

A ‘MATCH–NO MATCH’ NUMERICAL AND GRAPHICAL KERNEL DENSITY APPROACH TO INTERPRETING LEAD ISOTOPE SIGNATURES OF ANCIENT ARTEFACTS*

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A new method for interpreting lead isotope ratios of artefacts is presented: a numerical and graphical ‘match–no match’ with possible raw materials. By calculating the definite integral under the kernel density estimate plot of different mining districts, using open-access software and legacy data, the relative probability that an object is made of an ore is indicated. A match with the reference data set may indicate the true origin, while no match indicates an unknown origin, that is, not present in the data set of mineral resources. Likewise, the composite or recycled nature of artefacts can be investigated in a probabilistic manner.

KEYWORDS: PROVENANCE, RECYCLING, MIXING, KERNEL DENSITY ESTIMATE, LEGACY DATA, LEAD, SILVER

INTRODUCTION

The history of man-made materials has always been an essential part of archaeology. Ancient artefacts are direct proxies of how technology evolved. Especially the origin of the raw materials used, their transformation into objects, and the exchange and trade of objects have been topics of investigation. The better we understand the different contexts in which people produced and used objects, the more likely we are to understand the underlying practices of everyday life. Scientific analysis has been used for decades to answer a range of questions in this domain, of which ‘how was it made’, ‘what was it used for’ and ‘where was it made’ are the most obvious.

Man-made materials can be studied using a range of analytical equipment, from the naked eye or a hand lens to very expensive laboratory tools. Each of these will provide different information of contrasting or, in an ideal scenario, complementary accuracy and reliability (Degryse and Bentley 2017). In the analysis of ancient objects, there tends to be a hierarchy of techniques used, starting simply and cheaply with the bulk of the samples, and selecting smaller numbers at each stage to be subjected to analysis by more lengthy and expensive techniques (Shortland and Degryse 2017). Many techniques from the geosciences have been explored for provenancing purposes applied to archaeological materials.

Although providing a substantial amount of information on their own, the simultaneous use of

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several methods provides a more robust framework for interpreting ancient material culture, as overlaps are common when potential sources are compared (Kempe and Harvey 1983). The provenance determination of, for example, stone and ceramics is often accomplished through chemical and mineralogical–petrographic analysis (Degryse and Braekmans 2014). More generally, to study the source of raw materials for ancient craft production, in particular trace element profiles and isotope ratio analysis, has proven to be indispensable (Degryse and Bentley 2017). Nearly every element of the periodic system of the elements has been used at some point to compare the typical composition of the source materials with that of archaeological artefacts, or to assign compositional groupings to archaeological objects.

Depending on the nature of the materials analysed (e.g., clay compared with ceramic, sand with glass, ore with metal, etc.), certain elements are chosen for their occurrence as major or trace elements and for the nature of the geological–mineralogical information they provide. However, smelting, melting or mixing may have a significant impact on the concentration or even the presence of many elements, and the relationship between the mineral raw material and artefact may be obscured. Such issues have been addressed using heavy, often radiogenic, isotopes, of which lead (Pb) isotopes are the most (in)famous example. Some elements, such as Pb, show natural variations in their isotopic composition as a consequence of one or more of their isotopes being the end product of the decay of naturally occurring, long-lived radionuclides. The isotopic composition of such elements varies depending on the initial parent/daughter ratio and the time elapsed since the formation of the resource. Other elements display natural variations in their isotopic composition as a consequence of kinetic and/or thermodynamic isotope fractionation effects (but are not relevant to this paper). Moreover, especially isotopes with relatively high masses and low internal relative mass differences, such as Pb, are not fractionated between ore and artefact during technical processing, even at the temperatures attained in pre-industrial pyrotechnology. A transformation such as smelting an ore into a metal has no effect (or too little an effect to be measurable) on the Pb isotope ratio of any of the materials involved. Except for a scenario in which raw materials are mixed or contaminated, the isotopic composition of an artefact will thus be identical, within analytical error, to the raw material from which it was derived. The signatures of different raw materials may differ, however, based on their geological age and origin (Brill and Wampler 1965). Lead isotope ratio analysis (LIA) has been used to trace the raw materials for craft production since the 1960s, in particular to investigate the provenance of lead, silver and bronze, mainly from the Mediterranean Bronze Age. The technique has subsequently been applied to the study of a range of inorganic archaeological materials (e.g., Degryse 2012).

The use of LIA has not been without controversy, however. One criticism of its use in archaeology has been the tendency to use isotope ratios as simple numbers that characterize an ore source, while these ratios evolve according to laws of radioactive decay and geochemistry (Albarède *et al.* 2012). Linear trends or even straight lines in isotope biplots involving a common denominator represent either *isochrons* recording the geological age of a set of cogenetic samples or binary mixing lines reflecting mixtures of two end components in different proportions (Faure 1986). Some attempts have been made to use such isochrons to calculate the age of parent deposits of metal artefacts, but although this is standard practice in geochemistry, the application to archaeological objects has proven much more complicated as anthropogenic mixing and recycling of commodities from multiple deposits are possible, providing invalid ages. Overall, there is an additional layer of complexity in archaeological objects, which is not accounted for in conventional geological approaches. Pb isotopes may be better understood, and limitations to the technique overcome, by asking different questions than merely provenance, for example: Do two artefacts derive from the same source or not? How do sources vary through time and

space? Bray and Pollard (2012), Pollard and Bray (2015) and Bray *et al.* (2015) have proposed a method that outlines the dynamic nature of metal in circulation. Rather than extracting a precise provenance from compositional information, the method determines the timing and origin of the new input of fresh materials into a system. In this way, Pb isotope data can provide a powerful tool with which to understand the circulation of materials, focusing on detecting change in the archaeological record. A different provenance then becomes only one option to explain chemical changes, next to and equivalent with other options such as the input and mixing of fresh materials, changes in technological practice or an increased recycling of materials (Pollard 2018).

Pollard (2009) summarized three key limitations when using Pb isotopic analysis for provenance studies in archaeology: (1) there is usually an overlap in Pb isotope ratios from geographically geologically distinct sources, making a clear chemical distinction between those areas difficult; (2) there is usually a large range in isotope signature within a single source area; and (3) Pb isotope ratios from a given ore field usually show a non-normal distribution of individual analyses, making statistical processing complex, or at least not straightforward. In effect, Pb isotopic signatures of one source area can be 'broad', and isotope ratios from distinct ore districts may considerably overlap, or even be difficult to define (e.g., see the broad similarity of Pb isotope ratios across Europe in Pollard 2018, ch. 6, fig. 1, pp. 145–148). Baxter *et al.* (2000) stated that the assumption of normality of Pb isotope data leads to the use of misleading graphical presentations of ore fields in Pb isotope ratio biplots (with sharp boundaries that, in effect, have little meaning), and to the very real danger of incorrect probability calculations based on such data sets. The use of kernel density estimates (KDEs) was advocated in this matter, though it was indicated that larger sample sizes of analysis of ore sources were necessary (i.e., several dozen), and that technical problems with regards the choice of smoothing and orientation parameters exist (Baxter *et al.* 1997, 2000).

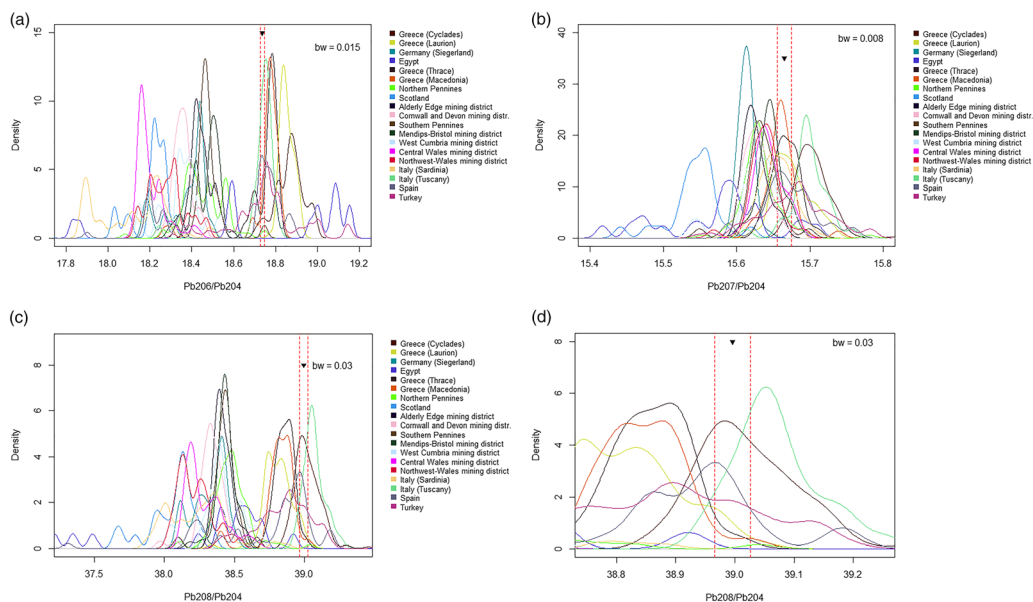


Figure 1 Kernel density estimate (KDE) distributions, calculated from legacy data, for all mining districts included in the study, presented for lead (Pb) isotope ratios (a) $^{206}\text{Pb}/^{204}\text{Pb}$, (b) $^{207}\text{Pb}/^{204}\text{Pb}$ and (c) $^{208}\text{Pb}/^{204}\text{Pb}$, next to (d) a detail of (c). Triangles indicate the isotope ratio of the first artefact studied, a Roman lead ingot (Trincherini *et al.* 2009); and red dashed lines indicate this value $-\sigma$ and σ . [Colour figure can be viewed at wileyonlinelibrary.com]

However, while the use of a simple graphical inspection of Pb isotope ratio biplots has been advocated for assessing provenance (Scaife *et al.* 1996), an approach most used in archaeological Pb isotope studies until today, KDE approaches provide a valid alternative for data representation and statistical calculations. Nevertheless, though this approach has been fully integrated in geographical information systems, this approach is currently still rare in chemistry and archaeological science in general (e.g., Bronk *et al.* 2015; Bidegaray and Pollard 2018), and even more so in Pb isotope studies (Beardah and Baxter 1999; Scaife *et al.* 1999; Hsu *et al.* 2018), this likely because of the complex calculations and programming involved.

While issues of accuracy and precision of instrumentation and fractionation of Pb isotopes in technical processing have been resolved, either by extensive testing or by the technological developments in new instrumentation such as multi-collector (MC) inductively coupled plasma-mass spectrometry (ICP-MS) (Degryse 2012), the relevance of sampling strategies, the definition of ore fields and the presentation of data still demand effort. The goal of the present paper is to present an innovative method to interpret Pb isotope ratio data in archaeology, based on the three fundamental ratios measured in laboratory studies ($^{208}\text{Pb}/^{204}\text{Pb}$, $^{207}\text{Pb}/^{204}\text{Pb}$ and $^{206}\text{Pb}/^{204}\text{Pb}$), and interpreting these with kernel density techniques, moving towards a more probabilistic model. This approach provides a numerical and graphical ‘match–no match’ with the isotopic signature of possible raw materials, taken from legacy data, using open-access software.

METHODOLOGY

Starting from a database of Pb isotope ratios measured on lead (mostly galena) ores, published over the last five decades and organized by mining district rather than modern geographical subdivisions (see Appendix 1 in the additional supporting information for sources), a kernel density plot is drawn per ratio measured ($^{208}\text{Pb}/^{204}\text{Pb}$, $^{207}\text{Pb}/^{204}\text{Pb}$ and $^{206}\text{Pb}/^{204}\text{Pb}$). A KDE is a non-parametric way to estimate the probability density function of a random, continuous variable, so that inferences about the population can be made, based on a finite data sample. A key capability of KDEs is their ability to test the degree of overlap between distributions, answering questions on (dis)similarities between assemblages (e.g., Bronk *et al.* 2015). KDEs do not assume normal distribution of the data and produce more smooth distributions compared with conventional histograms (which are very dependent on chosen bin width and start/end points; Baxter *et al.* 1997). Such a KDE is built up by drawing a kernel, that is, a non-negative function that integrates to 1, on each of the data points. These kernels are then summed to make the kernel density plot. The estimated density plot is thus dependent on the selected function for the kernels and the bandwidth of the kernels (for details of calculations, see Bronk *et al.* 2015; and Hsu *et al.* 2018). The choice of bandwidth is crucial for the shape of the KDE and subsequent interpretations, finding a good balance between over-smoothing the data and hiding detail versus showing discontinuities. There is, however, no well-defined method to establish the optimal kernel density bandwidth for multidimensional data. According to Zhang *et al.* (2006), a commonly used approximation for the optimal bandwidth of the multidimensional kernel is:

$$h = \{4/(d + 2)n\}^{1/(d+4)}$$

where d is the number of dimensions (parameters measured); and n is the number of samples in the data set. This estimate approaches optimality as the data approach multivariate normality. In a first proof-of-principle application, the chosen kernel here is a normal distribution approximation or Gaussian approximation, also called Silverman’s (1986) rule of thumb, and a uniform bandwidth

was chosen for all regions per Pb isotope ratio. The bandwidth chosen, however, will need to be further evaluated. For each mining district, one plot per ratio is made based on all ore data available.

The measured LIA of artefacts in an assemblage can then be compared with the kernel density plots. The sum of probability under a normalized density curve is always equal to 1, or 100%. Since probability is the area under the curve, we can then specify a range to calculate the probability within this range. The relative probability that the isotope values of the measurement(s) lie in the interval of 10 times the standard deviation (SD) for all possible sources is calculated by integrating each density function over that interval. The results are shown in bar plots per region, and these relative probabilities are only to be compared within the sample studied. The region with the highest relative percentages on the three isotope ratios is the most probable source area. If no match is found, either the source area is unknown, that is, not incorporated in the reference data set of possible mineral resources used, or the Pb isotopic composition of the artefact or assemblage is a mixture of several sources. A match with an ore source can indicate a true origin of the artefact, or can be a false match where a mixture of sources gives an accidental correspondence to another ore source. All calculations and plots are generated using RStudio, a free and open-source integrated development environment for R, a programming language for statistical computing and graphics. For the R code used for the calculations in this paper, see Appendix 2 in the additional supporting information.

For the proof-of-principle application presented here, lead and silver artefacts are examined. While the method is applicable to any material with Pb isotopic data, for example, copper alloys or glass, the metals looked at here have the advantage that apart from mixing of ore sources or recycling of objects, the Pb present in the artefact will originate from a single ore only. Conversely, the origin of the Pb present in glass and copper alloy can be debated, possibly originating from several raw materials such as the silica source and (de)colorant, or the copper and tin ore used. This would complicate matters for the sake of this demonstration.

Only silver and lead mining districts with data available from 20 or more ore samples are included. For each mining district, one KDE plot per ratio is made, based on all ore data available. Lines 1–59 of the R code draw the $^{206}\text{Pb}/^{204}\text{Pb}$ plot; lines 61–118 the $^{207}\text{Pb}/^{204}\text{Pb}$ plot; and lines 120–176 the $^{208}\text{Pb}/^{204}\text{Pb}$ plot. First, the kernel density plot is drawn with the `plot(density())` function (lines 3–5) and the other KDEs are added with the `lines(density())` function (lines 24–59). The other lines of code add the legend and provide layout. The result is shown in Figure 1. While the combined plots are difficult to read as such, they do show that every mining district has a distinct kernel density signature, moreover one that can be mathematically calculated and separated.

RESULTS

To demonstrate the method proposed, a random artefact was chosen with a known provenance from one of the mining areas discussed in this data set: a Roman ingot that was epigraphically identified as manufactured in the Cartagena ore district in Spain, analysed by Trincherini *et al.* (2009). The three measured Pb isotope ratios and their SDs are given in Table 1; the position of the object is added to the KDE plots in Figure 1.

To calculate the relative probability that the artefact has its origin in a certain mining district, the area under the KDE of that district and delimited by the artefacts isotope ratio $\pm 10 \times \text{SD}$ (Fig. 1, d) is calculated with the definite integral of the KDE. This is done for every mining district separately.

Table 1 Lead (Pb) isotope ratios for a lead ingot with Spanish origin (after Trincherini et al. 2009)

$^{206}\text{Pb}/^{204}\text{Pb}$	$\sigma^{206}\text{Pb}/^{204}\text{Pb}$	$^{207}\text{Pb}/^{204}\text{Pb}$	$\sigma^{207}\text{Pb}/^{204}\text{Pb}$	$^{208}\text{Pb}/^{204}\text{Pb}$	$\sigma^{208}\text{Pb}/^{204}\text{Pb}$
18.737	0.001	15.665	0.001	38.996	0.003

In the R code, these probabilities are calculated for the $^{206}\text{Pb}/^{204}\text{Pb}$ KDEs in lines 179–255, for the $^{207}\text{Pb}/^{204}\text{Pb}$ KDEs in lines 257–330, and for the $^{208}\text{Pb}/^{204}\text{Pb}$ KDEs in lines 332–405, with the `integrate.xy` function. The `try()` if(‘try-error’) function makes sure that if the SD interval does not intersect with the KDE, the probability will be 0. The obtained probability percentages are visualized in the bar charts in Figure 2.

The combined relative probabilities are represented in Figure 2 (d). For the percentages for the particular object under study, see Appendix 3 in the additional supporting information. These indicate the relative probability galena has been mined in the Cyclades, Thrace, Macedonia, Tuscany, Spain or Turkey. The regions where one or more of the values are (close to) 0 are deleted from the list. Moreover, this can also be clearly seen on the bar chart in Figure 2 (d). Here it is clear that Spain has the highest combined relative probability to be the origin of the ore used

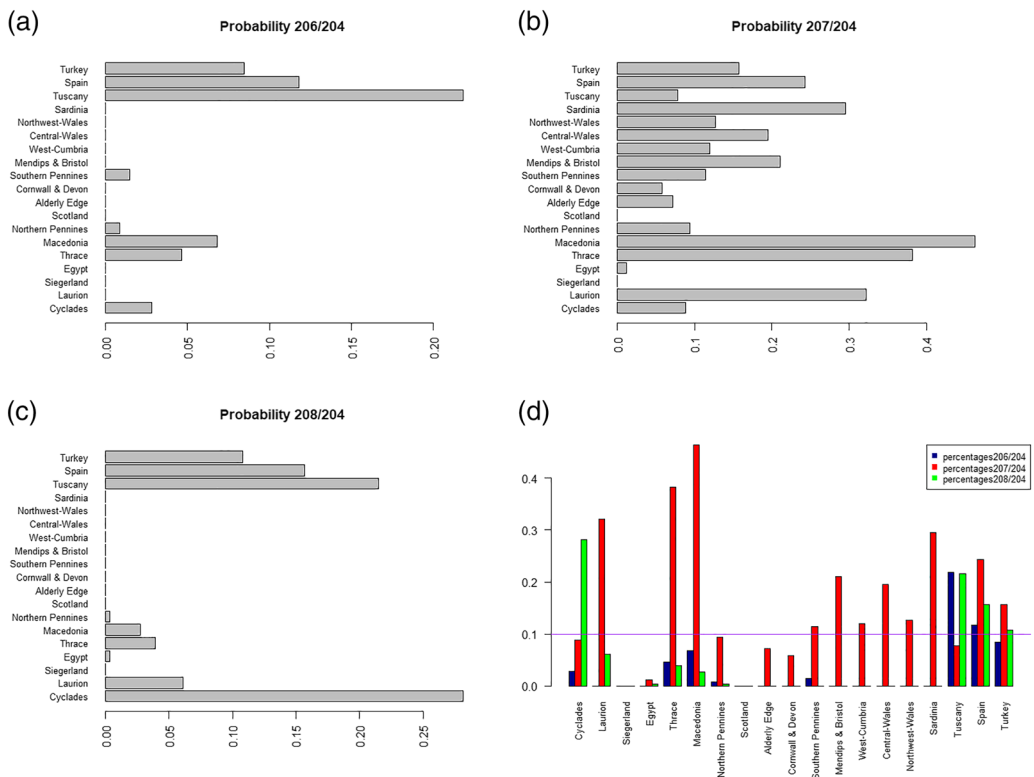


Figure 2 Bar charts with relative probabilities for lead (Pb) isotope ratios (a) $^{206}\text{Pb}/^{204}\text{Pb}$, (b) $^{207}\text{Pb}/^{204}\text{Pb}$ and (c) $^{208}\text{Pb}/^{204}\text{Pb}$ per mining district, and (d) the combined relative probability percentages for each mining district, for a Roman lead ingot from Cartagena (Trincherini et al. 2009). [Colour figure can be viewed at wileyonlinelibrary.com]

to manufacture the ingot. Although it could be argued that the different Spanish mining districts should have separated KDEs, there are not yet enough data in this data set to draw two significant KDEs and it is not strictly necessary due to the properties of a KDE. Two different districts would give two maxima, which is the case for the Spanish data for the $^{206}\text{Pb}/^{204}\text{Pb}$ plot and the $^{208}\text{Pb}/^{204}\text{Pb}$.

In a second case study, following an identical process, relative probabilities were calculated for the Pb isotope ratios (Table 2) of a lead ingot with German origin, analysed by Bode *et al.* (2009). They matched the ingot with the Brilon mining district and the northern Eifel district with standard biplots. Epigraphic studies also indicate that the origin of this ingot is in Germany (Rothenhöfer 2003; Hanel and Rothenhöfer 2005). The bar plot in Figure 3 indeed clearly shows the German origin of the ingot.

In a third case study, the Pb isotopic composition of 88 samples of the Berthouville silver collection, a Roman silver hoard discovered through ploughing in 1830 in Normandy, France, and kept at the Bibliothèque Nationale in Paris (Degryse and Brems 2014; Lapatin 2014) is used to test this method further. For each of the samples analysed, the same calculations were made as in the former case studies. For the resulting bar charts, see appendix 4 in the additional supporting information. Different groups of objects can be distinguished. The first and largest

Table 2 Lead (Pb) isotope ratios for a lead ingot with German origin (after Bode *et al.* 2009)

$^{206}\text{Pb}/^{204}\text{Pb}$	$\sigma^{206}\text{Pb}/^{204}\text{Pb}$	$^{207}\text{Pb}/^{204}\text{Pb}$	$\sigma^{207}\text{Pb}/^{204}\text{Pb}$	$^{208}\text{Pb}/^{204}\text{Pb}$	$\sigma^{208}\text{Pb}/^{204}\text{Pb}$
18.164	0.004	15.602	0.002	38.108	0.007

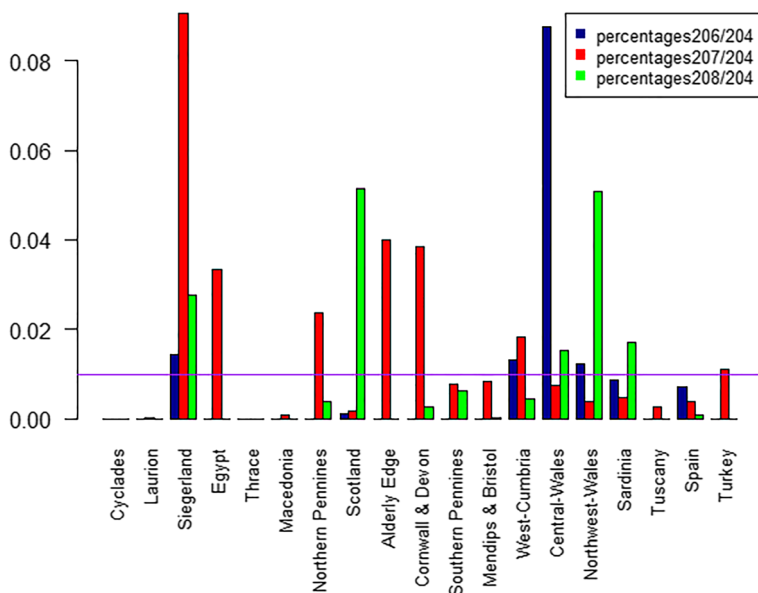


Figure 3 Combined relative probability percentages for each mining district of a lead ingot with a German origin (Bode *et al.* 2009). [Colour figure can be viewed at wileyonlinelibrary.com]

group comprises the samples that clearly indicate an origin in the south or the centre of Britain (Fig. 4, a). In a second group we find the samples that also point to a British origin, but corresponding to a different source region, mainly the Northern Pennines (Fig. 4, b). A total of 10 samples do not show a clear provenance, which could be the result of the use of an unknown ore (or a resource not mentioned in the reference database used). However, perhaps more likely, this lack of correspondence can also be an indication for mixing and/or recycling of the silver used. The 10 samples can be subdivided visually into two groups that have a clearly different signature/origin, for instance two different batches of mixed/recycled raw materials (Fig. 4, c). Outside of these groups, one exceptional sample (71.1) shows a likely Greek origin of the silver used (Fig. 4, d).

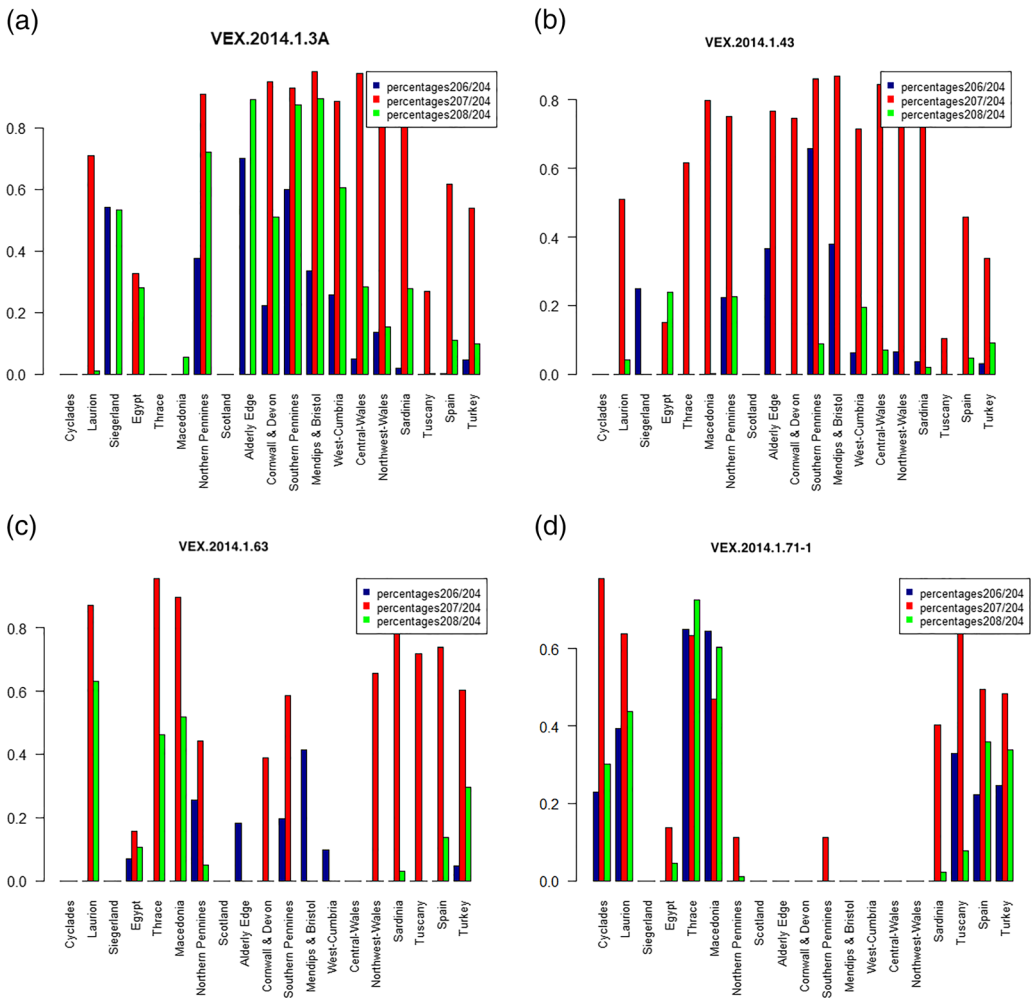


Figure 4 Combined relative probability percentages for each mining district of the Berthouville silver samples with (a) a likely origin in south-central Britain (first group), (b) a likely origin in the northern Pennines (second group), (c) a likely mixed and/or recycled origin and (d) a likely Greek origin. [Colour figure can be viewed at wileyonlinelibrary.com]

CONCLUSIONS

This study illustrates a new method to interpret Pb isotope analysis of archaeological artefacts using KDEs per mining district to provide a visual and a mathematical probabilistic approach to the question of not only provenance but also mixing and recycling of resources. This is performed by calculating the definite integral under different KDEs over an interval of $10 \times$ SD of the measured Pb isotope ratios, thus calculating the relative probability that the researched sample has its origin in each of the mining districts. In the first two proof-of-principle case studies, the method gives a valid result. The third case study, looking into the Pb isotope data from the Berthouville treasure, confirms the validity of this method and touches on a way to identify mixed or recycled raw materials.

The first exploration of this method looks promising, though the parameters used to draw the KDEs and calculate the relative probabilities need to be evaluated. In addition, results are (as always) dependent on the mining districts that have been added to the reference data set of mines and ore deposits. Nevertheless, this method offers a more correct statistical approach to Pb isotope ratios, as it is not dependent on the normal distribution of data sets. This approach also offers a graphical way in which to assess the correspondence of an artefact to several mining districts, such as with the use of biplots, while being based on more sound mathematical grounds to make such a comparison. Moreover, mixing of metals and possible recycling can be inferred, and approached in more quantitative ways, as can the composition of materials with possible multiple lead sources in their raw material mixture.

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SUPPORTING INFORMATION

Additional supporting information may be found online in the Supporting Information section at the end of the article.

Data S1. Appendix 1: Sources.

Data S2. Appendix 2: R Code.

Data S3. Appendix 3: Probability matrix for the Spanish ingot.

Data S4. Appendix 4: Bar charts of the Berthouville samples divided into four groups.