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

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To cite this version:

Laetitia Métreau, Florence Cattin, Igor Maria Villa, Patrick André, Carmela Chateau. Lead provenance for medieval decorated tile glazes from Brittany and Anjou (13th-14th c.). Journal of Archaeological Science: Reports, 2021, 38, pp.103037. 10.1016/j.jasrep.2021.103037 . halshs-03536836

Version of Record: https://www.sciencedirect.com/science/article/pii/S2352409X21002492
Manuscript_8d762546518f501a0103619a787dc876

TITLE

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DECLARATION OF INTEREST

Declarations of interest: none.

ABSTRACT

Medieval pavements composed of lead-glazed tiles decorated with a variety of techniques continue to inspire questions about the organisation of glaze manufacture, and the supply and origin of lead materials. The tiles analysed in this study are from Suscinio I (a 13th-century pavement) and Suscinio II (a 14th-century pavement), at the Château of Suscinio in Brittany, and also from the 14th-century pavement at the fortified manor house in Brain-sur-Allonnes, Anjou. Lead isotope analysis (LIA) was used to examine samples from 44 lead-glazed tiles, 29 of which are transparent, while 15 are tin-opacified (an exogenous technique in these regions during this period). Five out of the six LIA groups thus identified favour site-specific supply networks, while results for the remaining group, LIA5, indicate a multi-site supply network. After combining LIA results with archaeological and historical data, the most likely provenance for the lead materials in LIA5 is Derbyshire, in the British Isles. Both the importation of ready-to-use glazing mixtures and the use of lead from neighbouring mines can now therefore be discounted as plausible hypotheses for the production of the tin-opacified lead-glaze tiles analysed in this study.

HIGHLIGHTS

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

KEYWORDS

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

1. INTRODUCTION

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

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

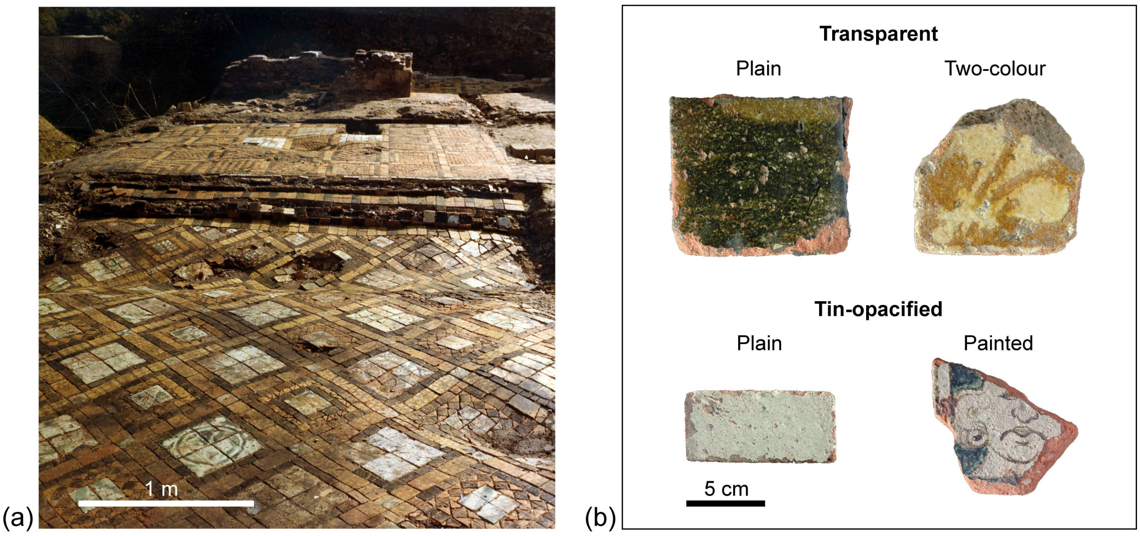

Figure 1. (a) The medieval pavement of Suscinio II, excavated in 1975 (© G. Chapuy, in Métreau et al., 2015). (b) The four types of decoration on the lead-glazed tiles (© L. Métreau, adapted from Métreau et al., 2017).

In medieval times, lead was used for many purposes (Benoit, 1985; Téreygeol, 2009). Galena (PbS)—also known by potters as *alquifoux* (Picon et al., 1995)—and litharge (PbO) were two of the lead compounds frequently used to lower the silica melting point in glazing mixtures (Abel, 1975; Tite et al., 1998). The lead present in archaeomaterials can be characterised by lead isotope analysis (LIA), a technique that has been used since the 1960s (Grögler et al., 1966; Brill and Wampler, 1967; Gale and Stos-Gale, 2000). Studies using LIA on glazes have generally focused on technological choices and specific distribution networks, as well as on lead sources (Mason et al., 1992; Wolf et al., 2003; Stos-Gale, 2004; Iñañez et al., 2010; Walton and Tite, 2010; Shen et al., 2019; Fornacelli et al., 2020; Medeghini et al., 2020; Chang et al., 2021).

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

2. MATERIALS AND METHODS

2.1 Decorated tile pavements

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

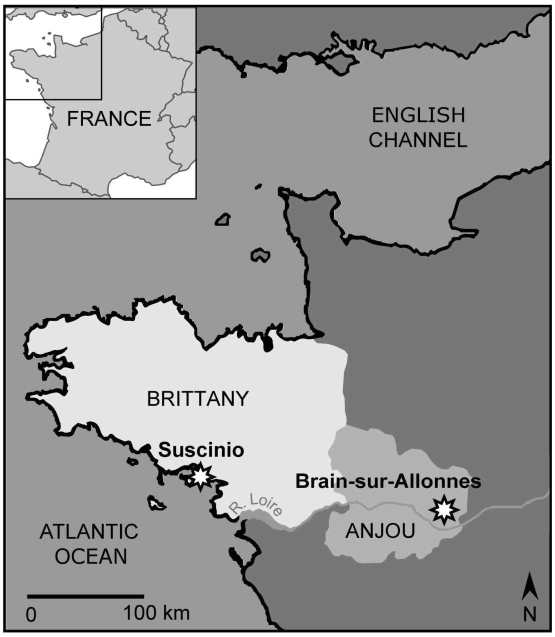

Figure 2. Map of the study area. Stars indicate pavement sites (L. Métreau, adapted from André and Le Pennec, 2003).

2.1.1 Suscinio I

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

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

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

2.1.2 Suscinio II

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

Most of the lead-glazed tiles are transparent (plain, ca. 62%; two-colour, ca. 36%), but some are tin-opacified (plain, ca. 0.5%; painted, ca. 1.5%). The main components of the glazing mixtures were silica and a lead compound (transparent = 50–68 wt% PbO; tin-opacified = 35–45 wt% PbO). Clay was added to the transparent glazing mixture, whereas tin oxide was used to produce the opaque glaze (Métreau et al., 2017). Some of the transparent lead-glazed tiles show signs of tin-oxide contamination, suggesting that both glazing processes took place in the same workshop. Technical flaws on some of the tin-opacified lead-glaze tiles may reflect a poorly controlled firing process. As with Suscinio I, detailed examination of the clay body has identified characteristics similar to those of medieval Breton ceramics. The area around the Château of Suscinio is the most likely source for the non-calcareous clayey raw materials (Appendix A).

The tiles from Suscinio II are stylistically similar to decorated tiles found in northern France (e.g. at the Abbey of St Bertin, St Omer), at a distance of over 600 km (André, 2003).

2.1.3 Brain-sur-Allonnes

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

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

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

2.2. Sample collection and LIA parameters

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

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

DECOR	PAVEMENT			Total
	Suscinio I	**Suscinio II**	**Brain-sur-Allonnes**	**Total**
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**

Table 1. Distribution of high lead glaze samples.

Before sampling, the glaze surface was cleaned with acetone and rinsed with distilled water. As it proved impossible to chip small fragments from the surface with a sharp scalpel, a 2 mm-diameter conical diamond tip (RADIOSPARES) connected to a multifunctional tool with adjustable rotation speed (DREMEL, 300 model) was used to collect powder samples (2–29 mg). The tip was cleaned for five minutes in an ultrasonic bath between samples to avoid contamination. As the glazes are rich in lead (35–68 wt% PbO), a 1 mg sample proved sufficient (Wolf et al., 2003). These powder samples were digested in 100 µl concentrated hydrofluoric acid at the Archéologie, Terre, Histoire et Sociétés Laboratory (ARTEHIS, CNRS, MC, University of Burgundy). The residue after evaporation at 105 °C was dissolved in 50 µl concentrated nitric acid to convert fluoride into nitrate, and evaporated again at 105 °C. Lead isotope ratios were then measured using a Nu Instrument^{TM} MC-ICP-MS and an Apex (TM) desolvating nebulizer at the Laboratory of Isotope Geology (University of Bern). For mass spectrometric analysis, the residue was dissolved in 0.5 M nitric acid, diluted to obtain a signal of ca. 20 pA for ^{208}Pb, and spiked with Tl to correct for instrumental mass fractionation. Mercury interference on mass 204 was corrected by measuring ^{202}Hg. Glaze sample measurements were interspersed with measurements of international standard reference material, NIST SRM 981. The measured Pb isotopic composition of the reference material (Table 2) overlaps with values reported by Rehkämper and Mezger (2000). Measured values are therefore presented without bias correction.

Reference	^{208}Pb/^{204}Pb	^{207}Pb/^{204}Pb	^{206}Pb/^{204}Pb	^{208}Pb/^{206}Pb	^{207}Pb/^{206}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

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

2.3 Lead isotope analysis (LIA) interpretation

The lead isotope results were then compared with an ore reference database, containing 138 datasets and data points (Appendix B), to eliminate the least plausible candidates and to highlight potential sources. The ore reference database was compiled for this study from the available geological and archaeological literature, and from the Oxford archaeological lead isotope database (OXALID), which is freely available online (Stos-Gale and Gale, 2009). It includes lead isotope ratios obtained on a variety of minerals, with lead ore as the priority.

Comparing LIA results for ores and archaeological samples will generally eliminate incompatible sources, but unequivocal identification of provenance is rarely possible (Cattin et al., 2010). The complexity of geological history may result in vast isotopic fields, with great diversity within the same ore body. By contrast, ore bodies many hundreds of kilometres apart may have similar isotopic compositions, making further discrimination impossible. In addition, ore reference data may be limited in number, even for major mining regions. Old mining works are not always dated; mines may not all have been identified; medieval mines may even have been destroyed (Téreygeol, 2009). Lead ores from different sources may have been mixed together, and lead metal was frequently reused over time (Boni et al., 2000). Mixing of sources can only occur during glaze preparation, because the lead in finished glazes is non-recyclable (Mason et al., 1992).

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

3. RESULTS AND DISCUSSION

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

3.1 Lead isotope data

Table 3 presents the range of values for lead isotope ratios, organised by type of glaze and by decorative technique. Based on these results, six groups can be proposed (LIA1 to LIA6), leaving four outliers. Although LIA2 and LIA3 lie close together, they are treated as separate groups because of the slight difference in isotopic values between plain and two-colour transparent lead-glazed tiles. No such logical subdivision is apparent for the data points in LIA5, despite the broad range of isotope ratios in this group.

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

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. Analytical error for lead isotope ratios is shown as 2 standard errors of the mean.

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

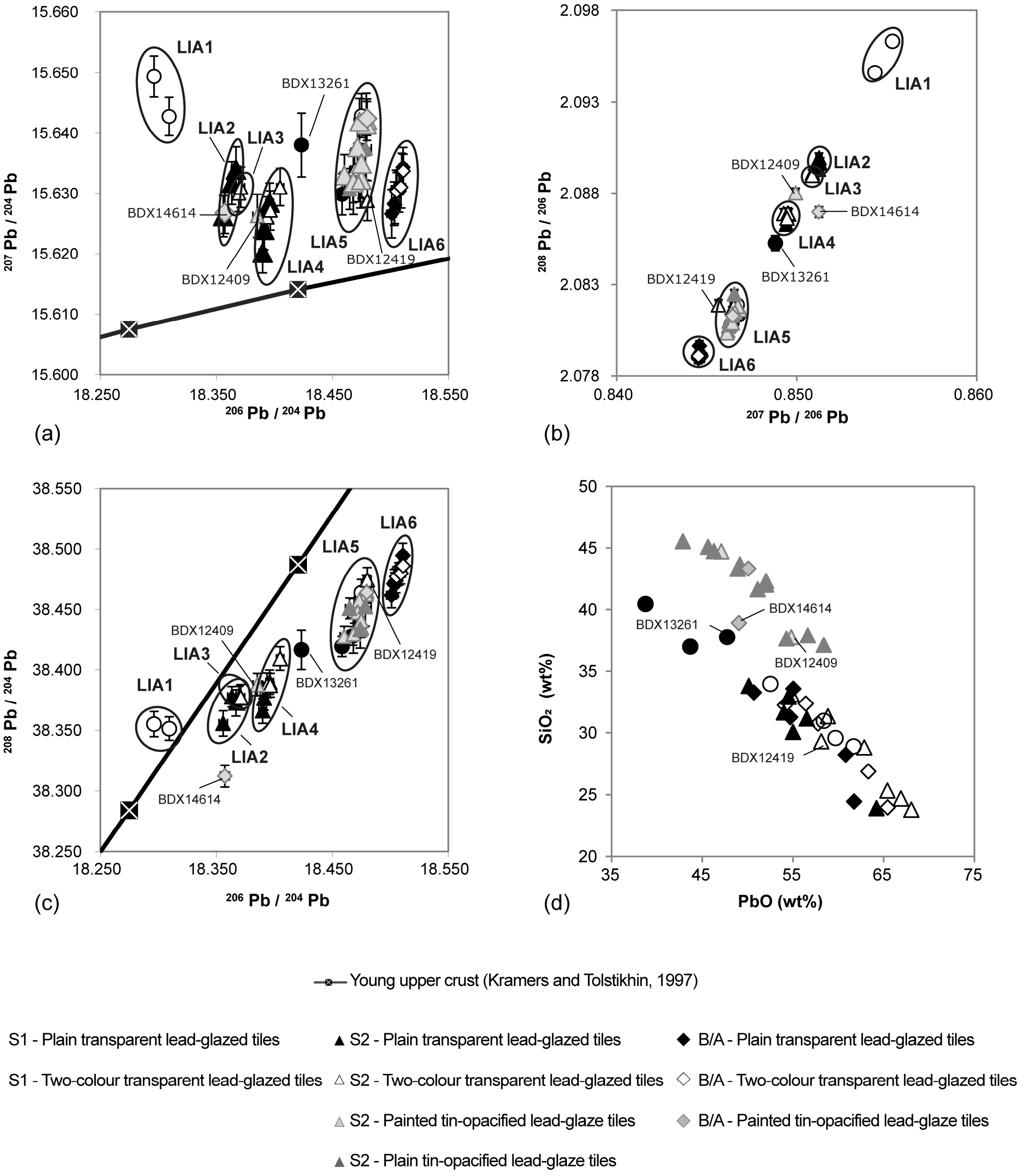

Figure 3. Lead isotope ratios and elemental composition of glaze samples from S1–Suscinio I, S2–Suscinio II, and B/A–Brain-sur-Allonnes: (a) ^{207}Pb/^{204}Pb vs. ^{206}Pb/^{204}Pb; (b) ^{208}Pb/^{206}Pb vs. ^{207}Pb/^{206}Pb; (c) ^{208}Pb/^{204}Pb vs. ^{206}Pb/^{204}Pb; (d) SiO_2 vs. PbO wt% concentration.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

3.2 Organisation of glaze manufacture and lead supply strategies

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

	LIA GROUPS AND PAVEMENTS											
	LIA1	**LIA2**	**LIA3**	**LIA4**	**LIA5**			**LIA6**	**Outliers**			**Total**
DECOR	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**

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

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

Almost all the tin-opacified lead-glaze tiles are found in LIA5, apart from two outliers (BDX12409 from Suscinio II, and BDX14614 from Brain-sur-Allonnes). This group also includes four transparent tiles, one from Suscinio II, and three from the earlier pavement, Suscinio I. The tin-oxide contamination of two transparent glaze samples from Suscinio II, BDX12415 (LIA5) and BDX12419 (the closest outlier to LIA5), suggests that these two-colour tiles must therefore have been produced in the same workshop as the tin-opacified lead-glaze tiles (Métreau et al., 2017). Medieval pottery workshops producing a variety of wares are known to have existed in the late 13th century, in the faubourg Sainte-Barbe, Marseille, France, for example (Thiriot, 1995).

3.3 Potential sources of lead materials

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

	LIA GROUPS AND PAVEMENTS					
	LIA1 S1	**LIA2** S2	**LIA3** S2	**LIA4** S2	**LIA5** S1+S2+B/A	**LIA6** 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	

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

Possible sources present some drawbacks, because the dataset contains outliers, consists of a small number of data points, or covers a broad isotopic field. Plausible sources are more likely candidates, with a narrow isotopic field. Good matches potentially indicate the most likely sources. To evaluate potentially compatible sources for the lead in the glazes, relevant archaeological and historical arguments must be explored. For this purpose, the ore reference database (Appendix B) was organised into six major geographic zones, further subdivided into 34 subzones (Figure 4).

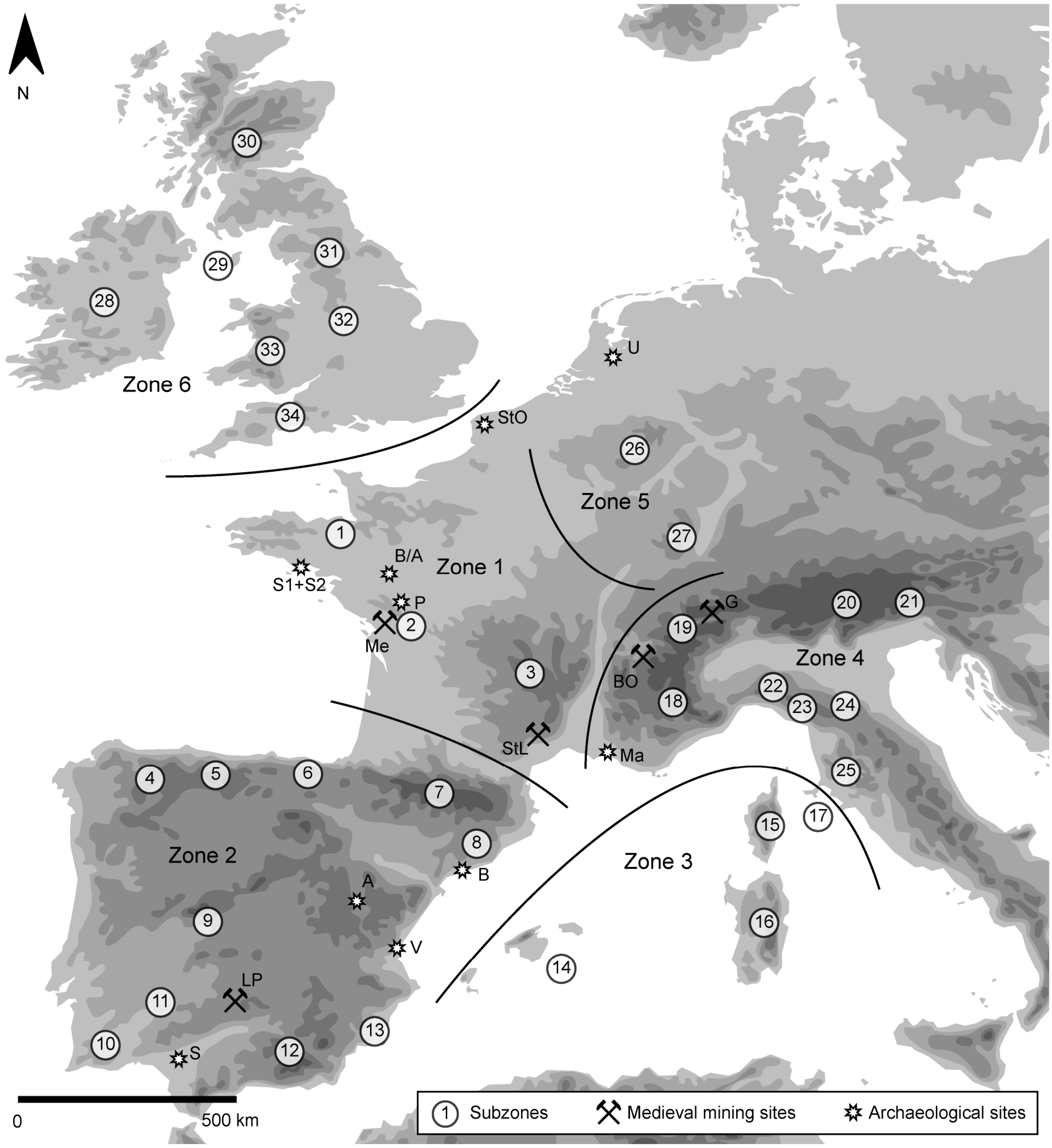

Figure 4. Map of the ore reference database (Appendix B); **circle**, subzone: **Zone 1**: **1**-Armorican Massif, **2**-Seuil du Poitou, **3**-Massif Central; **Zone 2**: **4**-West Asturian Leonese, **5**-Cantabrian, **6**-Cantabrian-Basque, **7**-Pyrenees, **8**-Catalonian Coastal Range, **9**-Central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

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

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

	LIA GROUPS AND PAVEMENTS					
Sources	**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	•

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

3.4 The local or regional source hypothesis (Zone 1)

Archival documents (1384–1386) attest that lead for some of the tin-opacified lead-glaze tiles was mined at a distance of 45 km from the Palace of Poitiers, for the Duke de Berry (Bon, 1992). The local provenance hypothesis for Suscinio I and II would therefore be the Armorican Massif (Subzone 1), where mines for various metals have been exploited since the Bronze Age. Medieval mining activity nevertheless remains difficult to evaluate (Domergue et al., 2006; Le Carlier de Veslud and Jouanet-Aldous, 2015; Le Carlier de Veslud et al., 2017).

Figure 5 presents detailed results for Subzone 1, divided into three categories. Although nine plausible matches are identified (Appendix B), the error margin is greater than the match, making these sources appear less likely. Another potential drawback is that there is often only one data point for each ore body in the Armorican Massif, so that comparison between LIA groups and the database is less robust.

The only plausible neighbouring source (less than 50 km from Suscinio) is Plumelin, a data point that matches LIA4. The lead isotopic data for LIA2, LIA3, and LIA4 (Suscinio II) are in the same range as the ore bodies south-west of Pontivy, at a distance of about 75 km, while La Villeneuve, about 80 km from Suscinio, is the only dataset compatible with LIA5. Huelgoat, a lead mine about 120 km north-west of Suscinio, which may have been exploited earlier than 1540 (Coativy, 2006), is a plausible match for LIA1 (Suscinio I), but with only one data point. Plélauff, a much earlier mine about 100 km away, is a plausible match for LIA2 (Suscinio II), but again with only one data point.

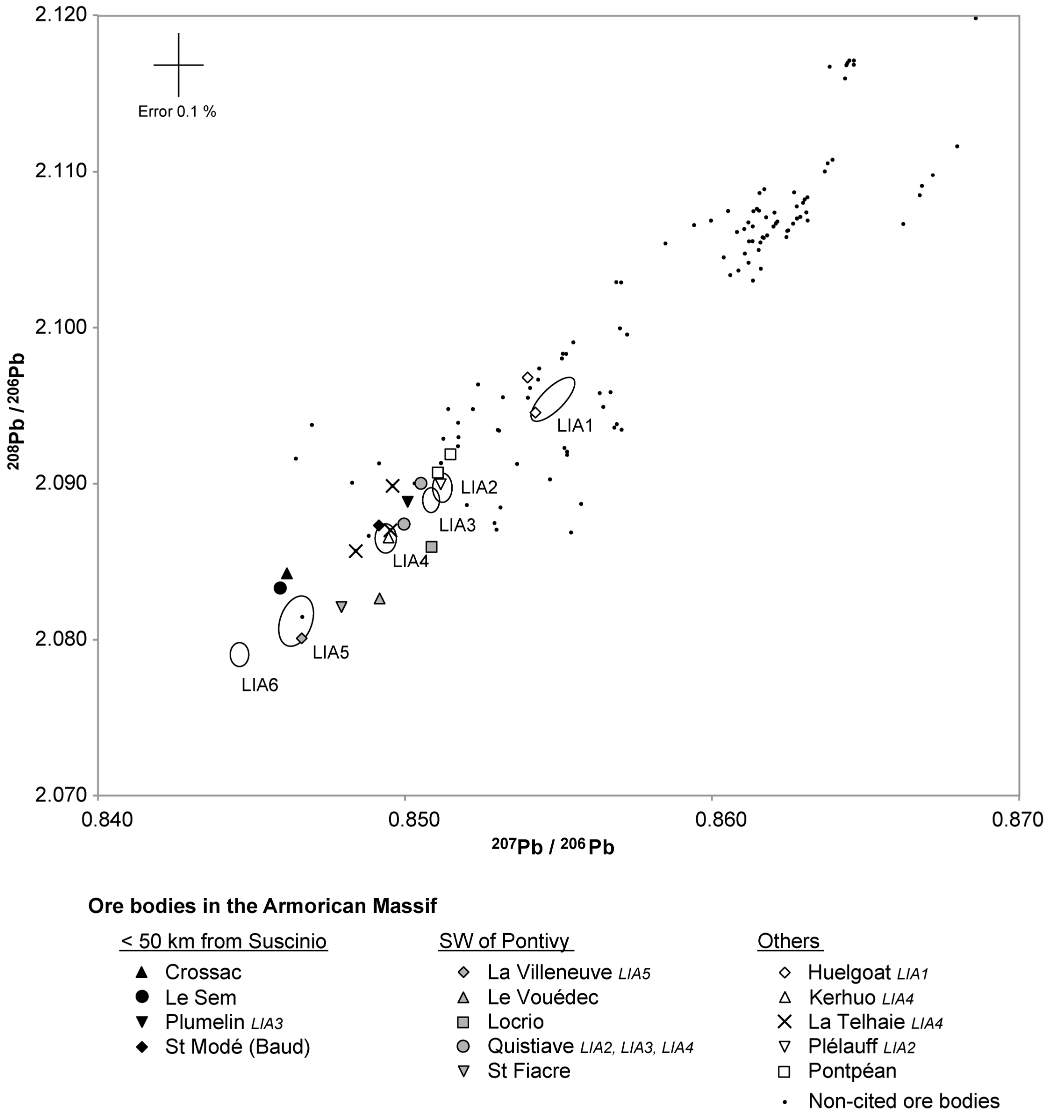

Figure 5. Plot of LIA groups (black ellipses) and ore data from the Armorican Massif (Appendix B).

At a distance of 145 km from Brain-sur-Allonnes, Melle (in Subzone 2) presents a possible match, but this mine was only exploited from the 6th to the 10th century (Téreygeol et al., 2010). Eleven of the transparent lead-glaze samples from Suscino II (LIA2, LIA3, and LIA4) present a plausible match with Saint-Laurent-le-Minier (in Subzone 3), much further south in the Massif Central (Appendix B).

3.5 The imported glazing mixture hypothesis (Zone 2)

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

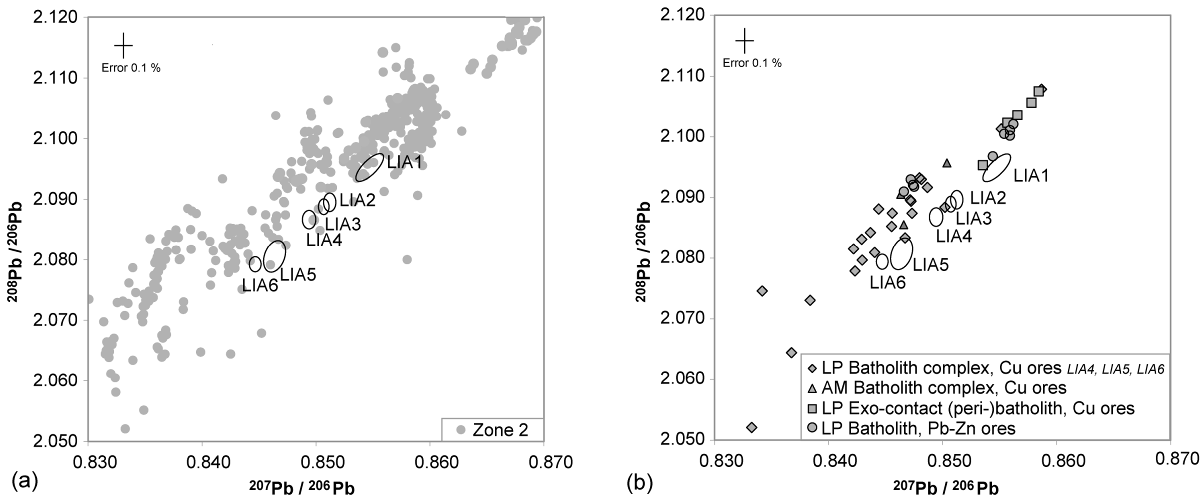

Figure 6 (a). Data for Zone 2, showing LIA groups; (b) Subzone 9, data from Los Pedroches, showing LIA groups (Appendix B).

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

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

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

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

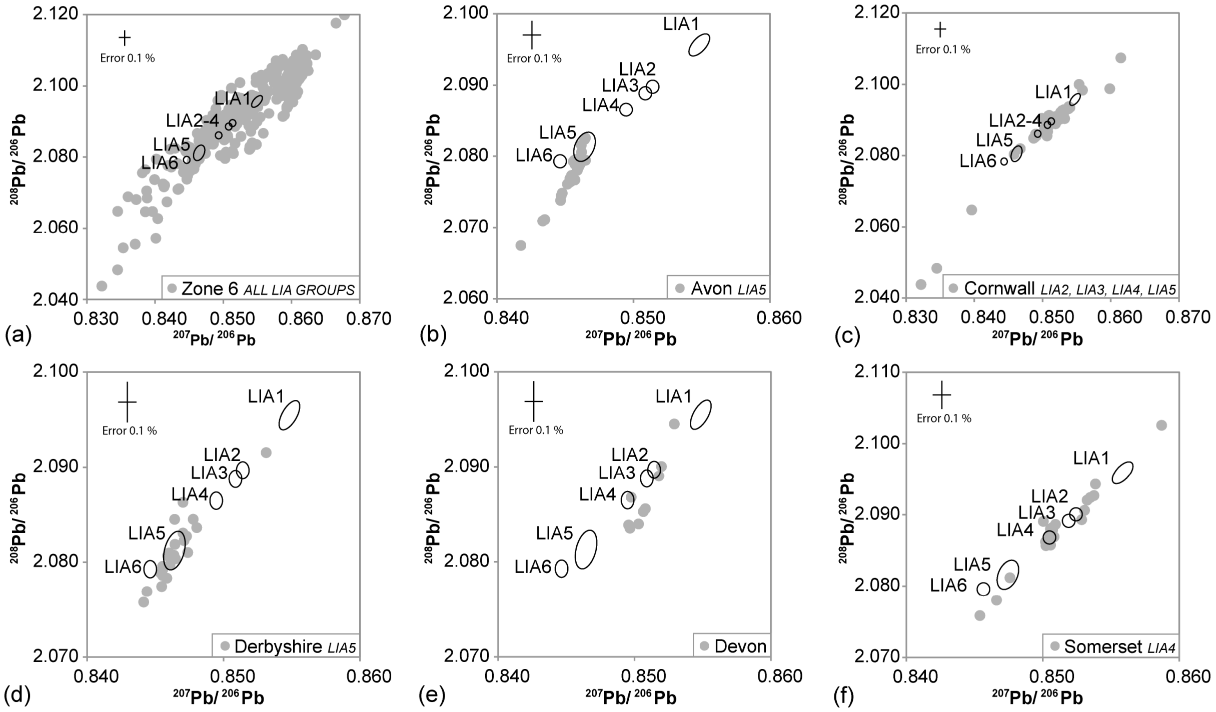

Figure 7. LIA groups (black ellipses) plotted against ore data from Zone 6 (Appendix B).

Results for Zone 6 also indicate two good matches, Avon and Derbyshire, as potential provenance for LIA5 (Appendix B, Figure 7). A British origin for the lead in the tin-opacified glazes challenges two previously explored hypotheses: the opening of mines in neighbouring regions (Section 3.4; Figure 5), and the importation of ready-to-use glazing mixtures from the Iberian Peninsula (Section 3.5; Figure 6). At the turn of the 14th century, Derbyshire was the main lead-producing area in England and Wales, exporting lead to Northern Europe, mainly via Hanseatic trade routes (Blanchard, 2005), and thus appears to be a more historically plausible solution than Avon. Both Suscinio, on the coast of Brittany, and Brain-sur-Allonnes, in the Loire Valley, had easy access to Atlantic trade routes. Stylistic and technical affinities between Suscinio II and contemporaneous Flemish pavements suggest other routes, along the English Channel, to the Abbey of St Bertin, in St Omer, and to sites around Utrecht, where a workshop producing tin-opacified lead-glaze tiles has been identified (de Groot and Pot, 1985).

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

4. CONCLUDING REMARKS AND PERSPECTIVES

This exploratory study will contribute to the growing body of knowledge about the history of decorated tile pavements, and the tin-glazed earthenware technique, or *faïence*. The corpus of 44 tiles, representative of three medieval pavements and four decorative techniques, is large enough to produce meaningful results. Lead isotope analysis of glaze samples, when combined with archaeological and historical evidence, can provide important information about lead supply networks, notably the provenance of the lead in the tin-opacified glazes from Suscinio II and Brain-sur-Allonnes (14th century). The plausible matches between Zone 6 and all the LIA groups, and the good match with English lead for LIA5, together suggest a rather unexpected supply route. As the same lead was used for the tin-opacified glazes, and for some of the transparent lead-glazed tiles from Suscinio, the Derbyshire provenance for LIA5 tends to discount the hypothesis of an imported ready-to-use glazing mixture for the opaque tin glazes. The positive identification of the British Isles as a source of imported lead could be the starting point for future investigations of other medieval tin-opacified lead-glaze products. At other sites, written sources may have been preserved that could provide more information about the motivations underlying the choice of specific types of lead for such products. Identifying the supply routes for both lead and tin is among the ultimate goals for such studies.

ACKNOWLEDGEMENTS

The authors would like to thank the following colleagues, whose assistance and judicious comments made this study possible: Françoise Bechtel, Adelphine Bonneau, Michel Boucher-Bredoux (A.R.E.G.H.A.T), Peter Davey, Joseph Gauthier, Bernard Gratuze, Barbara Guénette-Beck, Fanny Madeline, Fabrice Monna, Jean Rosen, Florian Téreygeol, and Sophie Wolf. The revised manuscript benefitted from constructive suggestions by two anonymous reviewers, whose help is gratefully acknowledged.

FUNDING SOURCES

Financial support for data collection, analysis, and interpretation, together with the writing of the report, and the decision to submit the article for publication was provided by the following bodies: Bordeaux Montaigne University, the French Ministry of Higher Education and Research (MESR), the Centre of Physics Applied to Archaeology at the Institute of Research into Archaeomaterials (IRAMAT-CRP2A, CNRS, Bordeaux Montaigne University), and the Archéologie, Terre, Histoire et Sociétés Laboratory (ARTEHIS, CNRS, MC, University of Burgundy). Publication in English was made possible by a grant from the Fonds Vidal, MSH Dijon USR3516 CNRS-Université de Bourgogne.

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