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A numismatic study of Roman coins through X-ray fluorescence and X-ray computed µ-tomography analysis

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ABSTRACT
Thirteen of a group of thirty Roman coins, found in the so-called "Grotta delle Ninfe" near Cerchiara di Calabria (Calabria, Italy) and preserved in the Brettii and Enotri Museum showcase in Cosenza (Calabria, Italy) have been under archaeometric investigation. The coins have followed a degradation process due to the sulphurous water source near the discovery site. Due to a thick layer of corrosion products, the inscriptions are entirely unreadable. This paper aims to know the constituent material and find hidden signs or inscriptions on the coins using X-ray fluorescence (XRF) and X-ray microtomography (µ-CT). Employing the X-ray µ-CT, we made some inscriptions readable, and through a numismatic study, we learned the provenance of the coins and their period of manufacture.

Section: RESEARCH PAPER

Keywords: Roman coins; X-ray µ-CT; XRF spectroscopy; archaeometry; non-destructive investigations

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1. INTRODUCTION

At the base of Mount Sellaro, a limestone relief of the Pollino massif, there is the "Grotta delle Ninfe" (Figure 1), a cave of natural origin (Cerchiara di Calabria, Calabria, Italy). "Grotta delle Ninfe" is an ancient source of sulphurous water flowing at a nearly constant temperature of 30 °C. Thanks to the therapeutic effect of sulphurous water, this cave is used for healing baths, also known as "Grotta dei Bagni".

Archaeological studies [1] carried out at the "Grotta delle Ninfe" have demonstrated that in this place there was a continuity of use and attendance since Prehistory. This is confirmed by literary sources [1] and the natural context, which is characterised by caves and water sources. The cave looks like a votive deposit and a sacred place. Furthermore, the Sanctuary of the "Madonna delle Armi" (whose origin goes back to the 10th century and whose name recalls the presence of caves) indicates the persistence of worship in this area.

In 1905, a large group of oil lamps and coins from the Imperial Roman age were found in the "Grotta delle Ninfe" cave. These archaeological finds are now preserved at the Brettii and Enotri Museum in Cosenza (Calabria, Italy), and the coins are catalogued in the Coins Collection Catalogue [2], [3]. The type of oil lamps found is among the most widespread in Italy and the Mediterranean region between the Augustan age (44 BC - 14 AD) and the 2nd century AD [4]. At first, it was possible to relate the coins to either the Julio-Claudian dynasty (27 BC - 68 AD) or the Flavian dynasty (69 AD - 96 AD) based on the contextual discovery of the lamps. The long permanence of the Roman coins in the "Grotta delle Ninfe", characterised by a sulphurous

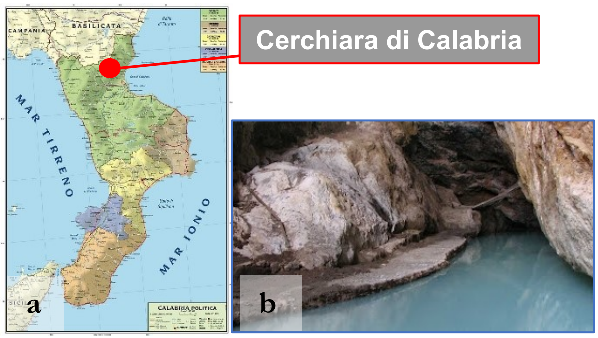

Figure 1. The red point represents the places where the coins under investigation were found (a); the interior of the "Grotta delle Ninfe" (b).

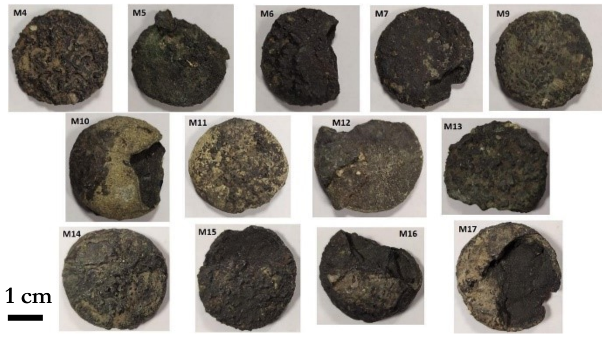

Figure 2. Coins investigated. It is clearly visible that the thick layer of degradation products covers the coins' surface.

water source, caused several chemical processes that formed a thick layer of black crust presumably made by copper sulphide minerals, like Chalcocite and Brochantite [5], [6]. Due to these thick layers of corrosion products, the inscriptions were entirely unreadable.

Through this study, we propose to identify better the minting period of the coins. To better understand these samples, it is worth clarifying that, due to devaluation and reforms made throughout history, the Roman monetary system underwent several changes. These changes concerned the nominal, their weight and the raw material used to mint them [7]. In this respect, Octavian inherited a highly complex economic system at the beginning of his principate. The long circulation of bronze coins, also of old denominations, and the standard changes of the *as* did not allow to have a guarantee on the weight. Moreover, the sestertius was struck in silver with reduced dimensions and weight, which often caused difficulty in the transactions. The production and circulation of gold and silver coins were encouraged to mitigate the situation. Still, due to the conflicts within the Res Publica and the mobile mints in the wake of the generals, these suffered devaluations [7]. After that, the princeps were thus forced to launch a radical monetary reform to restore solidity to a system in crisis. A "quadrimetallic" system has been imposed with gold, silver, orichalcum, and copper emissions [8]. The *as* (10.90 g) and its sub-multiples, previously struck in bronze, were replaced by pure copper emissions, while the multiples (sestertius 27.28 g and dupondius 13.64 g) were produced in orichalcum. The Senate was entrusted with the control of copper-based emissions and the *princeps* with the noble metal emissions. Very soon (about 12. A.D.), the names of the magistrates disappeared from the noble metal issues; later (about 4 BC), they were omitted from the copper-based ones but kept the initials S.C. *(Senato consultum)* [8]. The Augustan coinage [9] was an integral part of the propaganda, the emperor's portrait spread on the obverse of almost all issues, and the choice of reverse types was always well thought out to convey precise messages. The mint of Lugdunum specialised in producing noble metal specimens, while the mint of Rome oversaw copper-based ones. During 64 BC Nero was forced to implement a new monetary reform: the weight of the gold (Aureus) and silver (Denarius) emissions were substantially reduced and the dupondius introduced the iconography of the emperor with a radiate head, probably to better distinguish it from the ***as*,** seen the similar weight and diameter (the successors will maintain this custom) [8]. Later, under Vespasian and Titus, the Neronian system was maintained, while under Domitian, instead, there was a return to the Augustan standard. The Flavians' production focused on dynastic portraiture and architecture [10]. After the Domitian, Trajan restored the Neronian post-reform model.

In this work, we apply X-ray fluorescence spectroscopy (XRF) to identify constituent elements of the coins and employ the X-ray micro computed tomography (X-ray µ-CT) to determine the dimensions of coins and to individuate possible inscriptions.

2. MATERIALS AND METHODS

Non-invasive archaeometric investigations can better identify the coins' typology, characterise the alteration processes, and analyse the constituent materials of the coins. Chemical and morphological studies were conducted using an *X-ray fluorescence* portable spectrometer and an *X-ray microtomography* apparatus but at first, we carried out a visual investigation. We focused on thirteen coins in the collection mentioned above. The samples were labelled as: M4, M5, M6, M7, M9, M10, M11, M12, M13, M14, M15, M16, and M17 (see Figure 2).

Visual testing assessed that the coins were made in pure copper or copper alloy. The extended exposure of these specimens to sulphur-rich water springs has resulted in the formation of a thick layer of black crust through various chemical processes that make the inscriptions on the coins illegible, while deep cracks can be observed on some of them. Thus, the state of conservation of the recovered coins is so degraded that any sign or inscription is not visible to the naked eye.

The diameter, thickness, and weight of the coins are reported in the Table 1.

The XRF investigations presented in this paper were carried out by a "Tracer 4-sd" portable X-ray spectrometer from Bruker

Table 1. Diameter, thickness, and weight of coins.

Sample	Diameter ± 1 (mm)	Thickness ± 1 (mm)	Weight ± 1 (g)
M4	34	9	21
M5	30	7	14
M6	32	10	14
M7	33	8	14
M9	30	9	15
M10	31	9	13
M11	30	8	12
M12	31	8	11
M13	32	10	14
M14	31	10	18
M15	31	10	13
M16	30	11	13
M17	31	9	11

at the X-ray and Raman Spectroscopy laboratory of the Department of Biology, Ecology and Earth Sciences (University of Calabria). This technique is one of the most common and straightforward used in archaeometry studies [11], [12]. The XRF chemical investigation allows us to detect the coin's chemical nature and to understand the alteration processes. The parameters of the XRF measurements were set as follows: voltage 15 kV, current 35 μA, vacuum > 17 Torr, exposition time 100 s, and spot size 4 mm.

The objective of the XRF survey is to assess and compare the chemical composition of different samples, aiming to determine whether the coins are crafted from pure copper or a copper alloy. To make this determination, it's crucial to identify the presence or absence of secondary metals (like tin or zinc in bronze or orichalcum alloy, respectively) and estimate their approximate and relative quantities. It worth pointing out that the precision of the chemical composition analysis may be influenced by the presence of a corrosion layer, as well as the surface effects related to copper loss and zinc loss through decuprification and dezincification. [13], [14], [15].

The coins were first gently washed with a brush and distilled water and then with ethanol in order to remove the residual soil and to expose the coin superficial layer. Thus, three measurement points were acquired on each coin and the results of each measurement group were averaged to produce a more reliable data set. The samples were positioned directly on the detector windows.

We made both qualitative and semi-quantitative analyses. The concentration estimate of chemical elements can be calculated using direct comparison of count rates after determining calibration factor using Standard References (SUS, approximate composition of RC 36/16, high alloy, Cu-Base provided by Bruker). We relied on the Spectra 7 software (a Bruker proprietary software).

X-ray μ-CT, one of the most powerful imaging techniques for inspecting the internal morphology of an object, is a diagnostic method frequently used in different fields of science [16], [17], [18]. In particular, X-ray μ-CT plays an important role in the cultural heritage domain because it is a non-destructive and conservative technique [19], [20], [21], [22]. One of the advantages of X-ray μ-CT is that it does not require sample preparation. In addition, this technique allows us to inspect and characterise in three dimensions and with a micrometric resolution [23], [24], [25]. Microtomographic investigations were carried out at the μTomo1 experimental station @STAR Lab at the University of Calabria. The μTomo1 operation sequence to conduct the measurement, the virtual sample reconstruction and the data visualisation are described in M. Lopez-Prat et al. [20]. The X-ray μ-CT measurement parameters were set to: voltage 120 kV, current 83 μA, the exposure time for each projection of 2 s, and scaled pixel size 25 μm. The tomographic reconstructions were carried out by correcting the ring artefact and setting the beam hardening parameters [26], [27], [28].

3. RESULTS

Table 2 shows the sulphur and metallic element relative concentration determined on the coin's surface by XRF semi-quantitative analysis. The surface measurements revealed copper and sulphur presence on all coins except one, whose relative weights are between 80.2 % – 93.3 % and 4.7 % – 10.0 %, respectively, and other trace elements. Moreover, we have evidence that sample M12 is the only coin showing a significant percentage of Zinc (about 2.3 %).

Table 2. Values of concentration of the elements detected on coins' surface.

Sample	S (con. %)	Fe (con. %)	Cu (con. %)	Zn (con. %)
M4	6.5 ± 0.5	1.3 ± 0.3	92 ± 4	0.2 ± 0.1
M5	4.7 ± 0.4	1.4 ± 0.5	93 ± 3	0.3 ± 0.1
M6	6.7 ± 0.7	0.5 ± 0.0	92 ± 2	0.2 ± 0.1
M7	8.1 ± 0.7	0.3 ± 0.2	91 ± 4	0.2 ± 0.1
M9	5.5 ± 0.6	1.4 ± 0.1	92 ± 1	0.6 ± 0.1
M10	6.6 ± 0.7	0.5 ± 0.1	92 ± 4	0.2 ± 0.2
M11	5.3 ± 1.3	1.6 ± 0.5	93 ± 1	0.4 ± 0.2
M12	10 ± 0.3	6.4 ± 0.1	80 ± 2	2.3 ± 0.6
M13	6.6 ± 0.5	2.6 ± 0.4	90 ± 3	0.3 ± 0.1
M14	6.2 ± 0.2	0.5 ± 0.1	93 ± 3	0.3 ± 0.1
M15	5.6 ± 0.4	0.7 ± 0.2	93 ± 2	0.2 ± 0.2
M16	7.1 ± 0.5	0.3 ± 0.0	92 ± 2	0.1 ± 0.0
M17	7.6 ± 0.8	2.6 ± 0.2	89 ± 1	0.3 ± 0.1

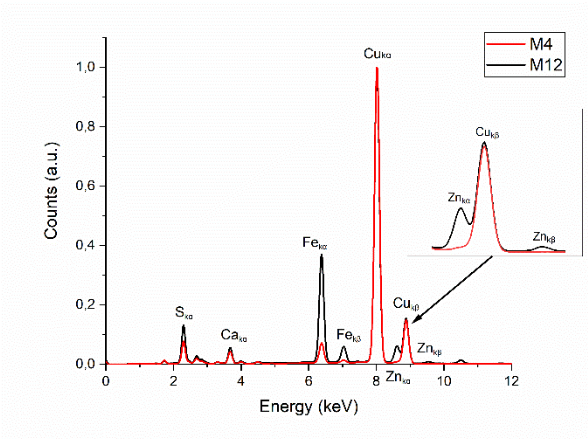

Figure 3. XRF spectrum of the M4 (red line) and M12 (black line) coins. Zoom of the zinc region is shown in the box.

Because the XRF spectra of all copper coins are very similar, we show in Figure 3 only those of the M4 coin (red line) and M12 coin (black line), where the zinc peak is clearly visible.

In the 3D virtual reconstruction of coins, obtained through X-ray μ-CT survey, shows the presence of a thick oxidation layer. Figure 4 shows an example of the transverse virtual sections of the M4, and Figure 5 shows a longitudinal virtual section of M6 coins. The outer sulphur-rich layer that covers the bulk of the coins is discernible. By X-ray μ-CT surveys, it was possible to measure the original thickness and diameters of the coins, which are reported in Table 3.

Moreover, the X-ray μ-CT surveys revealed signs and inscriptions under the outer sulphur-rich layer on only four coins: M4, M6, M7, and M9. Studying these inscriptions allowed us to recognise the type of coins.

4. DISCUSSION

The XRF survey allowed us to determine the raw material used to make the coins. Therefore, the presence of zinc in the M12 coin indicates that it is composed of an orichalcum alloy [29], while the M4, M5, M6, M7, M8, M9, M10, M11, M13, M14, M15, M16, and M17 coins are confirmed to be made of copper. Knowing the raw materials allowed us to focus the next work of the image comparison only on the reference coins with similar composition.

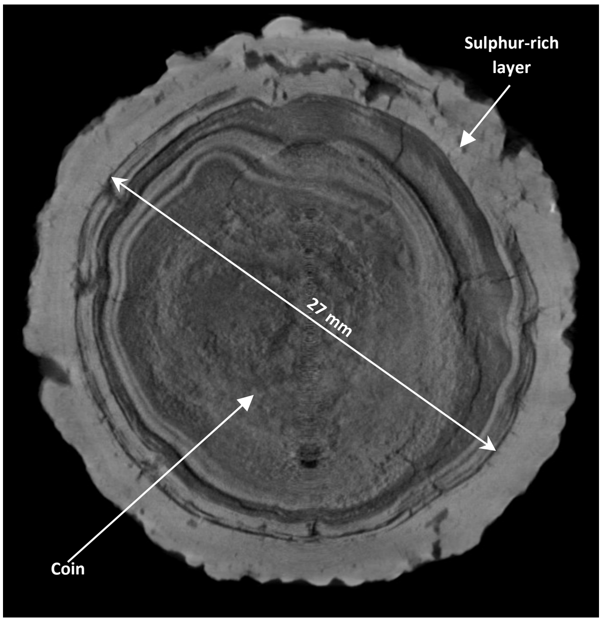

Figure 4. Transverse grayscale slice of the M4 coin.

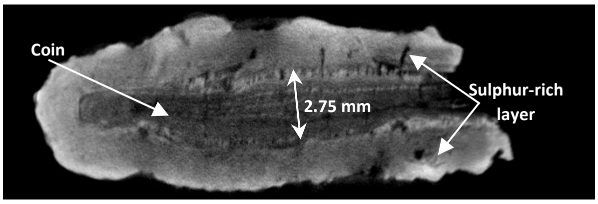

Figure 5. Longitudinal grayscale slice of the M6 coin.

Table 3. Diameter and thickness of coins without external crust.

Sample	Diameter ± 0.3 (mm)	Thickness ± 0.3 (mm)
M4	25.0	2.0
M5	26.6	1.6
M6	27.0	1.8
M7	28.4	4.4
M9	25.3	1.9
M10	26.7	2.5
M11	26.4	1.5
M12	27.0	2.0
M13	27.3	3.0
M14	26.3	2.0
M15	23.9	1.5
M16	24.0	2.3
M17	25.4	3.6

Figure 6 a and b show two grayscale virtual sections of the obverse (a portion) and reverse of M4 coin, respectively, in which several letters are visible. On the obverse, we can read "VNIC" (Figure 6a blue frame), while on the reverse side, we can read "FFPLVRIVSA" (Figure 6b red frame). A sign indicated by a yellow arrow in Figure 6b is also visible on the reverse side. Similar signs and letters are present on Augustus As coin, classified as Augustus n° 428 in RIC I classification date 7 BC [30], shown in Figure 6c. On the obverse is the head of August turned to the left and there is the inscription [CAESAR AVGVST PONT MAX TRI]BVNIC [POT]. On the reverse there is the inscription LVRIVS A[GRIPPA IIIVIR AAA] FFP, SC in the field.

Figure 7a shows grayscale virtual sections of a collage of different slices at different depths of the M6 coin's obverse. Figure 7b shows grayscale virtual sections of the reverse of the M6 coin. On the obverse, we can read "E[RVA] TRA[IA]N AVG GERM" (Figure 7a blue frame), while on the reverse, we can read "COS II" (Figure 7b red frame). The letters in square brackets are inferred by the distance between other letters and comparison with reference coins. These findings suggest that the M6 coin was manufactured during the reign of Trajan between 98–101 AD, and then it would be a Trajan As. The legend present on the obverse of this coin is typically "IMP CAES NERVA TRAIAN AVG GERM", while on the reverse is present "COS II" or "COS III". So, the reference coins could be Trajan n° 392-395-396-402-406-407-408 (for Cos II) or Trajan N°410-416-417-422-423-424-425 (for Cos III) in RIC II classification [30].

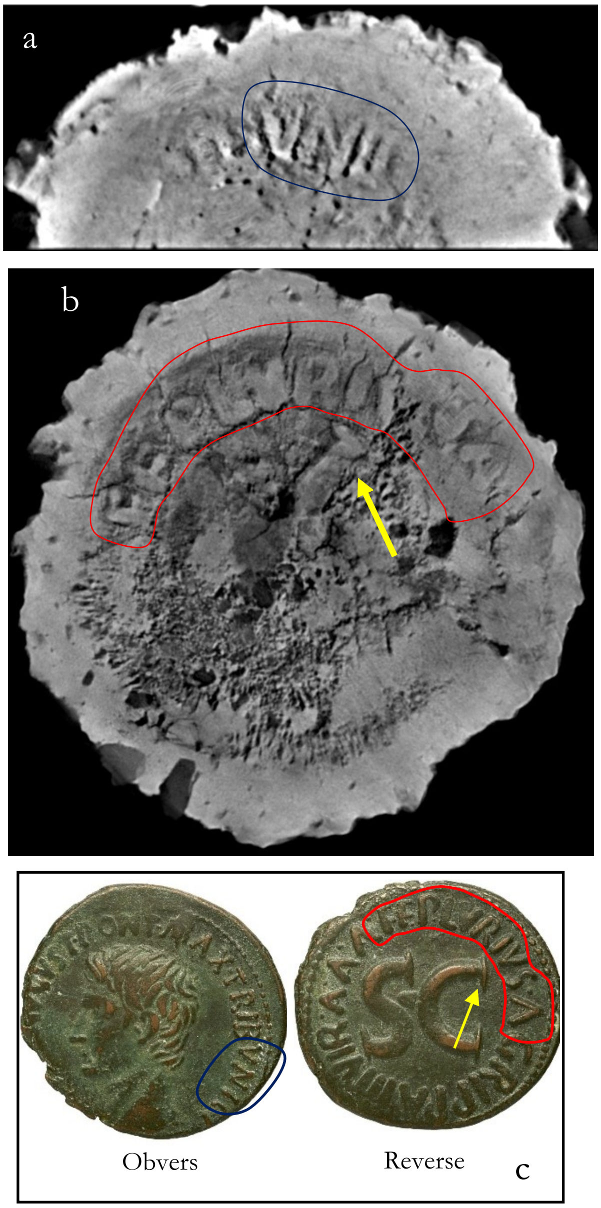

Figure 6. Grayscale virtual sections of the obverse (a) and reverse (b) of the M4 coin and reference coin such as Augustus As (c). The blue square (Figure 6a-c) indicates the letters "VNIC", the red square (Figure 6b-c) indicates the letters "FFPLVRIVSA" and the yellow arrow (Figure 6a) indicates part of a sign present on the reference coin too (Figure 6c).

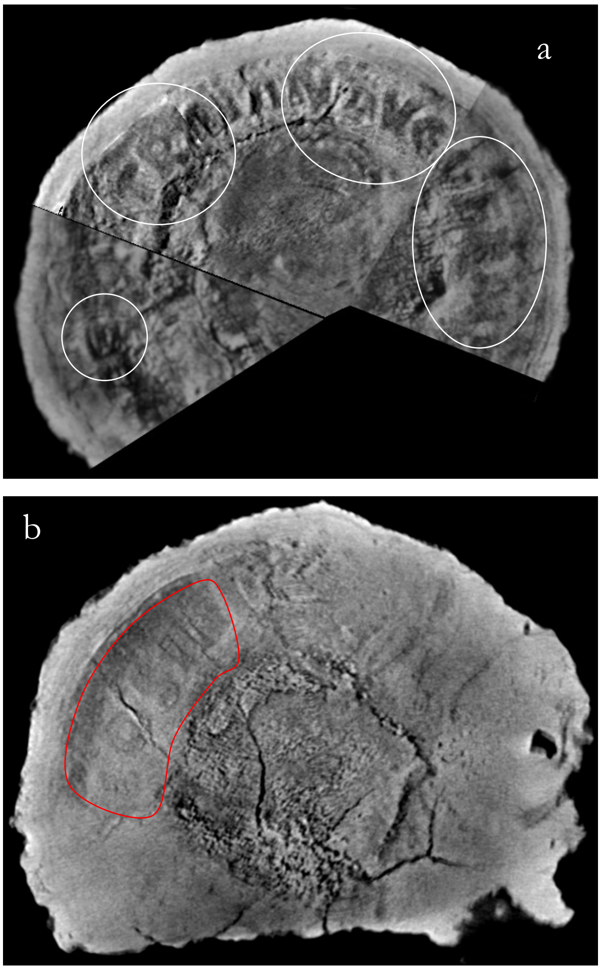

Figure 7. Grayscale virtual sections of a collage of different slices at different depths of the M6 coin's obverse (a) and grayscale virtual sections of the reverse of the M6 coin (b). In figure a, the white circles indicate the letters "E", "TRA", "NAVG" and "GERM". In figure b, the red square indicates the letters "COSIII".

Figure 8 shows grayscale virtual sections of a collage of different slices at different depths of the M7 coin's obverse in which we can read "IMP C[AES] DOMIT AV[G] [G]E[RM]" (Figure 8 red frames). The letters in square brackets are inferred by the distance between another letter. These letters suggest that the M7 coin was manufactured during the reign of Domitian between 84–96 AD, and then it would be a Domitian As.

Figure 9a shows grayscale virtual sections of the obverse of M9 coin in which a head profile turned to the right is visible. This profile is very similar to the figure of the Vespasian laureate head (see coin shown in Figure 9b). Being made of copper, coin M9 is probably a Vespasian As.

5. CONCLUSIONS

Thirteen Roman coins preserved in the Brettii and Enotri Museum in Cosenza have been studied by complementary physical methodologies such as X-ray μ-CT and XRF portable spectroscopy. These non-invasive and non-destructive methods have allowed us to reveal constituent materials, determine the original dimensions of coins, and find hidden signs or inscriptions.

Twelve coins are made of copper, while one is made of orichalcum. The coins have followed an oxidation process due to the sulphur spring near the discovery site. Oxidation layers made the coins' inscriptions unreadable. Through the morphological studies by X-ray μ-CT, we could find signs and inscriptions compatible with the inscriptions on the reference coins.

Figure 8. Grayscale virtual sections of the obverse of the M4 coin (a) and obverse of the reference coin such as Vespasian As (b). In both figures, the same head profile is visible.

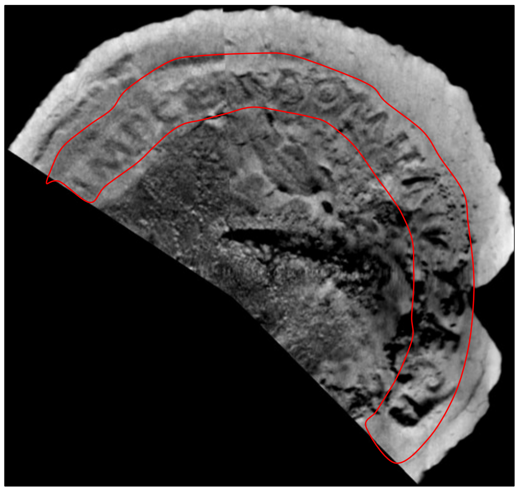

Figure 9. Grayscale virtual sections of a collage of different slices at different depths of the M7 coin's obverse. The red line indicates the letters "IMP CAES DOMIT AVG GERM".

Therefore, we can conclude that the coins analysed are of various types. Four of these have been associated with four different coins belonging to different historical periods, ranging from 7 BC to 101 AD. These are specimens minted during the reign of Augustus, Vespasian, Domitian and Trajan, the imperial era coinage.

Knowing the chemical composition of coins and comparing the tomography virtual reconstructed data with references coins, we can conclude that the M4 coin is an August As, the M6 coin is a Trajan As, the M7 coin is a Domitian As, and the M9 coin is Vespasian As.

Our findings provided insights into both the timeframe and origin of the artifacts, while also bringing to light concealed elements, including legible inscriptions that were previously inaccessible using alternative techniques.

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