Stylistic and technical affinities between

491 Suscinio II and contemporaneous Flemish pavements suggest other routes, along the English

492 Channel, to the Abbey of St Bertin, in St Omer, and to sites around Utrecht, where a workshop

493 producing tin-opacified lead-glaze tiles has been identified (de Groot and Pot, 1985).

494 The quality, accessibility, and economy of English tin might explain why lead from foreign
495 sources was preferred (Muhly, 1985; Tylecote, 1987). Piccolpasso's 16th-century treatise on
496 Italian maiolica (1556-1557) mentions that tin from Flanders was recommended for opacifying
497 lead glazes, but the source of this tin was south-west England (Lhôte, 2007; McSweeney, 2011).
498 The use of an oxidised lead-tin alloy (termed *calcine*) ensures homogeneous distribution of the
499 opacifying tin oxide (SnO_2) throughout the glaze. To act as an opacifier, the tin-oxide content of
500 tin-opacified lead glazes should be at least 5 wt% SnO_2 (Mason and Tite, 1997). A tin-oxide
501 content ranging from 20 to 25 wt% SnO_2 has been observed on high-quality Mediterranean wares
502 (Picon et al., 1995). A ready-to-use lead-tin alloy thus seems very unlikely, since the craftsmen
503 would not be able to control the quality of the *calcine*. For the tin-opacified lead-glaze tiles of
504 Suscinio II, a single supply route from England for both lead and tin (possibly in the form of
505 ingots), thus appears to be the most plausible hypothesis.

506

507 4. CONCLUDING REMARKS AND PERSPECTIVES

508 This exploratory study will contribute to the growing body of knowledge about the history of
509 decorated tile pavements, and the tin-glazed earthenware technique, or *faïence*. The corpus of
510 44 tiles, representative of three medieval pavements and four decorative techniques, is large
511 enough to produce meaningful results. Lead isotope analysis of glaze samples, when combined
512 with archaeological and historical evidence, can provide important information about lead supply
513 networks, notably the provenance of the lead in the tin-opacified glazes from Suscinio II and
514 Brain-sur-Allonnes (14th century). The plausible matches between Zone 6 and all the LIA
515 groups, and the good match with English lead for LIA5, together suggest a rather unexpected
516 supply route. As the same lead was used for the tin-opacified glazes, and for some of the
517 transparent lead-glazed tiles from Suscinio, the Derbyshire provenance for LIA5 tends to

518 discount the hypothesis of an imported ready-to-use glazing mixture for the opaque tin glazes.
519 The positive identification of the British Isles as a source of imported lead could be the starting
520 point for future investigations of other medieval tin-opacified lead-glaze products. At other sites,
521 written sources may have been preserved that could provide more information about the
522 motivations underlying the choice of specific types of lead for such products. Identifying the
523 supply routes for both lead and tin is among the ultimate goals for such studies.

524

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532

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