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# Provenance studies using lead isotopy: contribution of the consideration of geological contexts in archaeological databases

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**Abstract** – The identification of mineral supply sources and trade routes are at the heart of the archaeological issues. The tracing of sources of metal production *via* lead isotopy has been used since the 1980s to identify the deposits from which the metal constituting the archaeological objects came. Such studies are based on mineral signature repositories and archaeologists have thus built up databases containing thousands of ore deposit analyses. The databases, however, only very rarely include geological information and are limited to geographic information. But considering only geographical data leads to many limitations of studies, including the overlapping of signatures between remote regions. This problem could nevertheless be circumvented by taking into account precise ore deposit data that enables to think in terms of restricted mineralized subsets. We illustrate this through the example of data collected in the Alps by Marcoux (1986) and Nimis *et al.* (2012). Taking into account geological data (and more specifically, geological data) could thus considerably improve the accuracy of provenance interpretations and enable multivariate statistical processing to be carried out (these statistical treatments are inconclusive if they are carried out on purely geographical data). However, remains the problem of the thousands of ore analyses carried out without having defined their mineralization context. Although still imperfect (some contexts are not individualized), the use of a statistical treatment could nevertheless be used to identify the geological contexts.

**Keywords:** lead isotopy / provenance studies / archaeology / databases / statistics / geology

**Résumé** – **Traçage de provenances par isotopie du plomb: apport de la prise en compte des contextes géologiques dans les bases de données archéologiques.** L'identification des sources d'approvisionnement en minerais et des circuits d'échanges est au cœur des problématiques archéologiques. Le traçage de sources de productions métalliques *via* l'isotopie du plomb est ainsi utilisé depuis les années 1980 pour identifier les gisements dont est issu le métal composant les objets archéologiques. De telles études s'appuient sur des référentiels de signatures de minerais et les archéologues ont ainsi constitué des bases de données, riches de milliers d'analyses de gisements. Cependant, ces bases de données n'intègrent que très rarement des informations géologiques et se limitent à une information d'ordre géographique. Or, ne considérer que des données géographiques conduit à de nombreuses limitations des études dont l'*overlaps* de signatures entre des régions éloignées. Ce problème pourrait néanmoins être contourné en prenant en compte des données de gisements précises permettant de réfléchir en termes de sous-ensembles minéralisés restreints. Nous illustrons ceci à travers l'exemple de données récoltées dans les Alpes par Marcoux (1986) et Nimis *et al.* (2012). La prise en compte de données géologique (et plus précisément géologiques) pourrait ainsi permettre d'améliorer considérablement la précision des interprétations de provenance et de proposer des traitements statistiques multivariées (ces mêmes traitements étant peu concluants s'ils sont réalisés sur des données uniquement géographiques). Cependant, reste le problème des milliers d'analyses de minerais effectuées sans avoir défini leur contexte de minéralisation. Bien qu'encore imparfaite (certains contextes ne s'individualisant pas), recourir à un traitement statistique pourrait néanmoins permettre de retrouver les contextes géologiques.

**Mots clés** : isotopie du plomb / traçage de sources de production / archéologie / bases de données / statistiques / géologie

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## 1 Introduction

Lead has four stable isotopes: the isotope  $^{204}\text{Pb}$  which is primitive;  $^{206}\text{Pb}$ ,  $^{207}\text{Pb}$  and  $^{208}\text{Pb}$  which are radiogenic<sup>1</sup>. These radiogenic isotopes are the final forms of radioactive decay of Uranium and Thorium:  $^{235}\text{U} \Rightarrow ^{207}\text{Pb}$ ;  $^{238}\text{U} \Rightarrow ^{206}\text{Pb}$ ;  $^{232}\text{Th} \Rightarrow ^{208}\text{Pb}$ . However, metallic copper, silver artifacts, but also pigments, contain lead in very small quantities. This presence of lead in the form of traces comes from the ore that was used to shape the object and its (lead) isotopic signature can be researched. As there is no lead fractionation during the metallurgical process (Cui and Wu, 2011; Pernicka, 2014), the isotopic lead signature of an artifact is comparable to that of the ore that was used to shape it, and can allow to identify the ore deposit used to craft the archaeological object regardless of the period of manufacture<sup>2</sup>. Lead isotopic studies have so been used since the late 1980s to identify the source of the metal used to produce an artifact found in an archaeological context. This type of study is commonly referred to as provenance studies and lead isotopic analyses are an important part of the analyses carried out in the archaeological studies.

The importance of tracing provenance is such that thousands of mineral analyses have been carried out as part of archaeological studies, including the Oxalid<sup>3</sup> database (the database includes analyses performed on protohistoric artefacts and minerals from the Mediterranean, British Isles and Balkans) or the database compiled by B. Scaife<sup>4</sup> (including analyses of ores from Egypt, Levant, Maghreb). Nevertheless, although archaeologists recognise that the isotopic signature of lead is specific to each mineralization (Stos-Gale, 1992), they do not take into account the geological contexts in their provenance studies. Indeed, as already pointed out by Ixer (1999), Guénette-Beck and Serneels (2010) and Baron *et al.* (2014), archaeological databases contain only geographical information and lack geological<sup>5</sup> information. The nature of the minerals analyzed is not always indicated and the complexity of the deposits from which they are derived is generally poorly known. In addition, ores are classified by large geographical areas in which mineralization of very different ages and types can be found. Finally, some samples sometimes even not correspond to ore deposit: they are host rocks completely disconnected from artifacts (chromites in Oxalid data base for example).

In addition, provenance studies generally do not take into account data collected by geologists. Used as metallogenic tracers<sup>6</sup>, the “geological” data are often considered to be insufficiently sampled to be used to answer the question of the origin of archaeological artefacts. Indeed, geologists carry few measurements per mine but perform measurements on several

deposits affected by the same mineralizing episode. On the contrary, archaeologists favour numerous measurements per mine in order to obtain the most complete possible spectrum of the signature of the mine being investigated.

However, could lead isotopic measurements from geological research be integrated into archaeological provenance studies? What could be the contribution of taking into account geological contexts (and not just a geographical origin) for provenance studies? Could it improve the tracing of production sources?

## 2 Archaeological provenance studies

The study of provenances *via* lead isotopy is a complex operation, requiring many factors to be taken into account.

First of all, defining the geographical origin of the metals making up an artifact requires knowing the chemical composition of the artifacts. The origin of an object made of a copper alloy can only be traced through the use of lead isotopes if the quantity of lead in the artefact is known. While the thresholds affecting signatures are as yet poorly known, it is nevertheless clear that if lead is added as an alloying element, the signature of the lead deposit blurs that of the copper deposit.

It is also important to consider the period in which the artifact was made, the sources known to have been used at the time, and the extent of trade networks, as these are valuable clues for deciding between several possible sources of production (the artifact typology can also be used to obtain valuable information about the area of origin of the objects).

However, even with these precautions, the use of lead isotopes as the only source tracer is regularly criticized (Baron and Coustures, 2018) because of two main factors: sources mixing and statistical overlaps.

(i) Possible mixtures of sources and recycling may occur during the manufacture of the objects. The mixtures can be of different types:

- Mixtures of metals from different deposits can be made, for example, by mixing ores from different sites to obtain an alloy with specific properties;
- Mixture signatures can also be the result of recycling: a metallic object (broken for example) can be remelted. Metallurgists can melt copper from a broken object and mix it with copper from a different origin to create a new object with a signature corresponding to the average of the two deposits<sup>7</sup> (Longman *et al.*, 2018).

(ii) Problems of statistical discrimination of signatures from certain metalliferous regions: the tracing of production sources is often hampered by the fact that statistical discrimination of geochemical signatures is sometimes impossible because some minerals from distant regions have a close lead isotopic signature. These overlapping signatures of remote mining districts are generally referred to as signature overlaps. For example, the lead isotopic signatures of the Taurus Mountains in Turkey are very similar to those of Cyprus (Yener *et al.*, 1991) but also to the Aegean region

<sup>1</sup> There are other isotopes of Pb, unstable and of a too short lifetime to be of interest for the study of geological events.

<sup>2</sup> This research is always linked to a coherent geographical context: American ores cannot have been used to make Roman objects. The deposit areas inserted in the isotope signature comparison model thus vary according to the framework of the different studies.

<sup>3</sup> <http://oxalid.arch.ox.ac.uk/The%20Database/TheDatabase.htm>.

<sup>4</sup> <http://brettscaife.net/lead/data/index.html>.

<sup>5</sup> Characterization of ore deposits.

<sup>6</sup> To study the genesis of metal ore deposits.

<sup>7</sup> If the origins are identical, the signature will be identical.

(Eshel *et al.*, 2019). Consequently, in some studies, several possibilities of metal supply sources can be proposed without it being possible to decide in favour of one or the other without any other elements of distinction. Typological and/or geochemical data (trace elements) of the artifacts are then sometimes used in addition to lead isotopes.

If solutions to identify source mixtures are still lacking<sup>8</sup>, the solution found to reduce the overlaps is to characterize the full extent of the variability of the isotopic signature of the sampled mines. Archaeological databases therefore generally include several measurements per mine: the median number of analyses per mine is 17 in the Oxalid database; Stos-Gale and Gale (2009) recommends to conduct 30 to 50 analyses per mine. Thus, some historical mines such as the Laurion (Oxalid database) or the mining district of Mitterberg in Germany (Pernicka *et al.*, 2016) were characterized by hundreds of mineral analyses.

Once the mines and large production areas have been delineated, provenance studies can be carried out. These methods have evolved considerably: during the 1980s, archaeologists compared the signatures of ores and artifacts by simple reading on two separate graphs  $^{208}\text{Pb}/^{206}\text{Pb}$  vs  $^{207}\text{Pb}/^{206}\text{Pb}$  and  $^{206}\text{Pb}/^{204}\text{Pb}$  vs  $^{207}\text{Pb}/^{206}\text{Pb}$  (see Stos-Gale and Gale (2009) for a precise description of the evolution of tracing process). In the early 1990s, this graphical reading was facilitated by the tracing of 95% confidence ellipsoids highlighting the signatures of the various mining districts. During the 2000s, these ellipsoids began to be traced by the Euclidean distance method and Kernel density, making their contours sharper (and statistically more accurate). However, these discriminations of different groups of mine populations are problematic: on mineral databases disconnected from any geological context, it seems almost impossible to differentiate some regions (Pampaloni, 2017).

Another limitation can be pointed: not all producing areas are present in the archaeological databases. A striking example is the southern part of the Massif Central at the end of Prehistory: although many mining sites and metallurgical workshops have been discovered, no isotope analysis has been carried out there to date outside the Cabrières site. The same is true for French Brittany, where there are many indications of copper exploitation of the early Bronze Age (Pailler *et al.*, 2016) but for which archaeologists have not conducted any sampling. These areas which still lack data in archaeological databases have been analyzed by the BRGM (*Bureau de Recherches Géologiques et Minières*). Besides, some areas which do not show any evidence of ancient extraction are not necessarily devoid of exploitation sites: archaeological surveys are difficult to carry out in these areas, which are generally mountainous, wooded and little explored by archaeologists. Indeed, the identification of paleo-pollutions from local protohistoric (Bronze Age and late Latenian period) mining operations in the Morvan Mountains (Monna *et al.*, 2004), has led to the identification of latenian mines in Bibracte; Bronze Age mines remain to be identified.

<sup>8</sup> Among recent work, Bray and Pollard (2012) and Longman *et al.* (2018) proposing statistical treatments in order to try to identify possible mixtures but they are still in their inception stages.

The use of analyses carried out by geologists can therefore be useful in filling gaps in archaeological databases. Moreover, the way geologists approach sampling could provide a new way of processing data and avoid the effect of signature overlaps.

### 3 Contribution of geological/geological data

One of the reasons given by archaeologists for not taking into account lead isotopic data from geological work is that they consider that these areas have too few analyses per deposit. Indeed, in geology, only one or two measurements are performed for simple and monogenic deposits (and therefore very homogeneous), and 7 to 10 for complex polygenic deposits (Marcoux, 1986, p.288). However, this low number of analyses per mine is compensated by a wider coverage of the mineralized zone, which is then defined in its entirety and therefore in all its variability of signatures.

In other words, geological studies draw valuable information from the analyses performed because they characterize the fine variations of signatures within the same metallogenic province. The lack of analysis of small deposits in archaeological studies may in fact prove to be a limitation for the tracing of sources.

Very early used as geochronometers (Stacey and Kramers, 1975; Cumming and Richards, 1975<sup>9</sup>), lead isotopes were fast utilized to study mineralization phenomena. In 1981, Zartman and Doe (1981) demonstrated that powerful orogenic and/or hydrothermal phenomena erase lead isotopic signatures from the rocks involved to create new closed systems<sup>10</sup>. Thus, isotopic signature of a deposit depends on 3 factors (Marcoux, 1986):

- the signature of the remobilized source(s);
- the age of the mineralization (radioactive decomposition occurs at the end of the mineralization process);
- the mineralizing fluid phase (two mineralizations of the same age but involving different fluids and/or rocks from different geological contexts will therefore have a different isotopic signature).

In more detail, signatures from deposits of different types but from the same region are closer together than synchronous deposits of the same type, scattered over a vast geological area. Indeed, the isotopic composition of lead seems to be more related to the geological environment than to the typology or chronology of mineral deposits: this phenomenon is called “regionalism” (Marcoux, 1986, p.99).

This regionalism, as Marcoux (1986) pointed out, implies that an orogenic belt does not have a homogeneous isotopic signature<sup>11</sup>. Thus, at the scale of a large orogenic belt, the

<sup>9</sup> Those works offer isochronous evolution curves still used today to date geological contexts *via* radioactive decay laws.

<sup>10</sup> The isotopic signatures of these systems correspond to a mixture of several deposition environments involved. An isotopic signature of lead is therefore a combination of multi-stage lead (from geological contexts of different ages). Marcoux (1986) characterizes this phenomenon under the term “lead mixture”.

<sup>11</sup> Only very large deposits and undisturbed hydrothermal-sedimentary deposits have homogeneous signatures over long distances.

variation in lead isotope ratios varies between 1 and 2%. Nevertheless, work on the scale of small geological sub-belts makes it possible to obtain a significant gain in terms of signature homogeneity: the variation in isotope ratios is then around 0.4%.

Geologists thus use the variability of lead isotopic signatures to study the relationships of filiation of deposits in terms of age or metallogenic sources<sup>12</sup> in a same mining district. The purpose here is therefore not to accurately characterize a mine but to compare the signatures of several deposits in the same mineralized belt. This fine description of the contexts makes it possible to finely separate the mineral populations and avoid the effects of overlaps that occur if we consider information without geological data. This phenomenon can be illustrated by case studies, here we have chosen the Western Alps.

#### 4 Case study: The contribution of geological data on lead isotopes to archaeological research in the Alps

In order to illustrate the contribution of geological analyses to archaeological issues, we take the example of the Western Alps. Although prehistoric copper mining sites are known in the Alps (in Saint Véran: Bourgarit *et al.*, 2010; in the Massif des Rousses: Moulin *et al.*, 2010 or even at Clue de Roua: Rostan and Mari, 2005), lead isotopy data from geological studies, such as Marcoux's in 1986 or Nimis *et al.*, 2012, are almost never used in prehistoric copper production source tracing studies.

In the French Alps, E. Marcoux distinguishes 4 contexts of mineralization:

- Veins of the Mesozoic alpine cover (8 analyses);
- Alpine Pb-Zn-Cu veins of the hercynian basement (16 analyses);
- Mesozoic alpine stratiform deposits (3 analyses);
- Skarnoids<sup>13</sup> of the Alps (2 analyses).

In the Italian Alps sector of the South Tyrol region, Nimis *et al.* (2012) distinguish 8:

- Variscan basement and Permian intrusives – Sulphide-rich vein system (8 analyses);
- Permian volcanics (lower group) – Sulphide-rich vein (5 analyses);
- Variscan basement – Stratiform massive sulphide (23 analyses);
- Variscan basement – Remobilization veins (7 analyses);
- Ladinian volcanics – Mineralized tectonic breccia (5 analyses);
- Variscan basement – Breccia pipe (4 analyses).
- Variscan basement – Fluorite-rich vein (2 analyses)
- U. Permian-L. Triassic carbonates – Stratabound veinlets/disseminated (2 analyses)

<sup>12</sup> Volcanogenic massive sulphide deposits from a same metallogenic region but whose formation ages are shifted over time thus have different signatures.

<sup>13</sup> Deposits created in contact with magmatic (hot) and carbonate (cold) rocks.

The geological contexts do not show a precise spatial distribution (Fig. 1).

Using those lead isotopes data, we can try to assign a geographical origin to a theoretical object with the following signature:  $^{206}\text{Pb}/^{204}\text{Pb}$ : 18.35,  $^{208}\text{Pb}/^{204}\text{Pb}$ : 38.57,  $^{207}\text{Pb}/^{204}\text{Pb}$ : 15.65,  $^{208}\text{Pb}/^{206}\text{Pb}$ : 2.1 et  $^{207}\text{Pb}/^{206}\text{Pb}$ : 0.853 (this signature was chosen because it clearly doesn't correspond to a given geographical origin nor to a specific type of ore deposit in bivariate diagrams).

A bivariate diagram drawn without any geological information doesn't make it possible to discriminate the French Alps (round) and the Italian Tyrol (triangles). Besides, tracing 95% confidence ellipsoids doesn't allow a distinction to be made between the western Alps and the Italian Tyrol (Fig. 2). The presentation of the results by Euclidean distances would also not give a convincing result: the signature of the object being located at an equal distance from French and Italian ores.

Moreover, if here no conclusive provenance results can be obtained by graphical reading, the same is true if multivariate statistical treatments are used. As already mentioned by Sayre *et al.* (2001) (for Anatolia), the use of Gaussian mixtures does not lead to the creation of mineral population groups representative of reality. This can be explained by the fact that the distribution of lead isotopic signatures of deposits is not initially normal. A Shapiro-Wilk type normality test can be used to highlight this fact. Carried out on Variscan basement – Stratiform massive sulphide (which has the clearest distribution), the test gives a result of non-normality with an error threshold of less than 2%.

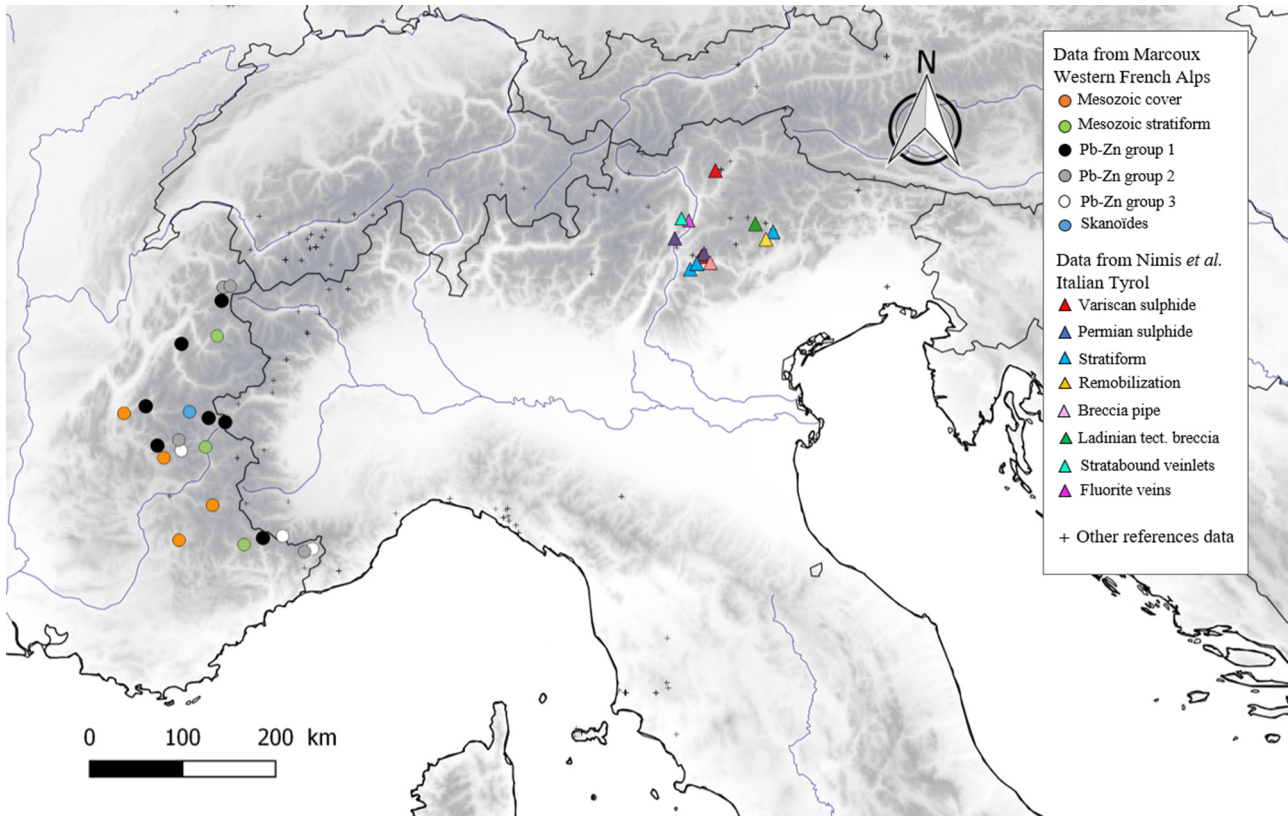
A solution allowing a clear distinction between these contexts can nevertheless be provided through the use of geological data. Indeed, taking under account those data allow a more accurate response to our provenance study. This can be seen in a bivariate diagram: if we draw the diagram  $^{208}\text{Pb}/^{204}\text{Pb}$  vs  $^{206}\text{Pb}/^{204}\text{Pb}$  (in order to visualize the geological contexts<sup>14</sup>), we can note that the geological contexts are well individualized (Fig. 3): the different types of ore deposits can be underlined by their alignment along regression curves.

Three other bivariate diagrams can be traced. The geographical identification of the source of the theoretical object then appears obvious: the object comes from the French Alps and more precisely, from a Pb-Zn-Cu type 1 vein<sup>15</sup> (Fig. 4).

This example illustrates the fact that taking into account the geological contexts makes it possible to differentiate deposits and therefore metal sources through the observation of linear regressions in diagrams including  $^{208}\text{Pb}$ . In the absence of geological data, and by having only geographical data (as is the case in the Oxalid database for example), it is impossible to distinguish regions that are geologically different. It should also be noted that a finer geographical division would also not have enabled the different source areas to be distinguished: geological clustering over an area produces sub-populations whose variances are lower than the variance of the area as a whole.

<sup>14</sup> As previously mentioned, this is due to the fact that the latter is the result of the disintegration of thorium.

<sup>15</sup> According to our division.



**Fig. 1.** Location of mineral data used in this study: the French Alps (Marcoux, 1986) are represented by circles and the Italian Tyrol (Nimis *et al.*, 2012) by triangles. Each colour represents a type of ore deposit. These are not the only isotope analyses carried out on alpine ores, the other analyses are indicated by black crosses (data from Höppner *et al.*, 2005; Cattin, 2008; Giunti, 2011; Artioli *et al.*, 2016; Pernicka *et al.*, 2016).

Moreover, thinking in terms of type of mineralization rather than considering each mine seems more coherent because some mines are linked to the exploitation of complex mineralization (linked to remobilization phenomena or successions of hydrothermal phases). As example, a wide dispersion of the signatures is observed for the French Alps Saint Véran copper deposit (data from Giunti (2011) and Cattin (2008)) (Fig. 5). The mineralization presents important remobilization phases linked to the succession of 4 intense tectonic phases (Ance! *et al.*, 2006).

It would thus appear more interesting to give an origin of precise geological context to an artefact much more than a purely geographical origin. Perhaps should we rethink our reading of lead isotopes to return to what they characterize: a geological history. Admittedly it could be that the geographical precision of the origins is less precise but we would have a more accurate vision of the sources with less overlaps (but always the limit of source mixtures).

## 5 Perspectives: More precise interpretations possible by taking into account geological data

As previously mentioned, in bivariate reading groupings carried out in the absence of geological data create heterogeneous population groups with wide ranges of isotopic signatures. The integration of new data can only create

additional overlaps in these heterogeneous groups. Taking into account geological contexts therefore makes it possible both to reduce the effect of overlaps and to obtain a very fine signature of mining districts (signature ranges are restricted and metallogenic contexts are highlighted by  $^{208}\text{Pb}$ ).

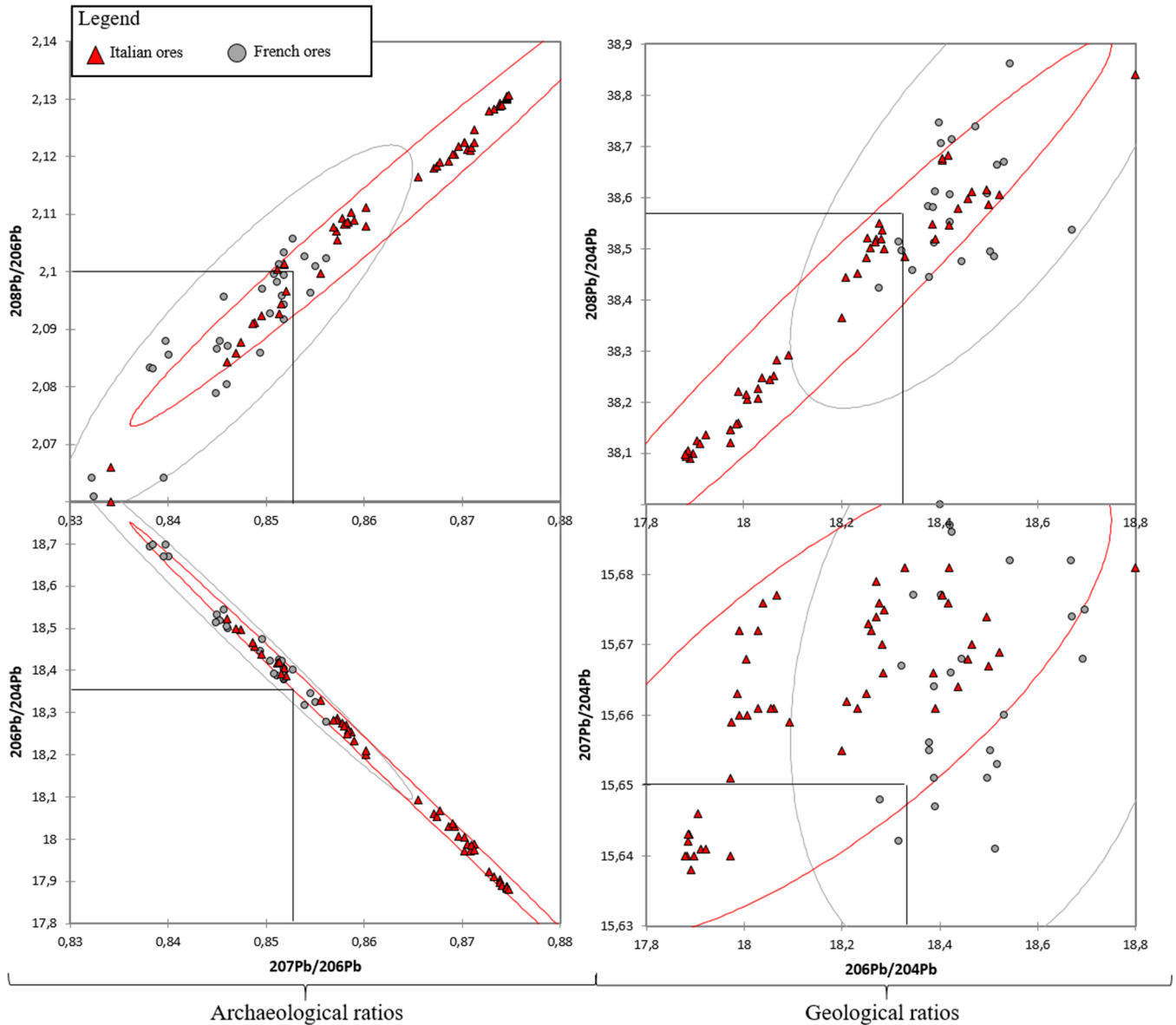
Thinking in terms of geology makes it possible to rethink the use of multivariate statistics which, as mentioned above, have not been used with only geographical data. Several types of statistical treatments can be applied.

### 5.1 Statistical treatments applicable to lead isotopic data supported by geological information

Since mid-2010, the team at the Padua laboratory has been providing archaeological tracing work accompanied by geological data. (Artioli *et al.*, 2016). This same team presents provenance studies in which Euclidean distances are traced in 3D (Artioli *et al.*, 2014) which makes it possible to observe the variations of 3 ratios on the same graph<sup>16</sup>.

However, in order to assess the 3D data and propose relevant groupings of lead isotopic signatures, precise geological data are required (the finer is the geological division, the narrower is the range of signatures per deposit type).

<sup>16</sup> In addition, this team standardizes isotopic ratios on  $^{204}\text{Pb}$  and not on  $^{206}\text{Pb}$  to better distinguish groups of mineral populations.



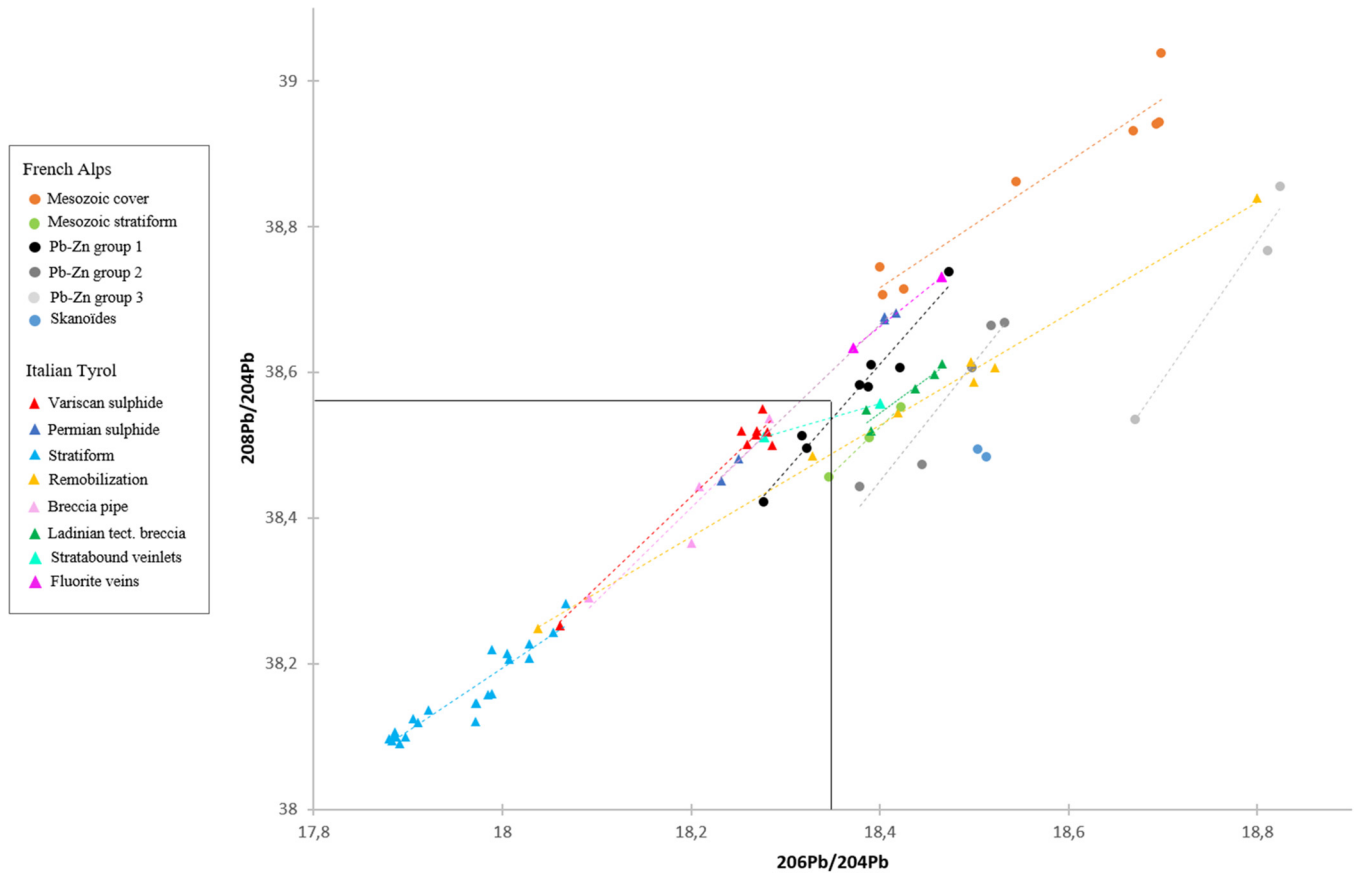
**Fig. 2.** Location of samples from the French Alps (circles) and the Italian Alps (triangles) in the four types of diagrams, commonly used by archaeologists (left: x-axis =  $^{207}\text{Pb}/^{206}\text{Pb}$ ) and by geologists (right:  $^{206}\text{Pb}/^{204}\text{Pb}$  x-axis). The 95% confidence ellipsoids do not allow to differentiate the deposits in the French Alps from the deposits in the Italian Tyrol. The hypothetical object (identified by the crossing of the lines) does not spatially approximate either of the two ore population groups except in the diagram  $^{207}\text{Pb}/^{204}\text{Pb}$  vs  $^{206}\text{Pb}/^{204}\text{Pb}$  where it can be interpreted as coming from the French Alps.

This type of treatment is very effective in known geological contexts and defines production zones with precision at the scale of a mining district or even the mine.

But this is not the only type of statistical treatment possible. If a MANOVA is not possible because the populations are not perfectly Gaussian, a treatment associating PCA then HCA (presented below) or a Discriminant Factorial Analysis, could be envisaged to determine the sources (Tomczyk *et al.*, 2019). A Discriminant Factorial Analysis has the advantage of blocking known contexts. However, it requires that the population groups

have a high inter-class variability, a low intra-class variability and that these groupings be represented by sufficient samples (each geological context must be represented by at least 3 to 7 data depending on the type of deposit).

In addition, there is still a limit to the processing of the thousands of isotopic data already present in archaeological databases and lacking any geological context. In addition to tracking production sources, HCA treatments give good results and can propose classes that are fairly consistent with the geological contexts.



**Fig. 3.** Visualization of geological contexts in a bivariate diagram  $^{208}\text{Pb}/^{204}\text{Pb}$  vs  $^{206}\text{Pb}/^{204}\text{Pb}$ . The margin of error of the measurements is smaller than the diameter of the point. The Pb-Zn-Cu veins (in shades of grey) can be redistributed by Gaussian mixtures models: three coherent groups are then obtained. Variscan and Permian sulphide veins line up along the same regression curve and could therefore be grouped into a single group. Only the remobilizations create an overlaps effect of signatures with the Ladinian tectonic breccias and stratiform Mesozoic deposits.

## 5.2 Multivariate statistics taking into account the 5 isotopic ratios

We are thus applying a multivariate statistical treatments taking into account all the 5 isotopic ratios of lead commonly used by archaeologists and geologists ( $^{207}\text{Pb}/^{206}\text{Pb}$ ,  $^{208}\text{Pb}/^{206}\text{Pb}$ ,  $^{206}\text{Pb}/^{204}\text{Pb}$ ,  $^{207}\text{Pb}/^{204}\text{Pb}$  and  $^{208}\text{Pb}/^{204}\text{Pb}$ )<sup>17</sup>. Our goal is to test if a multivariate treatment allows to find the geological contexts in addition to testing the origin of the theoretical object (and to verify that the result is the same as the one obtained by graphical reading). Indeed, taking into account the 5 ratios stretches the statistical model (Fig. 6).

A Hierarchical Cluster Analysis (HCA) performed on the coordinates of an PCA involving the 5 reports (standardized) allows to observe the similarity of the isotopic signatures of the different geological contexts<sup>18</sup>.

The consideration of 5 isotopic ratios underlines a fairly clear distinction between the different geological contexts

<sup>17</sup> The bivariate diagrams show that the normalized  $^{206}\text{Pb}$  data complement the normalized  $^{204}\text{Pb}$ : some groups are more distinguished according to the diagrams (Fig. 4).

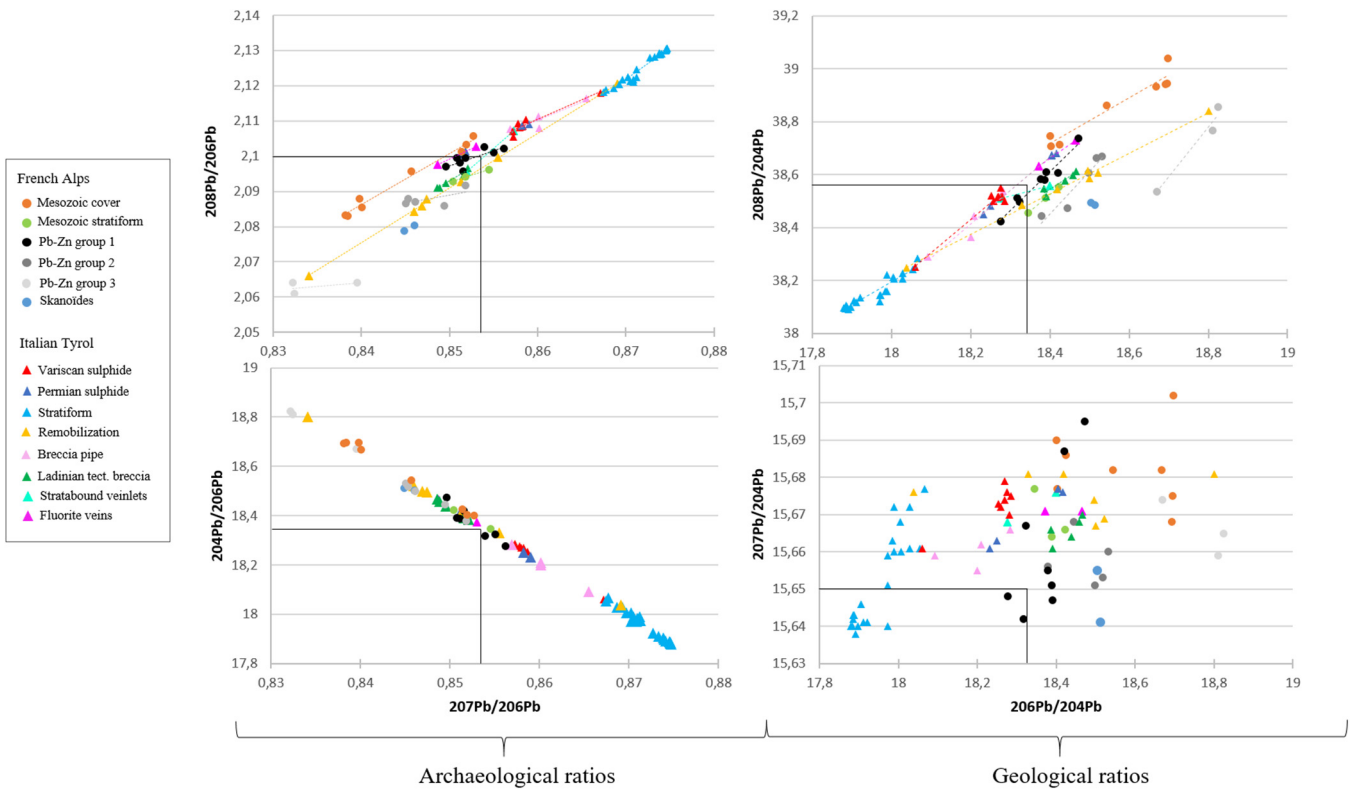
<sup>18</sup> For the sake of consistency, we did not take into account the contexts represented by two analyses only.

(Fig. 7). The Italian Alps ore deposits are mainly distributed in a large statistical class. The geological contexts that appear clear in graphic reading (Stratiform or Variscan sulphides deposits for example) are also clearly distinguished in the sub-classes of the dendrogram. The proposed statistical treatment thus makes it possible to find the contexts when they do not present important overlaps. Contexts with very similar signatures are distributed in the same subclass (Ladinian, remobilization and Mozoic stratiform deposits are grouped in the same subclass; this is also the case for part of the Mozoic cover and Permian sulphide ore deposits). The use of a HCA thus seems to be a reliable reflection of the geological contexts. It could be used when the latter are missing, but nevertheless allows only little to go beyond the signature overlaps.

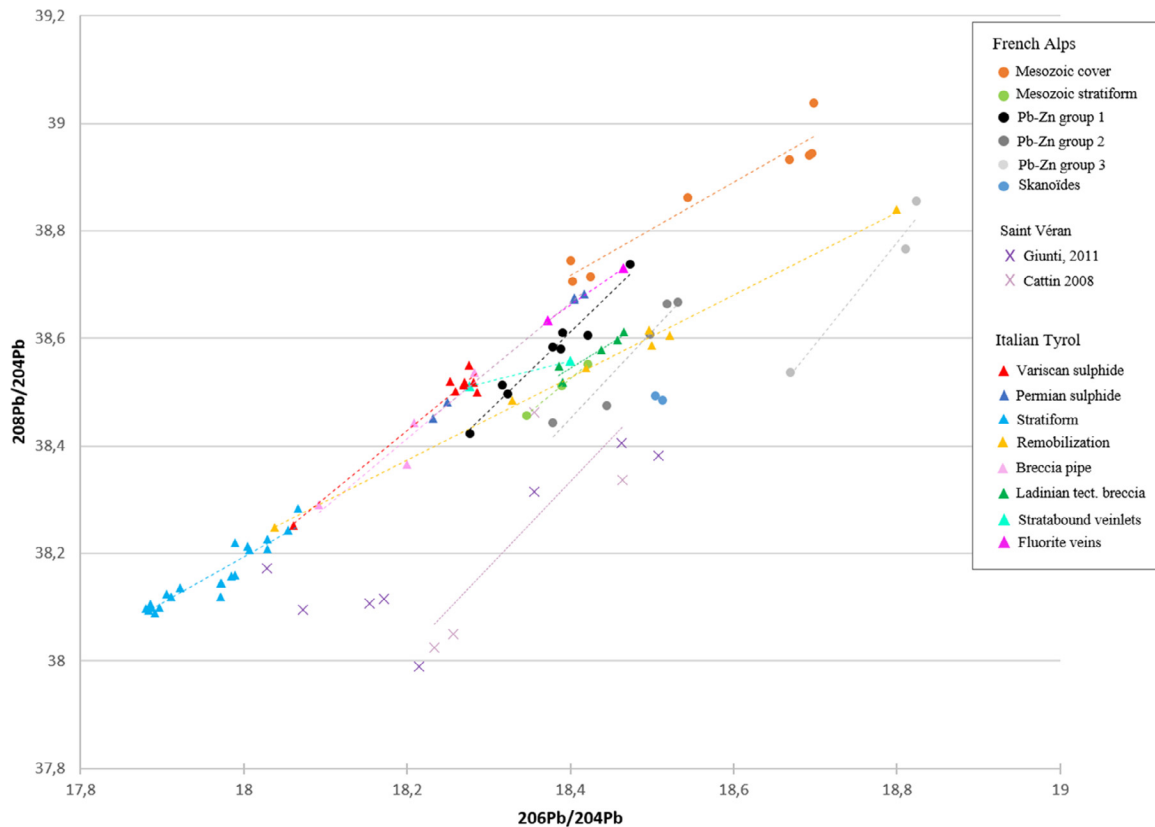
Moreover, if one seeks to affiliate the theoretical object, it is possible to assume the geological context from which the metal used to produce the theoretical object originates: the object is located in a fairly well-defined class corresponding to the Pb-Zn-Cu type 1 veins (as previously identified).

Thus, far from being a perfect solution to retrieve metallogenic contexts, the use of multivariate statistics could provide an approximation of the geological reality when the latter is missing. However, it doesn't worth an extensive field

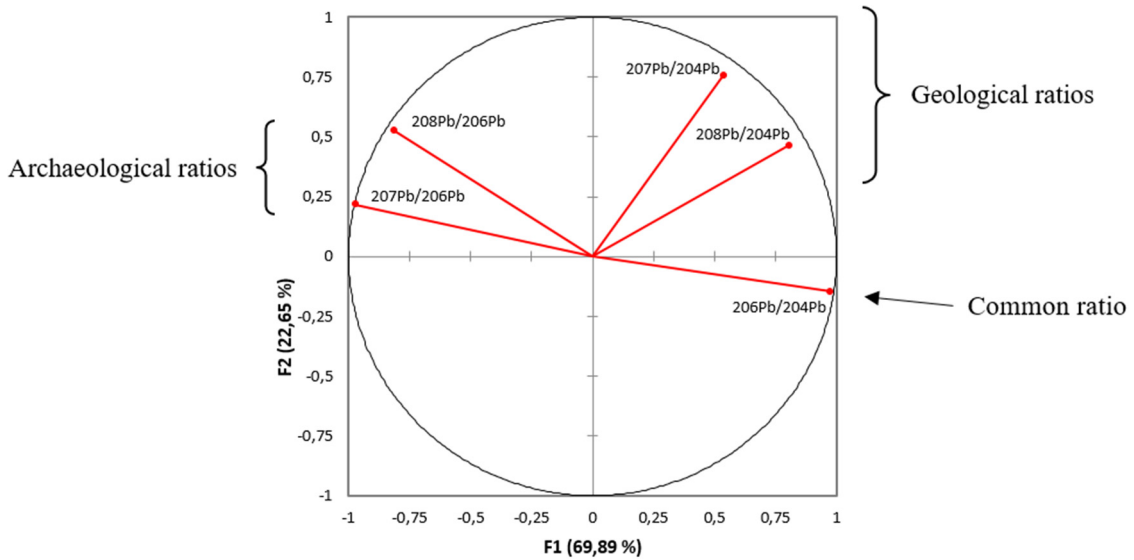




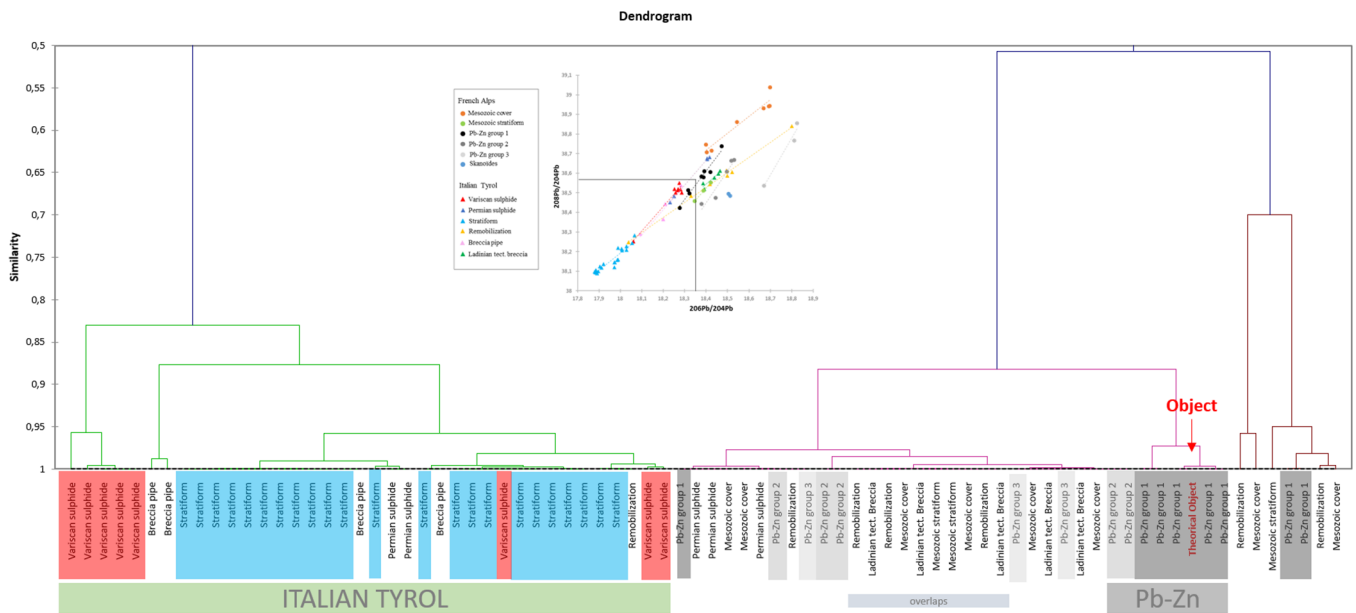
**Fig. 4.** Bivariate diagrams with indications of geological contexts. It is interesting to note that the geological contexts are marked here especially in diagrams involving  $^{208}\text{Pb}$ : they are aligned along regression lines.



**Fig. 5.** Addition of the Saint V éran mine data (cross) in the bivariate diagram  $^{208}\text{Pb}/^{204}\text{Pb}$  vs  $^{206}\text{Pb}/^{204}\text{Pb}$ . The wide dispersal of the mine's signatures reflects the complexity of the deposit.



**Fig. 6.** Correlation circle of the Component Analysis illustrating the contribution of taking into account 5 isotopic ratios (based on data from Marcoux (1986) and Nimis *et al.* (2012)):  $^{206}\text{Pb}/^{204}\text{Pb}$  is strongly anti-correlated with the ratios normalized on  $^{206}\text{Pb}$ , which allows the angle to be opened. The ratios normalized on  $^{206}\text{Pb}$  are then the extremes of the model.



**Fig. 7.** Hierarchical Cluster Analysis (HCA) using the 5 lead isotopes ratios perform on Marcoux (1986) and Nimis *et al.* (2012) data. Certain types of deposits are subdivided into different subclasses. Some are widely dispersed and lose cohesion (*e.g.* remobilization). The theoretical object is associated with one of the two subclasses of the Pb-Zn-Cu type 1 veins.

study and (re)sampling with clear identification of the mineralized contexts.

Finally, it is important to point out that in the case where geological information is present and not researched, the latter could be integrated into a statistical treatment allowing simultaneous processing of quantitative (isotope ratios) and qualitative (geological) data. One possibility could consist in

the use of a multiple correspondence analysis (MCA) performed on a table of variables with qualitative modalities gathering the variable (natively qualitative) of the geology as well as the variables of isotopic ratios (put in classes so that they can be treated as qualitative modalities). This type of approach is quite classical for data from the social sciences and humanities where the problem of simultaneous treatment of

quantitative and qualitative data frequently arises. A recent example where this type of treatment has been used is Vanlandeghem *et al.* (2020).

## 6 Conclusion

A detailed characterization of geological contexts makes it possible to refine statistical groupings of ore data and avoid the effects of signature overlaps. Far from throwing a blur because of the small number of samples per mine, taking into account the geological contexts therefore makes it possible to improve lead isotopic source tracing. It allows a better discrimination of mineral populations and reduces signature overlaps. It would thus be appropriate to rethink the interpretations of provenance by first defining potential (and incompatible) type of deposits and then, in a second stage, by proposing hypotheses concerning geographical origins.

There is still a limit to the processing of the thousands of isotopic data already present in archaeological databases and lacking any geological context. If statistics could be used to generate statistical groupings close to the real contexts, as we just showed in this paper, it is however important to update these databases when possible and, moreover, to consider the geological contexts in the scope of future mineral sampling.

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