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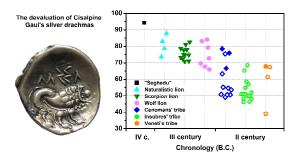
#### **Abstract**

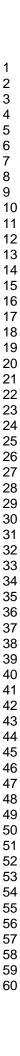
The Cisalpine Gaul's coinage has been produced by different tribes settled in northern Italy between the 4<sup>th</sup> and the 1<sup>st</sup> century B.C. During this wide chronological range several types of silver drachms have been emitted, nowadays classified by numismatists in different typologies. To verify the presence of a debasement along the years and to investigate the exchange ratios among different drachmas, the rich collection of the Hungarian National Museum in Budapest has been analysed. Measurements have been performed at the Budapest Neutron Centre with the Prompt Gamma Activation Analysis (PGAA), a bulk technique which enables to overcome surface enriched layers and alterations. This technique allows silver and copper to be quantified, while to check the presence of tin and other minor elements the X-ray fluorescence (XRF) has been used. Results show that the silver content falls from the 94% of the first emissions up to the 50% of the Cenomans' and Insubres' tribes late typologies (Fig. 1). This strong debasement takes place between the III and the II century B.C. and could be related with the military efforts in the decades around the second Punic war. At the same time, we observe the transition from a binary silver-copper alloy to a ternary one, made of silver, copper and tin.

#### Text: one sentence, of maximum 20 words, highlighting the novelty of the work

This work, through PGAA and XRF measurements, enabled to detect silver debasement and alloy changes in pre-Roman age coins.

#### Colour graphic: maximum size 8 cm x 4 cm





100 The devaluation of Cisalpine Gaul's silver drachmas 90 80 70 60 "Seghedu" Naturalistic lion 50 Scorpion lion Wolf lion Cenomans' tribe 40 Insubres' tribe Veneti's tribe 30 IV c. III century II century Chronology (B.C.)

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## Journal Name

# **RSCPublishing**

#### **ARTICLE**

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# Compositional analysis of a historical collection of Cisalpine Gaul's coins kept at the Hungarian National Museum

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The Cisalpine Gaul's coinage has been produced by different tribes settled in northern Italy between the 4<sup>th</sup> and the 1<sup>st</sup> century B.C. During this wide chronological range several types of silver drachms have been emitted, nowadays classified by numismatists in different typologies. To verify the presence of a debasement along the years and to investigate the exchange ratios among different drachmas, the rich collection of the Hungarian National Museum in Budapest has been analysed. Measurements have been performed at the Budapest Neutron Centre with the Prompt Gamma Activation Analysis (PGAA), a bulk technique which enables to overcome surface enriched layers and alterations. This technique allows silver and copper to be quantified, while to check the presence of tin and other minor elements the X-ray fluorescence (XRF) has been used. Results show that the silver content falls from the 94% of the first emissions up to the 50% of the Cenomans' and Insubres' tribes late typologies. This strong debasement takes place between the III and the II century B.C. and could be related with the military efforts in the decades around the second Punic war. At the same time, we observe the transition from a binary silver-copper alloy to a ternary one, made of silver, copper and tin.

#### Introduction

The application of scientific analyses in the numismatics field dates back at the 18th century, when the first chemical -and destructive- compositional measurements have been performed on ancient coins. Nowadays, many different techniques are available to answer specific questions concerning historical issues faced by numismatists. Compositional analyses intended to provide the quantification of the main alloying elements remain however the most important and useful in this specific research field. In particular, compositional data of major elements are specifically important for the study of devaluation in antiquity, which is a common feature for economically developed societies. Nowadays, the average level of inflation has in general no impact on the circulating coins of contemporary currencies, being them produced with non-precious metals. Nonetheless, in ancient times devaluation processes had an immediate effect on the coin weight and/or on the so-called fineness, which is the content of precious metal (gold or silver).

For this reason compositional analyses, along with a careful analysis of average weights, are particularly suited to provide useful data for the study of economic changes in antiquity. Once these technical data are available, metrological relationships among coins of a certain denomination produced in different periods or among foreign currencies can be studied. A very famous example is provided by the study on the devaluation of the Roman silver denarius.1 However, debasement is a rather common practice and can be found in different historical periods and geographic areas. Other experimentally proven debasements are e.g. those concerning the Visigothic gold coinage,<sup>2</sup> the gold dinars issued in the Iberian peninsula,<sup>3</sup> the gold Aksumite coinage,<sup>4</sup> the Carolingian silver denarii<sup>5</sup> and the Roman-age tetradrachms from Alexandria.6 To gain compositional data, several techniques have been employed along the years. While between the '50s and the '70s the most used were those based on activation<sup>8</sup> or on chemical analyses,<sup>9</sup> in the last two decades an approach based on mass spectroscopy (e.g. ICP-MS) prevailed. 10 Nevertheless, requiring the latter approach sampling or microsampling, a neutron technique has been preferred for this work.

## Journal Name

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Fig. 1. A selection of Cisalpine coins from the HNM analysed in this work (for each coin obverse and reverse are shown). From top-left: (a) "Seghedu" type; (b) naturalistic lion type; (c) scorpion lion type; (d) wolf lion type; (e) Cenomans' tribe type; (f) Insubres' tribe type; (g) Veneti's tribe type; (h) Veragres' tribe type. The reference bar is 2 cm long.

In the last years, a wide plan of scientific analyses has been developed to fully characterize, from a chemo-physical point of view, the so called Celtic coinage from northern Italy. These coins, also known as Cisalpine coins or "Celtic coins of the Po valley", had been produced by the human communities settled between the French region of Provence and northern Italy. They minted almost only silver coins based on the weight standard of the drachm of Massalia, a Greek colony founded on the southern coasts of France. These coins, produced in a wide chronological range (between the beginning of 4th and the 1st century B.C.), reproduce mainly the iconography of the "heavy" drachm of Massalia, which depicts the head of the goddess Artemis on obverse and a lion with the ethnic abbreviation  $MA\Sigma\Sigma A$  on reverse. They are usually easily distinguishable from the official emissions thanks to different stylistic features, which enable numismatists to define several typologies (Fig. 1). This coinage is still affected by many historical issues, although some studies have attempted to solve some of them, e.g. attribution to specific tribes, datings and localization of emission centres. 11-13 In this work we intend in particular to deal with the variation of silver content in different typologies of drachmas. Indeed, the few existing measurements carried out in the '60s with chemical analysis on three late coins of different typology<sup>11</sup> suggest the existence of a strong devaluation among the first and the latter emissions. To have a first general idea on the evolution of the Celtic coinage from northern Italy we chose to analyse the rich collection kept at Hungarian National Museum in Budapest, which counts several pieces of the main typologies.

#### **Samples**

The 58 drachmas from northern Italy analysed in this work come all from the numismatic collection of the Hungarian National Museum (HNM) in Budapest. A first group of these coins has been published within a numismatic study about 20 years ago<sup>14</sup>

and republished few years later on the *Sylloge Nummorum Graecorum* series, together with a second group recovered in the meanwhile. <sup>15</sup> The specimens entered in the collection in different moments along the years. Among them, several come from different bequests by important Hungarian numismatists of the 19<sup>th</sup> and 20<sup>th</sup> century, while others have been directly purchased by the Museum.

20 out of the 58 Massalian imitations nowadays in the collection were bequeathed to the National Museum by Count Miklós Dessewffy, who was an important scholar of Celtic and "barbarian" coins. The Count had contacts with Italian scholars and dealers as well, since the majority of his Massalian imitation's drachms were found in Verona, Verdello (near Bergamo) and in Abbiategrasso (near Milan). He might have visited Italy probably between 1910 and 1914, because those coins are not represented in the first catalogue of his collection<sup>16</sup> but are published in the following years. 17-19 His collaboration with the Coin Cabinet of the HNM might have enriched the HNM collection of Cisalpine coins, since the coins from the Verdello hoard acquired directly by the Museum entered in the collection in the same years. Finally, other Massalian-imitations coins come from the collections of István Delhaes and Ferenc Kiss, but they do not have any recorded provenance.

The remarkable importance of the HNM nucleus of Cisalpine coins is given by the number of the specimens, which is rather high for such a local coinage in a foreign coin cabinet, but especially because some coins still bear the indication of the provenance. Moreover, all the major typologies of drachmas are represented by several samples: for this reason we can state that the 58 samples considered in this work are representative of the entire Cisalpine Gaul's coinage, covering almost all the typologies produced between 4<sup>th</sup> and 1<sup>st</sup> century B.C.

A complete list of the coins analysed is reported in Table 1, along with all inventory codes and some technical details. Following is a short description of the main streams of Celtic coinage from northern Italy:

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- "Seghedu" drachm (Arslan type IV). Minted on the weight standard of the "heavy" drachm of Massalia, this coin is one of the only five specimens known. Despite the presence of a different reverse, which do not depicts the lion, and a legend written in northern Etruscan characters, is considered as part of the Celtic coinage because of a die-link with the first imitations.<sup>20</sup>
- 2. "Naturalistic lion" and "scorpion lion" drachmas (Arslan types V-VI and VII-VIII). Commonly found together in hoards, they are the most ancient drachmas (3<sup>rd</sup> century B.C.) minted in the regions of northern Italy. They have been minted at first with an average weight of 3.0 grams, but an internal evolution probably exists for the "scorpion lion" type, as suggested by some stylistic changes and weight loss.
- 3. "Wolf lion" drachmas (Arslan types XVI-XVII). Produced in an area comprised between Piedmont and Lombardy regions from the end of 3<sup>rd</sup> century B.C.
- Cenomans' tribe drachmas (Arslan types IX-X). Minted by tribes settled in the area around Brescia, probably the Cenomans, during the 2<sup>nd</sup> century B.C.
- Insubres' tribe drachmas (Arslan types XI-XII-XIII-XV). Attributed to the leading tribe settled in Lombardy, the Insubres, these coins have been emitted by the half of 2<sup>nd</sup> century B.C.
- 6. Veneti's tribe drachmas (Arslan types XVIII-XIX-XX). Emitted by the Veneti, a tribe of non-Celtic origin, during the 2<sup>nd</sup> century B.C.
- 7. Other minor typologies. Among these are coins attributed to minor tribes, such as those living in the Trentino area (Arslan type XXI) and the Veragres from the Vallese area, Switzerland (Arslan type XXII).

All the historical data and classifications presented above refer to the studies of E.A. Arslan<sup>14</sup>, and are the most recent data available on this specific topic. Nonetheless, many of them (e.g. dating and attributions) are still proposals, which have not been exhaustively confirmed by archaeological research yet.

#### **Experimental**

When scientific measurements on archaeological and artistic objects are necessary, several aspects have to be taken into account to develop a proper strategy of analysis. Indeed, these kinds of materials need special requirements as concerns the non-destructivity of the employed technique, the representativeness of the analysed volume compared to the entire object and finally the statistical significance of the number of measurements performed. For ancient coins, this means that the analytical technique should not leave traces after the measurements, that results needs to be representative of the total volume, and finally that several samples must be analysed if one wishes to draw general conclusions on an entire emission or coinage. Before starting any analysis, we carefully evaluated all these factors to choose the best approach to investigate the issues regarding the silver debasement.

To this aim, at first, we sectioned a drachm (from a private collection) attributed to the Veneti, a tribe settled in the northeastern part of Italy. This coin is generally considered as a late production of the Celtic coinage, dated to the 2<sup>nd</sup> century B.C., and chosen for this work because believed to be severely debased compared to previous emissions. The sample has been cut into two halves with a rotating diamond wheel and then one piece was

embedded into resin. Afterwards, the section was polished with papers having different grit and also with a diamond crystal paste (with grains of 6-3-1  $\mu m$ ). Observation at the optical microscope took place soon after the polishing procedure; a picture is shown in Fig. 2. The presence of alteration layers on surface can be clearly seen: this phaenomenon is rather common in ancient coins  $^{12}$ , due either to intentional processes or to the natural soil corrosion. Microanalysis (EDX) measurements carried out with a Cambridge Stereoscan S360 equipped with an Oxford Inca X-act SDD detector gave the results reported in Table 2 (for the areas indicated in Fig.2).



Fig. 2. Section of a late Celtic drachm attributed to the Veneti's tribe (coin coming from a private collection). A clear alteration layer can be seen on the coin surface.

Area of analysis	Ag (wt. %)	Cu (wt. %)	Sn (wt. %)	O (wt. %)
1	$54.2 \pm 0.4$	5.1 ± 0.2	$16.6 \pm 0.3$	$24.0 \pm 0.5$
2	26.5 ± 0.2	$66.2 \pm 0.3$	4.8 ± 0.2	$2.5 \pm 0.2$
3	$25.0 \pm 0.2$	56.4 ± 0.3	5.9 ± 0.2	12.7 ± 0.3

Table 2. Microanalysis results obtained on the section. The areas are those indicated in Fig. 2.

It is therefore apparent that the presence of this layer, and its incredibly variable thickness within the sample, dismisses the use of surface techniques which could not provide representative compositional results for the bulk. For this reason, a neutronbased technique, i.e. the Prompt Gamma Activation Analysis (PGAA) has been chosen for this work. Indeed, PGAA provides average values of the entire volume and therefore the contribution of corroded areas is minimized. This technique has already been used for compositional analysis of ancient coins in the same facility.<sup>22,23</sup> PGAA is based on the radiative neutron capture process, i.e. the immediate emission of energetic gamma rays from the compound nucleus formed after the neutron is captured in the atomic nucleus. The emitted  $\gamma$ -radiation is characteristic, so qualitative and quantitative element analysis can be done using the energy and intensity of the radiation. The gamma-rays with MeV energies are not significantly absorbed within the sample and therefore can be detected with semiconductor HPGe detectors. In PGAA the detection of the gamma rays takes place during the irradiation with neutrons. When applying PGAA, one has to assess the potential risk of the samples activation due to neutron irradiation. Under the present measurement conditions (beam flux and time), however, the induced activity decays well below the clearance levels within a few hours and therefore is not a limiting factor for the analysis of cultural heritage artefacts.

On the other hand, complementary X-ray fluorescence measurements have been carried out to check the presence of tin (or other minor elements), as due to the highly different sensitivities the detection of this metal inside a silver matrix is rather difficult with PGAA. It must be stated that XRF results are not representative of the entire volume of a coin, because of

enrichment, alterations and the limited penetration depth of fluorescence photons. Despite these limitations, the aim was to simply indicate either the presence or absence of tin in the alloy: this is the reason why arbitrary units (a.u.) for tin instead of percent values are reported.

#### **Prompt Gamma Activation Analysis**

The PGAA measurements took place at the neutron-induced prompt gamma spectroscopy (NIPS) facility<sup>24,25</sup> at the Budapest Neutron Centre (Hungary). Here 6-7 samples were placed vertically into pockets made of Teflon foil fixed on an Al sample frame. A computer controlled sample stage made possible to run unattended batch runs with predefined counting times. The first set of measurements were done with a Compton-supressed 23% relative efficiency HPGe detector inside lead passive shielding, whereas the second set was done using a low energy germanium detector (LEGe). This latter has a significantly better energy resolution, but lower efficiency for gammas above 1 MeV. Its application for the Ag-Cu matrix is advantageous, as both Ag and Cu elements have their most intensive prompt-gamma lines at low energies, so reliable analysis can be done using this region of the spectrum only. Moreover, the complicated spectral interferences can be much better resolved. In both cases the maximum available dimensions of the incident neutron beam were 400×400 mm<sup>2</sup>. In order to achieve optimal counting rates with the detector electronics, different neutron apertures were applied. In the case of the first setup 5-10 mm<sup>2</sup> spots were irradiated on the coins along their total depths, while with the LEGe detector the beam could be increased to analyse the whole coin volume (60-270 mm<sup>2</sup> areas). The collimation of the gamma photons was made with a cylindrical lead collimator of 30 mm diameter.

The PGAA analytical signal comes from the radiative neutron capture process. The copper was analysed using the 278 keV gamma ray emerging from the  $^{63}\text{Cu}(n,\gamma)^{64}\text{Cu}$  reaction, whereas the silver content was derived from those of the  $^{109}\text{Ag}(n,\gamma)^{110}\text{Ag}$  reaction. In case of silver, the most important line was at the 198 keV, but several lines could be involved in the calculation to make the results more reliable. Spectrum fitting was done by a dedicated software based on the algorithms of HYPERMET PC<sup>26</sup>, allowing library-driven fit of preselected regions for a set of similar spectra. The elemental concentrations were determined by ProSpeRo Excel macro utility.  $^{27}$ 

#### X-ray fluorescence measurements

The XRF measurements were carried out with an Innov-X Olympus Delta Premium type handheld XRF spectrometer, which is ideally suited for non-destructive and instant alloy composition analysis. This XRF analyser is equipped with a 4W Rh anode X-ray tube. The appropriate working temperature is provided by thermoelectric Peltier cooling system. The fluorescent X-ray photons are collected with a large area silicon drift detector (SDD).

For the analysis of the silver coins, Alloy Plus mode with Precious metals addition was used. In this operating mode, the device uses two beams with different voltages, the first of 40 kV and 100  $\mu A,$  followed by a second beam with 8 kV and 200  $\mu A.$  With the alloy mode, metals between Ti and Bi can be analysed. For the quantitative determination, Olympus' own peak-fitting and de Jongh algorithms were employed via the embedded

software of the Innov-X equipment, utilizing the Fundamental Parameters approach.

During the XRF measurement session, the same size of X-ray beam (collimated, 3 mm diameter) was used, and the possibly most flat surfaces of the coins (woman profile faces) were chosen for elemental analysis.

#### Results and discussion

The coins from the HNM have been measured in two different sessions with two different approaches and detectors. The good efficiency of HPGe allowed to place a <sup>6</sup>Li plastic collimator to reduce the irradiated area of the coin (5 or 10 mm²). Nonetheless, the suspected heterogeneity of some samples led us to change our approach. To this aim, we analysed a second group of coins and a selection of those previously measured with the LEGe detector: in this modality, almost all the volume of the coins was irradiated, thus allowing to get an average result.

#### Homogeneity of samples

To check the homogeneity of the samples, we analysed a small group of coins twice, with the two different approaches. A comparison among results obtained with the two detectors is reported in Fig. 3. We found out that the volume analysed with the HPGe detector (a cylinder with a base diameter of 5 or 10 mm²) is representative of the total volume, measured with the LEGe detector. This is valid in all the cases, even for the samples suspected to be heavily silver-enriched on the surface. For this reason the difference among the results obtained with the two detectors will not be highlighted in the rest of the paper.

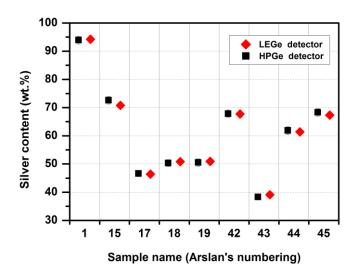


Fig. 3. Comparison of PGAA results obtained irradiating a small volume (HPGe detector) and the entire coin (LEGe detector). The numbering of samples is that of E.A. Arslan. <sup>5</sup> The error bars are in some cases smaller than the symbols.

#### **Quantitative analysis**

The analysis of PGAA spectra enabled the quantification of two of the most common metals used for coinage's alloys, i.e. silver and copper. All the results presented here are normalized and

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consider exclusively the presence of these two elements. The quantification method used in this work is based on the ko principle, where cross section ratios or mass ratios are calculated from the ratios of the peak areas. This model is valid for ideally thin samples, when a direct proportionality between  $\gamma$ -rays peak areas and masses is proved. In our case an additional correction for thicknesses and densities<sup>27</sup> has been applied, using an average value of density and the thickness measured directly on each single specimen. Since the density on the ancient coins has not been measured, we used a coin (25 cents, Netherlands, 1928) of mint-certified composition (Ag: 64%; Cu: 36%) and known thickness (1.8 mm) and density (9.94 g/cm<sup>3</sup>) to make some estimates. We considered different values of density, e.g. pure silver and pure copper, along with the lowest values measured on a group of ancient silver-copper alloy coins from a private collection (it does not lie between the pure elements density because of the presence of alterations phases and voids). We finally observed (Table 3) that these two physical parameters have a certain influence on the calculation of the composition. This enabled to define the absolute uncertainties for our samples, considering both the extrapolated<sup>28</sup> uncertainties for silver and copper (statistical error) and the contribution of thickness and density (systematic error). The latter is equal to the semidifference of the maximum and the minimum calculated value reported in Table 3. The total absolute uncertainty used for the ancient coins (Table 1) is therefore given by the square root of the quadratic sum of statistical and systematic errors.

	Lowest density for ancient samples (7.00 g/cm <sup>3</sup> )	Min. density  – pure copper (8.96 g/cm³)	Max. density – pure silver (10.49 g/cm³)	Real density (9.94 g/cm <sup>3</sup> )
Min. thickness (1.3 mm)	63.0 ± 0.6	63.2 ± 0.6	64.1 ± 0.6	$63.8 \pm 0.6$
Max. thickness (2.5 mm)	$63.4 \pm 0.6$	$63.6 \pm 0.6$	64.7 ± 0.6	64.5 ± 0.6
Real thickness (1.8 mm)	63.2 ± 0.6	$63.4 \pm 0.6$	64.4 ± 0.6	64.1 ± 0.6

Table 3. PGAA silver content (wt.%) and its absolute uncertainty of a mint-certified coin (Ag 64 wt.%) assuming different thicknesses and densities. Uncertainties reported in this case are the simple statistical errors.

#### Presence of tin

The aim of the present work was the quantification of silver content in the Cisalpine coins, in order to compare and investigate the devaluation among different emissions. The quantitative analysis has been performed considering a binary alloy, made of silver and copper. Nonetheless, the few compositional data available in literature 11 and some preliminary measurements on Cisalpine coins from other museums suggested the presence of non-negligible contents of a third element, the tin. Because of the difficulties in the determination of tin with PGAA technique, an XRF analysis was performed. In Fig. 4 the content of tin for different series is reported. The unit (a.u.) is an arbitrary one because these data can only give an order of

magnitude for the tin content, since XRF results are affected by enriched layers and alterations.

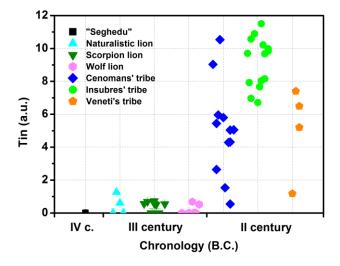


Fig. 4. Tin content (a.u.) for different Cisalpine emissions. The error bars are in some cases smaller than the symbols.

The XRF analysis also revealed, in all the samples, the presence of small amounts of gold and lead, commonly associated to the silver. Sulphur, phosphorous and iron have been detected only on some coins, and should be referred to alterations and soil deposits.

#### Composition of Cisalpine Gaul's coins

Results concerning the coins from the HNM are all listed in Table 1. These compositional results obtained with PGAA technique show very clearly the presence of a general silver debasement in the Cisalpine Gaul's coinage. In Fig. 5 the coins are grouped by typology and in a chronological order. Although we could not rescale the silver and copper content according to the XRF results for tin, which are surface values, the coins with a high amount of this last metal have been represented with empty points. Nevertheless, we can state that, according to the composition of the sectioned coin listed in Table 2 and the compositional data of three similar coins reported by Pautasso<sup>11</sup>, the average tin content varies between 4.8 and 9.6 wt.%. Considering these values, both silver and copper could be therefore lowered of quantities comprised between once or twice the experimental uncertainties.

The first imitations of the Massalian drachm, here represent by the "Seghedu" type drachm, show a substantial alignment with the official denomination of the Greek city<sup>29</sup> and therefore a ratio exchange of 1:1. The subsequent emissions, i.e. "naturalistic" and "scorpion lion" drachmas, which have some stylistic features in common, show comparable silver content, between 71 and 88 wt.%. A more careful stylistic analysis and a wider set of compositional data could even help to discriminate different productions within the two types. Moreover, the average weights of drachms of these typologies in different hoards are not consistent, and vary between 3.06 g (Casale and Serra Riccò hoards) and 2.76 g (Sassello hoard).<sup>30</sup>

The two following series, the "wolf lion" and the Cenomans' tribe types present values which are very spread: for the first one,

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specimens range from 84 towards 66 wt.%, while for the second from 78 down to 49 wt.%. This anomalous feature is particularly interesting, because it is highly improbable that a mint could have produced at the same time drachms of a same typology but with different silver content. These values must then be considered as an indication of the long-lasting production of some series, which suffered of an internal involution (compositional and stylistic as well). The Insubres' tribe emissions display all very similar compositions, grouped around an average value of 50% of silver, very close to the composition found by Pautasso. The Veneti's tribe coinage seem to have gone through an evolution and a devaluation as well, even if the number of specimens analysed id too small to draw general conclusions.

The last three coins, not shown in Fig. 5, belong to people settled in the Alpine areas. The coin Dess. 1038 represents a sub-group of the "scorpion lion" type, whose findings are concentrated in the Trentino region (north-east of Italy); the silver content and a low tin amount confirm their contemporaneity. Coins Dess. 1047 and Dess. 1048 are attributed to the Veragres' tribe, set in the Vallese area (south-west Switzerland). They display completely different compositional values (respectively 74% of Ag, 15 a.u. of Sn and Ag<LOD, 9.5 a.u. of Sn), indicating a deep evolution of this type, but also demonstrating an early and long-lasting diffusion of the monetary instrument even within tribes living in marginal areas of the ancient world.

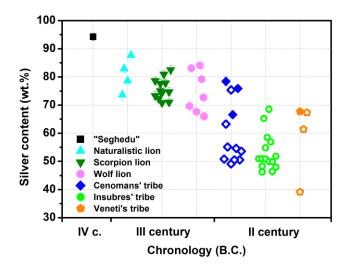


Fig. 5. Drachms grouped according to silver content and typology. Empty points indicate a high tin content (>4 a.u.).

#### **Conclusions**

 In this work 58 drachmas emitted by the pre-Roman tribes settled in northern Italy have been analysed. In order to get reliable data, a non-invasive bulk technique like PGAA has been used. Nevertheless, being the tin rather difficult to detect within a silver matrix, its presence has been checked with the XRF technique. The PGAA demonstrated to be a valuable technique for the analysis of silver alloys, providing fast and reliable results on a large number of specimens.

The compositional measurements on the HNM collection of Cisalpine Gaul's coins provided very important results from a historical point of view. For the first time, we observed the incredible variability among the different emissions. Our results show that the silver content falls from the 94% of the first emissions up to the 50% of the Cenomans' and Insubres' tribes late typologies. The compositional devaluation came also with a weight loss of around 45%, between the "naturalistic" and "scorpion lion" (3.06 g, Casale and Serra Riccò hoards) and the Insubres's tribe (1.65 g, Arslan's type XV, Treviglio hoard) drachms. This strong debasement took place between the III and the II century B.C., in a period of important political changes in northern Italy. It could be in particular put in relation with the military efforts of the Celtic tribes together or against Rome in the decades around the second Punic war.

At the same time, we observed the transition from the use of a binary silver-copper alloy to a ternary one, made of silver, copper and tin. The presence of tin can therefore be used to discriminate between the III and the II century B.C. productions. Moreover, this feature is particularly interesting for our knowledge of Celtic technological skills. Indeed, we can state that the introduction of tin is likely due to the colour properties of the ternary alloy, where the tin is used to compensate the reddish colour linked to the increase of copper.

In conclusion, all this information brought new light for the numismatics research, but are also useful to investigate economic processes during Iron Age, which are still nowadays largely unknown.

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#### **Notes and references**

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- D. R. Walker, *The Metrology of the Roman Silver Coinage*, British Archaeological Reports, Oxford, 1976.
- J. M. P. Cabral, M. G. Marques, M. F. Araujo, and J. R. Marinho, Metallurgy in Numismatics, 1998, 4, 472–482.
- 3. W. A. Oddy, *Metallurgy in Numismatics*, 1980, **1**, 99–118.
- W. A. Oddy and S. C. Munro-Hay, Metallurgy in Numismatics, 1980, 1, 73–82.
- M. Blet-lemarquand, G. Sarah, B. Gratuze, and J.-N. Barrandon, Cercetări Numismatice, 2009, XV, 43–56.
- E. R. Caley, in *Centennial Publication of the American Numismatic Society*, ed. H. Ingholt, New York, NY, 1958, pp. 167–180.

**Journal Name** 

- 7. M. F. Guerra, in *Radiation in Art and Archeometry*, eds. D. C. Creagh and D. A. Bradley, 2000, pp. 378–416.
- 8. A. A. Gordus, Archaeometry, 1967, 10, 78–86.
- 9. E. R. Caley, The Ohio Journal of Science, 1950, 50, 107–120.
- G. Sarah, B. Gratuze, and J.-N. Barrandon, J. Anal. At. Spectrom., 2007, 22, 1163–1167.
- A. Pautasso, Le monete preromane dell'Italia settentrionale, Centro di Studi Preistorici e Archeologici di Varese, Varese, 1966.
- 12. E. A. Arslan, Sibrium, 1995, XXII, 179-215.
- 13. E. A. Arslan, Études Celtiques, 1990, XXVII, 71–97.
- 14. E. A. Arslan, Numizmatikai Közlöny, XC-XCI, 9–33.
- M. Torbágyi, Sylloge Nummorum Graecorum. Hungary, Budapest, Magyar Nemzeti Múzeum, Vol. I: Hispania - Sicilia, Ennerre, Milano, 1992.
- 16. M. Dessewffy, Barbár pénzei I, Budapest, 1910.
- 17. M. Dessewffy, Barbár pénzei IV, Budapest, 1915.
- 18. M. Dessewffy, Revue Numismatique, 1914, 18, 170–180.
- 19. M. Dessewffy, Barbár pénzei III, Budapest, 1913.
- 20. J. Corsi and F. Barello, *De Nummis Gallicis. Mélanges de numismatique celtique offerts à Louis-Pol Delestrée*, 2013, 31–37.
- L. Beck, S. Bosonnet, S. Réveillon, D. Eliot, and F. Pilon, Nucl. Instruments Methods Phys. Res. Sect. B, 2004, 226, 153–162.
- Z. Kasztovszky, E. Panczyk, W. Fedorowicz, and Z. Révay, J. Radioanal. Nucl. Chem., 2005, 265, 193–199.
- 23. Z. Kasztovszky, Z. Révay, T. Belgya, and G. L. Molnár, *J. Radioanal. Nucl. Chem.*, 2000, **244**, 379–382.
- L. Szentmiklósi, Z. Kis, T. Belgya and A. N. Berlizov, J. Radioanal. Nucl. Chem. 2013, 298, 1605-1611.
- L. Szentmiklósi, T. Belgya, Z. Révay, and Z. Kis, *J. Radioanal. Nucl. Chem.*, 2010, 286, 501–505.
- B. Fazekas, J. Östör, Z. Kiss, A. Simonits and G. L. Molnár, J. Radioanal. Nucl. Chem. 1998, 215, 271-277.
- 27. Z. Révay, Anal. Chem. 2009, 81, 6851-6859.
- Z. Révay, Nucl. Instruments Methods Phys. Res. Sect. A, 2006, 564, 688–697.
- 29. J.-N. Barrandon and C. Brenot, *Mélanges l'École française de Rome*. *Antiquité*, 1978, **90**, 637–668.
- 30. J. Corsi, Numismatica Lovaniensia, 21, in press.

# Journal Name

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# **ARTICLE**

Description of HNM coins					Compositional results			
Sample code	Arslan's number	SNG number	Arlan's type	Main stream	Weight (g)	Ag (wt.%)	Cu (wt.%)	Tin
Dess. 1049	1	-	IV	"Seghedu"	3.63	94.2 ± 1.3	$5.8 \pm 0.2$	
Kiss 34	2	218	V	Naturalistic lion	3.66	82.9 ± 1.5	17.1 ± 1.1	
R.I. 4089	3	219	V	Naturalistic lion	3.21	87.7 ± 1.7	12.3 ± 1.3	
Kiss 34a	4	220	V	Naturalistic lion	3.05	$78.6 \pm 1.2$	$21.4 \pm 0.8$	
Dess. 34	5	221	VI	Naturalistic lion	2.32	$73.6 \pm 2.0$	$26.4 \pm 1.8$	
19.A 1914.2	6	231	VII	Scorpion lion	3.24	74.7 ± 1.3	25.3 ± 1.1	
Dess. 25	7	232	VII	Scorpion lion	3.21	71.0 ± 1.2	$29.0 \pm 1.0$	
19A.1914.1	8	233	VII	Scorpion lion	3.17	$72.3 \pm 1.3$	$27.7 \pm 1.1$	
Dess. 23	9	234	VII	Scorpion lion	3.14	$73.3 \pm 1.5$	$26.7 \pm 1.3$	
51A.1912.6	10	237	VII	Scorpion lion	3.05	$78.6 \pm 1.3$	21.4 ± 1.0	
Dess. 32	11	235	VII	Scorpion lion	3.04	77.8 ± 1.2	$22.2 \pm 0.8$	
51A.1912.1a	12	238	VII	Scorpion lion	2.99	74.4 ± 1.1	$25.6 \pm 0.8$	
Delhaes 12	13	239	VII	Scorpion lion	3.00	82.5 ± 1.6	17.5 ± 1.3	
Dess. 27	14	240	VII	Scorpion lion	2.96	80.9 ± 1.4	19.1 ± 1.0	
44.1949.10	15	236	VII	Scorpion lion	2.91	70.8 ± 1.1	29.2 ± 0.8	
19A.1914.5	16	241	VII	Scorpion lion	2.71	77.6 ± 1.4	22.4 ± 1.1	
17A.1991.1	-	-	VII	Scorpion lion	3.31	75.0 ± 1.1	$25.0 \pm 0.8$	
Dess. 1041	23	248	IX	Cenomans' tribe	2.71	54.6 ± 1.1	45.4 ± 1.4	•
22A.1914.1	24	249	IX	Cenomans' tribe	2.68	50.8 ± 1.0	49.2 ± 1.4	•
Delhaes 8	25	250	IX	Cenomans' tribe	2.65	53.6 ± 0.9	46.4 ± 1.3	•
22A.1914.3	26	251	IX	Cenomans' tribe	2.29	55.1 ± 1.2	44.9 ± 1.5	•
185A.1913.3	27	252	IX	Cenomans' tribe	2.25	50.5 ± 1.2	49.5 ± 1.5	•
22A.1914.2	28	253	IX	Cenomans' tribe	2.11	49.1 ± 1.0	50.9 ± 1.4	•
Delhaes 9	29	254	IX	Cenomans' tribe	1.76	50.5 ± 0.9	49.5 ± 1.3	•
Dess. 1046	30	255	IX	Cenomans' tribe	1.42	75.4 ± 1.1	24.6 ± 0.7	•
51A.1912.1b	31	256	X	Cenomans' tribe	2.55	75.9 ± 1.1	24.1 ± 0.7	
Dess. 24	32	257	X	Cenomans' tribe	2.17	63.2 ± 1.3	36.8 ± 1.3	•
95A.1907.176	33	258	X	Cenomans' tribe	2.16	66.6 ± 1.3	33.4 ± 1.2	
Delhaes 10	34	259	X	Cenomans' tribe	1.97	78.4 ± 1.6	21.6 ± 1.4	
Dess. 1054	17	242	XI	Insubres' tribe	2.42	46.4 ± 0.9	53.6 ± 1.4	•
22A.1914.6	18	243	XI	Insubres' tribe	2.27	50.8 ± 0.9	49.2 ± 1.3	•
Dess. 1055	19	244	XI	Insubres' tribe	2.18	50.9 ± 0.9	49.1 ± 1.3	•
22A.1914.7	20	245	XI	Insubres' tribe	1.95	50.9 ± 0.9	49.1 ± 1.3	•
22A.1914.8	21	246	XI	Insubres' tribe	1.90	54.8 ± 1.0	45.2 ± 1.2	•
Dess. 1057	22	247	XI	Insubres' tribe	1.78	68.5 ± 1.3	$31.5 \pm 1.2$	•
Dess. 1059	-	223	XI	Insubres' tribe	2.09	58.5 ± 1.0	$41.5 \pm 1.2$	•
Dess. 1058	-	222	XI	Insubres' tribe	2.15	48.3 ± 0.9	$51.7 \pm 1.4$	•
185A.1913.1	-	227	XIII	Insubres' tribe	1.93	51.8 ± 0.9	$48.2 \pm 1.3$	•
22A.1914.4	-	228	XIII	Insubres' tribe	1.85	$46.2 \pm 0.9$	53.8 ± 1.4	•
53A.1913.1		224	XIII	Insubres' tribe	2.43	49.9 ± 0.9	50.1 ± 1.3	•
22A.1914.5	-	225	XIII	Insubres' tribe	2.43	49.9 ± 0.9 47.9 ± 1.1	50.1 ± 1.5	•
R.I.4092	-	226	XIII	Insubres' tribe	1.98	$47.9 \pm 1.1$ $56.9 \pm 1.0$	$32.1 \pm 1.3$ $43.1 \pm 1.2$	•
		229					ļ l	•
185A.1913.2	-		XIII	Insubres' tribe	1.59	$65.2 \pm 1.0$	34.8 ± 1.0	
95A.1907.177	- 25	230	XV	Insubres' tribe	1.86	49.8 ± 0.9	50.2 ± 1.3	•
53A.1913.2	35	260	XVI	Wolf lion	2.91	72.7 ± 1.1	27.3 ± 0.8	
19A.1914.3	36	261	XVI	Wolf lion	2.73	69.6 ± 1.8	30.4 ± 1.7	
Dess. 1039	37	262	XVI	Wolf lion	2.71	$67.6 \pm 1.0$	$32.4 \pm 0.9$	

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53A.1913.3	38	263	XVII	Wolf lion	3.23	$79.2 \pm 1.1$	$20.8 \pm 0.6$	
19A.1914.4	39	264	XVII	Wolf lion	2.78	66.0 ± 1.4	$34.0 \pm 1.4$	
Dess. 1040	40	265	XVII	Wolf lion	2.23	83.1 ± 1.2	$16.9 \pm 0.5$	
Delhaes 11	41	266	XVII	Wolf lion	1.86	84.1 ± 1.2	$15.9 \pm 0.5$	
R.I.4088	42	267	XVIII	Veneti's tribe	2.50	67.7 ± 1.3	$32.3 \pm 0.9$	•
Delhaes 704	43	270	XIX	Veneti's tribe	2.51	$39.1 \pm 0.8$	$60.9 \pm 1.5$	•
Dess. 1042	44	269	XIX	Veneti's tribe	2.22	61.4 ± 1.0	$38.6 \pm 1.1$	•
Delhaes 13	45	268	XX	Veneti's tribe	1.93	67.3 ± 1.0	$32.7 \pm 0.9$	•
Dess. 1038	46	271	XXI	Tribe from Trentino ?	2.46	$79.0 \pm 1.4$	$21.0 \pm 1.1$	
Dess. 1047	47	272	XXII	Veragres' tribe	1.43	74.3 ± 1.7	$25.7 \pm 1.6$	•
Dess. 1048	-	-	XXII	Veragres' tribe	1.53	0 *	100	•

Table. 1. A complete list of the coins from the HNM analysed in this work, with museum and publications codes, technical details and Ag-Cu contents with absolute uncertainties. The symbol  $\bullet$  indicates a tin amount > 4 a.u. The second column refers to the E.A. Arslan numbering<sup>5</sup>, while the third column reports the Sylloge Nummorum Graecorum (SNG) numbering.<sup>6</sup> For the last coin (\*) the Ag content is below LOD.