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Title: High resolution sourcing of pottery demonstrates long-distance mobility in the North Western Mediterranean during the Neolithic transition

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Abstract: The Neolithisation of the North-Western Mediterranean is still an open issue. New data recently enriched the chronological and cultural archaeological framework, bringing more precise absolute dates and showing a new and more complex process of expansion of farming in Southern Europe.
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Dear Editor,

Would you please find our proposal of a manuscript submission to Journal of Archaeological Science.

There are four main reasons that allow us to think that this manuscript can be interesting for a possible publication in Journal of Archaeological Science:

- We report a new accurate analytical approach for Neolithic pottery sourcing.
- We were able to precisely circumscribe the source area for pottery production through discriminant geochemical proxies.
- Our petrographic and geochemical results on Impressa Neolithic potteries from two distant well-dated sites (France and Italy) provides the first evidence for interregional relationships over a span of more than 1000 km in the Western Mediterranean.
- These results allow us to propose an unexpected milestone in the first Neolithic migration path from Southern Italy, towards the Central and High Tyrrhenian, and further to the Mediterranean Languedoc.

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We thank you in advance for considering our proposal,

With our best regards,

Marzia Gabriele

Highlights

- High informative potential of multi-analytical approach for pottery sourcing
- Geochemical proxies precisely circumscribe the source area for pottery raw material
- The results provide the evidence for Neolithic interregional relationships
- Unexpected milestone in the first Neolithic migration in the NW Mediterranean

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Title

High resolution sourcing of pottery demonstrates long-distance mobility in the North Western Mediterranean during the Neolithic transition

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Abstract

The Neolithisation of the North-Western Mediterranean is still an open issue. New data recently enriched the chronological and cultural archaeological framework, bringing more precise absolute dates and showing a new and more complex process of expansion of farming in Southern Europe.

The Mediterranean route of colonization (6000-5600 BCE), is characterised by the so-called Impressed Wares (IW) or Impresso-Cardial Complex (ICC) showing a huge internal diversity in material culture, notably in pottery style and technology. This polythetic imprint of the ICC is intimately linked to dynamics of raw materials exploitation (such as obsidian) and interconnections within circulation and exchange networks of goods.

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Keywords

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1 Introduction

1.1 Tracking the farming pioneers in the N.W. Mediterranean

The spread of farming and neolithic ways of life from the Eastern Mediterranean and the Aegean towards Western Europe is well known for having followed two main routes (Childe, 1925). The continental one, through the Central Balkans and the Danube valley, which was at the origins of the Linearbandkeramik Complex (LBK), reached Northern France after 5350 BCE (Whittle, 2018). The Mediterranean route, linked to the Impressed Wares or Impresso-Cardial complex (ICC), reached Southern France at least five centuries before, c. 5850 BCE (Binder et al., 2017). Many issues are related to the social dynamics at the origin of erratic dispersal of the very first ICC farming communities in the Western Mediterranean, which highly contrasts with the LBK pioneer front.

In both cases, the role played by migrants within these processes has been demonstrated by genomics (Mathieson et al., 2018). Concerning the ICC, the modelling of a large set of audited radiocarbon dates currently places its formative stage in Southern Italy and Dalmatia during the very beginning of the 6th millennium BCE, mostly after 5950 BCE (Binder et al., 2017; McClure et al., 2014). Few genomic data are currently available for the earliest ICC aspects, i.e. in Croatia, from Zemunica cave between 6000 and 5750 BCE and from Kargadur, between 5670 and 5560 BCE: they strengthen the idea of a genetic connection with the Balkans, Aegean and Anatolia, regarding the maternal and paternal lineages (Mathieson et al., 2018). These evidences raise new issues about the possible roots of the ICC in the second half of the 7th millennium BCE, within the Southern Balkans and the Aegean, in the contexts of the Monochrome or Proto-Sesklo Pottery which punctually reached the Ionian sea (Berger et al., 2014).

North and westwards, in Italy and France, analyses of Neolithic DNA are very rare and mostly concern later periods (Lacan et al., 2011; Rivollat et al., 2017). In this area, peopling dynamics are mainly demonstrated by transfers of material culture and domestic taxa. For instance, it is now well known that domestic animals and crops are exogenous (mainly sheep, goat, wheat and barley) and originated from Southwest Asia (Rowley-Conwy et al., 2013). This enables to study the spread of animal breeding and agriculture and to observe their rhythms and pathways. In this framework, systemic studies of material culture also offers the possibility to track the trajectories of the first farmers (Bernabeu Auban et al., 2017; Ibáñez-Estévez et al., 2017).

From each part of the Italian Apennine chain, peopling dynamics seem to be diverse regarding the cultural connections as well as the diffusion tempo. On the Adriatic side, the ICC settlements kept highly concentrated in Apulia, Basilicate and East Calabria during c. two centuries, crossing over the Tavoliere towards Central Italy at a rather late period: c. 5750 BCE in the Abruzzo and c. 5600 BCE in the Marche. In contrast, on the Tyrrhenian side, the meshing of earliest settlements appears very sparse while the diffusion speed appears to be very fast: actually the far Ligurian and French coasts were reached as soon as c. 5850 BCE (Binder et al., 2017). Similarly, the first data on pottery technology indicate that different communities of practice occurred in the Adriatic and Tyrrhenian sides. In the Adriatic area, the forming methods using coils and long slabs were clearly related to the Balkans' tradition, whereas a distinctive Spiralled patchwork technology (SPT) was in use in the Tyrrhenian (Gomart et al., 2017).

Obsidian is well known for having played a great role during the earliest Eastern Mediterranean Neolithic (Dixon et al., 1968), and especially a symbolic one (Cauvin, 1998). Totally ignored by the Late Hunter-Gatherers from Western Mediterranean, its use has been transferred to the West as part of the Neolithic package. Most of the attractive sources of obsidian are located in the western islands (Pantelleria, Lipari, Palmarola and Sardinia) where this glass was exploited and spread there from the earliest stage of the ICC (Ammerman and Andrefsky, 1982; Muntoni, 2012; Tykot et al., 2013). Although there is currently no evidence of early ICC settlements located close to the obsidian sources, tools made of obsidian from Palmarola and Sardinia have been identified in the earliest ICC from the North-Western Mediterranean, especially at Arene-Candide in Liguria (Ammerman and Polglase, 1997), Peiro Signado and Pont de Roque-Haute in the Mediterranean Languedoc (Briois et al., 2009; Binder et al., 2012).

Until now the obsidian transfers through the Western Mediterranean have been considered as the main evidence of maritime voyaging since geochemical analyses provided unquestionable results for linking distant sites or people, compared to the simple analogies suggested by pottery styles.

These data shed light on the Tyrrhenian as a specific cultural landscape where the sea could have played a central role during part of the ICC. One of the key-issues in this context is related to the range and the regime of maritime mobility. Researches carried out in the last couple of decades have shown that the Mediterranean area is a hot spot of cultural diversity (Rigaud et al., 2018). Furthermore, this maritime area probably offered the possibility of multidirectional movements, but also different forms of mobility (pioneering, travelling, interactions and exchanges) (Manen et al., 2018). Consequently it is still difficult to precisely identify circulation routes, cultural filiations and origin of the incoming farmers. In this study a multi analytical approach to pottery provenance through petrographic and geochemical analyses has been implemented to provide insight into the crucial question of Neolithic dispersal routes.

1.2 Pottery pastes as a proxy for human trajectories

In this work we demonstrate that pottery sourcing analysis provides a major contribution for tracking ICC networking dynamics in the Western Mediterranean.

Pottery studies have been developed for long in the Mediterranean Neolithic contexts (Capelli et al., 2017, 2008; Convertini, 2010, 2007; Echallier, 1991; Ferraris and Ottomano, 1997; Gabriele, 2014, 2015; Gabriele and Boschian, 2009; Manen et al., 2010; Martini et al., 1996; Muntoni, 2003; Paolini-Saez, 2010; Spataro, 2002; Ucelli Gnesutta and Bertagnini, 1993). In most cases, analyses have indicated a local production of ICC pottery, while non-local ones are exceptions. The limited range of such transfers (10 to 100km) generally suggests that the pottery trade was embedded in functional networks, illustrating the logistical mobility of the first ICC farmers (Binder, 1991a; Capelli et al., 2017; Manen and Convertini, 2012).

However, previous studies have already highlighted the possibility of long distance pottery transfers in the Mediterranean Languedoc (Convertini, 2010, 2007), the Liguro-Provençal arch and Tuscany (Capelli et al., 2017, 2008; Gabriele, 2014, 2015), especially with regard to the presence of volcanic component of the paste (hereinafter referred as volcanic pottery and paste). The latter offer specific petrographic markers and geochemical features of a very high resolution, as demonstrated by a large set of pottery studies from distinct regions (Comodi et al., 2006; Barone et al., 2010; Brunelli et al., 2013; Palumbi et al., 2014; Belfiore et al., 2014; Scarpelli et al., 2015; La Marca et al., 2017). Furthermore, recent applications of in situ geochemical methods on non-volcanic mineral inclusions allow to enhance the accuracy and reliability of provenance analyses (Gehres and Querré, 2018).

Here, we provide a multi analytical comparative approach to pottery characterization and provenance through petrographic and geochemical analysis. Studying non-local volcanic pottery from the two ICC sites of Portiragnes – Pont de Roque-Haute (Languedoc, France) (hereinafter referred as PRH) (Guilaine et al., 2007) and Giglio - Le Secche (Tuscany, Tuscan Archipelago, Italy) (hereinafter referred as GLS) (Brandaglia, 2002) (Fig. 1), we demonstrate long-distance circulation of pottery – more than 1000 km following the coast or 600 km as a bird flies - between Central Italy and Languedoc.

2 Materials and Methods

2.1 Sites and samples

The open-air site of PRH, which offered a set of pits dug in a fluvial terrace, was interpreted as a short duration settlement in a ria (Guilaine et al., 2007). The modelled age of this occupation is estimated between 5860-5710 and 5800-5680 BCE, i.e. one of the earliest ICC settlements currently known in the Western Mediterranean (Binder et al., 2017). Together with domestic remains (mammals, seashells and tools including obsidian from Palmarola) a series of ca 603 sherds (at least 55 individuals) (Manen and Guilaine, 2007) indicates a local pottery production exploiting reworked alluvial Pliocene deposits (Convertini, 2010, 2007). Few individuals are characterized by volcanic aplastic components and among them one pot is impressed with the umbo of a *Cardidae* shell (Fig. 2A).

The site of GLS is a shelter close to a north-western beach of Giglio Island. Rich deposits of well preserved pottery (Brandaglia, 1991) associated to a large set of Palmarola obsidian tools (Barone et al., 1996; Brandaglia, 1987) demonstrated its long attendance, starting during the earliest stage of ICC (5840-5540 BCE) and lasting at least during the second half of the 6th

millennium BCE (Binder et al., 2017). Most of the earliest pottery was built using local residual deposits on granite formation, with the notable exception of few vessels shaped in a volcanic paste and decorated with the ventral margin of a *Cardidae* shell (Fig. 2B,C) (Gabriele, 2014).

2.2 Analytical methods

Petrographic and chemical methods were performed through different scales of observations on each specimen *via* different supports in order to have comparable and complementary data.

First, petrographic analysis were obtained by stereomicroscopy directly on the tree pottery fragments and by standard optical microscopy on six thin sections, whit support of scanner images, to characterized a-plastic inclusions and fabric textural features. For each archaeological samples two thin section are available, whose one covered and one uncovered, for elemental analysis too. Description of textures of inclusions, pores and matrix were performed following guidelines of soils micromorphology (Stoops, 2003) and ceramic description proposals (Quinn, 2013; Whitbread, 1989). The examinations were carried out at CEPAM's laboratory (CNRS, Nice, France).

Subsequently, to be able to verify the real compositional correspondence between potteries of both sites, chemical analysis on majors and trace elements were carried out to determine composition of whole pottery and mineral inclusions, such as clinopyroxene. Microchemical in-situ analysis on clinopyroxene are based on the assumption that its chemical composition is a marker of chemical composition of parental magma (Barone et al., 2010; Leterrier et al., 1982). Indeed, crystal-chemistry of clinopyroxene is related to different geochemical and petrological magma affinities (Cellai et al., 1994; Cundari and Salviulo, 1987; Gentili et al., 2014). Finally, for discerning our hypothetically petrographic and geochemical possible sources, data available in scientific literature are used.

The bulk pottery compositions were obtained by Inductively Plasma Atomic Emission Spectrometry (ICP-AES) and Inductively Coupled Mass Spectrometry (ICP-MS), for major and trace elements respectively on two pottery powders at the Geochemical and Petrographical Research Center in Nancy (SARM laboratory, CNRS-CRPG; Supplementary dataset) following the procedure described in Carignan et al. (2001).

Chemical analysis by environmental scanning electron microscope (FEI PHILIPS XL30 ESEM) equipped with an Energy Dispersive Spectroscopy (EDS) system for X-ray microanalysis (Quantax XFLASH6/30 silicon drift 10mm²) have been applied on 74 clinopyroxene and 53 K-feldspar (sanidine) selected single crystal grain minerals found as inclusions in polished thin section and mapping on scanner images. The analysis were carried out at the laboratory of the Centre for Material Forming (CEMEF, Ecoles des Mines de Paris, Sophia Antipolis, France).

Laser Ablation Inductively Coupled Plasma Mass Spectrometry (LA-ICP-MS) examinations for estimating major and trace elements have been applied on clinopyroxene single crystals found as inclusions in the epoxy impregnated ceramic samples, left from the processing of thin sections, by stereomicroscopy observations and mapping on scanner images. LA-ICP-MS analysis were undertaken on 78 selected clinopyroxene grain minerals, other than those of SEM-EDS investigations (Supplementary dataset). The largest clinopyroxenes were selected in order to avoid possible contamination by other mineral species or clay paste from the ceramic during the ablation process. To prevent any pollution of the argon/helium carrier gas flow during the ablation process, the sherds were cleaned in an ultrasonic bath to remove the microscopic dust particles produced by polishing. LA-ICP-MS analysis was conducted at the laboratory of Centre Ernest-Babelon of the IRAMAT (Orléans, France).

The analytical protocol (pit ablation mode) developed for obsidian inclusions analysis (Palumbi et al., 2014) has been adapted to the analysis of clinopyroxenes by measuring magnesium on 25Mg instead of 24Mg. This allows sampling the clinopyroxenes with a larger laser beam diameter (up to 100 µm according to the mineral grain size) without saturation of the detector by this element and thus improving detection limits of elements such as the rare earths.

As encountered with the analysis of obsidian inclusions, one of the critical parameters of this type of analysis (pit mode) is the thickness of the analysed clinopyroxene grains, owing to the fact that they were inserted in a ceramic paste and that they may contain other mineral species included in their structure.

Consequently, in order to avoid overshooting the inclusions and to maintain a high signal level, a 10 Hz laser pulse frequency was used and the analytical time was reduced from 55 to 25 seconds (8 seconds for pre-ablation and 17 seconds for analysis), that is 8 mass scans from lithium to uranium.

To ensure that the measured signal is not perturbed by the presence of other mineral species its evolution is systematically checked during the whole ablation. If other mineral phases are encountered, the calculation protocol developed to study concentration profiles in glass is applied to calculate the clinopyroxene composition and to identify the other mineral phase if it is possible (Gratuze, 2016).

However, the contribution or the modification brought by another mineral species to the signal measured for a clinopyroxene may not be always easy to detect if the composition of both species is fairly similar or if the proportion of the perturbating mineral specie in the whole signal is weak. It is thus only when the chemical contrast between both species is important that the correction of the signal is possible, as illustrated by the presence of a zircon grain in one of the recorded spectrum or by à transition between a clinopyroxene and a feldspar. For most of the other cases the presence of another mineral specie may not be detected and will just increase the dispersion or the variability of the calculated compositions. To avoid clay contaminations, the analysis were carried out in the middle of the clinopyroxene grains. When possible the largest grains were selected for the analysis, however, analysis of very small grains were also carried out by adapting the laser beam diameter.

External calibration was performed using the National Institute of Standards and Technology Standard Reference Materials 610 (NIST SRM610), along with Corning reference glasses B and D. 28Si was used as an internal standard. Concentrations were calculated according to the protocol detailed in Gratuze (2016). Detection limits range from 0.01% to 0.1% for major elements, and from 20 to 500 ppb for minor and trace elements. Compatibility of data is monitored by the regular analysis of reference materials NIST SRM612 as unknown sample.

3 Results and discussion of the comparative study

3.1 Petrographic analysis of pottery pastes

At a stereo-microscopic scale, pottery pastes are significantly characterised by sub-rounded/rounded green and dark-green clinopyroxene and colourless or whitish feldspar inclusions (Fig. 3A, B), up to very coarse sand size. However, more heterometric and larger lithic inclusions are also observable. Pastes are friable and not homogeneous in colours (Fig. 3A, B).

At thin sections optical-microscopic scale, porosity is characterised by meso- and macro planes and few macro vughs. The porosity distribution and orientation are well expressed in the GLS samples, where have been recognised concentric features and parallel, inclined and bow-like bands of oriented planes. A-plastic inclusions are common, mostly sub-rounded and rounded, moderately sorted on fine-medium sand size, within elements more or less larger. Grain distribution and orientations are weakly-moderately expressed, up to single- and double-spaced relative distance. Clinopyroxene and K-feldspar (sanidine) are the most common minerals (Fig. 3C, D). Clinopyroxene is frequently rounded, coloured whit green pleochroism and twinned (Fig. 3C-F). Sanidine is generally less rounded and larger (up to very coarse sand size) than clinopyroxene, fresh and clear, Carlsbad twinned (Fig. 3C-F). Other mineral grains are identified in different proportions and size, within oxides, quartz, plagioclase, black and white micas. Lithic inclusions are generally rounded, heterometric, up to very coarse sand and very fine gravel size. Lithoclasts are identified as alkaline volcanic rocks (Fig. 3G, H), sandstone, siliceous sedimentary rocks, and quartz-metamorphic rocks. Matrix is optical active in GLS samples with stippled-speckled b-fabric and striated b-fabric. Colour is heterogeneous, linked to Fe reduction on the margins and Fe oxidation on the core of the fragments.

3.1.1 Petrographic possible volcanic source areas

Because of petrographic and archaeological considerations, most likely source of raw materials are the Italian Miocene-Quaternary potassic and ultrapotassic volcanic rocks from the so-colled Volcanic Provinces (hereinafter referred as VP) part of the Magmatic Provinces of the Tyrrenian region (Fig. 1) (Conticelli et al., 2004; Peccerillo, 2017). Furthermore, petrographic pottery data, namely characteristic association of sanidine and green clinopyroxene minerals with minor amount of volcanic, sedimentary and metamorphic lithoclasts, suggests considering volcanic formations hydrographically or geomorphologically linked with ones of different geological origins. In this perspective, more suitable volcanic centres are the Monte Amiata in the Tuscany VP (hereinafter referred as TVP) (Conticelli et al., 2015; Cristiani and Mazzuoli, 2003), and Vulsini (Barton et al., 1982; Holm, 1982; Palladino et al., 2014), Vico (Barbieri et al., 1988; Palladino et al., 2014; Perini et al., 2004; Perini and Conticelli, 2002) and Sabatini (Conticelli et al., 1997; Del Bello et al., 2014; Palladino et al., 2014) districts in the Roman VP (hereinafter referred as RVP). We cannot a priori exclude Roccamonfina (Ghiara et al., 1979), Phlegrean Fields (Armienti et al., 1983; Belkin et al., 2016; Civetta et al., 1997; Fedele et al., 2009; Mollo et al., 2016) and Somma-Vesuvius (Bertagnini et al., 1998) districts in the Campania VP (hereinafter referred as CVP) (in this paper Roccamonfina volcanic district is considered part of the CVP, Fig. 1; moreover for the CVP we haven't been considered data on eruptions youngers than 8 ka).

In addition, for comparison we can consider same volcanic districts that can bring a similar K-feldspar-clinopyroxene mineralogical association than archaeological pottery samples. For example, the Italian Miocene-Quaternary volcanic rocks of San Vincenzo (Feldstein et al., 1994; Ferrara et al., 1989; Poli and Perugini, 2003a) and Monte Cimino districts (Perugini and Poli, 2003; Conticelli et al., 2013) in the TVP, Monte Vulture Volcano (Bindi et al., 1999) in the Apulian VP (hereinafter referred as AVP); the Miocene-Quaternary Monte Arci (Dostal et al., 1982) district and the Oligo-Miocene Bosa-Alghero, Anglona and Logudoro districts (Guarino et al., 2011) of the Sardinia VP (hereinafter referred as SVP) (in this paper the different Sardinian volcanic districts are considered in the same VP, Fig.1).

Conversely, because of their entirely volcanic origin, some thyrrenian islands such as Capraia (Tuscan archipelago, Tuscany) (Chelazzi et al., 2006; Poli and Perugini, 2003b), Ponza (Pontine archipelago, Latium) (Conte and Dolfi, 2002; Paone, 2013) and Vulcano (Aeolian archipelago, Sicily) (Faraone et al., 1988) are unsuitable, even if they can bring a K-feldspar-clinopyroxene mineralogical association. At the same time, the basaltic volcanic formations near the site of PRH can be excluded, mainly due to the lack of K-feldspar phenocrysts in this rock type (Dautria et al., 2010). For the same reasons others French and Italian volcanic districts, as Cap d'Ail, Alban hills (Boari et al., 2009) and Monti Ernici (Boari and Conticelli, 2007; Frezzotti et al., 2007), are not considered.

3.2 Major elements of single mineral inclusions

Data of SEM-EDS analysis show alkali-feldspar minerals compositionally homogenous whit Or_{67} to Or_{85}, only one case with Or_{50} in GLS02 sample. Alkali-feldspar classification is represented in ternary diagram in supplementary figure 1.

Clinopyroxenes are predominantly composed of diopside and Fe-rich diopside; augite to Mg-rich augite and CaFe-rich clinopyroxene are also presents (Supplementary dataset). Also in case of LA-ICP-MS analysis, clinopyroxenes are predominantly composed of diopside and Fe-rich diopside with Fs_{13} to Fs_{20}; augite ($Wo_{43}En_{54}Fs_4$) to Mg-rich augite ($Wo_{44}En_{33}Fs_{23}$) and CaFe-rich diopside ($Wo_{51}En_{38}Fs_{11}$ to $Wo_{52}En_{28}Fs_{20}$) are also presents (Supplementary dataset). Clinopyroxene classification is represented in the QUAD diagram referring to Morimoto (1988) in supplementary figure 2. Major-element chemical composition of clinopyroxene available in scientific literature allow us to differentiate within previously indicated petrographic possible sources in the TVP (Aulinas et al., 2011; Conticelli et al., 2015, 2013; Feldstein et al., 1994), RVP (Barton et al., 1982; Comodi et al., 2006; Conticelli et al., 1997; Cundari, 1975; Dal Negro et al., 1985; Del Bello et al., 2014; Gentili et al., 2014; Holm, 1982; Kamenetsky et al., 1995; Palladino et al., 2014; Perini, 2000; Perini et al., 2004; Perini and Conticelli, 2002), CVP (Armienti et al., 1983; Aulinas et al., 2008; Belkin et al., 2016; Civetta et al., 1997; Fedele et al., 2009; Ghiara et al., 1979; Mollo et al., 2016; Pappalardo et al., 2008), AVP (Bindi et al., 1999; Caggianelli et al., 1990), SVP (Dostal et al., 1982; Guarino et al., 2011). The Quad diagrams show substantially correspondence between Mg-rich augite and diopside composition of clinopyroxenes in pottery

(Fig. 4A) and volcanic rocks of RVP, CVP and AVP (Fig. 4B, C, E). Instead, partially correspondence with rocks of TVP and SVP, especially due to the lack of clinopyroxene with augite composition in pottery pastes (Fig. 4D, F). Moreover, pottery clinopyroxenes are characterized by limited compositional variations in major elements, considered as cationic values. In Ti_{tot} vs Al_{tot} binary diagrams (Supplementary Figure 3), the cluster of pottery clinopyroxene composition fits in the field of clinopyroxenes of the RVP, TVP and CVP (Supplementary Figure 3B, C, D), instead partially fits in the clinopyroxene compositionally fields of the AVP and SVP (Supplementary Figure 3E, F).

3.3 Trace Element Analysis of Whole Pottery

We realized ICP-MS trace element analysis on two bulk ceramics samples from the two archaeological sites. A soil sample from the PRH site (Sedimentary Pliocene deposits) was also analyzed. Results were reported in Supplementary Dataset. In the spider diagram (Fig. 5A) PRH and GLS potteries are geochemically indistinguishable. Their spectra display the same Large Ion Lithophile Elements (LILE) enrichment, the same high negative Ta and Ti anomalies, and the same slight Sr anomaly. Furthermore, trace element contents of rocks from the Languedoc Volcanic Province (Agde volcano and lava at the PRH site) do not display Ta, Sr and Ti anomalies (Fig. 5A), suggesting that volcanic minerals of ceramics are not derived from southern France. Indeed, Languedoc Volcanic Province corresponds to homogeneous alkali basaltic geochemistry (Dautria et al., 2010), different from typical calc-alkaline geochemistry of the subduction zones (Italian Volcanic Provinces) (Peccerillo, 2017; Gasperini et al., 2002).

PRH soil shows the same pattern than the potteries excepted for Sr with a major negative anomaly. Furthermore, PRH soil spectrum is different from the regional lavas (Fig. 5A). The PRH alluvial soil geochemistry can be interpreted as a mixing of sedimentary, metamorphic, plutonic and volcanic rocks. The absence of sanidine mineral grains suggesting that it was not used for PRH and GLS pottery.

Trace element contents from rocks of RVP, TVP and CVP are also reported (Fig. 5B). Although PRH and GLS ceramic samples match Italian Volcanic Province spectra, differences remain apparent especially for Sr, High Rare Earth Elements (HREE, i.e. Tb, Dy, Ho, Tm, Yb) and High Field Strength Elements (HFSE, i.e. Ta, Zr, Hf). Significant negative Ta and Ti anomalies are present as well in Italian Magmatic Provinces and in bulk archaeological ceramics, supporting Italian volcanic rocks as potential sources for the archaeological materials. The CVP and TVP display strong negative Sr anomaly unlike the RVP. Taking into account the Sr contents, ceramic samples are more in agreement with the RVP. Furthermore, archaeological samples display a depleted HREE content like the RVP and TVP, while the CVP provides slight HREE enrichment.

3.4 Trace Element Analysis of Clinopyroxene inclusions

LA-ICP-MS trace element analysis were performed on clinopyroxenes included in pottery paste from the PRH and GLS sites. Trace element contents are reported in Supplementary Dataset. Our data were confronted with data available in literature (i.e. trace element contents from RVP (Comodi et al., 2006; Gentili et al., 2014; Scarpelli et al., 2015) and CVP (Arienzo et al., 2009; Civetta et al., 1997; Fedele et al., 2009; Mollo et al., 2016; Pappalardo et al., 2008; Scarpelli et al., 2015) pyroxenes. In the spider diagram, PRH and GLS ceramics display the same spectra pattern, with pronounced Ta, Sr, Zr and Ti negative anomalies (Fig. 6A). Although clinopyroxenes from RVP and CVP show also similar spectra, small variance appears for Sr, Light Rare Earth Elements (LREE, La, Ce, Pr) and HREE contents (Fig. 6B). The RVP pyroxenes reach higher values for LREE, while the CVP pyroxenes can reach higher values for HREE and smaller values for Sr contents. However, spectra of archaeological pyroxene chemistry do not allow us to decipher the volcanic source accurately. For further, we investigated precise trace element contents which could be specific proxies for the sourcing. First, in the diagram Eu* vs Sm_N, we reported our data and those of the Italian Volcanic Provinces (Supplementary Figure 4). The pyroxenes of the PRH and GSL sites display similar variability and indistinguishable Eu* or Sm_N values, strengthening an identical geological source for the ceramics. Although the ceramic pyroxenes fit better with the geochemical field of the RVP, we reliably cannot exclude the potential provenance of the archaeological pyroxenes from the CVP. Further tests were made in order to find geochemical discriminant parameters (Fig. 7; Supplementary Figures 5; 6). Finally, many content data on pyroxenes demonstrate the pottery pyroxenes origin and confirm their equivalent composition. The Nd/Lu vs Ce/Lu, Sm/Yb vs La/Yb, Nd/Tm vs Ce/Tm and Zr/Y vs Ce/Y, diagrams allow to discriminate the geochemical field of RVP and CVP pyroxenes (Supplementary Figures 5B-E; 6B). For our study, the most accurate diagram is Y vs Ce where the pyroxenes from archaeological samples match the unique geochemical field of the RVP pyroxenes (Fig. 7B).

3.5 A unique source area for long distance exogenous pottery

Our study shows a clear correspondence between the three archaeological pottery samples at each levels of each method of analysis. This petrographic and chemical evenness suggests the exact same provenance for the volcanic pottery of both sites of PRH and GLS, confirming the non-local origin of the vessels. The basaltic volcanic formations near the site of PRH can be excluded, both through petrographic and geochemical analyses. Moreover, Giglio Island is not a suitable source due to the exclusive presence of granitic and metamorphic formations and the absence of volcanic formations (Capponi et al., 1997; Westerman et al., 2003). Petrographic investigations allow us to highpoint mineralogical and roundness textural features of the a-plastic inclusions of pottery pastes match of both sites. Instead, the diversity visible in the other textural features of the fabric elements (i.e. granulometry) may depend on the internal variability of the deposits used as raw material. The roundness of inclusion shows, indeed, that secondary sedimentary deposits have been used for pottery production (Capelli et al., 2008; Convertini, 2007; Gabriele, 2014).

The correspondence between elemental compositions of pottery pastes of both sites is clearly demonstrated by the results of chemical analysis of whole pottery and especially of a-plastic single mineral inclusions. In the ternary and binary diagrams the clusters of pottery clinopyroxene composition in both major and trace elements matching the same field and trend of evolution. LA-ICP-MS trace-element data of clinopyroxenes in pottery compared with literature data for clinopyroxenes in rocks of Roman and Campanian VPs allow us to distinguish between the more likely sources areas for pottery production. The correspondence between clinopyroxene compositions of archaeological and geological data is clearly demonstrated in Y vs Ce binary diagrams (Fig. 7), where trends of distribution concentration of trace elements matching each other with Roman VP. Conversely, there is no match with the cluster of Campanian VP.

These results well demonstrate the efficiency and reliability of our methodology based on consequently and complementary step of analysis. Petrography (both macro and micro observations) is the first and essential step, and must be confirmed and detailed with subsequent chemical analysis, in order to circumscribe real source areas of raw materials.

4 Unravelling early farming dynamics in the Western Mediterranean

This comparative and multi-analytical study provides the first evidence for interregional relationships over a span of more than 1000 km in the Western Mediterranean early Neolithic, through the circulation of pottery. We were moreover able to precisely circumscribe the source area for this pottery production, between the Fiora and the Tiber river basins in the Southern Toscany and Northern Latium.

These results show how pottery raw materials can act as a powerful proxy to grasp early Farmers strategies and dynamics. Such long-distance pottery transfers are embedded in a wider framework during the very first stage of the W. Mediterranean Neolithic dispersal. Its fast spread is interpreted as part of a pioneering colonization model based on the use of maritime routes, but whose social drivers are still misunderstood.

This model is suggested to be at the origin of the settlement of small Neolithic seafaring groups far from their origins. Through this process, the whole Neolithic practices and know-how were progressively transferred to an extended region. By this way, the technical traditions newly implemented in the North-Western Mediterranean are expected to be very similar to those of the origin area which is still controversial. However, PRH potters clearly belong to the community of practices developed west to the

Apennine and then significantly differ from the Adriatic and Balkans tradition (Gomart et al., 2017). Similar connections are observed in the field of cropping practices based on hulled wheats and barley and moreover in animal husbandry since PRH ewes exhibit the same morphology than most of the Tyrrhenian ones (Guilaine et al., 2007).

Together with obsidian from Palmarola, the pottery originating from Latium can help to identify an unexpected milestone in the first Neolithic migration path from Southern Italy, towards the Central and High Tyrrhenian, and further to the Mediterranean Languedoc.

As a paradox, volcanic pastes and obsidian sources exploited during the earliest Impressa stages are situated in areas of Central Italy where dwelling sites are poorly identified; the closest and earliest sites are in Latium, Settecannelle cave (Ucelli Gnesutta, 2002) in the Fiora Valley and La Marmotta on the banks of the Bracciano Lake (Fugazzola Delpino, 2002), and in Umbria, Panicarola (De Angelis, 2003) on those of the Trasimene Lake (Fig. 8). A similar situation can be observed about the Sardinian obsidian exploitation: despite the trade of Monte Arci glass towards Liguria (Arene Candide) (Ammerman and Polglase, 1997; Maggi, 1997) and Languedoc (Pont-de-Roque-Haute and Peiro Signado) (Briois et al., 2009; De Francesco and Crisci, 2007) appears from 5850-5750 BCE, only one early dwelling place has been recognized on this island and suspected, with question marks, to be contemporary (Su Coloru) (Lugliè, 2018; Sarti et al., 2012). Similarly, evidences of the earliest impressed wares are very rare in Corsica (Campu Stefanu, Cesari et al., 2014; Albertini rock-shelter, Binder and Nonza-Micaelli in press)

Considering this scarcity, one could suspect that the area where raw materials have been collected was in some way *terra incognita* for the Neolithic pioneer groups. But the same lack of data could indicate as well that the territorial meshing of the early farmers is severely underestimated today, due to various hazards, as littoral submersion, sedimentary covering, sites destructions or research weaknesses… At least, these pottery analyses reveal invisible parts of the original meshing as well as pollen revealing very early cropping within areas where Neolithic sites are currently unknown (Branch et al., 2014; Guillon et al., 2010). This observation suggests a peopling discontinuity between Southern-Italy and the Franco-Ligurian region and lead to reassess the question of leapfrog dispersal (Zilhão, 2014).

Among the issues which are opened by these results, a burning one concerns the nature and temporality of the processes occurring for acquiring various raw materials and for transferring pots or other goods at long-distance. This questions both the mobility regimes and the social interactions at the beginning of the Neolithic transition in the Western Mediterranean.

The hypothesis of short-term voyaging episodes, connecting the Northern Latium, the Tuscan Archipelago and the Mediterranean Languedoc, is toughly supported by the data. Indeed, the chronological resolution of radiocarbon dating, as well as the vagueness of stylistic comparisons, cannot allow linking those three regions throughout sole pioneer events. Actually, recent literature evokes a long duration of the production and trade of pottery from RVP, for instance in northern Latium (Settecannelle) (Ucelli Gnesutta and Bertagnini, 1993), Tuscan archipelago (Cala Giovanna Piano, Pianosa island) (Gabriele and Boschian, 2009), and Liguria (Pian del Ciliegio) (Capelli et al., 2017, 2008). At the same time, all along the 6th millennium BCE, in the whole Tyrrhenian area and Liguria, several networks are developed at a smaller range as highlighted for example by movements of wares with low pressure ophiolitic components (Capelli et al., 2017; Gabriele and Boschian, 2009; Martini et al., 1996). In Provencal area, the site of Nice - Caucade is a good example of regional multidirectional exploitation (early Impressa stage) (Convertini, 2010; Manen et al., 2006).

The multipolarity of the transfers observed for a large set of raw materials and goods have been considered as a strong argument for indirect acquisition and for the early setting of social networks (Binder and Perlès, 1990; Perlès, 2012). In the context of a pioneer colonization of the Western Mediterranean, this networking appears to be of a great spatial extension, which could indicate a very high level of maritime mobility, the development of sailing skills and durable connection.

Surprisingly, during the following stage of the ICC, after 5500 BCE, this extended network seems to have collapsed. This is highlighted for instance by the general disappearance of the obsidian trade throughout Provence and Languedoc (Binder et al., 2012), by the increasing of the polymorphism of pottery styles (Manen, 2002), and by the diversification of economic patterns giving a wider place to hunting activities (Binder, 1991b). This break could be the result of an increasing admixture between

Farmers and local Hunter-Gatherers or of an economic and social reorganization of communities to face new environments and a specific declension of the Neolithic Paradigm (Guilaine, 2018).

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Author contributions

M.G., F.C., C.V. and D.B. designed research. M.G., C.V. and D.B. wrote the paper with B.G., L.G. and C.M. M.G., F.C., C.V., B.G., S.J. and G.B. performed research and analyzed data. M.G. and G.D. made the iconographic apparatus with C.V. and F.C. All authors revising the work and approval the final version to be published.

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Figure captions

Figure 1

Map of north western Mediterranean studied area showing distribution of archaeological sites and geological formations of the Volcanic Provinces considered for this study.

Figure 2

Archaeological studied potteries from (A) Pont de Roque-Haute (drawn by J. Coularou in Manen and Guilaine 2007, fig. 49) and from (B-C) Le Secche (B macrophotography and C stereo-microphotography).

Figure 3

Microphotography comparison of pottery samples. Arrows point out the main mineral components of pottery pastes: rounded clinopyroxene (Cpx), K-feldspath (Kfs) and volcanic rock (VR). A, C, E and G from Le Secche (GLS); B, D, F, H from Pont de Roque-Haute (PRH). A-B stereomicroscopic observations; C-H thin section microscopic observations.

Figure 4

QUAD classification diagram of Wollastonite (Wo), Enstatite (En), Ferrosilite (Fs) for (A) clinopyroxenes from archaeological samples analysed by SEM-EDS and LA-ICP-MS and for (B-F) archaeological samples and selected Italian volcanic provinces (Armienti et al., 1983; Aulinas et al., 2008; Barton et al., 1982; Belkin et al., 2016; Bindi et al., 1999; Caggianelli et al., 1990; Civetta et al., 1997; Comodi et al., 2006; Conticelli et al., 2015, 2013, 1997; Del Bello et al., 2014; Dostal et al., 1982; Fedele et al., 2009; Feldstein et al., 1994; Ghiara et al., 1979; Guarino et al., 2011; Holm, 1982; Mollo et al., 2016; Palladino et al., 2014; Pappalardo et al., 2008; Perini et al., 2004; Perini and Conticelli, 2002). A to F diagrams corresponds to the enlarged part of the QUAD diagram (grey coloured area).

Figure 5

Primitive mantle normalised trace-element spider diagram for (A) bulk archaeological samples, PRH soil and Languedoc volcanic formations (Dautria et al., 2010) and for (B) bulk archaeological samples and selected Italian volcanic formations (Gasperini et al., 2002; Peccerillo, 2017). Normalisation values from McDonough and Sun (1995).

Figure 6

Primitive mantle normalised trace-element spider diagram for (A) clinopyroxenes from archaeological samples analysed by LA-ICP-MS and for (B) clinopyroxenes from archaeological samples and selected Italian volcanic provinces (Arienzo et al., 2009; Civetta et al., 1997; Comodi et al., 2006; Fedele et al., 2009; Gentili et al., 2014; Mollo et al., 2016; Pappalardo et al., 2008; Scarpelli et al., 2015). Normalisation values from McDonough and Sun (1995).

Figure 7

Binary diagram Y vs Ce where concentration in ppm are reported for (A) clinopyroxenes of archaeological samples and for (B) clinopyroxenes of archaeological samples and selected Italian volcanic provinces (Arienzo et al., 2009; Civetta et al., 1997; Comodi et al., 2006; Fedele et al., 2009; Gentili et al., 2014; Mollo et al., 2016; Pappalardo et al., 2008; Scarpelli et al., 2015).

Figure 8

Map of north western Mediterranean studied area showing the identified volcanic source area for pottery provenance and location of neolithic archaeological sites. Tyrrhenian geological obsidian outcrops are also reported.

Figure
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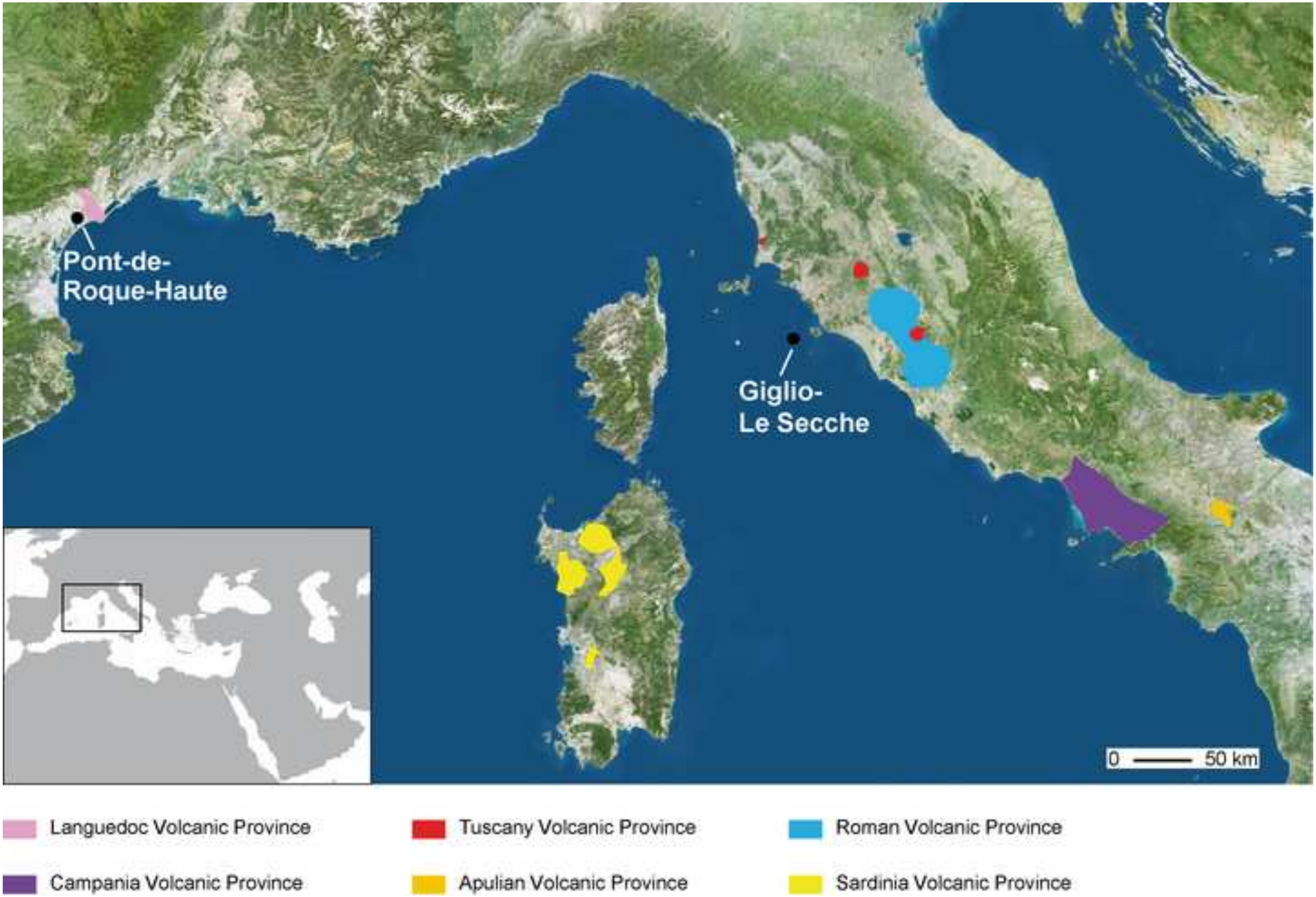

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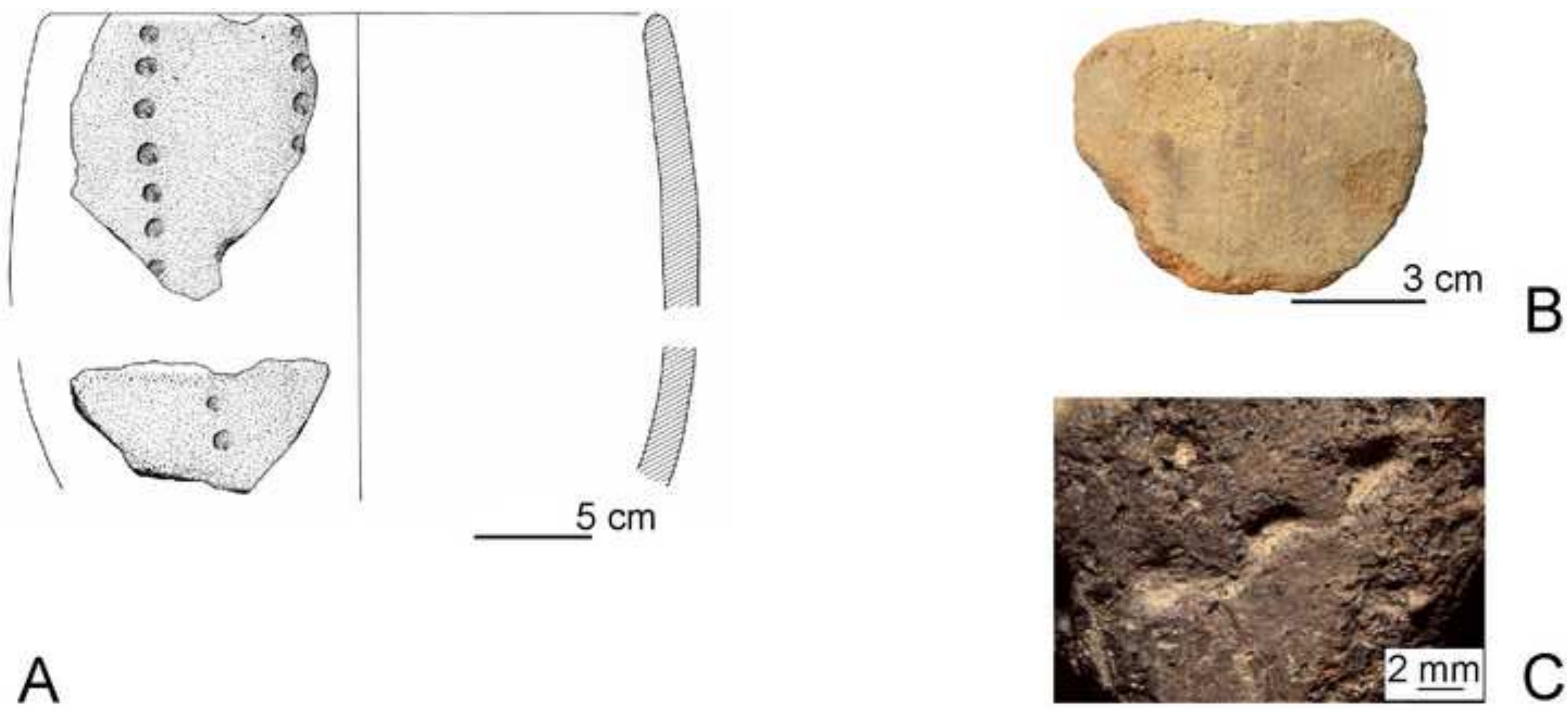

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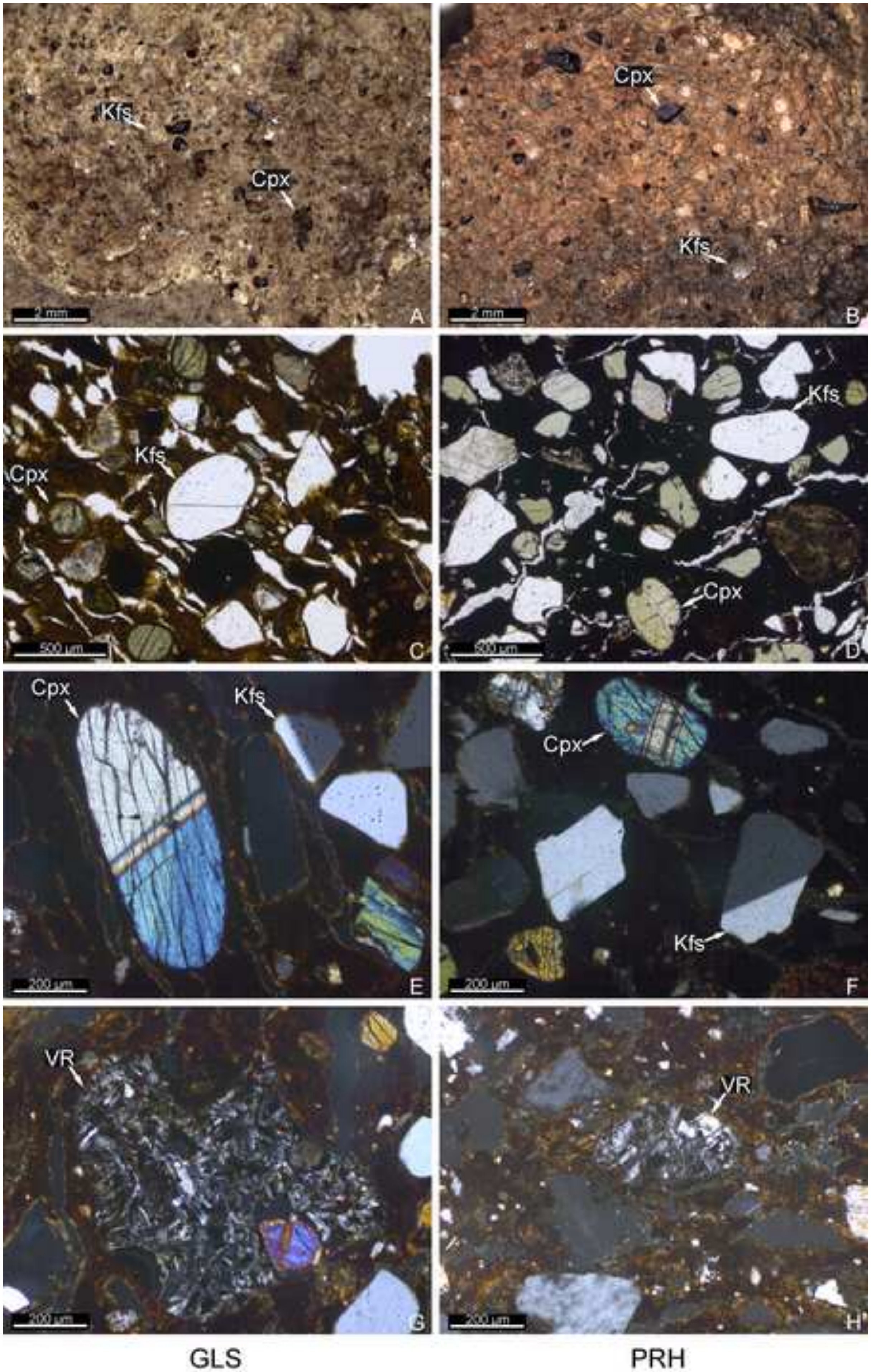
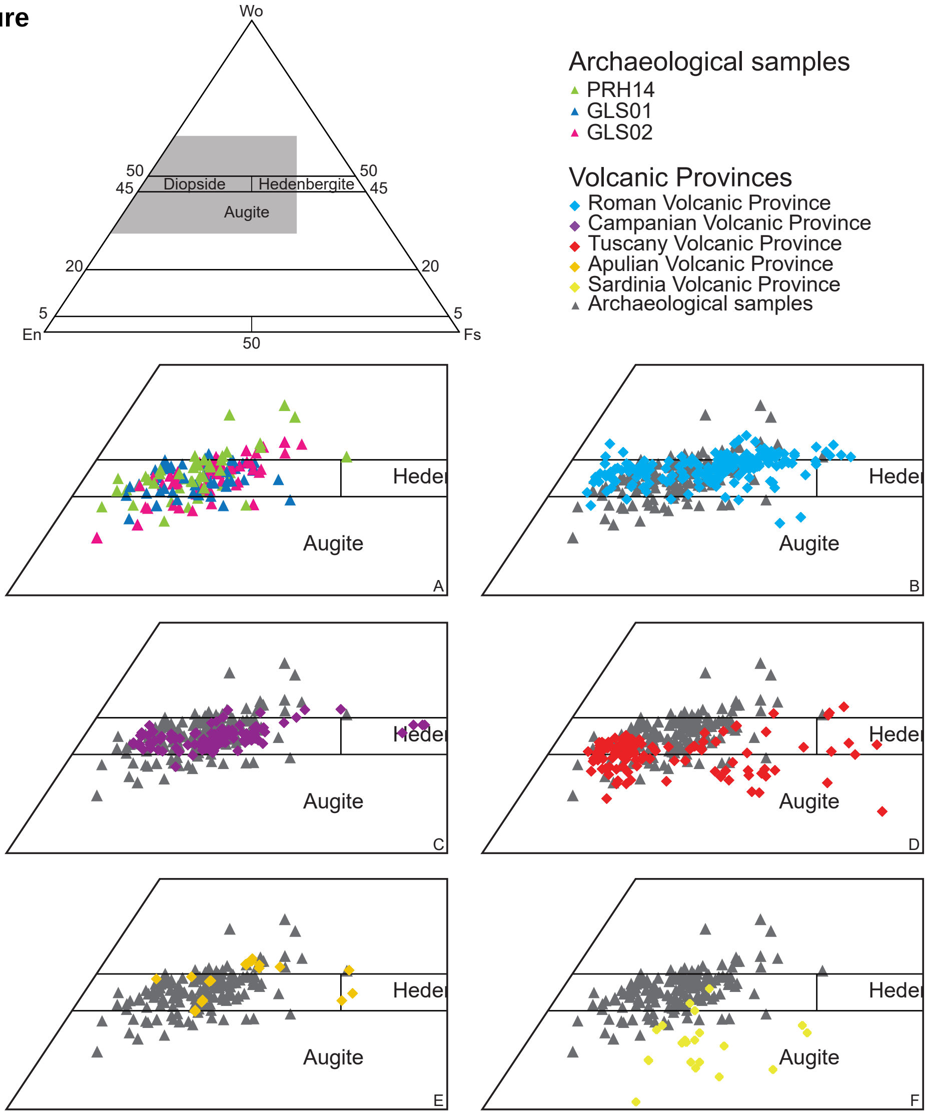


Figure



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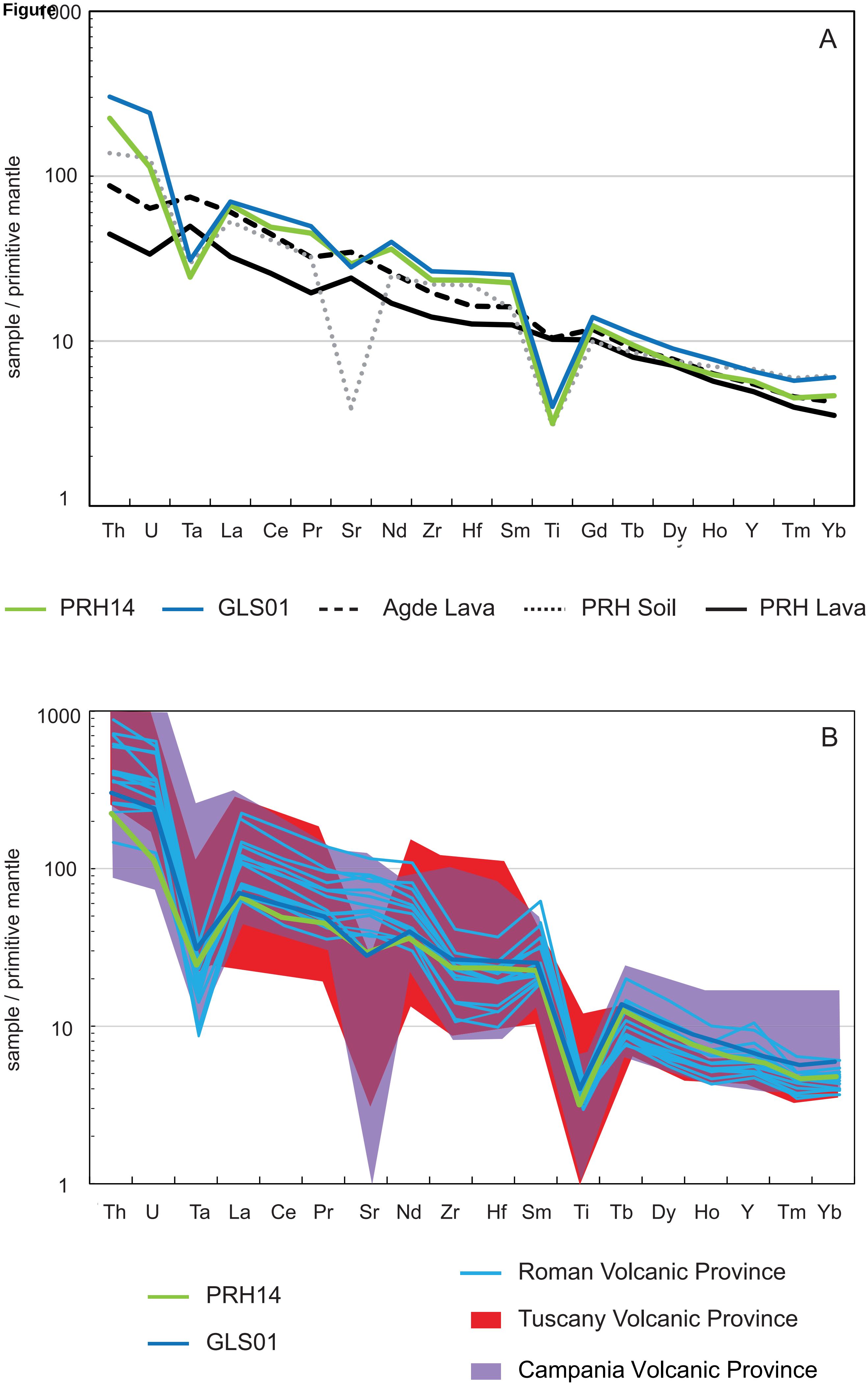
Archaeological samples
PRH14
GLS01
GLS02
Volcanic Provinces
Roman Volcanic Province
Campanian Volcanic Province
Tuscany Volcanic Province
Apulian Volcanic Province
Sardinia Volcanic Province
Archaeological samples
Heder
Augite
A
B
C
D
E
F

Figure



A
sample / primitive mantle
1000
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Th U Ta La Ce Pr Sr Nd Zr Hf Sm Ti Gd Tb Dy Ho Y Tm Yb
PRH14
GLS01
Agde Lava
PRH Soil
PRH Lava
B
sample / primitive mantle
1000
100
10
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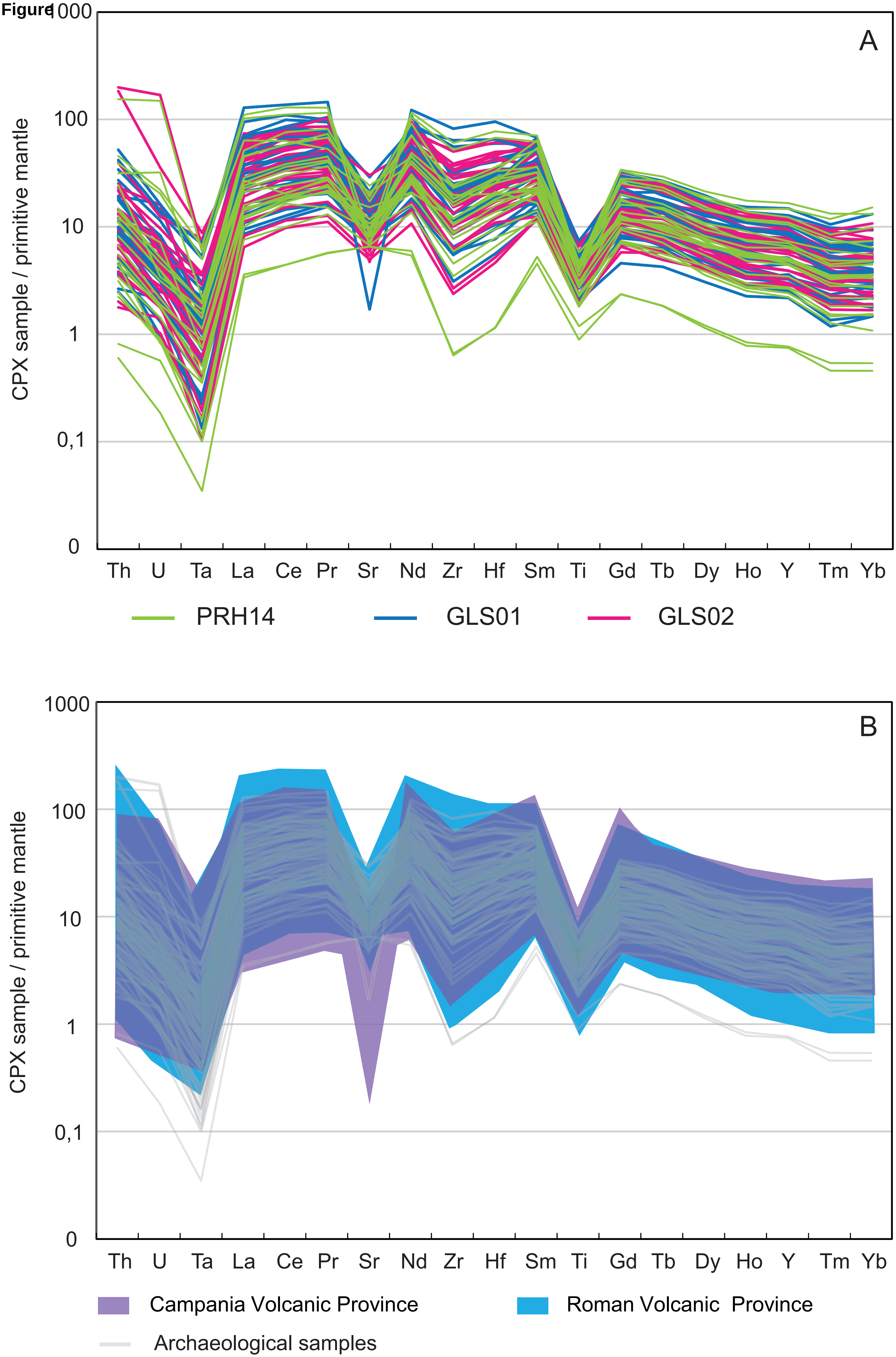
Th U Ta La Ce Pr Sr Nd Zr Hf Sm Ti Tb Dy Ho Y Tm Yb
PRH14
GLS01
Roman Volcanic Province
Tuscany Volcanic Province
Campania Volcanic Province

Figure



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CPX sample / primitive mantle
Th U Ta La Ce Pr Sr Nd Zr Hf Sm Ti Gd Tb Dy Ho Y Tm Yb
PRH14
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GLS02
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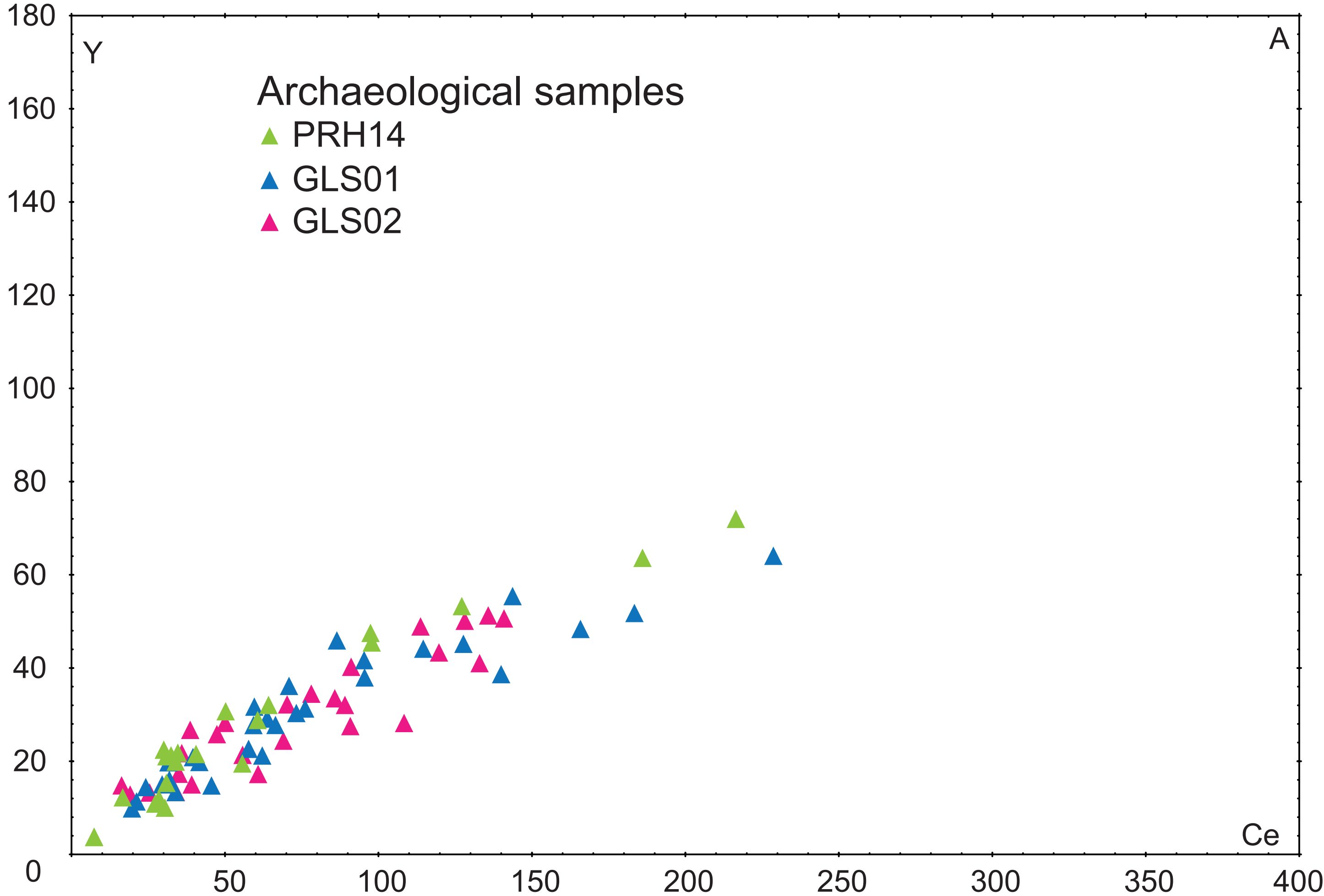
CPX sample / primitive mantle
Th U Ta La Ce Pr Sr Nd Zr Hf Sm Ti Gd Tb Dy Ho Y Tm Yb
Campania Volcanic Province
Roman Volcanic Province
Archaeological samples

Figure



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Archaeological samples
PRH14
GLS01
GLS02
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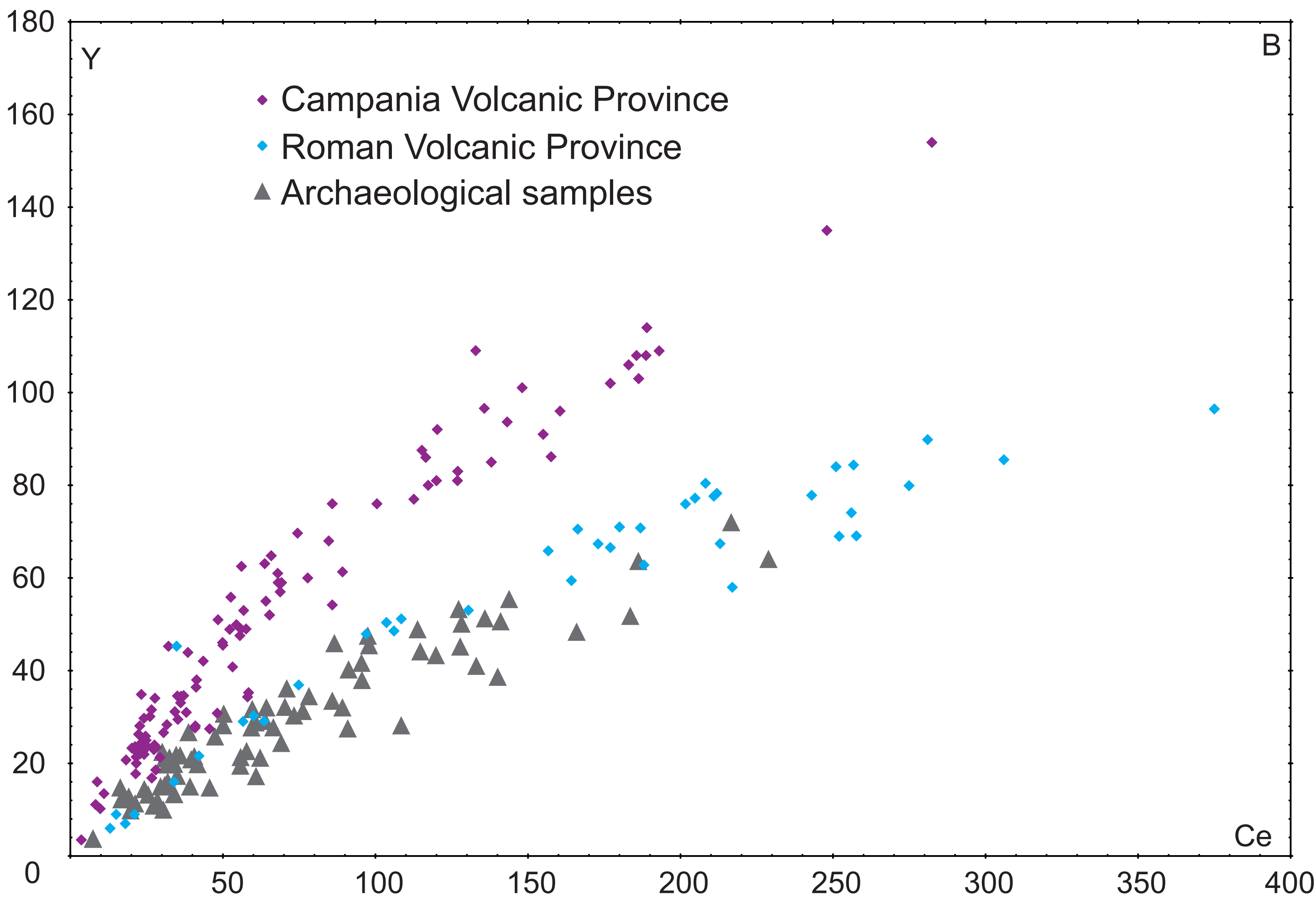

180
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Campania Volcanic Province
Roman Volcanic Province
Archaeological samples
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Figure

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Supplementary Material
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*Declaration of Interest Statement

Title

High resolution sourcing of pottery demonstrates long-distance mobility in the North Western Mediterranean during the Neolithic transition

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