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Continuity and Innovation in Pottery Technology: The Karst Region (North-East Italy) from Neolithic to Early Bronze Age

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Abstract: This paper explores the development of pottery technology in the Trieste Karst region (North-East Italy) from the Neolithic to the Early Bronze Age (EBA). It also seeks to identify cultural links with other areas by examining potentially imported vessels. Archaeometric analyses (X-ray diffraction and optical microscopy) reveal significant differences between Neolithic ceramics (Danilo-Vlaška Group) and the majority of Late Copper Age (LCA)/Early Bronze Age (EBA) pottery (primarily associated with the Ljubljana Culture and a few with the Cetina Culture). Neolithic pottery displays consistent characteristics across all vessel types, including coarse grain, prevalent sparry calcite temper, and the absence of grog. In contrast, most LCA and EBA vessels exhibit distinct features such as very fine-grained paste, no sparry calcite, notable use of grog temper, higher quartz, muscovite, and flint content. Notably, from a technological perspective, the analyzed Cetina vessels bear a strong resemblance to the majority of LCA ceramics. The differences between Neolithic and LCA/EBA vessels clearly suggest the use of new raw materials, recipes, and techniques, likely reflecting changes in cultural and social contexts and potential connections with the core area of the Ljubljana Culture.

Keywords: pottery; Northeastern Italy; Neolithic; Copper Age; Early Bronze Age; X-ray diffraction; optical microscopy; technology



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1. Introduction

The Trieste Karst region (North-East Italy) occupies the southwest part of the Classical Karst, at the northernmost edge of the Eastern Adriatic coast (Figure 1). It is a plateau of low rounded hills and low mountains ranging from 100–200 m to about 700 m above sea level, with a few major peaks reaching the maximum height. The outcropping rocks are mainly limestone with limited dolostone occurrences [1], bordered to the east by the Brkini Flysch and to the south-east by the Istrian basin Flysch. Apart from marls, both the Brkini and the Istrian Flysch are mainly constituted by lithic greywackes, where the main constituents are quartz and calcite, followed by feldspar and clay minerals [2,3]. In the flysch, calcite is mainly detrital, not sparry. Perennial surface waters are few, while rain percolates through natural holes and caves.

The number of natural caves is high, about 3000, and this feature certainly conditioned the use of the area during Prehistory, but it was also the target of field investigations that began in the last decades of the 19th century. Indeed, both professionals and speleologists/amateurs were attracted by these caves, with two main consequences: archaeological remains have been found in c. 180 caves, while only a few open-air sites are known until

the beginning of the Bronze Age [4,5] when massive stone structures—called *castellieri*—were built on top of many hills in Karst and Istria; the quality of the recovered data is non-homogenous, as it reflects the different preparation of the researchers.

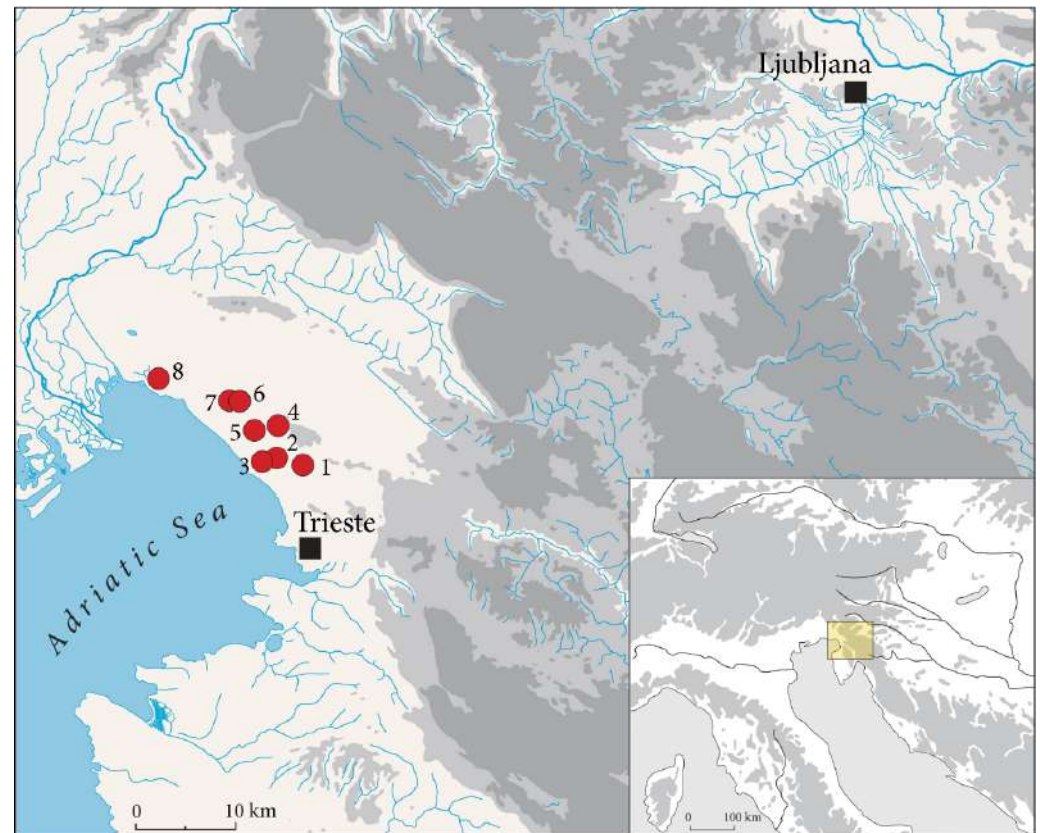


Figure 1. The Karst region and the archaeological sites where the studied ceramics were discovered: 1: Ciclami Cave; 2—Zingari Cave; 3—Tartaruga Cave; 4—Cotariova Cave; 5—Pettine Cave; 6—Edera Cave; 7—Caterina Cave; 8—Mitreo Cave.

Moreover, a large part of the findings from these investigations had remained unpublished until the early 1990s, when—almost in parallel with a sensible decrease and virtual stop of field activities (with only a few notable exceptions)—started critical reviews and complete editions of materials found in sites of particular interest (e.g., [6–9]). These reviews, combined with close examinations of the literature and archival documents, have produced a considerable increase in the number of artifacts and data, which reveal differences from site to site that may be naturally as well as culturally determined. It is now evident, however, that, except for a few cases, no cave has been totally excavated: caution is consequently needed when one tries to reconstruct the chronology and the characteristics of the human presence in each cavity and over the whole area.

As a basically archaeological approach is not sufficient to try to discover to what extent the physical environment might have influenced the anthropic use of caves in Prehistory (and beyond), in the late 1990s, an interdisciplinary project was launched to check the physiographic characteristics of the archaeological cavities and their surroundings. The necessity of storing and managing the increasing quantity of data brought to the creation of a dedicated geo-referenced database—C.R.I.G.A.: *Catasto Ragionato Informativo delle Grotte Archeologiche*—later transformed into a webGIS and put online in 2011 [10]. More recently, the project has been extended to all the caves of the Friuli Venezia Giulia region: the updated database and a volume that summarizes the present state of the art are now visible on the Regional Speleological Cadaster website (<https://catastogrotte.regione.fvg.it/pagina/100/criga>, accessed on 22 January 2024).

Besides C.R.I.G.A. and its evolution, other lines of research have significantly contributed to generating the present state of the art: in the Karst area in particular, sedimentological and soil micromorphological analyses of cave deposits, on the one hand [11–13], and archaeometric analyses of lithic and ceramic artifacts, on the other (for stone axes see: [14]; for pottery see: [15–18]).

Boschian [11,12] and Boschian and Montagnari Kokelj [13] carried out a study on Karst cave deposits using sedimentological and soil micromorphological analyses. The results suggest a rather intensive use of caves during the Neolithic, declining into the Bronze Age. According to the model proposed first by Brochier in the early 1980s [19], several Karst caves might have been used as grottes–bergeries rather than habitats–bergeries [13] (pp. 348–350), but certainly, this hypothesis is limited due to incomplete excavation of post-Mesolithic levels.

Archaeometric analyses began in the late 1990s as part of the Italian project aimed at integrating science and archaeology in studying prehistoric polished industries in Northern Italy. This initiative led to a long-term project focused on the territories known as Caput Adriae, including Northeastern Italy, Central and Western Slovenia, and Northwestern Croatia. Initially, attention was given to shaft-hole axes due to lithological differences observed in artifacts from the Karst area and Friuli compared to Neolithic samples, suggesting changes in raw material provenance [20]. Subsequent studies extended to all types of polished stone axes, confirming previous hypotheses and identifying main raw material sources ([14] and references therein) Later, on the basis of previous results, archaeometric analyses were applied to Neolithic, Copper, and Early Bronze Ages (hereafter EBA) pottery from Karst caves and Slovenian sites of the Ljubljansko barje [15–18], where axes and pots of peculiar forms had been found.

However, limited findings have been published regarding only small groups of Neolithic and Copper Age pottery from the Karst region. This paper aims to address this gap by presenting findings from the analysis of about 70 vessels from eight Karst caves characterized by important stratigraphic sequences (for a detailed description of the contexts, see references in Table S1 [6–9,21–24]). This study seeks to trace the development of pottery technology from the Neolithic to the EBA and identify cultural connections through the examination of potentially imported vessels.

1.1. Archaeological Context

In 2014, the Congress “Preistoria e Protostoria del Caput Adriae”—XLIX Scientific Meeting of the Italian Institute of Prehistory and Protohistory—took place in Friuli Venezia Giulia, 24 years after the previous one held in 1990. It represented an opportunity to update regional data with the results of both field works and other studies, like the ones mentioned, and to contextualize them in a wider scenario, including the adjacent regions of Austria, Slovenia, and Croatia.

The first part of the proceedings, published in 2018, contains general essays written by multiple authors on the four macro-periods in which Prehistory and Protohistory are conventionally divided. These essays still remain good reference points to understand the wider chronological and cultural background of the artifacts analyzed in the present paper and of the sites where they come from [25], but as a closer view of the Karst area may be of use, we will focus on the Neolithic and Copper Ages, updated with the latest studies.

1.1.1. Neolithic

Only a few ceramic fragments attributed to the Impressed Ware Culture seem to have been found in some caves in the Trieste Karst [26–30] (p. 494). However, the available data indicate that a fully Neolithic Culture only emerged later, around the mid-sixth millennium BC, with the development of the Danilo–Vlaška Culture or Gruppo dei Vasi a Coppa, whose connections with the Danilo Culture, distributed mainly over Central and Southern Dalmatia, have been emphasized by various authors [25,28–33]. Materials attributed to this group are present in more than 40 caves in total, but in highly variable numbers, from

one to many hundreds, presumably due to the nature of discovery (occasional findings vs. systematic excavations), on the one hand, and of ancient use, on the other (see <https://catastogrotte.regione.fvg.it/pagina/100/criga>, accessed on 22 January 2024). The reasons why Northern Istria and the Karst region were not directly involved in the spread of the Impressed Ware Culture and the role played by the last Mesolithic groups in this process are still debated [33].

The Danilo–Vlaška Culture, distributed over the Karst and Istrian regions, takes one of its names from the most common ceramic vessel, characterized by a tulip-shaped form set on a pedestal base, often decorated with incised lines intersecting to form hanging triangles below the rim, the so-called *vaso a coppa*. The sites in the Karst region are all in caves or rock shelters, and it appears that their use is associated with specific economic activities, primarily pastoralism [34]. The analysis of deposits in some caves has indeed demonstrated that they were often used for the stabling of livestock [11–13]. Archaeometric analyses of polished stone artifacts have contributed to identifying the extent of their movements and hypothesizing the exchange mechanisms involved [14], in which the availability of sea salt along the coast could have played a significant role [35].

The ceramic assemblage, while connected to that typical of the Danilo Culture, does not exhibit the same variety of forms and decorations found in sites in Dalmatia and also in the Friulian Plain [25,36]. Based on the available data, it is not easy to define the role of the Karst region in relation to Friulian sites and the Istrian and Dalmatian areas. In this regard, it should be noted that ceramic remains with decorations related to the Danilo environment and some fragments of rhyta, probable evidence of an open-air site, were discovered in Sermin, in the bay of Koper in Northern Istria [37–39]. This raises the question of the relationship between coastal open-air sites located near watercourses and arable land and the Karst interior, which is characterized by a rocky environment without a surface hydrographic network.

Revisions of archaeological materials from old excavations have allowed for a precise definition of the most common ceramic forms within the Danilo–Vlaška Group. The *vaso a coppa*—i.e., hemisphere bowls showing variability in the restriction of the upper part and the depth of the body, as well as in the decoration, mainly incised with a geometric design, is the most common element (ranging from 65% to 90% of assemblages); it often appears associated with open bowls with raised rims, carinated bowls/biconical plates, more generic bowls and rhyta—vessels with four legs, asymmetrical body, forward-facing mouth, and high curving handle [9,25].

The flaked stone artifacts associated with Danilo–Vlaška Culture are usually few and of non-local origin. Available data show that jade axes originating in the Western Alps reached the Neolithic groups of Friuli Venezia Giulia and coastal Istria as early as the second half of the sixth millennium BC, during the Danilo–Vlaška Culture. The exchange of this and other classes of lithic artifacts testifies that, in this period, this area was fully integrated into long-distance exchange systems that used mainly coastal routes [14].

The archaeological materials from the Karst sites are largely the result of old investigations. While the available data allow for a fairly reliable picture of the early Neolithic, we are not able to recognize a secure chronological–cultural sequence for the subsequent periods [25,40]. This difficulty in defining the post-Vlaška phases is a common challenge in both the Karst and Istrian regions. Some scholars have observed that this may partly be due to the absence of a drastic change in ceramic forms compared to those of the Danilo–Vlaška Culture, which would continue to be produced with the cessation of a limited number of types [25,27,28].

However, it seems that some ceramic materials from over a dozen sites can be typologically attributed to the cultural sphere of Hvar, mainly present in Dalmatia and its hinterland. These are mostly keeled bowls with distinct rims and rare fragments of painted ceramics [9] (pp. 49–51), [25]. Without stratigraphic indications, the classification of these materials cannot be considered secure. Individual finds have been attributed to the Square Mouthed

Pottery Culture, such as some ceramic fragments from the Gallerie and the Ciclami caves [6] (p. 155), [7] (p. 184).

Archaeometric data about polished stone axes show that coastal connections, already active during the Vlaška Culture, continued in the fifth millennium BC, as indicated by a few oversized jade axe blades and other materials. Far from the coast, jade axes entered Central Slovenia, probably reaching sites of the Sava Group of the Lengyel Culture in the first half of the fifth millennium BC. In roughly the same period, shaft-hole axes made of Bohemian metabasites spread over Central and Southeastern Europe, crossed the Alps, and reached Karst and Northern Italy [14].

1.1.2. Copper Age

Available archaeological data are insufficient to define the cultural aspects that occurred in the Karst region from the late Neolithic to the full Copper Age. The same applies to the chronological and cultural development of the Eneolithic in the Friuli region [40]. The Eneolithic complexes found in caves usually have many cultural components mixed together in the same deposit. It is, therefore, difficult to define clearly the relevance of these components in each complex since diagnostic elements are generally few and often mixed with other, less significant materials. It is only possible to identify some typological elements that could be characteristic of different phases without being able to connect them to the context in which they circulated. For example, it has been suggested that a particular treatment of the surfaces of some containers, locally defined as Besenstrich, may be linked to an early phase of the Copper Age [6,41], but similar pottery surface treatment probably started already in the Neolithic and remained in use till the beginning of the Bronze Age. From a typological point of view, single ceramic materials from a few caves could be compared to Nakovana-like forms [9] (p. 52), [42] (p. 379). In Dalmatia, the Nakovana style replaced late Hvar pottery at the beginning of the fourth millennium BC, and it is considered characteristic of the Early Copper Age (hereafter ECA) [42–44]. The results of research in the pile-dwelling sites of Ljubljansko barje provide important chronological and cultural data for comparison, which could allow for a deeper understanding of the Karst cultural aspects in the fourth millennium BC [45,46].

It is only with the beginning of the third millennium BC that we are once again able to place the Karst region within a network of connections, primarily looking to the east. This is the period when radical changes, such as the emergence of social ranking and a new social order, emerged at the European scale, probably in connection with the arrival of new populations [47,48].

The relationships between Karst and central Slovenia, already active during the preceding millennium, reached their peak. Inland Slovenia, more specifically, the area of the Ljubljansko barje, is likely the center of diffusion for a series of archaeological elements, probably impulses and cultural influences, that are more difficult to define, toward the Adriatic regions, especially Karst and the Southeastern Friulian Plain [6] (pp. 155–162), [7] (pp. 118–122), [8] (pp. 85–94), [14,25,49]. This process is exemplified by the widespread distribution of the so-called Ljubljana-type shaft-hole axes. These axes were crafted from raw materials sourced in the Karavanke mountain range in the Eastern Alps. They were highly prevalent in the Ljubljansko barje region during the final centuries of the fourth millennium BC. Their use persisted into the first half of the third millennium BC, and their presence extended to the Karst region, parts of the Friulian Plain, and Istria [14].

The cultural sequence in the Karst region tentatively unfolds through the development of various cultural aspects: the pre-Ljubljana elements, which are mainly associated with the Vučedol Culture of the first half of the third millennium BC, and the Ljubljana elements, which are attributed to the Ljubljana Culture, which is dated to the middle of the third millennium BC [7,8,50,51]. In the first half of the third millennium BC, the influence of the Vučedol Culture most likely reached the Karst region from the area of the Ljubljansko barje in Central Slovenia. A network of connections also extended across this area to distant regions such as Central Europe, as evidenced in particular by the characteristic

cross-footed bowls decorated with cord impressions [18]. With the emergence of the Ljubljana Culture, the connection between the Ljubljansko barje and the Karst region reached its peak. The Ljubljana Culture, which takes its name from the original area of Ljubljansko barje, is characterized by globular vessels with funnel necks and hemispherical bowls with thickened rims decorated with so-called barbed wire decoration (in Slovenian *odtisi niti, navitih na ploščice*; French *barbelé*) (for the decoration see [52]). Such vessels are widespread, not only in the Karst region but throughout the entire Adriatic region ([51] and references therein). In the second half of the third millennium BC, elements of the Cetina, Gata–Wieselburg, and Polada Cultures can also be found in a few sites of Trieste Karst [7,8]. Moreover, distinctive ceramic forms and decorations have been identified, indicating diverse cultural contexts, such as those involved in the so-called Bell Beaker phenomenon. These findings extend not only to materials retrieved from caves but also those discovered on the Doberdò del Lago plateau [8] (pp. 85–94), [53], [54] (p. 60).

There are numerous aspects of the Late Copper Age (hereafter, LCA) in Karst and its surrounding regions that remain elusive. The pottery discovered in Karst caves, which are similar to the ceramics of the Ljubljana Culture, has been categorized by several scholars as part of the Adriatic type of the Ljubljana Culture [55] (pp. 10–12), [56] (pp. 41–43), more recently called Ljubljana–Adriatic pottery style by [57]. This classification encompasses findings from the Karst region to Dalmatia. However, the materials from the Karst region bear closer typological resemblance to those from Ljubljansko barje than to those from Dalmatia (see, most recently, [51]), and the controversial comparability is further complicated by a chronological issue. The Ljubljana Culture is, indeed, dated to the middle of the third millennium BC (precisely, late 26th and the 25th century BC), while the so-called Ljubljana–Adriatic pottery style would have emerged already in the initial half of the third millennium, potentially extending until 2400 BC [51,57].

Moreover, recent research has focused on the unclear chronological relationship between the so-called Ljubljana and Cetina styles, as the latter would have appeared around the middle of the third millennium and lasted until approximately 1900 BC [57]. This ambiguity persists despite available C14 dates and archaeological evidence. Consequently, the possibility that these styles coexisted around the mid-third millennium BC cannot be definitively confirmed or ruled out.

As far as polished stone axes are concerned, during the fourth millennium BC, the exchange networks of Caput Adriae are increasingly influenced by the Eastern Alpine and Balkan worlds, where the raw material sources of the main groups of shaft-hole axes are located. The association of the rocks used for axe production and copper ore suggests that the changes in raw material exploitation strategies during the Copper Age were probably related to the development of the first metallurgy [14].

2. Materials and Methods

2.1. Materials

In this study, we present previously unpublished archaeometric data concerning a total of 45 investigated vessels from 8 cave sites in the Trieste Karst area (see Figure 1). Among these, there are 8 vessels dated to the Neolithic period, attributed mainly to the so-called Danilo–Vlaška Group, 2 vessels attributed to the ECA, 35 vessels dated to the LCA/EBA, of which 30 are assigned to the Ljubljana Culture and 5 to the Cetina Culture (refer to Table S1 for details; Figures 2–7). Additionally, we have included 13 Neolithic and 8 LCA vessels from the same geographic area, previously analyzed and documented, in Table S1. In Figures 2–7, all the available drawings of the analyzed vessels have been reproduced with the exception of the recently analyzed LCA decorated cross-footed bowls [16,18] and 2 small fragments from Caterina cave and 1 from Zingari cave.

As previously said (see Section 1), archaeological findings come from c. 180 caves, but regular excavations have been carried out in less than 50 of them. Revisions of unpublished materials and studies of the last decades have focused on data shortcomings and interpretation issues. The long transitional phase after the Danilo–Vlaška Neolithic until the Late

Eneolithic, although documented in c. 20 sites, is still rather obscure, and this bears upon the possible recognition of continuity in human presence and, consequently, continuity and/or innovation in pottery technology in the area under study.

On these bases, the selection of vessels has been carried out with the aim of including both finer, often decorated ceramics, as well as coarser ceramics that can be stratigraphically and/or typologically attributed to a specific cultural and chronological phase. Additionally, the stratigraphic position and cultural significance of the vessels have been taken into consideration when making the choice. In the case of the findings attributed to the Ljubljana Culture, the selection was also based on already selected samples of the same culture from the area of Ljubljansko barje. The aim was to analyze, on the one hand, finely crafted finds decorated in the manner typical of the Ljubljana Culture (i.e., with barbed wire decoration) and, on the other hand, those belonging to the so-called common ware, which is of a coarser facture [51].

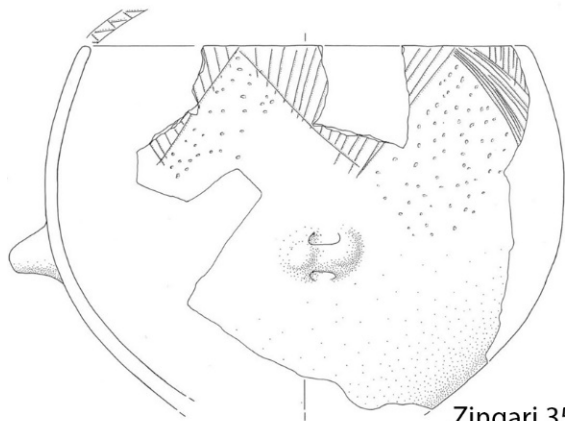
The investigated Neolithic vessels encompass various types. Among the most common Neolithic vessels selected in our study are the so-called *vasi a coppa* vessels (Figure 2; Figure 3: 3540, NP, 3545). These bowls are spherical with an inturned or vertical rim and usually have a small pedestal. Very often, they have small vertically perforated lugs and incised geometric decorations, mainly in the upper part of the recipient. Some of the analyzed bowls also have repair holes (Figure 2: 3548, 3534; Figure 3: 3540).

Besides the globular-shaped bowls, we also analyzed 3 carinated bowls, of which one had a raised rim and two a slightly everted rim (Figure 3: 53511; Figure 4: 2658, 16888). In one case, the bowl had a preserved high foot (Figure 4: 2658). These types of open bowls are not so common among the Neolithic ceramic repertoire in Karst, and they are similar to the vessels of the Danilo Culture in Dalmatia [6] (p. 153).

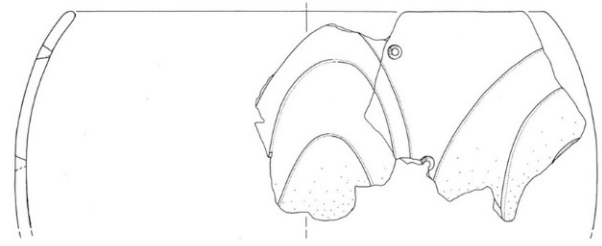
Two small bowls from the Tartaruga Cave, decorated with incisions, and a small pedestal were included in our study (Figure 4: 2618, 2706, 2676).

In addition to the more characteristic and common Neolithic vessels, we have also analyzed some vessels that are less frequently represented in terms of form and/or decoration in Karst Cave contexts. We analyzed a beaker from the Zingari Cave decorated with vertically perforated lugs and rhombic motifs all over the recipient and a globular vessel with an everted rim (Figure 3: no numb. 136, no numb. 159). Both vessels are mostly compared within the framework of the Danilo Culture [7] (p. 115), [29] (T.46: 1), [58] (T.LXXXIV: 11).

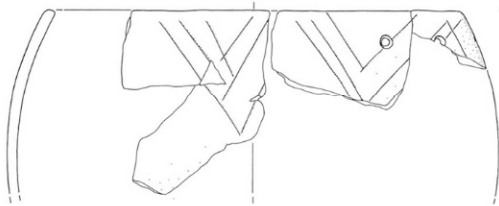
Other finds examined as part of our research include a jug with a large handle decorated with parallel vertical ribs and two vessels with vertical subcutaneous handles, one of which has vertical grooves, discovered in the Ciclami Cave (Figure 4: 5880, 5914, no numb. 726). The first vessel, the ribbed jug, recalls vessels characteristic of the Neolithic Culture of Fiorano [6] (p. 153 and references therein), while the two vessels with subcutaneous handles and a decoration consisting of shallow grooves are similar to vessels of the ECA Nakovana Culture [59] (T.XLVIII).



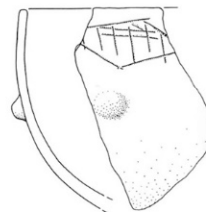
Zingari 3533



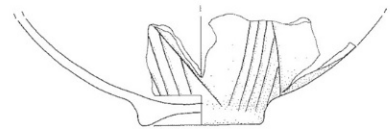
Zingari 3548



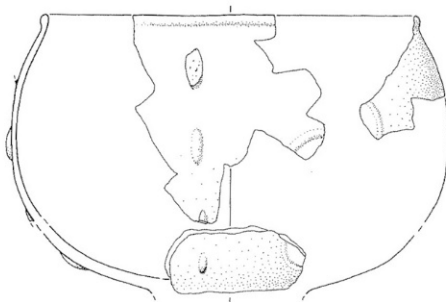
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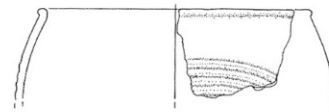
Zingari 3541



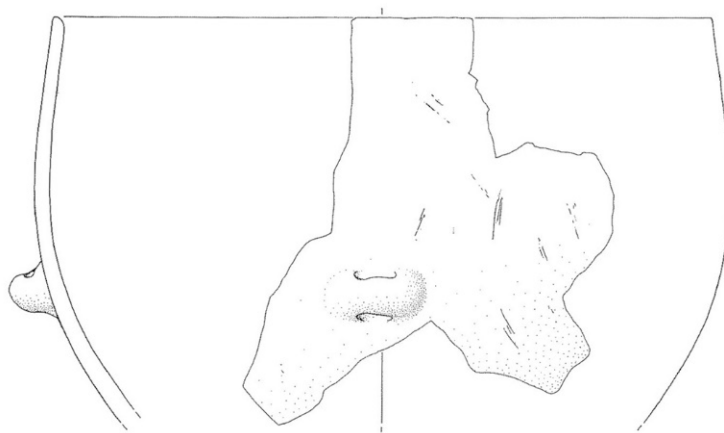
Zingari 3523



Zingari 3546



Zingari 3543



Zingari 3556

Figure 2. Analyzed Neolithic vessels from Zingari Cave. Scale 1:4 (for references, see Table S1).

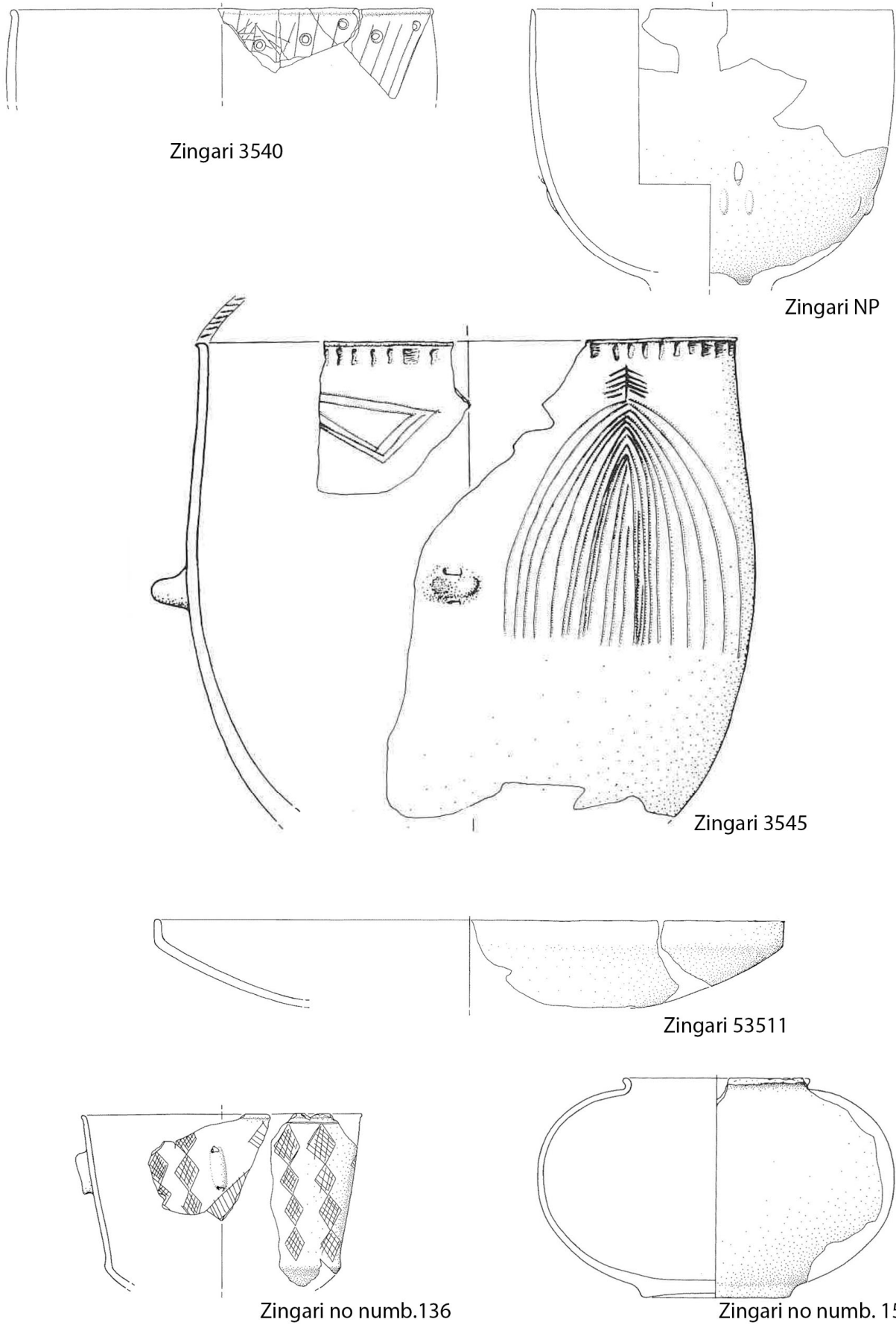


Figure 3. Analyzed Neolithic vessels from Zingari Cave. Scale 1:4 (for references, see Table S1).

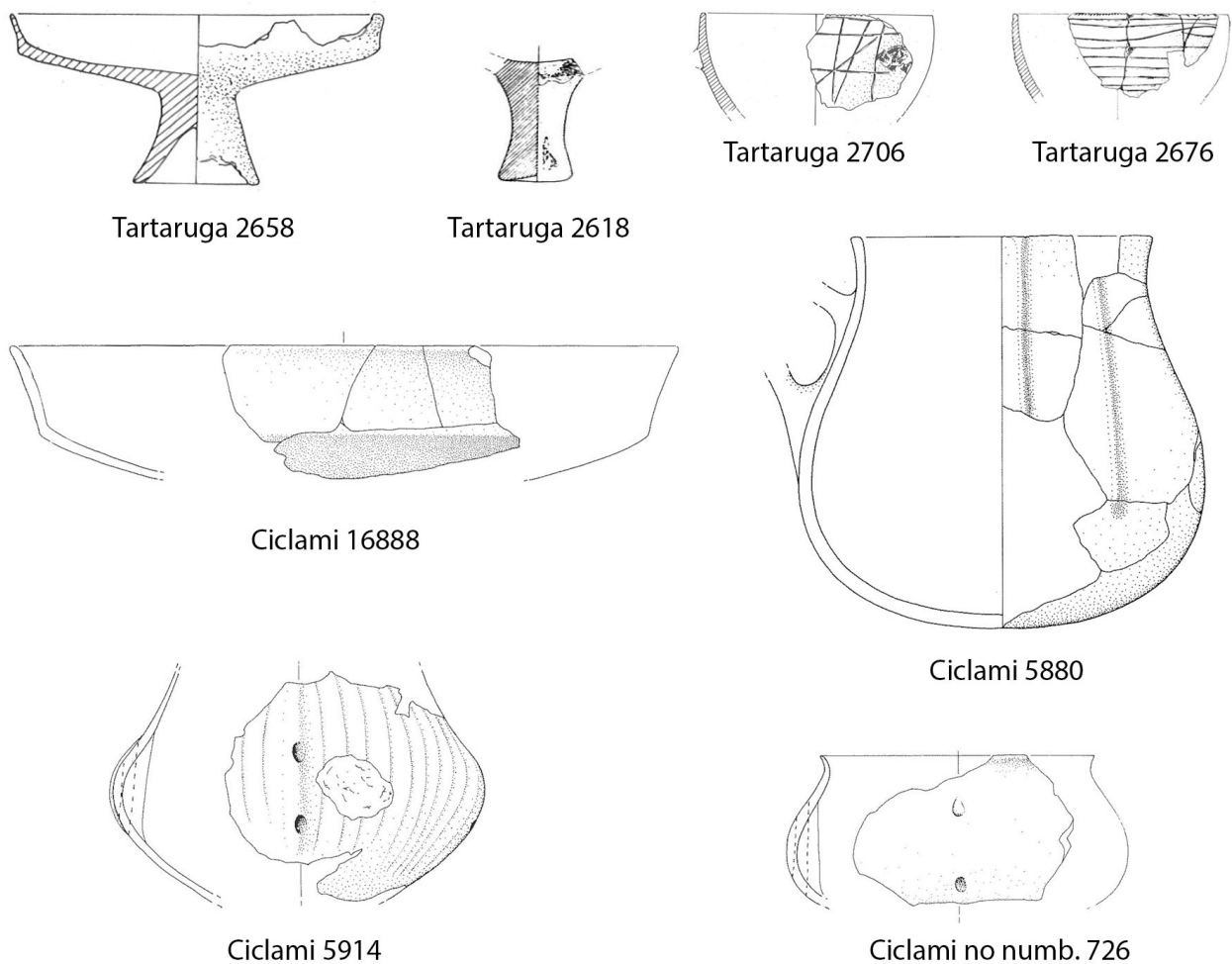


Figure 4. Analyzed Neolithic and Early Copper Age (Ciclami 5914 and Ciclami no num. 726) vessels from Tartaruga and Ciclami Caves. Scale 1:4 (for references, see Table S1).

The examined LCA vessels, mostly attributed to the Ljubljana Culture, include both finer vessels and coarser ceramics. The finer vessels include hemispherical bowls with a flat, thickened rim (Figure 5: 3458, 48963; Figure 6: SN), globular vessels with a funnel-shaped neck and handle (Figure 5: 3467; Figure 6: 17164, 11667, 11668, 1827), a fragment of a platter or baking pan (Figure 6: 3149), and several decorated vessel fragments (Figure 5: 3457, 3470; 6: SN1, 16434, 16162, etc.). The coarser pottery includes fragments of larger vessels with a thickened rim decorated with finger and/or nail impressions (Figure 5: 348796–348798, 348791–348792, 48853–48857, 3418). The finer pottery mostly has barbed wire decorations. This type of decoration technique also occurs in the context of the Vučedol Culture and the Bell Beaker Culture [51,52]. The vessels analyzed in our study are most comparable to the pottery of the Ljubljana Culture [50].

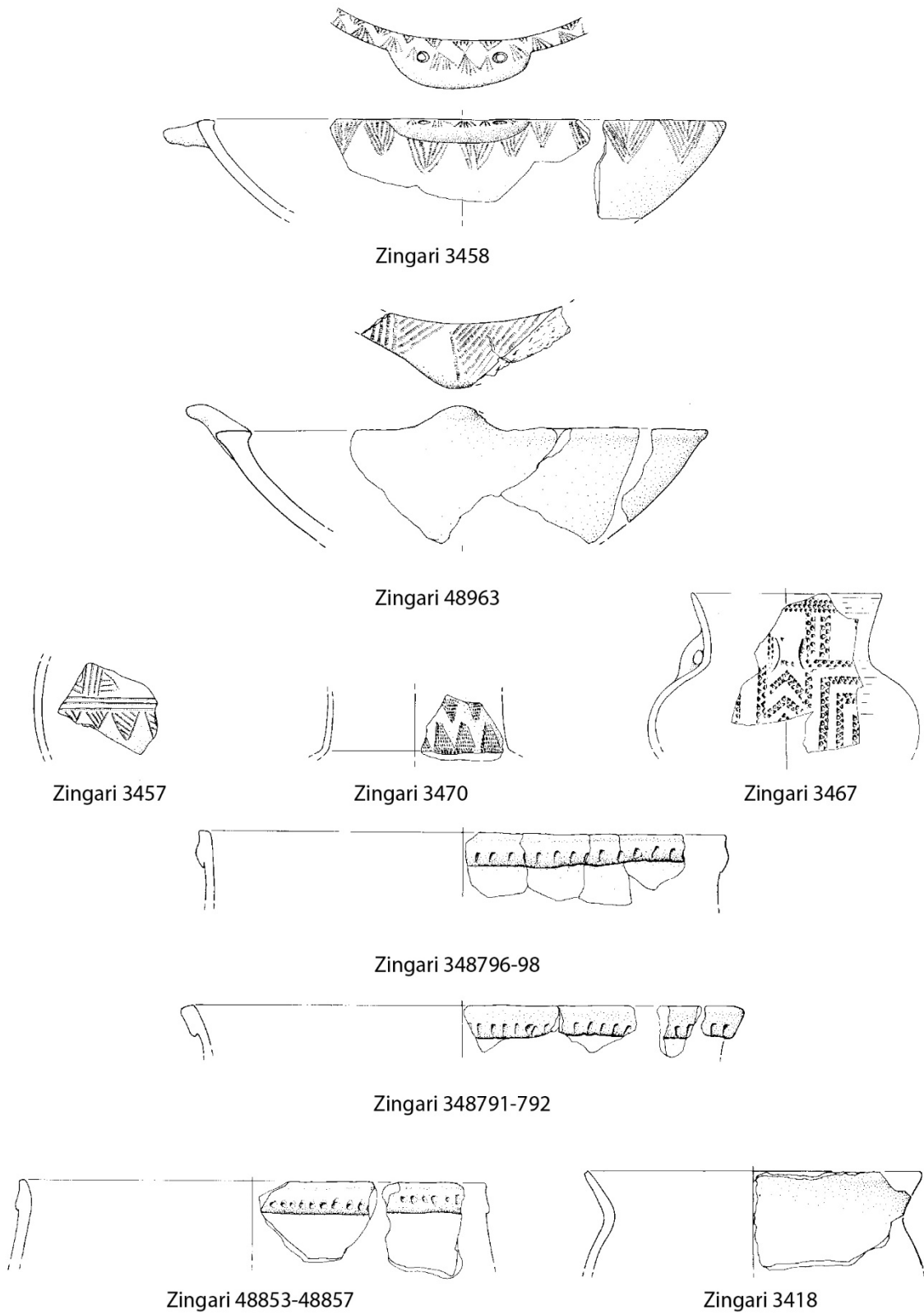


Figure 5. Analyzed Late Copper Age vessels from Zingari Cave. Scale 1:3 (for references, see Table S1).

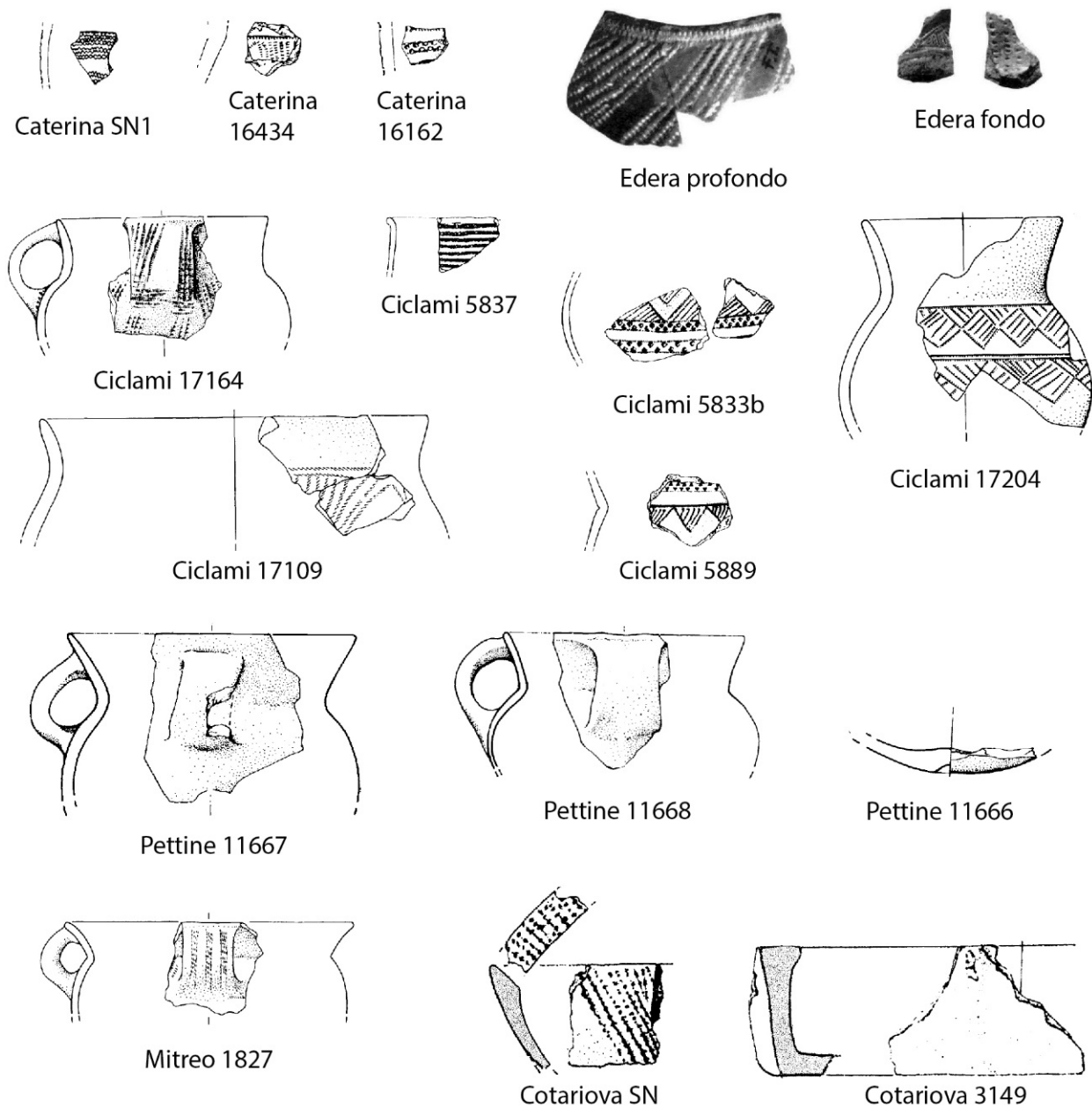


Figure 6. Analyzed Late Copper Age vessels from Caterina, Edera, Ciclami, Pettine, Mitreo, and Cotariova Caves. Scale 1:3 (for references, see Table S1).

In addition to the vessels attributed to the Ljubljana Culture, we have selected some examples of the Cetina Culture, which date from the second half of the 3rd millennium BC and were found in the caves of Ciclami and Zingari. From the Ciclami Cave, two jugs with a sharp-edged rim and a handle attached to the rim (Figure 7: 17020a, 17143), fragments of a vessel that probably belonged to a similar jug (Figure 7: 17020b), and a fragment of a hemispherical vessel decorated with impressions on a flat rim (Figure 7: 17210) were analyzed. From the Zingari Cave, we analyzed two fragments of two very similar vessels, probably jugs, with incised decoration and impressions (Figure 7: 3471, 3468-35).

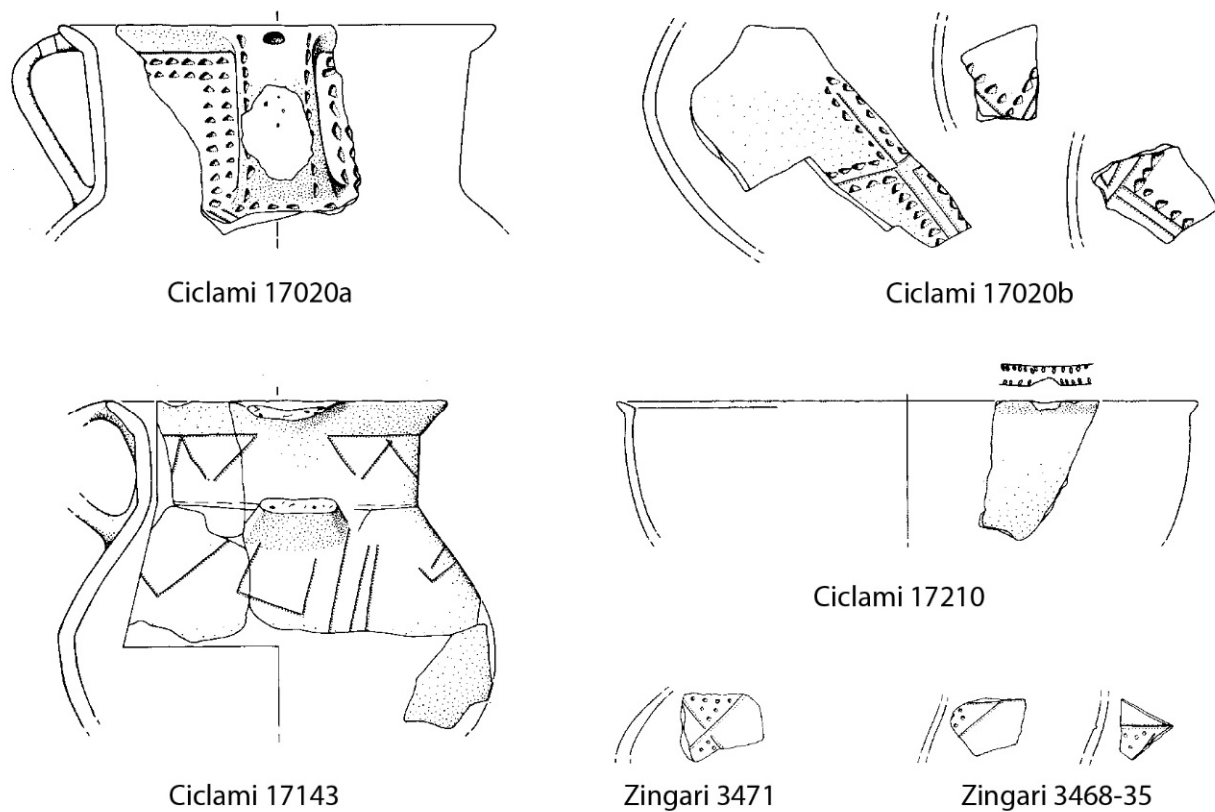


Figure 7. Analyzed vessels attributed to Cetina Culture from Ciclami and Zingari Caves. Scale 1:3 (for references, see Table S1).

2.2. Methods

2.2.1. Macroscopic Observation

The surface of all samples has been observed using a stereomicroscope and/or a magnifying glass in order to identify visible lithic grains.

2.2.2. X-ray Diffraction

Small powdered samples taken from the vases (generally less than 0.5 g) were analyzed by XRD at the Department of Mathematics, Informatics, and Geosciences of Trieste University using an STOE D 500 (Siemens, Munich, Germany) X-ray diffractometer at room temperature. $\text{CuK}\alpha$ radiation was used through a flat graphite crystal monochromator. The current used was 20 mA, and the voltage was set at 40 kV. The 2θ scanning angle ranged from 2 to 60° , with 0.1° steps and a counting time of 2 s/step. Mineral phases have been recognized by using the Hanawalt Manual.

2.2.3. Statistical Analysis of XRD Results

Principal component analysis (PCA) enables the extraction of relevant information hidden in XRD data, disclosing the possibility of automatic data processing even in the absence of a priori structural knowledge [60]. Even if its current applications in archaeometry are mainly based on the analysis of quantitative data, such as those derived from powder diffraction–quantitative phase analysis [61], X-ray powder diffraction profiles can be directly analyzed, providing both qualitative and quantitative results, as commonly performed in the analytical chemistry field [60].

In the present study, we have directly applied PCA on X-ray spectra in order to achieve a qualitative evaluation of the mineralogical composition of the investigated vessels. PCA of XRD spectra was calculated with Python via the Scikit-learn library [62]. Prior to computing statistical analyses on X-ray intensity spectra, data were \log_{10} -transformed and filtered,

isolating only 2θ values associated with main peak areas; then, the intensity values of each sample were normalized for their maximum intensity values. XRD profiles of pure calcite, quartz, and dolomite, the main components of the lithologies present in the area [1–3], have been downloaded from the RRUFF Project website (<https://rruff.info/>, accessed on 12 October 2023) and analyzed following the same procedure. Tests were successfully performed to determine if data were suited for factory analysis: Barlett's sphericity test results successfully with $p < 0.001$ and KMO = 0.70 (Kaiser-Meyer-Olkin test; acceptable values for KMO > 0.5).

2.2.4. Optical Microscopy

Thirty-seven small pottery fragments were taken from the artifacts, and after impregnation by epoxy resin, thin sections were prepared at the Department of Geosciences of Padua University. The thin sections have been examined under a polarizing microscope at the Department of Mathematics, Informatics, and Geosciences of Trieste University in order to define their mineralogical and textural features (Table S1). Fourteen additional thin sections of other vessels, produced and studied in recent years, have been included in this study and listed in Table S1.

2.2.5. X-ray Computed Micro-Tomography

Thirty-three vessels were imaged by X-ray computed micro-tomography (hereafter, microCT) at the Multidisciplinary Laboratory of the Abdus Salam International Centre for Theoretical Physics (Trieste, Italy) (Table S1), using a system [63] specifically designed for the study of archaeological and paleoanthropological materials [15–17,63–65].

The microCT acquisitions were carried out by using a sealed X-ray source (Hamamatsu L8121-03 by Hamamatsu Photonics K.K., Japan) at a voltage of 110 kV, at a current of 90 μ A, and with a focal spot size of 5 μ m. The X-ray beam was filtered by a 0.05 mm-thick copper absorber. A set of 1440 or 1800 projections of the artifacts was recorded over a total scan angle of 360° by a flat panel detector (Hamamatsu C7942SK-25 by Hamamatsu Photonics K.K., Japan; pixel size of 50 μ m). The resulting microCT slices were reconstructed using the commercial software Digi XCT 3.0.7 (Digisens) in 32-bit format and with isotropic voxel sizes from about 10 to 40 μ m.

Additional 21 vessels of similar chronology and finding area were already analyzed using the same microCT station [16,18] (Table S1).

In this study, microCT data have been mainly used to provide a qualitative evaluation of the pottery microstructure when destructive techniques were not applied for preservation reasons. Detailed analysis of microCT data is out of the scope of the present paper.

3. Results

3.1. Macroscopic Description

In most of the Neolithic and ECA samples, sparry calcite and limestone clasts have been identified on the surface of the vessels and along the fractures of the shards. The same mineral has been recognized only in a small number of the LCA vessels (3149, 3469, 3470).

3.2. X-ray Diffraction

X-ray diffraction has enabled the identification of quartz and calcite as the primary minerals, with calcite being predominant in the majority of Neolithic samples. In rare instances, only quartz has been detected. Feldspar has been identified in approximately 20% of the samples, while clay minerals/phyllosilicates are present in trace amounts in about 80% of the samples (Figure 8).

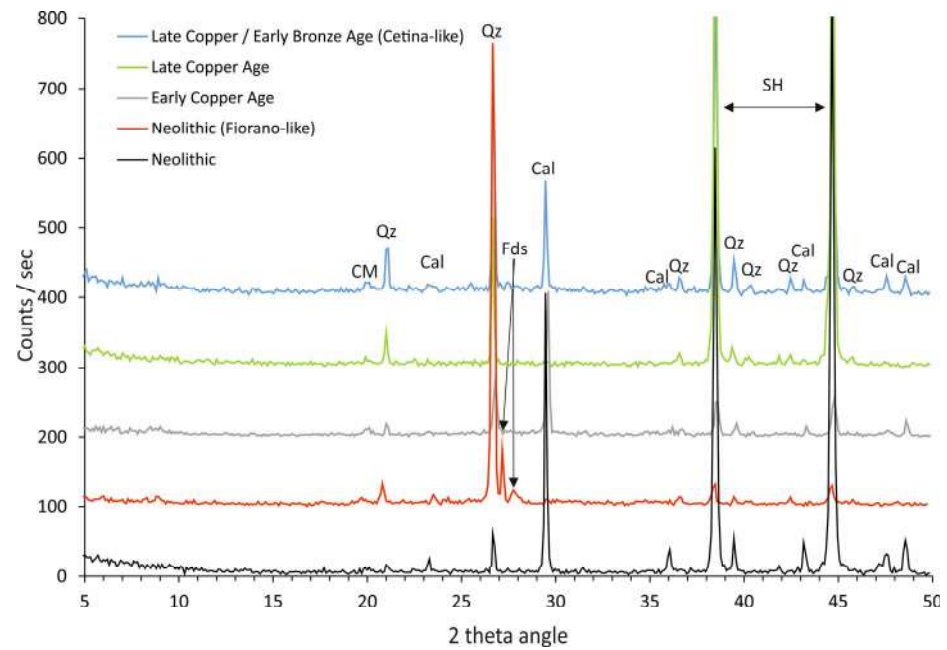


Figure 8. XRD of some selected samples. CM: clay minerals; Qz: quartz; Cal: calcite; Fds: feldspar; SH: sample holder.

3.3. PCA of XRD Data

The principal component analysis of the XRD spectra has provided interesting results, allowing us to visualize in a single diagram and at the same time the main mineralogical features of the analyzed assemblage (Figure 9), as it is not possible just comparing the diffractograms. In Figure 9, two main clusters are visible; to allow for an easier interpretation of the diagram, XRD data of pure calcite, dolomite, and quartz have been processed and plotted, too.

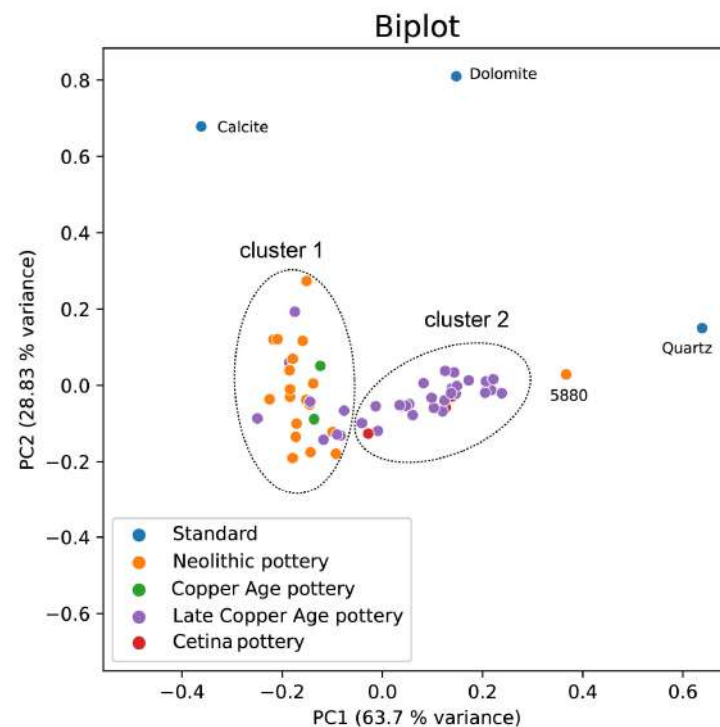


Figure 9. PCA of X-ray powder diffraction profiles.

The first cluster (labeled 1) shows an almost vertical orientation pointing to pure calcite and includes both Neolithic and Copper Age samples characterized by very abundant sparry calcite used as temper material (Figures 9 and 10). The Fiorano-style flask is the only Neolithic vessel that is well-separated from Cluster 1 and is close to the pure quartz (Figures 9 and 11).

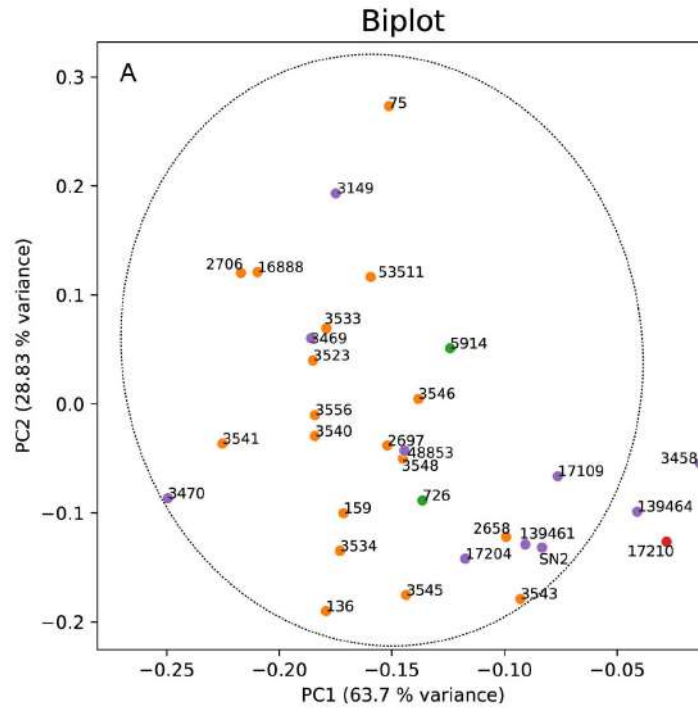


Figure 10. Zoomed view of Cluster 1 of Figure 9. For the symbols, see Figure 9.

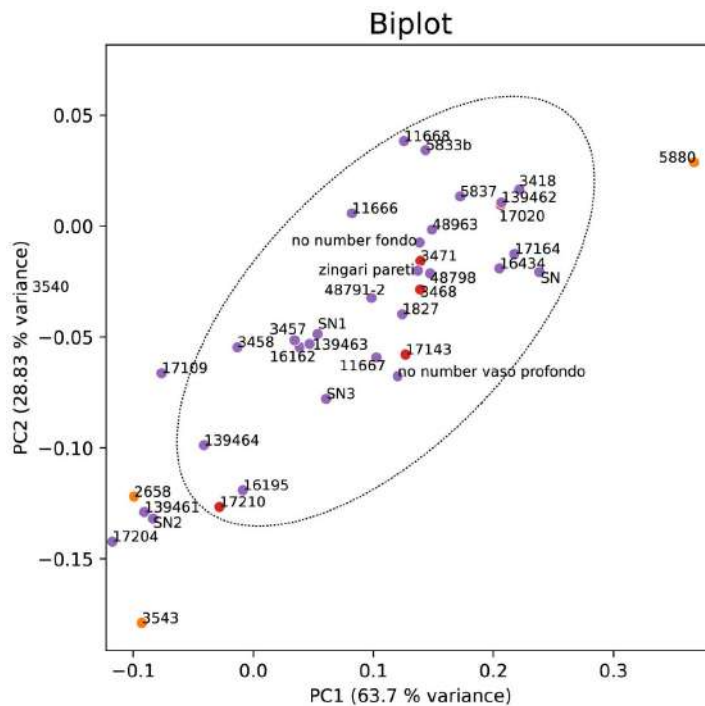


Figure 11. Zoomed view of Cluster 2 of Figure 9. For the symbols, see Figure 9.

The second cluster (labeled 2) shows a flatter direction pointing toward the pure quartz and, interestingly enough, includes only LCA/EBA ceramics. Among the Cetina-style

vessels, four fall very close to the right side of Cluster 2, while sample 17210 falls to its lowest margins, not very far from Cluster 1 (Figure 11).

3.4. Optical Microscopy

Observation of thin sections confirms the results of XRD analysis and provides additional information about the pastes of the Neolithic, ECA, LCA, and EBA investigated vessels (Table S2).

3.4.1. Neolithic Vessels

Most of the Neolithic vessels, including *vasi a coppa*, carinated bowls, bowls, and a decorated beaker, show very similar features (Figures 2–4; 12). They are characterized by very abundant sparry calcite (up to 40%) and less common carbonate rock fragments as temper material and a fine-grained matrix, containing angular quartz grains with silt- to very fine-sand-size and, occasionally, clay pellets (Figure 12). The globular vessel analyzed (Figure 3, no numb. 159), whose typology is not typical of the Vlaška Culture, is similar to the previous ones, but it additionally contains a grog fragment and a fossil, probably an echinoid fragment.

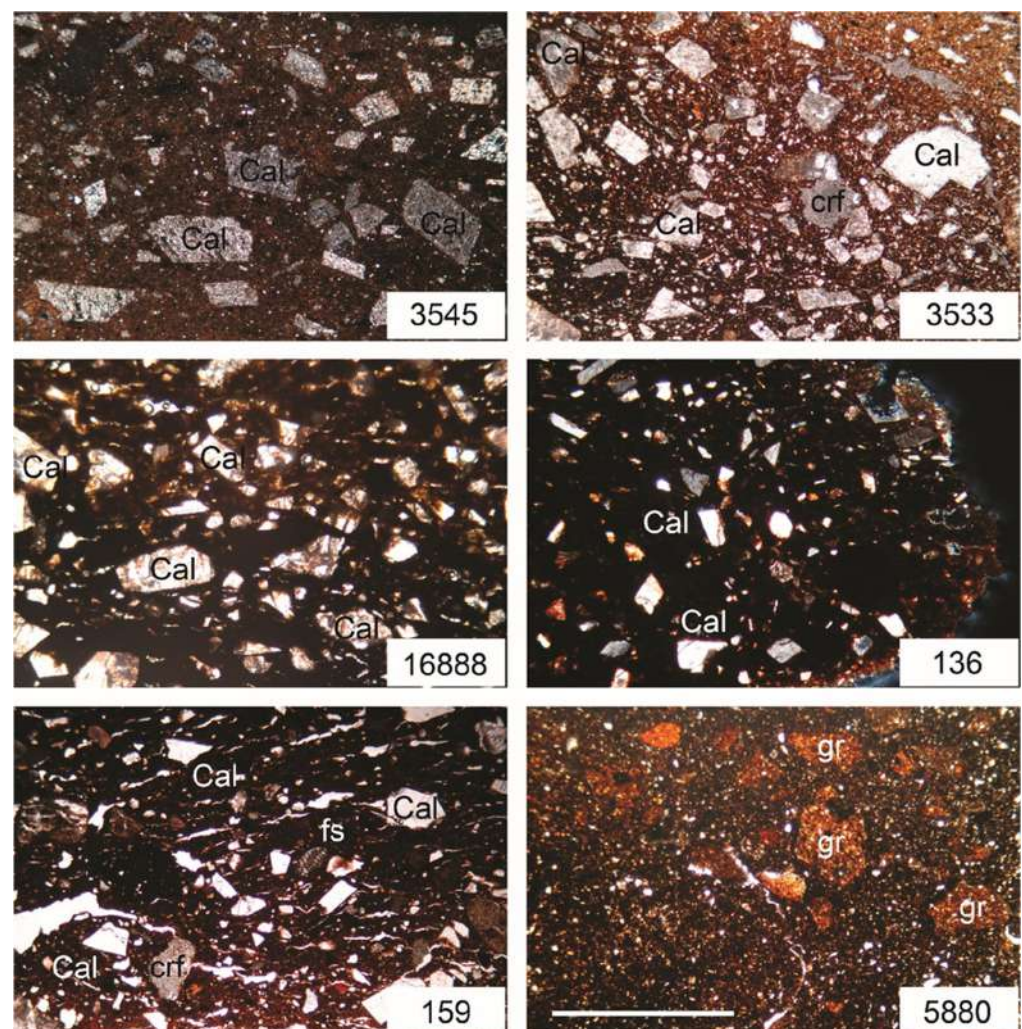


Figure 12. Thin-section photomicrographs of selected Neolithic vessels: 2 *vasi a coppa* (3545, 3533; crossed-polarised and plane-polarised light, respectively); 1 carinated bowl (16888; plane-polarised light); 1 decorated beaker (136 = no numb. 136; crossed-polarised light); 1 globular vessel (159 = no numb. 159; plane-polarised light); and 1 Fiorano-like flask (5880; plane-polarised light). Cal: calcite; crf: carbonate rock fragment; gr: grog; fs: fossil. Scale bar for all images: 2 mm.

The calcite was probably obtained by grinding speleothems, which are very common in the Karst environment where the archaeological sites—all caves—are located [15,66,67]. Given the excellent preservation of the calcite [68], it is reasonable to infer firing temperatures below 700 °C for the majority of the pottery, as previously suggested for other Danilo–Vlaška ceramics analyzed in the past [15,66,67].

Considering their mineralogical composition, the studied Vlaška artifacts correspond to Cluster 1 of the PCA diagram in Figures 9 and 10.

As already pointed out by [15], the Fiorano-like jug 5880 features a completely different fine-grained paste (Figure 12), small sub-angular quartz, and minor feldspar grains (optically plagioclase) with silt- to very fine-sand-size. Calcite is completely absent. Moreover, it contains abundant grog fragments (0.5–2 mm size), which resemble, in terms of fabric and mineral content, the other parts of the potsherd. Presumed dehydrated colloids and some fragments of phyllosilicates are present. The higher quantity of oxidized material and degassing bubbles (oxidized firing bubbles) suggest the use of higher firing temperatures.

Such a different paste compared to the Vlaška ceramics is reflected in the PCA analysis of XRD data in Figures 9–11, where the sample falls very far from Cluster 1 but also outside Cluster 2, including post-Neolithic ceramics.

3.4.2. Early Copper Age Pottery

The two vessels analyzed, showing a typology resembling Nakovana materials, are indistinguishable from the Neolithic Vlaška ceramics from a technological point of view (Figure 13). Both vessels are characterized by very abundant sparry calcite within a fine-grained matrix containing angular quartz grains with silt- to very fine-sand-size and occasionally clay pellets.

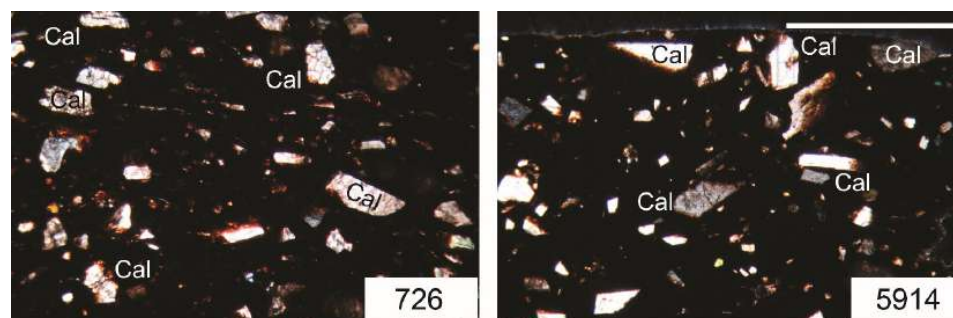


Figure 13. Thin-section photomicrographs of the analyzed Early Copper Age vessels. Cal: Calcite; crossed-polarised light; scale bar: 2 mm.

3.4.3. Late Copper Age Pottery

Petrographic and microCT data confirm significant differences in the fabric and lithic components of most investigated samples compared to the analyzed Neolithic ceramics (Figure 14). This is not surprising, considering the wide temporal gap between the early Neolithic and the third millennium BC of the Karst region.

With a few exceptions, LCA vessels exhibit a finely grained paste rich in grog temper and frequently contain flint. Additionally, small muscovite crystals and abundant quartz are present in the matrix, accompanied by less common feldspar. Occasional small lithic fragments, mostly from metamorphic rocks and sandstones, have also been identified (Table S2; Figure 14). Furthermore, the presence of some partially decomposed carbonate grains identified in SN1 (Figure 14) suggests the use of firing temperatures exceeding 700 °C, at least for some vessels. More specifically, in sample SN1, the remnants of a calcite grain exhibit a rounded shape with a potential reaction rim. Its shape, implying a volume increase and incomplete decomposition, suggests a firing temperature likely ranging between approximately 700 °C and 800 °C when the conversion of calcite into burnt lime is typically complete ([68] and references therein).

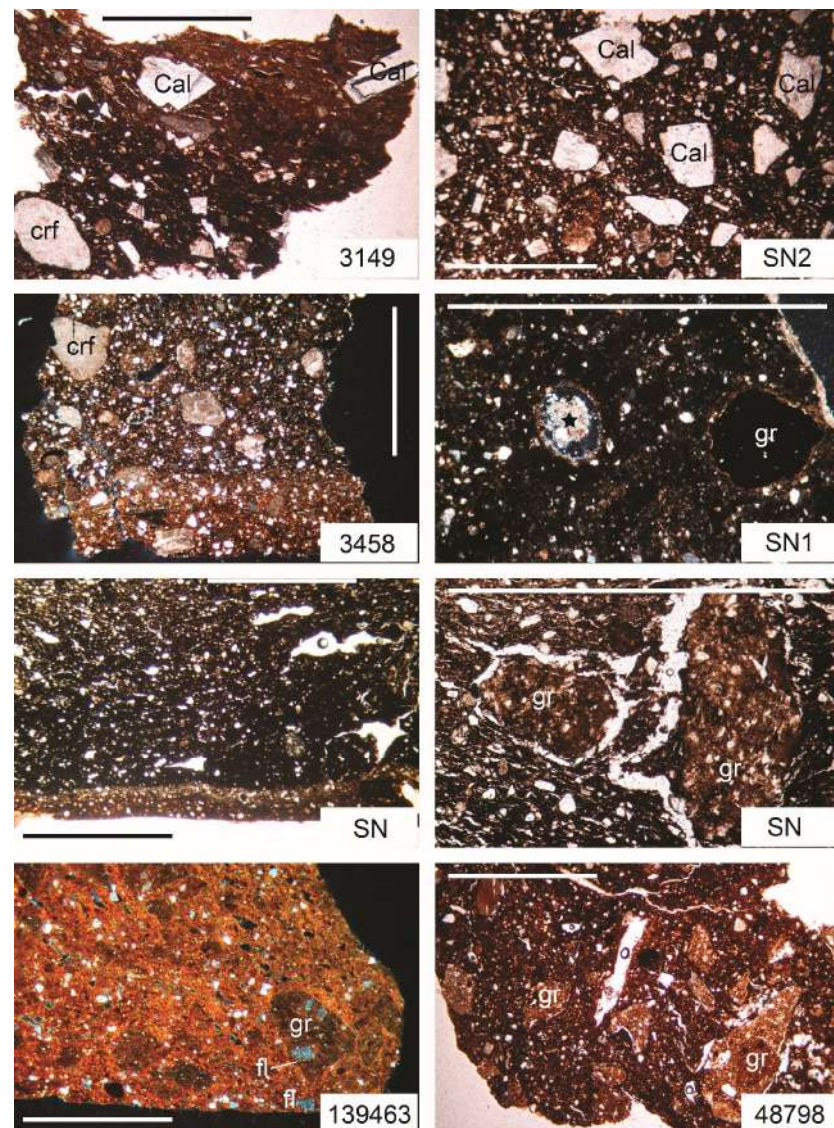


Figure 14. Thin-section photomicrographs of selected Late Copper Age vessels. Vessels 3149 and SN2 are relatively rich in sparry calcite akin to the Neolithic pottery, while others feature abundant grog without sparry calcite. Cal: calcite; crf: carbonate rock fragment; fl: flint; gr: grog; the black star indicates a partially decomposed carbonate. Plane-polarised light: 3149, SN2, SN, 48798; crossed-polarised: 3458, SN1, 139463. Scale bars: 2 mm.

However, it is worth noting that a few artifacts (3149, 3469, SN2, 405, 17204, 17109) stand out as exceptions. These artifacts exhibit a relatively high content of sparry calcite, showcasing a paste and petrographic composition comparable to that of Neolithic vessels (Figure 14). The presence of calcite in vessel 3469 has been ascertained by virtual sectioning of the microCT dataset.

Finally, some vessels exhibit an external band, possibly achieved by applying a thin layer of fine-grained clay (Figure 14, SN), while in others, it likely results from (controlled?) oxidizing firing conditions (Figure 14, 3458).

3.4.4. Cetina Pottery

The analyzed Cetina vessels demonstrate considerable similarity to the majority of LCA ceramics in terms of their very fine paste grain, the abundant presence of grog temper and flint, and the occurrence of feldspar in two thin sections (Table S2: 17020, 3471;

Figure 15). Three of our vessels show a thin external band (17020, 17143, 17210; Figure 15), most likely due to (controlled?) oxidizing firing conditions.

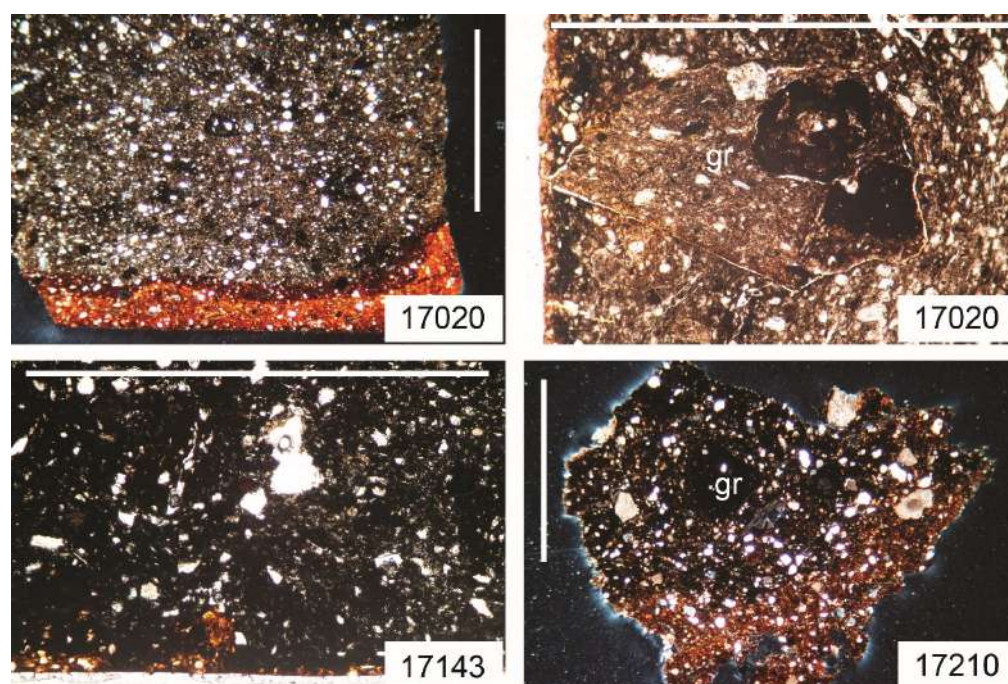


Figure 15. Thin-section photomicrographs of selected Cetina vessels. Plane-polarised light: 17020 right; crossed-polarised: all others. Most of the transparent lithic grains correspond to quartz. Scale bars: 2 mm.

4. Discussion and Conclusions

The mineralogical and petrographic data obtained from the analyzed vessels have revealed notable distinctions between Neolithic ceramics and the majority of LCA and Cetina pottery.

With the exception of the Fiorano-like flask, Neolithic pottery demonstrates uniform characteristics, notably the prevalence of sparry calcite and the absence of grog temper.

An interesting observation made possible by the new data is that not only the “classical” *vasi a coppa*, with recurring decoration or undecorated, but also *vasi a coppa* with unique motifs and vessels of different shapes—such as open bowls with raised rims, carinated bowls/biconical plates and more generic bowls—exhibit remarkably similar mineralogical and petrographic compositions (Figures 2–4).

On the one hand, this similarity was somewhat unexpected because the “classical” *vasi a coppa* and the other vessels are usually associated with Neolithic contexts, but the former is found in a much higher percentage than the latter. This imbalance, together with quite precise comparisons with Danilo pottery, might have led to the hypothesis of local production versus importation: a hypothesis that the results of our analyses would essentially question. On the other hand, previous analyses of rare artifacts culturally linked to the Dalmatian coast and the Danilo Culture, such as rhyta [66,67], had already indicated a mineralogical composition very similar to that of the much more common *vasi a coppa*, suggesting a shared local origin.

If the available data suggest a local production for all types of Danilo–Vlaška vessels, the scarcity of archaeometric investigation of Istrian and Dalmatian contemporary pottery and raw materials [69,70] presents a significant obstacle to their interpretation. This is especially true considering that the Eastern Adriatic coast mostly shares similar geology with the Karst region.

Conversely, the Fiorano-like flask (Figure 4, 5880) is highly probable to be an import, potentially originating from the Fiorano Culture in Northern Italy. Its non-local origin

is evident in a distinct typology and paste composition characterized by an abundance of grog fragments, absence of calcite, and a matrix containing small sub-angular quartz alongside minor feldspar grains, notably optically resembling plagioclase.

As far as the globular vessel analyzed (Figure 3, no numb. 159) is concerned, although its shape is not typical of the Vlaška Group, its basic paste characteristics are similar to those of the other Vlaška components, but it additionally contains a grog fragment and a fossil, probably an echinoid fragment. As no typological comparison for this pot has been found so far, with the exception of a few within the Danilo Culture (see the comparisons already mentioned above), these results are interesting but not easily explainable.

Moreover, another intriguing result is that the two vessels typologically comparable to Nakovana Culture materials [42–44] (Figure 4, 5914, no numb. 726), tentatively attributed to ECA, show technological features similar to those of Neolithic Danilo–Vlaška ceramics. Nakovana pottery has also been found in Istria, but the lack of unquestionable stratigraphic sequences in both areas, on the one hand, and the absence of archaeometric data regarding contemporary Istrian and Dalmatian pottery, on the other, prevents, also in this case, any definitive conclusion regarding local or non-local production.

Regarding the LCA ceramics, the current data suggest a probable local production primarily for a limited subset of potsherds. Specifically, this subset comprises those containing sparry calcite, which, according to the PCA diagram of XRD spectra, align within the same category as Neolithic materials (Cluster 1 of Figures 9 and 10).

On the contrary, the majority of LCA vessels exhibit distinct characteristics compared to their Neolithic counterparts. These distinctions include a very fine-grained paste, the absence of sparry calcite, a notable use of grog temper and flint, elevated quartz content, and a relatively abundant presence of muscovite in the matrix. Notably, in the PCA diagram of XRD data, these vessels cluster closely together in a specific region (Cluster 2 of Figures 9 and 11), confirming their relatively high quartz and low calcite contents. Moreover, the collected evidence suggests higher firing temperatures, likely ranging between approximately 700 °C and 800 °C, for at least some LCA vessels compared to those estimated for the Neolithic pottery.

At present, determining whether the mineralogical and petrographic uniformity observed in the majority of analyzed LCA ceramics stems solely from technological advancements compared to Neolithic production remains challenging. It is plausible that certain studied potsherds were not locally produced, considering the presence of components less commonly found in the Karst area (i.e., muscovite, flint, and metamorphic rock fragments) [15,66,67]. However, arriving at definitive conclusions would require a more extensive dataset, including an analysis of the chemical composition of both artifacts and natural raw materials. In this regard, the same research team responsible for this paper has already begun mineralogical, petrographic, and chemical analyses on around 50 vessels from the main LCA sites of Ljubljansko barje. The forthcoming results will hopefully enable a comparison, shedding light on whether the Karst region and present-day Central Slovenia not only shared strong cultural connections during the first half of the third millennium BC but also exchanged ceramics in addition to metal and stone artifacts [14].

According to the obtained mineralogical and petrographic data, four out of five analyzed Cetina vessels bear a good resemblance to the majority of LCA ceramics (Cluster 2 of Figure 9). They share common characteristics: a very fine-grained paste; use of grog temper; and use of flint. Moreover, some vessels of both Ljubljana and Cetina Groups show an external band, possibly achieved by applying a thin layer of fine-grained clay or due to (controlled?) oxidizing firing conditions.

Unfortunately, only a limited amount of preliminary archaeometric data regarding Cetina and Cetina-like ceramics from Dalmatia (Croatia) and other locations in the central Mediterranean is available. However, it is interesting to note that one of the predominant features observed in the few analyzed samples from Croatia is the presence of grog tempering [70,71], which is also evident in the Karst Cetina vessels discussed in this study.

In summary, archaeometric analyses reveal significant technological similarities between Danilo–Vlaška ceramics and the few artifacts attributed to the Nakovana Culture dated to the ECA. Concurrently, our findings indicate a marked technological disparity between the Neolithic and ECA materials and the bulk of LCA/EBA pottery, primarily linked to the Ljubljana Culture and a few to the Cetina Culture. These differences clearly suggest the use of new raw materials, recipes, and techniques, likely reflecting changes in cultural and social contexts and potential connections with the core area of the Ljubljana Culture. These changes may, in turn, be linked to the arrival of new populations on a European scale [47,48]. It is also particularly intriguing that the analyzed ceramics associated with the Ljubljana and Cetina Cultures exhibit a remarkably uniform technology, raising new questions about the relationship between these two pottery styles.

Supplementary Materials: The following supporting information can be downloaded at <https://www.mdpi.com/article/10.3390/heritage7060139/s1>, Table S1 List of analyzed samples and Table S2 Petrographic features of the vessels analyzed by optical microscopy.

Author Contributions: F.B. designed and led this study; F.B., E.L. and M.M.K. wrote the introduction, archaeological context, and conclusions; F.B., A.D.M., D.L., N.B. and M.V. wrote methods and discussion and conclusions; all authors reviewed this manuscript. All authors have read and agreed to the published version of the manuscript.

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Data Availability Statement: The data that support the findings of this study are included in this paper.

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Conflicts of Interest: The authors declare no conflicts of interest.

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