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Lead Isotope Ratios in Archaeological Provenance: LA-ICP-MS Comparative Analysis

Relações Isotópicas de Chumbo na Proveniência Arqueológica: Análise Comparativa por LA-ICP-MS

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ABSTRACT

The correlation between an element and its original source (geogenic or anthropogenic) can be studied through the isotopic composition of lead (Pb), which remains unchanged by physical or chemical processes. This study used laser ablation coupled with inductively coupled plasma mass spectrometry (LA-ICP-MS) to identify concentrations of lead isotopes $^{208}\mathrm{Pb}$, $^{207}\mathrm{Pb}$, $^{206}\mathrm{Pb}$, and $^{204}\mathrm{Pb}$ in three coins attributed to the Roman Empire. The objective was to identify the geographical origin of the raw materials utilized or that contaminated these artifacts. Pb isotope ratios were analyzed using Neptune and Element 2 instruments from Thermo™ and compared with the Oxford Archaeological Lead Isotope Database (OXALID). The calibration of the Neptune commenced with the NIST 610 standard, followed by 45 blank readings and subsequent sample readings. For the Element 2, 20 points per sample were selected, with measurement times of 25 seconds for the blank and 36 seconds for data acquisition, including the NIST 610 standard. The results show that the samples have residual lead isotopic ratios compatible with the OXALID database for the Roman Empire region, indicating possible authenticity. Both instruments showed good agreement, with the Element 2 model offering advantages in statistical analysis and lower sensitivity to sample contamination.

keywords isotopic composition, laser ablation, archaeological artifacts, lead isotopes, provenance analysis

RESUMO

A correlação entre um elemento e sua fonte original (geogênica ou antropogênica) pode ser estudada através da composição isotópica do chumbo (Pb), que permanece inalterada por processos físicos ou químicos. Este estudo utilizou ablação a laser acoplada à espectrometria de massa com plasma indutivamente acoplado (LA-ICP-MS) para identificar as concentrações de isótopos de chumbo $^{208}\mathrm{Pb}$, $^{207}\mathrm{Pb}$, $^{206}\mathrm{Pb}$ e $^{204}\mathrm{Pb}$ em três moedas atribuídas ao Império Romano. O objetivo foi determinar a provável origem geográfica da matéria-prima desses artefatos. As razões isotópicas do chumbo foram analisadas utilizando os instrumentos Neptune e Element 2 da Thermo™ e comparadas com o Banco de Dados de Isótopos de Chumbo Arqueológicos de Oxford (OXALID). As medições no Neptune começaram com a calibração usando o padrão NIST 610, seguidas por 45 leituras de branco e amostras. Para o Element 2, foram escolhidos 20 pontos por amostra, com tempos de medição de 25 segundos para o branco e 36 segundos para aquisição de dados, incluindo o padrão NIST 610. Os resultados mostram que as amostras têm proporções isotópicas residuais de chumbo compatíveis com o banco de dados OXALID para a região do Império Romano, indicando possível autenticidade. Ambos os instrumentos demonstraram boa concordância, com o Element 2 mostrando vantagens na abordagem estatística e menor sensibilidade à contaminação das amostras.

palavras-chave composição isotópica, ablação a laser, artefatos arqueológicos, isótopos de chumbo, análise de procedência

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Introduction

Long half-life radioactive decay systems are the only ones capable of providing reliable data for quantifying geological time and organizing the geological record coherently. Among these, the Uranium-Lead decay system has been particularly important for geological studies, as the half-life of natural uranium radionuclides is long enough to encompass the entire history of the Earth and short enough for radioactive and radiogenic isotopes to be quantified in uranium-concentrating minerals (Davis et al., 2003).

The isotopic composition of lead (Pb), which remains stable under most physical and chemical conditions, has proven to be a reliable proxy for investigating the provenance of archaeological materials, particularly in distinguishing geogenic from anthropogenic sources. In this study, three coins attributed to the Roman Empire, selected based on stylistic and archaeological criteria, were analyzed using Laser Ablation coupled with Inductively Coupled Plasma Mass Spectrometry (LA-ICP-MS), a technique recognized for its precision in trace element and isotopic ratio measurements (Košler & Sylvester, 2003; Nielsen & Harris, 2015).

The isotopic ratios $^{208}\mathrm{Pb}$, $^{207}\mathrm{Pb}$, $^{206}\mathrm{Pb}$ and $^{204}\mathrm{Pb}$ were quantified using two high-resolution instruments (Neptune and Element 2, both from Thermo™), and the results were compared with reference data from the Oxford Archaeological Lead Isotope Database (OXALID). Developed to support archaeometric research, OXALID compiles isotopic signatures from mining regions across Europe and the Middle East, with particular emphasis on Roman-era sites that produced metals for coinage, namely silver, gold, copper, and lead.

Given the frequent co-occurrence of lead in silver ores and the inclusion of select iron mines, the database offers a robust framework for provenance studies (Baxter et al., 2000; Mower et al., 2015). The widespread presence of lead as both a contaminant and a constituent in ancient artifacts further underscores the relevance of isotopic analysis in archaeological science (Albarède, 2009; Bollhöfer et al., 1999).

This research builds upon established methodologies by integrating advanced mass spectrometric techniques and comparative database evaluation to refine provenance analysis. The specific objectives are: (i) to determine the isotopic composition of Pb in Roman coins using LA-ICP-MS, (ii) to compare these signatures with those cataloged in OXALID to infer the geographic origin of the raw materials, and (iii) to contribute to the broader understanding of Roman metallurgical networks through high-resolution analytical approaches.

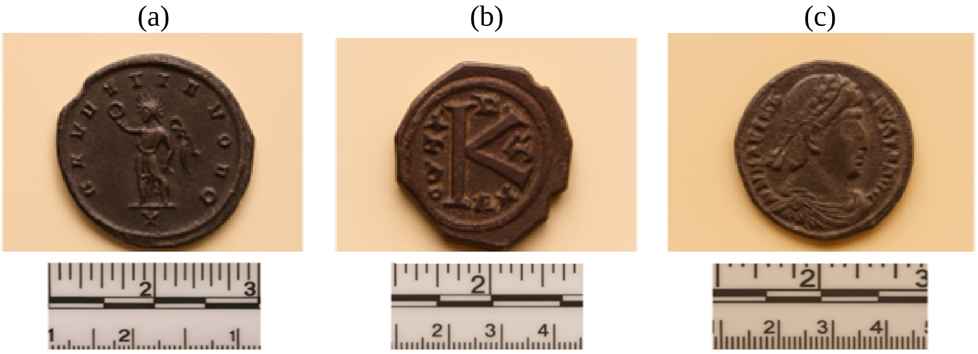
Materials and methods

The Roman coins analyzed in this study are part of a small set incorporated into the university's research collection. The artifacts have undergone internal evaluation by the responsible department and are not officially registered as heritage assets, having been made available exclusively for academic purposes.

The samples include specimens of Constantine II (bronze AE3, minted in Heraclea, 324 AD), Gallienus (silvered antoninianus, minted in Antioch, 260–268 AD), and Valens (bronze AE3, minted in Siscia, 364–367 AD). The pieces are in a good state of conservation (VF to cVF), presenting characteristic imperial iconography on the obverse and reverse.

The coins, dated between the 3rd and 4th centuries AD, are illustrated in Figure 1.

Figure 1 - Roman coins from the period of Constantine: (a) Constantino II (coin 1), (b) Gallienus (coin 2), and (c) Valens (coin 3).



A small portion of each coin specimen, Figure 1(a) to 1(c), was carefully scraped for analysis to remove surface oxides and contaminants, and subsequent analyses were targeted at these prepared regions.

Table 1 provides the characteristics of the coins, including parameters such as diameter, mass, and predominant composition. These variations in composition and dimensions are indicative of the diverse materials employed in coin minting during the Roman period. The historical data, including the approximate period and the issuing emperor for each coin, are crucial for contextualizing the lead isotopic signatures obtained.

Table 1 - Characteristics of the coins.

Coin	Constantine II	Gallienus	Valens
Smaller diameter (mm)	19.85	19.79	17.36
Larger diameter (mm)	21.48	21.75	17.89
Mass (g)	4.012	5.081	2.187
Main composition	Copper	Bronze	Copper
Probable time period	Fourth Century	Fourth Century	Fourth Century

The analyzed coins, identified based on the accompanying documentation, are described as follows:

- **Constantine II (coin 1):** Bronze AE 3, with a mass of 2.808 g and a diameter of 20.1 mm, minted in Heraclea (324 A.D.). The obverse (O: *CONSTANTINVS VN NOB C*) features a laureate, draped, and cuirassed bust right. The reverse (R: *DOMINOR NOSTROR CAESS, VOT V in wreath, SMHT in ex.*) displays the dedication. Approximate date: May 22, 337 – March/April 340 A.D. Conservation state: VF (Very Fine).
- **Gallienus (coin 2):** Silvered antoninianus, with a mass of 4.017 g and a diameter of 21.2 mm, minted in Antioch (260–268 A.D.). The obverse (O: *GALLIENVS AVG*) features a radiate, draped, and cuirassed bust right, seen from behind. The reverse (R: *VICTORIA AVG, Victory advancing left holding wreath in right hand and palm in left*) features the personification of Victory. Approximate date: August 253 – March 24, 268 A.D. Conservation state: VF (Very Fine).
- **Valens (coin 3):** Bronze AE 3, with a mass of 2.194 g and a diameter of 17.8 mm, minted in Siscia (364–367 A.D.). The obverse (O: *D N VALEN-S P F AVG*) features a pearl-diademed, draped, and cuirassed bust. The reverse (R: *GLORIA ROMANORVM, Valens advancing right, with labarum (Chi-Rho Standard) in left, dragging a captive*) presents the military theme. Approximate date: March 28, 364 – August 24, 367 A.D. Conservation state: cVF (Choice Very Fine).

The analysis of the artifacts, Figure 1, was based on the detailed properties of the coins, as well as the specifications of the G2 laser used for laser ablation, Figure 2.

Figure 2 - G2 laser ablation.

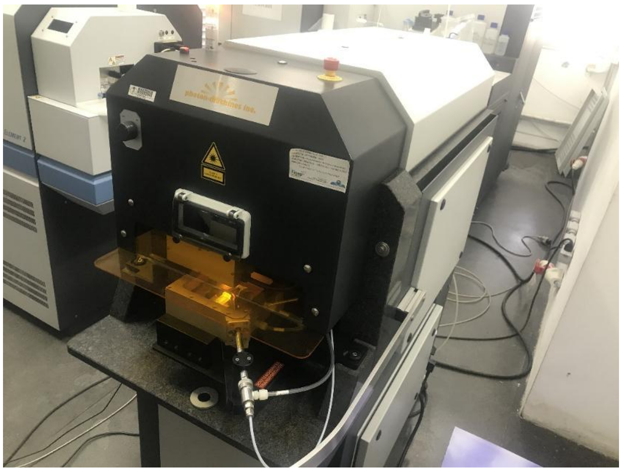


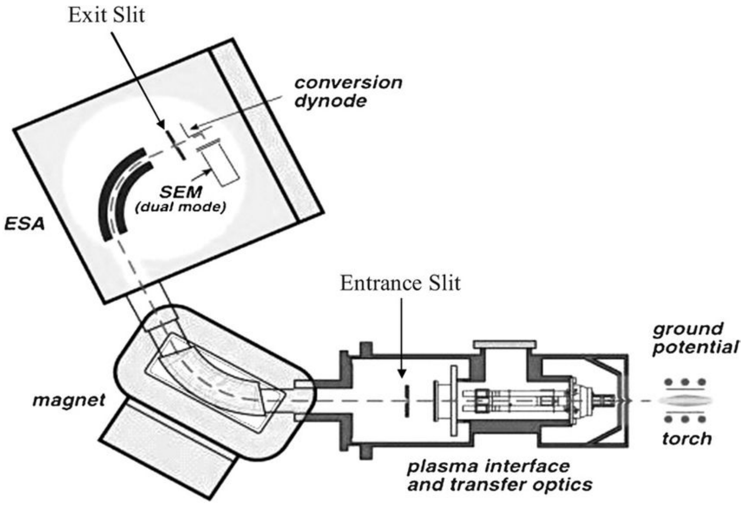
Table 2 describes the technical specifications of the G2 laser used for sample collection. Typical parameters used in this work were: ablation crater diameter of 50 μm; pulse repetition rate of 6 to 8 Hz; fluence of 2 J/cm^2; and irradiance of approximately 4×10^8 W/cm^2.

Table 2 - Characteristics of the G2 Laser.

Features	Specifications
Compact	Air-cooled
Pulse Length	< 5 ns
Pulse-to-Pulse Stability	$< 2\%$ RMS
Energy Density	Up to 15 J/cm^2
Repetition Rate	1 – 300 Hz
Firing Mode	Single, continuous shot, fixed dose

The Finnigan Element 2 is a high-performance Inductively Coupled Plasma Mass Spectrometry (ICP-MS) spectrometer, with a double-focusing magnetic sector field, for precise elemental analysis. Operating at 8000 V, it offers exceptional sensitivity, low detection limits, and automated tuning for reliability. Supporting multi-elemental analysis, it ensures interference-free measurements across the periodic table, making it ideal for advanced research and industrial applications, as illustrated in Figure 3.

Figure 3 - Element 2 architecture.



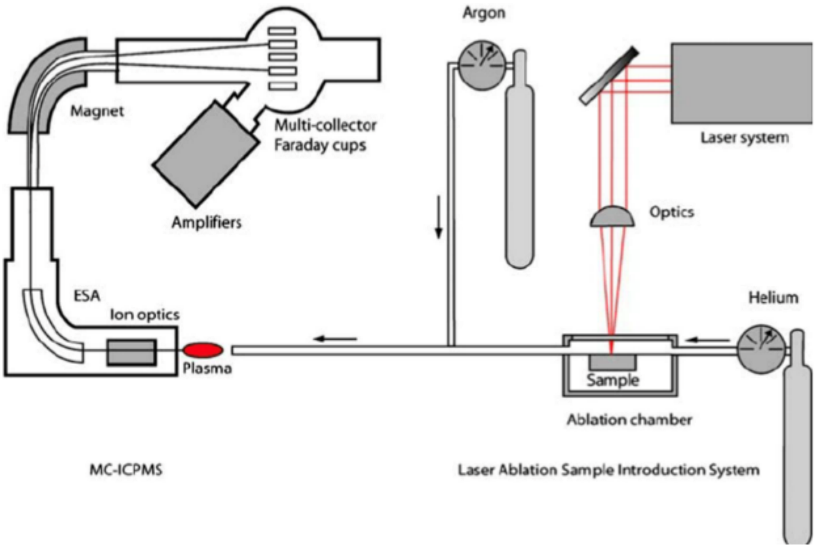
Note: Architecture of the Element 2 mass spectrometer. Schematic representation of the Element 2 system by Thermo™, used for high-resolution lead isotope analysis via inductively coupled plasma mass spectrometry (ICP-MS). The diagram highlights key components, including the sample introduction system, argon plasma source, double-focusing mass analyzer, and detector, illustrating the instrument's capability for precise isotopic measurements in archaeological materials. From "Architectural elements and examples", 2009, D. Large, J. Farmer. In D. Large, J. Farmer. Broadband cable access networks: the HFC plant (Cap. 10, pp. 237-266). Morgan Kaufmann.

The Neptune apparatus is basedon a so called Nier-Johnson reverse architecture, Figure 4, which is known for its ability to maximize efficiency and precision in isotopic and elemental analysis of samples. This architecture is particularly effective in multi-collector configurations, allowing the simultaneous collection of multiple isotopic ratios in a single measurement cycle.

This leads to a substantial enhancement in both speed and accuracy of measurements, which are essential when conducting high-complexity analyses, such as those conducted on archaeological artifacts and geological materials, which are crucial when dealing with high-complexity analyses, such as those performed on archaeological artifacts and geological materials (Calcagnile et al., 2005).

Additionally, the reverse ion optics of this architecture minimize interference and improve the reproducibility of data. This combination of factors makes the ICP-MS system with Nier-Johnson reverse architecture an ideal choice for studies that require detailed and reliable analysis of samples with low contamination or very

Figure 4 - Nier - Johnson reverse architecture.



Note: Nier–Johnson reverse geometry architecture. Schematic representation of the Nier–Johnson reverse geometry mass spectrometer, featuring a magnetic sector followed by an electrostatic analyzer. This configuration enhances mass resolution and energy focusing, making it particularly effective for precise isotopic measurements in high-sensitivity applications such as archaeometric analysis. From "Architectural elements and examples", 2009, D. Large, J. Farmer. In D. Large, J. Farmer. Broadband cable access networks: the HFC plant (Cap. 10, pp. 237-266). Morgan Kaufmann.

low concentrations of elements, such as those used in archaeometric and environmental studies.

ICP-MS in archaeometry

In addition to the individual advantages of each instrument, that is, the Neptune and the Element 2, the combination of their results provides a more comprehensive view of the different capabilities and limitations of each model (Albarède et al., 2020). This comparative approach is essential for a deeper and more detailed analysis of the samples, allowing for the maximization of the reliability of the lead isotopic results.

The Neptune, with its multi-collector configuration, is designed to measure isotopic ratios of trace elements in samples with high isotope concentrations or geologically complex compounds with high precision (Calcagnile et al., 2005). This equipment can perform rapid, simultaneous measurements of multiple isotopes, providing exceptional data resolution, which is crucial when tracing the geographical origin of metals used in ancient artifacts, such as Roman coins. However, due to its high sensitivity, the Neptune may be susceptible to interference from samples with contamination or signal variations, which could cause slight instability in measurements in a few cases.

On the other hand, the Element 2, a single-collector mass spectrometer, has the advantage of being more sensitive to low concentrations of elements and less susceptible to sample contamination, which is particularly important when dealing with fragile and potentially contaminated archaeological artifacts. Due to its single-collector configuration, this model performs measurements with complex data processing while providing greater accuracy in samples with higher concentrations and fewer interferences (Albarède, 2009).

The parallel application of both instruments and the direct comparison of the collected data allow for robust validation of the lead isotopic signatures in each sample. By using both systems, it is possible to optimize the precision of the measurements by leveraging the complementary strengths of both instruments. Neptune provides a more comprehensive analysis of isotopic signatures, while the Element 2 offers greater reliability and accuracy in samples with both higher and lower concentrations, minimizing errors due to contaminants or instrumental variations.

This process of validating data obtained from two distinct instruments not only strengthens the interpretation of the geological sources of lead used in Roman coins but also helps confirm the reproducibility of the results and the consistency of the measurements. This multi-instrument strategy is especially important in archaeometric studies, where the precision and reliability of data are critical to correctly interpreting the geographical origins of materials, shedding light on trade and production practices in the Roman Empire.

In summary, the use of two ICP-MS instruments allows for a multifaceted and highly precise analysis, enabling data combination for a more secure and robust interpretation of lead isotopic signatures. This approach also provides a solid foundation for future archaeometric investigations, where comparing results from diverse sources and technologies can broaden the scope and depth of conclusions (Ojha, 2013; Pearson et al., 2022).

Data acquisition procedure

The data acquisition procedure consists of a series of laser shots under the following conditions:

(i) Measurements with NIST 610 calibration standards: To correct the counts generated by the detection system.

(ii) Measurements without sample (Blanks): For counting chemical elements already present in the system, such as mercury in the helium gas used for sample transport; this is the background.

(iii) Measurements with sample.

The experimental analyses were performed using two instruments, the Neptune and the Element 2, each selected for its specific analytical capability, as described in the following sections.

Neptune (MC-ICP-MS)

Neptune, a multi-collector inductively coupled plasma mass spectrometer (MC-ICP-MS). This equipment, illustrated in Figure 5, is used for high-precision measurements of isotopic ratios of trace elements.

Figure 5 - Neptune equipment.

The main operating parameters and acquisition conditions are summarized in Table 3.

Table 3 - Operating parameters and acquisition conditions — Neptune

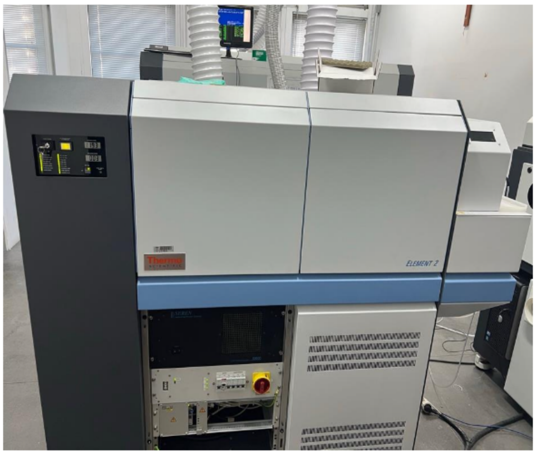
Parameter	Description
Calibration standard	NIST 610
Points per sample	14
Total analysis time	180 s (45 blanks + 45 samples)
Blank readings	45 readings, 2 s intervals
Sample readings	45 readings, 2 s intervals
Data format	Electron-volts, digitally converted to mass counts
Data processing	Excel spreadsheet (Farid model)

The values, shown in Table 3, were then consolidated in the equipment software for subsequent statistical analysis, using an Excel spreadsheet/Farid template.

Element 2 (ICP-MS)

Element 2, a single-collector inductively coupled plasma mass spectrometer (ICP-MS), is known for its high sensitivity and precision in analyzing elements at low concentrations, and its lower susceptibility to sample contamination, Figure 6.

Figure 6 - Element 2 equipment.



The corresponding operating parameters are listed in Table 4.

Table 4 - Operating parameters and acquisition conditions — Element 2

Parameter	Description
Points per sample	20
Blank acquisition time	25 s (average of 270 shots)
Sample acquisition time	36 s (average of 430 shots)
Data format	Electron-volts, digitally converted to mass counts
Data processing	Excel spreadsheet (Farid model)

The values, shown in Table 4, were then consolidated in the equipment's software for subsequent statistical analysis, using an Excel spreadsheet/Farid model.

Results and discussion

The proposed methodology offers dependable data for assessing lead isotope ratios in archaeometric samples. It seeks to resolve the challenges encountered with the NEPTUNE multi-collector equipment utilized in conjunction with the laser ablation system. In samples with high lead content and/or high concentrations of one of the lead isotopes, the apparatus's protection systems can momentarily deflect the ion beam (Calcagnile et al., 2005). This issue significantly increases measurement error, primarily occurring with laser ablation, which is the preferred and least invasive method for analyzing ancient object samples (Cabral, 1981).

This study compares the averages of Pb isotopic ratios obtained using two different instruments, Neptune and Element 2, with the values recorded in the Oxford Archaeological Lead Isotope Database (OXALID) and presented in Table 5. The comparison aims to verify the consistency and reliability of the isotopic measurements across different analytical platforms (Albarède et al., 2020). Both instruments showed good agreement with the OXALID database, validating the effectiveness of the laser ablation and ICP-MS techniques in analyzing Pb isotopic compositions in archaeological samples (Bliujienė et al., 2024; Bollhöfer et al., 1999).

Table 5 - Pb Isotopic Ratios for Each Point Analyzed in the Sample.

Instrument	**Point**	**Set 1**		**Set 2**		**Set 3**	
		208/206	207/206	208/206	207/206	208/206	207/206
Neptune	1	2.081	0.818	2.057	0.821	2.142	0.971
	2	2.093	0.860	2.123	0.832	2.351	0.881
	3	2.095	0.835	2.048	0.865	2.212	0.840
	4	2.067	0.851	2.021	0.827	2.167	0.817
	5	2.084	0.897	2.259	0.853	1.993	0.792
	6	2.181	0.850	1.989	0.848	2.139	0.852
	7	2.103	0.864	2.220	0.860	2.075	0.789
	8	2.059	0.850	1.997	0.795	1.957	0.781
	9	2.038	0.810	2.063	0.769	2.071	0.860
	10	2.087	0.868	2.081	0.812	2.098	0.847
	11	2.076	0.847	2.037	0.820	2.065	0.892
	12	2.083	0.881	2.054	0.885	2.059	0.879
	13	2.090	0.837	2.038	0.879	2.143	0.883
	14	2.110	0.863	2.079	0.941	2.027	0.978
Element 2	1	2.078	0.851	2.080	0.848	2.190	0.870
	2	2.107	0.895	2.092	0.854	2.173	0.878
	3	2.062	0.816	2.048	0.785	2.124	0.868
	4	2.090	0.894	2.072	0.831	2.101	0.859
	5	2.072	0.836	2.097	0.839	2.150	0.845
	6	2.050	0.827	2.076	0.845	2.168	0.837
	7	2.091	0.802	2.063	0.838	2.133	0.856
	8	2.085	0.823	2.062	0.832	2.141	0.888
	9	2.074	0.828	2.068	0.854	2.111	0.870
	10	2.092	0.836	2.069	0.852	2.070	0.879
	11	2.095	0.867	2.070	0.826	2.089	0.889
	12	2.091	0.885	2.067	0.889	2.060	0.872
	13	2.055	0.829	2.065	0.860	2.073	0.853
	14	2.085	0.852	2.058	0.833	2.031	0.879

Additionally, the results highlight the advantages and limitations of each instrument in terms of statistical analysis and sensitivity to sample contamination, as presented in Table 6.

Table 6 - Averages of Pb Isotopic Ratios for Each Coin.

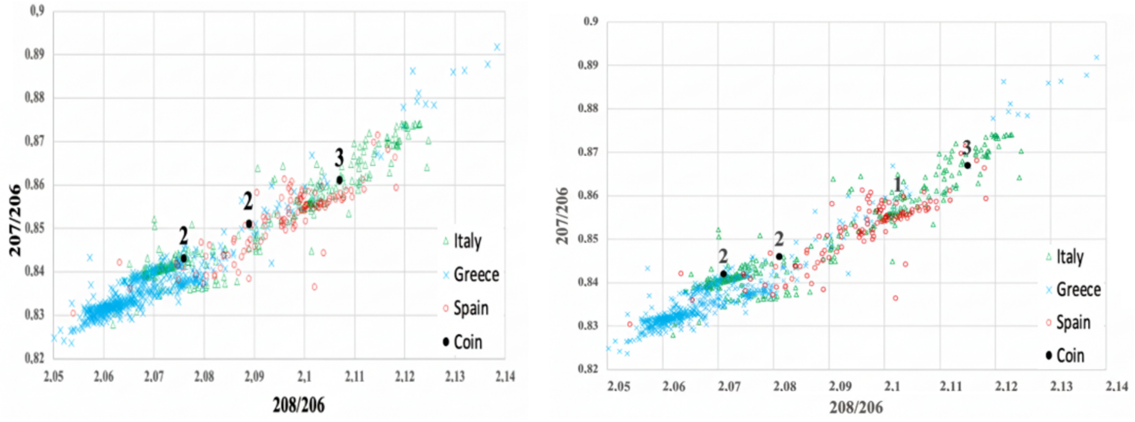
Coin	**Neptune**		**Element 2**	
	208/206	207/206	208/206	207/206
1	2.089 ± 0.032	0.852 ± 0.023	2.081 ± 0.016	0.846 ± 0.029
2	2.076 ± 0.077	0.843 ± 0.043	2.071 ± 0.013	0.842 ± 0.023
3	2.107 ± 0.098	0.862 ± 0.060	2.071 ± 0.016	0.867 ± 0.016

The slight variations in the isotopic ratios between the two instruments are expected, given the different configurations of the Neptune, Figure 5 (multi-collector) and Element 2 (single collector) instruments. The Neptune typically provides higher precision for high-concentration isotopic measurements, while the Element 2, Figure 6, is particularly sensitive to lower concentration samples and is less prone to contamination effects.

Comparison with OXALID database

Comparing the results, Table 5, with the OXALID database, which provides Pb isotopic signatures from Roman Empire-era mining regions, we observe a clear alignment of the isotopic ratios with known Roman mining regions, presented in Figure 7.

Figure 7 - Comparison of the Averages of Pb Isotopic Ratios Obtained on Thermo Neptune and Thermo Element 2 with the oxalid database: (a) Neptune, (b) Element 2.


(a) (b)

The values observed for both instruments fall within the expected range for materials associated with Roman-era metallurgy, which supports the authenticity of the coins (Albarède, 2009; Albarède et al., 2020).

The Pb isotopic ratios obtained for Coins 1, 2,and 3, corresponding respectively to Constantine II, Gallienus, and Valens, are consistent with the typical isotopic signatures found in regions active during the Roman Empire, such as the Iberian Peninsula and areas around the Mediterranean (Albarède et al., 2012; Bliujienė et al., 2024).

Comparison and interpretation of Pb isotopic ratios

The scatter plot of Pb isotopic ratios (208/206 vs. 207/206), shown in Figure 7(a) for Neptune and Figure 7(b) for Element 2, demonstrates the distribution of isotopic ratios for various regions, with the data points for the analyzed coins marked as 1, 2, and 3.

Both instruments show that the analyzed coins (represented by black circles) cluster within the expected range of isotopic ratios from regions known for Roman metallurgy, especially from the Iberian Peninsula (Spain) and Mediterranean areas (Italy and Greece) (Albarède, 2009; Albarède et al., 2020). This alignment with known Roman mining signatures reinforces the authenticity of the coins, Figure 1.

The graph further reveals how the coins' isotopic ratios compare to regional signatures, reinforcing the conclusion that the materials used in the production of the coins came from sources consistent with Roman-era trade routes.

The isotopic ratios for the coins are closely aligned with the data points for Spain and Italy, particularly for the 208/206 ratio, suggesting that these regions were likely sources for the metals used in the coins analyzed in this study.

While the variations in isotopic ratios between the two ICP-MS instruments may appear minor, they highlight the complementary strengths of using multiple instruments for comparative analysis (Korf et al., 2019). The Neptune provided a more comprehensive overview of the isotopic signatures, while the Element 2 ensured greater reliability and precision for both higher and lower concentration samples, minimizing errors due to contamination or instrument variations (Davis et al., 2003; Gilbert, 2023).

In summary, the results obtained for the three Roman coins, when compared with the OXALID database and plotted alongside data from known Roman mining regions, confirm the authenticity of the coins and their geographical origin. The consistency in the results from both ICP-MS instruments, when cross-referenced with the database, strengthens the conclusion that these coins were produced using raw materials from established Roman sources. This multi-instrument approach, supported by the graph and database comparison, significantly enhances the reliability and accuracy of the results, providing a deeper understanding of the materials used in ancient Roman coinage (Bollhöfer et al., 1999).

Conclusions

This study effectively demonstrated the application of lead (Pb) isotope analysis in determining the geographical origin of materials utilized in Roman coinage. The results obtained from the Neptune and Element 2 instruments exhibited strong concordance, aligning with the data from the Oxford Archaeological Lead Isotope Database (OXALID), thereby reinforcing the authenticity of the analyzed coins. By leveraging the complementary strengths of both instruments, the study capitalized on the Neptune's comprehensive isotopic signature analysis and the Element 2's precision, particularly for samples with lower concentrations or potential contamination.

The Pb isotopic ratios of the coins were consistent with typical signatures from recognized Roman regions, such as the Iberian Peninsula and Mediterranean areas, thereby confirming their authenticity. The scatter plot of Pb isotopic ratios (208/206 vs. 207/206), which compared the results from both instruments with OXALID data, further substantiated this conclusion. The visual comparison of the isotopic ratios with established regional signatures enhanced confidence in the validity of the results.

In conclusion, the integration of laser ablation with inductively coupled plasma mass spectrometry (LA-ICP-MS), along with the use of multiple high-precision instruments and graphical analysis, establishes a robust methodology for analyzing the geographical provenance of archaeological materials. This approach not only enhances our understanding of Roman trade and production practices but also provides valuable insights for future archaeometric studies and the preservation of historical artifacts.

Author Contributions

W. M. Canto; A. D. Tavares Junior; M. C. Geraldes; C. C. G. Leitão, and M. H. P. A. Coelho contributed equally to all stages of the work, including conceptualization, methodology, analysis, investigation, writing (original draft and review), visualization, and validation.

Conflicts of Interest

The authors declare that they have no known competing financial interests or personal relationships that could have influenced the work reported in this paper.

References

Albarède, F. (2009). *Geochemistry: An Introduction* (2nd ed.). Cambridge University Press.

Albarède, F., Blichert-Toft, J., Gentelli, L., Milot, J., Vaxevanopoulos, M., Klein, S., Westner, K., Birch, T., Davis, G., & Callataÿ, F. (2020). A miner's perspective on Pb isotope provenances in the Western and Central Mediterranean. *Journal of Archaeological Science*, *121*, 105194. https://doi.org/10.1016/j.jas.2020.105194

Albarède, F., Desaulty, A. M., & Blichert-Toft, J. (2012). A geological perspective on the use of Pb isotopes in archaeometry. *Archaeometry*, *54*(5), 853–867. https://doi.org/10.1111/j.1475-4754.2011.00653.x

Baxter, M. J., Beardah, C. C., & Westwood, S. (2000). Sample Size and Related Issues in the Analysis of Lead Isotope Data. *Journal of Archaeological Science*, *27*(10), 973–980. https://doi.org/10.1006/jasc.1999.0546

Bliujienė, A., Vybernaitė-Lubienė, I., Biveinytė, V., Pudžaitis, V., Babenskas, E., & Petrauskas, G. (2024). Lead isotopes in the context of the provenance of copper alloys and mutability processes in Lithuania from the second half of the 1st century to the 13th century AD. *Archaeologia Baltica*, *31*, 103671. https://doi.org/10.15181/ab.v31i0.2667

Bollhöfer, A., Chisholm, W., & Rosman, K. J. R. (1999). Sampling aerosols for lead isotopes on a global scale. *Analytica Chimica Acta*, *390*(1-3), 227–235. https://doi.org/10.1016/S0003-2670(99)00182-8

Cabral, J. M. P. (1981). Arqueologia e arqueometria. *Arqueologia – Especial Métodos*, *4*, 8–11.

Calcagnile, L., Quarta, G., & D'Elia, M. (2005). High-resolution accelerator-based mass spectrometry: precision, accuracy and background. *Applied Radiation and Isotopes*, *62*(4), 623–629. https://doi.org/10.1016/j.apradiso.2004.08.047

Davis, D. W., William, I. S., & Krogh, T. E. (2003). Historical development of zircon geochronology. In *Reviews in mineralogy and geochemistry* (pp. 145–181, Vol. 53). https://doi.org/10.2113/0530145

Gilbert, C. (2023). *Methodological developments in high resolution mass spectrometry-based proteomics for the analysis of museum and archaeological objects: trace analysis and structural characterization.* [Theses Docteur Sciences Chimiques, Université de Bordeaux]. https://theses.hal.science/tel-04269646/document

Korf, A., Hammann, S., Schmid, R., Froning, M., Hayen, H., & Cramp, L. J. (2019). Digging deeper: a new data mining workflow for improved processing and interpretation of high-resolution GC-Q-TOF MS data in archaeological research. *Scientific Reports*, *9*(1), 1–12. https://www.nature.com/articles/s41598-019-57154-8.pdf

Košler, J., & Sylvester, P. J. (2003). Present trends and the future of zircon in geochronology: laser ablation ICPMS. In J. M. Hancher & P. J. Sylvester (Eds.), *Laser ablation–icpms in the earth sciences: Current practices and outstanding issues* (pp. 243–275, Vol. 53). Mineralogical Society of America.

Large, D., & Farmer, J. (2009). Architectural elements and examples. In D. Large, J. Farmer. *Broadband cable access networks: the HFC plant* (Cap. 10, pp. 237-266). Morgan Kaufmann.

Mower, J., et al. (2015). High-fidelity quantum state evolution in imperfect photonic integrated circuits. *Physical Review A*, *92*(3), 032322. https://doi.org/10.1103/PhysRevA.92.032322

Nielsen, S. G., & Harris, M. (2015). Advances in in situ trace element and isotope analysis by laser ablation ICP-MS. *Geostandards and Geoanalytical Research*, *39*(4), 341–354. https://doi.org/10.1111/j.1751-908X.2015.00310.x

Ojha, S. S. (2013). AMS: a novel method of dating in archaeology. *International Journal of Humanities and Religion*, *2*(2), 37–43. https://www.academia.edu/3768380/AMS_A_Novel_method_of_dating_in_Archaeology

Pearson, P. N., Botticelli, M., Ericsson, J., Olender, J., & Spruženiece, L. (2022). Authenticating coins of the 'Roman emperor' Sponsian. *PLoS ONE*, *17*(11), e0274285. https://doi.org/10.1371/journal.pone.0274285