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Potentialities of X-ray fluorescence analysis in numismatics: the case study of pre-Roman coins from Cisalpine Gaul.

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ABSTRACT

X-ray fluorescence (XRF) is a well-known technique for the analysis of ancient metals. Thanks to the availability of portable instruments (p-XRF), it is extensively used for the chemical characterization of coins directly in museums. In this work, the potentialities of the technique have been investigated, through its application to a case study concerning the Cisalpine Gaul coinage. More than two hundreds drachmas have been analysed to discriminate different productions on the base of minor elements. Major elements, on the other hand, have been used to trace alloy changes through the centuries. As concerns the quantification of the silver content (fineness), XRF and neutron diffraction results have been compared.

Keywords: XRF; numismatics; coins; silver; archaeometry

INTRODUCTION

X-ray fluorescence (XRF) technique is a well-known analysis tool for the study of ancient metals. Nowadays, it is a valuable method to perform qualitative analyses as a first screening on cultural heritage objects, but also quantitative analysis if ideal conditions are met. Moreover, thanks to the availability of portable and handheld instruments that can be easily brought to museums, galleries and collections, the investigation of large number of objects directly in-situ has become possible, thus providing the opportunity to work with big datasets.

On the other hand, limits of X-ray fluorescence technique, when applied on untreated surfaces of archaeological artefacts, are well known as well. Several works have described how the corrosion, the presence of alteration layers on surface and the limited signal depth of fluorescence photons complicate its application on many metal artefacts. In these cases XRF analysis do not allow a quantitative computation of major elements, since these elements are non-homogenously distributed and therefore the analyzed volume would not be representative of the object under investigation. As concerns ancient silver-alloy coins, the presence of surface thick silver-enriched layers (Tate 1986; Beck 2004) is rather common and therefore a precise quantification of the fineness (precious metal) for debased coins cannot be achieved. Indeed, XRF analysis could overestimate the content of silver, being unable to analyze the inner core, making metrological studies extremely difficult or even impossible with this technique.

Nevertheless, XRF data could become extremely useful for numismatics studies for other research topics. Several works show, for instance, the possibility to discriminate different productions on the base of minor and trace elements usually linked both to silver and copper (e.g. Au, Pb, Bi, As) and sometimes interesting results have been gained from an historical point of view. The most interesting techniques which enable to gain such results are neutron activation (Gentner 1978) and, recently, mass spectrometry coupled with laser ablation (Blet-Lemarquand 2009; Sarah 2007). However, XRF technique has been used too in some cases to reach this goal (Civici 2007; Kantarelou 2011; Rodrigues 2001; Linke 2003) and often data obtained on minor elements were enough to discriminate different productions.

The aim of this paper is to investigate the potentialities of XRF technique in the field of ancient numismatics, trying to discriminate different productions of Cisalpine drachmas on the base of minor elements. For this purpose, more than two hundreds drachmas made of a silver-copper alloy have been analysed. At now, these poorly studied emissions from northern Italy have already been analysed by the authors in some previous works (Corsi 2015, Corsi 2016a) to investigate different issues.

CASE STUDY

The 227 silver drachmas analysed in this work were produced by tribes of Celtic or Celto-Ligurian culture settled in the regions of northern Italy, the so-called Cisalpine Gaul. The iconography of these coins reproduces the types of the silver drachm of Massalia, a Greek city on the coast of southern France which had an important influence in the surrounding Celtic-inhabited area. The production lasted between late III and I century B.C. in different mints spread between Piemonte and Veneto. Numismatists are used to identify different typologies of Cisalpine coins considering some stylistic features, such as the depiction of the lion.

Samples analysed in this work are kept in two different museums and come from three different hoards found in Liguria, Piemonte and Lombardia. The coins analyzed in this work are summarized in Table 1 and have been chosen specifically to be representative of five different typologies identified by numismatists (Fig. 1). According to the current classification, the oldest coins are the “naturalistic” and “scorpion lion” types, then the “wolf lion” type (attributed to the Lybians’ tribe) and finally the most recent Cenomans’ and Insubres’ types.

Two important hoards are kept in the Museo di Antichità in Torino: the treasure from Serra Riccò (Genova), consisting of 164 specimens (37 drachmas and 127 fractions, the latter not included in the present study), and the hoard of Biandrate (Novara), made of 40 coins. The discovery of Serra Riccò hoard (Barocelli 1926a; Rebora 2005) was very peculiar, since it happened in 1923 during construction works for the Genova - Casella railway, after an explosion induced by workers to remove some boulders. The coins found were at first shared among the workers and then dispersed, but some tens of them were recovered by authorities. Further digs organized by the “Sopraintendenza degli Scavi” led to the recovery of other specimens. Nonetheless, some scholars estimate the total number of coins close to 2000 (Gorini 2011). Some nuclei of these coins have then been sold in auction (Allen 1971; Fischer 1981) and bequeathed by museums or collectors (Orlandoni 1988). Dating of the burial is controversial and some scholars believe it was buried in the 1st century B.C. (Gorini 2011). However, weights and typologies of drachmas are of late 3rd century B.C. (Corsi 2016b; Crawford 1985), and this fact is explainable with a long-term accumulation votive deposit. The hoard from Biandrate was found in 1922 during agricultural works (Barocelli 1926b) and divided between the discoverer and the Regio Museo of Torino, even if a significant part of the discovery was not declared (Gambari 2013). The burial of the hoard is currently dated to 211-208 B.C., because of a *terminus post quem* offered by a Roman denarius buried with the drachmas.

The Manerbio hoard is the wider hoard containing Celtic coins from northern Italy (Arslan 2007). It has been found in 1955 in the area of Gravine Nuove, near Manerbio (Brescia), during excavation works. Some workers found, in a clay vase, more than 30 kilograms of coins, all soldered one to each other by corrosion products. More than 3 quarters of the hoard was dispersed, while the rest was recovered after the intervention of the local “Soprintendenza alle Antichità”. The treasure is constituted by 4194 Cisalpine drachms, nowadays shown in the Museo Civico di Santa Giulia in Brescia. Aside from the impressive number of coins, the importance of the hoard lies in the fact that only three typologies (attributed to Cenomans, Insubres and Lybians tribes) are present, and in equal parts (~ 33% for each typology). This peculiar feature led some scholars to consider the hoard as a federal fund, formed after a federal agreement among different tribes (Arslan 2007). The treasure was probably kept in a sanctuary, whose presence is not deduced by architectural evidences but supposed on the base of this finding and the silver *falere* (decorative disks for armours) found nearby (Arslan 2007). The hoard is currently dated to the years 150-135 B.C., on the base of the context.

EXPERIMENTAL

In this work, a handheld XRF instrument (p-XRF) was used. It is a commercial device Thermo Niton XL3t-900 GOLDD (Fig. 2) with a silver anode and an X-ray tube that can operate between 6 and 50 kV and between 40 and 200 µA. The detector is a SDD of 25 mm^2, with an energy resolution of 185 eV. The available beam diameter dimensions are 3 or 8 mm. A CCD camera is available to check the measured area.

XRF measurements were performed on the surface of untreated coins. The operative conditions used for the analyses of the samples were: beam energy of 40 keV (with an Al/Fe filter), current of 50 µA and beam spot of 3 mm. With a primary beam of maximum 40 keV, fluorescence lines of an energy range E < 40 keV can be excited, therefore enabling the detection of Ag, Cu, Pb, Au and Bi. Conversely, the filter used prevents the analysis of elements with a low Z, which comes with coin patinas and soil deposits, but are not relevant for the aims of this work. To conciliate available time in museums and meaningful analytical results, each measurement was set to 30 seconds per coin, performing one analysis per specimen on flat surfaces. In some cases, coins were measured twice in order to understand the compositional variability from point to point.

Since the aim of these measurements was not to provide quantitative data, we simply considered the intensity (counts) of peaks to perform the data analysis. Only in the case of silver content, for some samples, we calculated the composition using the WinAxil software, developed by Camberra on the Axil code (IAEA). A couple of MBH silver-copper alloy standards (respectively with 87.60 ± 0.06 and 94.40 ± 0.04 wt.% silver content; 12.26 ± 0.15 and 5.13 ± 0.03 wt.% copper content) has been used for this purpose. It must be stressed that due to the surface silver enrichments phenomena, results obtained in such way give a silver content that in the case of debased coins is overestimated.

RESULTS

XRF spectra (Fig. 3) show several peaks that can be attributed to different elements of the alloy. As concerns the major ones, XRF spectra reveal the presence, in all the drachmas, of silver and copper. The silver content determines the value of the coin, while the copper is intentionally added to debase the alloy or to reach a lower melting point. Moreover, it has been observed that on many specimens a third element, the tin, was present too (see Fig. 3, coin 87502). Minor elements detected with this technique are gold, lead, bismuth and iron. While the major elements are intentionally introduced in the alloy, minor elements are linked to the ore geology and/or to the supply sources of majors and could be used to identify groups with compositional similarity (Blet-Lemarquand 2014). Iron has not been considered because it could derive from soil deposits.

Silver content and fineness

One of the most important information for numismatists is the so called fineness, i.e. the content of precious metal that in this case is silver. As previously observed, XRF results for silver are usually affected by surface enrichments and alterations, especially for debased coins such as those described in this work. Some of these have been previously analyzed (Corsi 2016a) with a bulk technique, the time-of-flight neutron diffraction (ND), and both results are now reported in Table 2. In Fig. 4 the same data of Table 2 are plotted, to better show differences between XRF and ND results.

All the silver content obtained by means of XRF are higher (from 5% to 20%) than the values obtained by means of ND. The only exception is provided by sample no. 1 (see Table 2), which is a coin with a high fineness (about 93%) and of a different typology. Indeed, in cases where fineness is higher than 92%, there is no difference between surface and bulk silver composition, in agreement with the work of Beck (Beck 2004). Except for this coin, in all the samples the silver quantified with

XRF is constantly higher than 85%, without appreciable differences among the typologies considered. Nevertheless, ND data revealed a sharp split between the naturalistic/scorpion lion types (with an average of 82 wt.% Ag) and the later wolf lion type (with an average of 70 wt.% Ag). For this reason it is possible to suppose that a rather thick enriched layer is present in all the samples. These can be easily related with the presence of silver-enriched layers on surface, whose thickness can be relevant (up to 200 μm, see e.g. Zwicker 1993).

Presence of tin

We discovered that several samples show a high content of tin, along with silver and copper. The strong intensity of tin peaks clearly explain that this element is intentionally introduced in the alloy. Interestingly tin is present exclusively in all the coin typologies from the Manerbio hoard. In this way, it enables to discriminate different types and productions on the base either of its presence or its absence. Indeed, as can be seen from Fig. 5, the naturalistic lion type, the scorpion lion type and the wolf lion type from Biandrate are simply made of a binary silver-copper alloy, being tin well below LOD. On the other hand, the series belonging to the Cenomans and the Insubres tribes and the wolf lion type from Manerbio are made of a ternary silver-copper-tin alloy. It must be stressed that all the three typologies represented in the Manerbio hoard are made of ternary alloy.

These results confirm the chemical –destructive– analyses carried out on some coins from Manerbio in the '60s (Pautasso 1966), specifically a Cenoman drachm (Ag: 50.1%; Cu: 38.3%; Sn: 9.6%), a wolf lion drachm (Ag: 45.0%; Cu: 45.8%; Sn: 7.9%) and a Insubres drachm (Ag: 43.5%; Cu: 45.0%; Sn: 8.9%). They confirm also data obtained on a selection of coins kept at the Hungarian National Museum (Corsi 2015), for which tin was observed only in Cenomans and Insubres productions from the Verdello hoard (Bergamo, half of the II c. B.C).

The passage from a binary to a ternary alloy is of great interest, in particular because it can be traced also inside the same typology, i.e. the wolf lion. The importance of this result relies on the fact that the analyzed coins come from well-known hoards, thus their chronology could be put in relation with this change. The Biandrate hoard is currently dated to 211-208 B.C., while the Manerbio hoard was buried later, around 150-135 B.C. The introduction of tin in the coinage alloy should therefore have happened during the first half of the II century B.C.

Minor elements

As concerns minor elements, all the drachmas have small amounts of gold, lead and bismuth. These metals can be used to discriminate different productions. In this work, we chose to show these data using the following ratios: Au/Ag, Pb/Ag, Bi/Ag, because those elements are usually correlated with silver (Blet-Lemarquand 2014).

Results concerning lead and gold are displayed in Fig. 6a and show that naturalistic lion and scorpion lion types can be distinguished from the coins belonging to the wolf lion type present in the Biandrate hoard. Also drachmas from the Manerbio hoard show the presence of these elements (Fig. 6b), but spread on a larger scale of values which would completely superimpose the coins plotted in the previous graph (Fig. 6a). On the other hand, graph plotting bismuth and lead (Fig. 7) show some interesting features. First of all, a clear correlation between the two elements can be observed for the binary alloy coins and the ternary alloy coins. All the coins from the Manerbio hoard lie on the same line showing higher values of Pb but lower content of Bi (which is often below LOD). It must be also noticed that the three typologies (Cenomans' and Insubres' tribes types and wolf lion type) represented in the Manerbio hoard show very similar compositions, while coins from other hoards can be discriminated.

We finally evaluated the variability of XRF results on the same samples but in different areas. Indeed, the elemental distribution is not homogeneous, especially in ancient artefacts such as debased coins. A previous work (Kantarelou 2011) showed the highly variable spatial distribution of elements such as Au, Fe and Pb. We therefore measured some coins twice, both on the obverse and on the reverse. This enabled to calculate the relative percentage differences (Fig. 8) with the following formula: (Counts $_{Obverse}$ – Counts $_{Reverse}$ / Counts $_{Obverse}$) * 100. Absolute differences, due both to the inhomogeneity of the coins and to geometrical measurement factors, are on average of the 7% for Ag, 22% for Cu, 12% for Au and 22% for Pb. It is worth stating that these values do not affect the conclusions made on Fig. 5, 6 and 7.

CONCLUSIONS

In this work a total of 227 Cisalpine coins, belonging to five different typologies, has been analyzed with XRF technique to find out if elemental analysis could be useful for a chemical discrimination of different productions. To perform fast and *in situ* measurements, a handheld instrument (p-XRF) was used; the possibility to work directly in museums enabled to collect a wide and valid statistical sample without moving the objects towards scientific laboratories.

Our results enabled to distinguish, on the base of minor (gold, bismuth, lead) and major (tin) elements, the wolf lion type from the naturalistic and scorpion lion types, which cluster together. This discrimination of different coin productions, let us suppose that different supply sources or, at least, “stocks” were used by different emitting authorities. It is worth mentioning that minor elements alone cannot provide valuable information about the provenance of minerals used to extract the major elements, because of the influence of metallurgical processes and of the habits of metallurgists. For this reason the term “stock” is very appropriate, since it identifies the metal used in a particular age, in a defined area and with a specific metal refining procedure, thus keeping into account all the variables (including remelting) in a chronological perspective.

Moreover, we also uncovered that coins dated to the II century B.C. such as those from the Manerbio hoard are characterized by a ternary Ag-Cu-Sn alloy, while previous drachmas were struck with a simple binary Ag-Cu alloy. In particular, all the three typologies represented in the Manerbio hoard showed a very similar alloy: this resemblance could mean that the same raw material was used for their production. This appears to be very meaningful from an historical point view, since the composition (ternary alloy of silver, copper and tin) could have been decided in a federal agreement among the Insubres, Cenomans and Libyans tribes, and the silver could come from the same “stock” (considering the comparable amounts of gold, bismuth and lead). Moreover, it must be stressed that we observed the presence of a ternary alloy also in coins coming from the Verdello hoard (half of the II c. B.C.) kept in the Hungarian National Museum in Budapest (Corsi 2015), and this confirms that all the coins produced in this period have a certain amount of tin. The use of tin in silver coin is not limited to Cisalpine coins, but can be found in other Celtic coinages, e.g. those (Sarthre 1996) from the Atlantic Gaul (drachmas “*au cavalier ailé*”) and those (Thompson 1972) from the Armorican peninsula (drachmas with the “boar mint mark”). In both these cases, authors consider the presence of tin due to the use of scrap, recycled bronze objects. Adding copper to debase the coinage alloy was usual in the first emissions of the Cisalpine drachm, but the addition of scrap bronze could have occurred in the last ones.

In conclusion, this work show that a simple p-XRF fluorescence measurement on a meaningfully statistical sample could lead to significant results for numismatic studies. On the other hand, it should be stressed that only a good knowledge of both the analytical technique used and the structure of materials analyzed allow to get these important results.

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Fig. 1. Typologies of drachmas analysed in this work. (a) naturalistic lion type, (b) scorpion lion type, (c) wolf lion type (Biandrate), (d) wolf lion type (Manerbio), (e) Insubres' type, (f) Cenomans' type. Reference bar is 2 cm long.

Hoard	Naturalistic lion	Scorpion lion	Wolf lion (Lybians)	Cenomans' tribe type	Insubres' tribe type
Serra Riccò (Genova)	9	28	-	-	-
Biandrate (Novara)	2	3	35	-	-
Manerbio (Brescia)	-	-	50	50	50

Table 1. Number of specimens analysed with XRF technique for each typology of Cisalpine drachma.

Fig. 2. p-XRF setup during measuring session in museum.

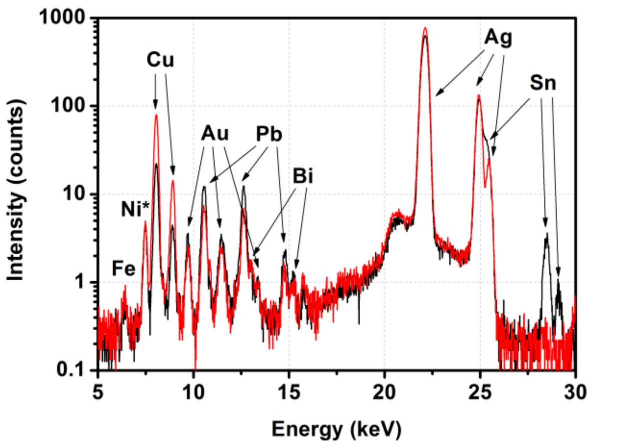


Fig. 3. XRF spectra of coins 155621 from the Manerbio hoard (black line) and 87502 from the Serra Riccò hoard (red line), with peaks labelled. Nickel is an instrumental peak.

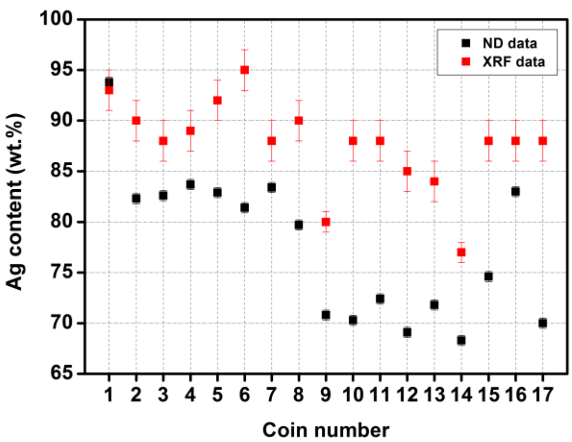


Fig. 4. Comparison of XRF and ND data [Corsi 2016].

Number	Inventory number	Provenance	Coin type	Ag wt.% (ND data)	Ag wt.% (XRF data)

1	22653	M.A. coll.	Massa beta	93.8 ± 0.5	93 ± 2
2	61851	Biandrate	Naturalistic lion	82.3 ± 0.5	90 ± 2
3	87502	Serra Riccò	Naturalistic lion	82.6 ± 0.5	88 ± 2
4	66500	Serra Riccò	Scorpion lion	83.7 ± 0.5	89 ± 2
5	66503	Serra Riccò	Scorpion lion	82.9 ± 0.5	92 ± 2
6	87509	Serra Riccò	Scorpion lion	81.4 ± 0.5	95 ± 2
7	66493	Serra Riccò	Scorpion lion	83.4 ± 0.5	88 ± 2
8	66489	Serra Riccò	Scorpion lion	79.7 ± 0.5	90 ± 2
9	66481	Biandrate	Late scorpion lion	70.8 ± 0.5	80 ± 1
10	66478	Biandrate	Wolf lion	70.3 ± 0.5	88 ± 2
11	61822	Biandrate	Wolf lion	72.4 ± 0.5	88 ± 2
12	61828	Biandrate	Wolf lion	69.1 ± 0.5	85 ± 2
13	61843	Biandrate	Wolf lion	71.8 ± 0.5	84 ± 2
14	66483	Biandrate	Wolf lion	68.3 ± 0.5	77 ± 1
15	61845	Biandrate	Wolf lion	74.6 ± 0.5	88 ± 2
16	66480	Biandrate	Wolf lion	83.0 ± 0.5	88 ± 2
17	66485	Biandrate	Wolf lion	70.0 ± 0.5	88 ± 2

Table 2. Silver content: XRF and ND data. Coin no. 1 comes from the historical collection of the Museo di Antichità (M.A.) of Torino and not from hoards.

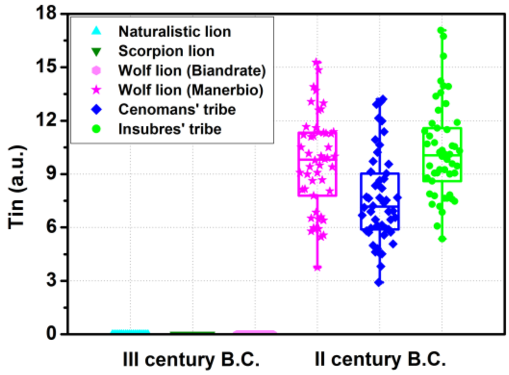
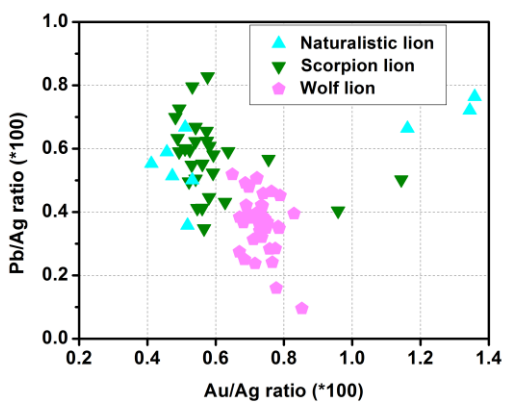


Fig. 5. Box-chart graph showing Sn presence in different typologies of Cisalpine drachmas. All the naturalistic lion, scorpion lion and wolf lion drachmas from Biandrate are below LOD of the instrument for Sn. Boxes include 50% of analysed points (from 25 to 75%) while the bars contain all the other points (from 0 to 25% and from 75 to 100%). Each box is separated in two parts by a line dividing the points in two identical groups (50 and 50%).



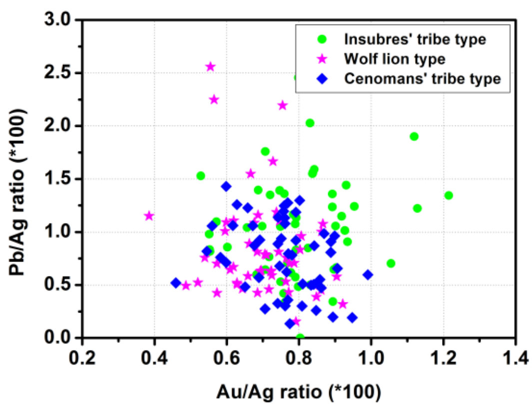


Fig. 6(a-b). Pb/Ag ratio vs. Au/Ag ratio in Cisalpine coins from (a) Serra Riccò and Biandrate hoards and (b) Manerbio hoard. Note: the Y axis scale is different.

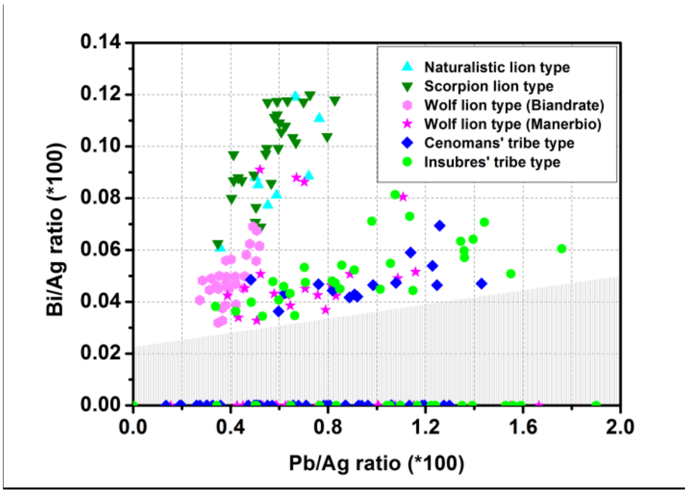


Fig. 7. Bi/Ag ratio vs. Pb/Ag ratio in coins from Serra Riccò, Biandrate and Manerbio hoards. For the last hoard, 85 coins (57% of the total) show Bi values below LOD (grey shaded area).

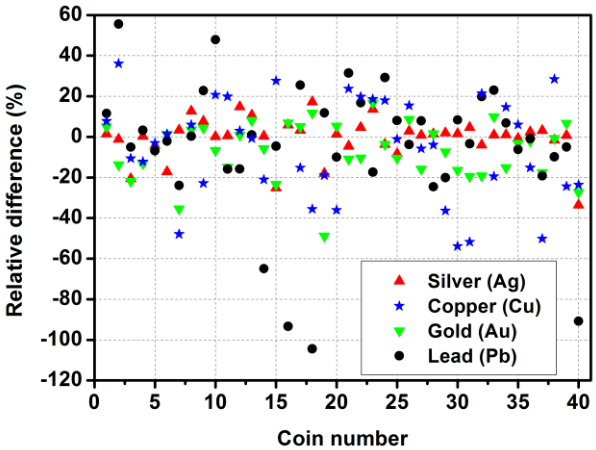


Fig. 8. Relative difference, for different elements, between measurements carried out on obverse and reverse. Coins in this graph come from the Biandrate hoard.