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## Long-distance mobility in the North-Western Mediterranean during the Neolithic transition using high resolution pottery sourcing

Marzia Gabriele, Fabien Convertini, Chrystèle Vérati, Bernard Gratuze, Suzanne Jacomet, Giovanni Boschian, Gilles Durrenmath, Jean Guilaine, Jean-Marc Lardeaux, Louise Gomart, et al.

### ► To cite this version:

Marzia Gabriele, Fabien Convertini, Chrystèle Vérati, Bernard Gratuze, Suzanne Jacomet, et al.. Long-distance mobility in the North-Western Mediterranean during the Neolithic transition using high resolution pottery sourcing. *Journal of Archaeological Science: Reports*, 2019, 28, pp.102050. 10.1016/j.jasrep.2019.102050 . hal-02373914

**HAL Id: hal-02373914**

**<https://hal.science/hal-02373914>**

Submitted on 21 Dec 2021

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1 **Title**

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40

41 **Abstract**

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

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

49 Through a comparative and multi-analytical approach to pottery characterization of entire assemblages from two distant sites in  
50 the North-Western Mediterranean, each characterized by predominantly local pottery production, we demonstrate long-distance  
51 mobility of non-local pottery vessels from the Tyrrhenian regions towards the Languedoc (up to 1000 km) during the Neolithic  
52 transition. Our study allows us to highlight this unexpected milestone in the first Neolithic migration in the North-Western  
53 Mediterranean.

54

55 **Keywords**

56 Pottery analysis, ceramic petrography, geochemistry, provenance study, early Neolithic, Impressed Ware

57

## 58 **1 Introduction**

### 59 *1.1 Tracking farming pioneers in the North-Western Mediterranean*

60 The spread of farming and Neolithic lifeways from the Eastern Mediterranean and the Aegean towards Western Europe is known  
61 to have followed two main routes (Childe, 1925). The continental route, which was at the origin of the Linearbandkeramik  
62 Complex (LBK), can be traced through the Central Balkans and the Danube valley, and reached Northern France after 5350 BCE  
63 (Whittle, 2018). The Mediterranean route, linked to the Impressed Wares or Impresso-Cardial complex (ICC), reached Southern  
64 France at least five centuries earlier, c. 5850 BCE (Binder et al., 2017). Many issues are related to the social dynamics at the origin  
65 of the erratic dispersal of the very first ICC farming communities in the Western Mediterranean, which highly contrasts with the  
66 LBK pioneer front.

67 In both cases, the role played by migrants within these processes has been demonstrated by genomics (Mathieson et al., 2018).  
68 Concerning the ICC, the modelling of a large set of audited radiocarbon dates currently places its formative stage in Southern Italy  
69 and Dalmatia during the very beginning of the 6th millennium BCE, mostly after 5950 BCE (Binder et al., 2017; McClure et al.,  
70 2014). Few genomic data are currently available for the earliest ICC aspects, i.e. between 6000 and 5750 BCE from Zemunica  
71 cave and between 5670 and 5560 BCE from Kargadur, both in Croatia. These data strengthen the idea of a genetic connection with  
72 the Balkans, Aegean and Anatolia, regarding both maternal and paternal lineages (Mathieson et al., 2018). This evidence raises  
73 new issues on the possible roots of the ICC in the second half of the 7<sup>th</sup> millennium BCE, and specifically in the Southern Balkans  
74 and Aegean regions in the context of the Monochrome or Proto-Sesklo Pottery which punctually reached the Ionian Sea (Berger et  
75 al., 2014).

76 North and westwards, in Italy and France, analyses of Neolithic DNA are very rare and mostly concern later periods (Lacan et al.,  
77 2011; Rivollat et al., 2017). In this area, peopling dynamics are mainly demonstrated by transfer of material culture and domestic  
78 taxa. For instance, it is now well known that domestic animals and crops were exogenous (mainly sheep, goat, wheat and barley)  
79 and originated from Southwest Asia (Rowley-Conwy et al., 2013). This enables us to study the spread of animal breeding and  
80 agriculture and to observe their rhythms and pathways. In this framework, systemic studies of material culture also offer the  
81 possibility to track the trajectories of the first farmers (Bernabeu Auban et al., 2017; Ibáñez-Estévez et al., 2017).

82 Peopling dynamics from each region of the Italian Apennine chain seem to have been diverse regarding cultural connections as  
83 well as diffusion tempo. On the Adriatic side, ICC settlements remained highly concentrated in Apulia, Basilicate and East  
84 Calabria during c. two centuries, crossing over the Tavoliere towards Central Italy at a rather late period: c. 5750 BCE in Abruzzo  
85 and c. 5600 BCE in Marche. In contrast, on the Tyrrhenian side, the meshing of the earliest settlements appears very sparse while  
86 the speed of diffusion appears to be very fast. In fact, early farmers reached the far Ligurian and French coasts as early as c. 5850  
87 BCE (Binder et al., 2017). Similarly, the first data on pottery technology indicate that different communities of practice occurred  
88 in the Adriatic and Tyrrhenian sides of the Apennine range. In the Adriatic area, pottery forming methods using coils and long  
89 slabs were clearly related to the Balkan tradition, whereas a distinctive Spiralled patchwork technology (SPT) was in use in the  
90 Tyrrhenian. In the SPT ceramics were constructed by juxtaposing circular patches, each formed by spiral coil (Gomart et al.,  
91 2017).

92 Obsidian is well known for having played an important role during the earliest Eastern Mediterranean Neolithic (Dixon et al.,  
93 1968), and notably a symbolic one (Cauvin, 1998). Obsidian was totally ignored by the Late Hunter-Gatherers of the Western  
94 Mediterranean, and its use was transferred westward as part of a Neolithic package. Most of the attractive obsidian sources are  
95 located in the western Mediterranean islands (Pantelleria, Lipari, Palmarola and Sardinia) where this glass was exploited and  
96 distributed from the earliest stages of the ICC (Ammerman and Andrefsky, 1982; Muntoni, 2012; Tykot et al., 2013). Although  
97 there is currently no evidence of early ICC settlements located close to these obsidian sources, tools made of obsidian from  
98 Palmarola and Sardinia have been identified in the earliest North-Western Mediterranean ICC, especially at the sites of Arene-  
99 Candide in Liguria (Ammerman and Polglase, 1997), and Peiro Signado and Pont de Roque-Haute in the Mediterranean  
100 Languedoc (Briouis et al., 2009; Binder et al., 2012).

101 Until now obsidian geochemical analysis has been the most effective and precise method for linking distant sites or people, and  
102 has provided the main evidence for voyaging and circulation throughout the Western Mediterranean compared to the simple  
103 analogies suggested by pottery styles.  
104 These data shed light on a specific Tyrrhenian cultural landscape where the sea likely played a central role during part of the ICC.  
105 The range and the regime of maritime mobility is one of the key issues in this context. Research carried out in the last couple of  
106 decades has shown that the Mediterranean is a hot spot of cultural diversity (Rigaud et al., 2018). Furthermore, this maritime zone  
107 offered the possibility of multidirectional movements, but also different forms of mobility (pioneering, travelling, interaction and  
108 exchange) (Manen et al., 2018). Consequently it is still difficult to identify precise circulation routes, cultural filiations and origins  
109 of the incoming farmers. In this study we implement a multi analytical approach to pottery provenance through petrographic and  
110 geochemical analyses to provide additional insight into the crucial question of Neolithic dispersal routes. We demonstrate that  
111 pottery sourcing analysis is both complementary, and can be as equally precise and pertinent as obsidian sourcing, for tracking  
112 ICC networking dynamics in the Western Mediterranean.

### 113 114 *1.2 Pottery pastes as a proxy for human trajectories*

115 Pottery studies have been developed for and applied to Mediterranean Neolithic contexts since the early 1990s (Capelli et al.,  
116 2017, 2008; Convertini, 2010, 2007; Echallier, 1991; Ferraris and Ottomano, 1997; Gabriele, 2014, 2015; Gabriele and Boschian,  
117 2009; Manen et al., 2010; Martini et al., 1996; Muntoni, 2003; Paolini-Saez, 2010; Spataro, 2002; Ucelli Gnesutta and Bertagnini,  
118 1993). In most of these studies, pottery analyses have indicated local production of ICC pottery, while non-local pottery  
119 production remains exceptional within assemblages. The limited range of non-local ceramic circulation (10 to 100 km) generally  
120 suggests that the pottery trade was embedded in functional networks, illustrating the logistical mobility of the first ICC farmers  
121 (Binder, 1991a; Capelli et al., 2017; Manen and Convertini, 2012).

122 However, previous studies have already highlighted the possibility of long distance pottery circulation in the Mediterranean  
123 Languedoc (Convertini, 2010, 2007), the Liguro-Provençal arch and Tuscany (Capelli et al., 2017, 2008; Gabriele, 2014, 2015),  
124 especially with regard to the presence of volcanic components in certain pottery pastes (hereinafter referred to as volcanic pottery  
125 and paste). The latter offer specific petrographic markers and geochemical features of a very high resolution, as demonstrated by a  
126 large set of pottery studies from distinct regions (Comodi et al., 2006; Barone et al., 2010; Brunelli et al., 2013; Palumbi et al.,  
127 2014; Belfiore et al., 2014; Scarpelli et al., 2015; La Marca et al., 2017). Furthermore, recent applications of in situ geochemical  
128 methods on non-volcanic mineral inclusions allow us to enhance the accuracy and reliability of provenance analyses (Gehres and  
129 Querré, 2018).

130 Here, we provide a multi analytical comparative approach to pottery characterization and provenance through petrographic and  
131 geochemical analyses. Through high-resolution study focused on non-local volcanic pottery with similar petrographic composition  
132 from the two ICC sites of Portiragnes - Pont de Roque-Haute (Languedoc, France) (hereinafter referred to as PRH) (Guilaine et  
133 al., 2007) and Giglio - Le Secche (Tuscany, Tuscan Archipelago, Italy) (hereinafter referred to as GLS) (Brandaglia, 2002) (Fig.  
134 1), we demonstrate that although most vessels were made locally, a few unique pottery vessels circulated at long distance between  
135 Central Italy and Languedoc, attaining distances exceeding 1000 km following the coast, or 600 km as the bird flies.

## 136 137 **2 Materials and Methods**

### 138 *2.1 Sites and samples*

139 The open-air site of PRH, which offered a set of pits dug into a fluvial terrace, was interpreted as a short duration settlement in a  
140 ria (Guilaine et al., 2007). The modelled age of this occupation is estimated between 5860-5710 and 5800-5680 BCE, i.e. one of  
141 the earliest ICC settlements currently known in the Western Mediterranean (Binder et al., 2017). Together with domestic remains  
142 (mammals, seashells and tools including obsidian from Palmarola) a series of ca. 603 sherds (pertaining to at least 55 individuals)  
143 (Manen and Guilaine, 2007), of which the totality was analyzed petrographically by the authors, indicates a dominant local pottery

144 production exploiting reworked alluvial Pliocene deposits (Convertini, 2010, 2007). Few individual pots (ca. three at least) are  
145 characterized by volcanic aplastic components and among them one pot (ca. two sherds) is made of non-local raw materials and  
146 impressed with the umbo of a *Cardidae* shell (Fig. 2A).

147 The site of GLS is a shelter close to a north-western beach of Giglio Island. Rich deposits of well preserved pottery (Brandaglia,  
148 1991) associated with a large set of Palmarola obsidian tools (Barone et al., 1996; Brandaglia, 1987) demonstrated its long  
149 occupation, starting during the earliest stage of ICC (5840-5540 BCE) and lasting at least until the second half of the 6<sup>th</sup>  
150 millennium BCE (Binder et al., 2017). Most of the earliest pottery (ca. 1091 sherds, pertaining to at least 60 individuals), of which  
151 the totality was analyzed petrographically by the authors, was made using local residual deposits of granite formation, with the  
152 exception of notable few vessels (ca. three sherds) shaped with a non-local volcanic paste and decorated with the ventral margin of  
153 a *Cardidae* shell (Fig. 2B, C) (Gabriele, 2014).

154 The remarkable presence of isolated non-local volcanic pottery within a homogeneous local production (ca. 99% of total sherds  
155 from both sites), has been demonstrated thanks to a systematic petrographic study of the entire pottery assemblage of both sites  
156 (Convertini, 2010, 2007; Gabriele, 2014). For the non-local pottery from both sites, the most likely sources of raw material are the  
157 volcanic provinces of central Italy. In order to verify this hypothesis and shed light on the earlier Mediterranean Neolithic  
158 scenario, three volcanic ceramic samples with similar petrographic composition from both sites (one from PRH and two from  
159 GLS), were further investigated using high performance geochemical methods in an integrated and comparative study perspective.  
160

## 161 2.2 Analytical methods

162 Petrographic and chemical methods were performed on each specimen in its various prepared forms (prepared as powder, thin-  
163 section, etc.) using different scales of observations in order to have comparable and complementary data. As ceramic pastes are  
164 heterogeneous by definition, painstaking care is taken in microscopically discriminating those minerals which are most sourceable  
165 and mapping them using imagery to prepare the samples for successive analysis of multiple mineral grains from each sample.  
166 While the number of sherds, representing at least two pots from two sites, is small, the number of in-situ analyses conducted on  
167 each sherd was gargantuan.

168 First, petrographic analysis was obtained by stereomicroscopy directly on the three pottery fragments, and by standard optical  
169 microscopy on six thin sections (two for each sample) with the support of scanner images to characterize a-plastic inclusions and  
170 fabric textural features. Description of textures of inclusions, pores and matrix were performed following the guidelines of soil  
171 micromorphology (Stoops, 2003) and ceramic description (Quinn, 2013; Whitbread, 1989). Analyses were carried out at the  
172 CEPAM laboratory (CNRS, Université Cote d'Azur) in Nice, France.

173 Subsequently, to be able to verify the real compositional correspondence between ceramics from both sites, chemical analysis of  
174 major and trace elements were carried out to determine composition of the bulk pottery and of single mineral grains, such as  
175 clinopyroxene. Microchemical in-situ analysis on clinopyroxene is based on the assumption that its chemical composition  
176 represents the chemical composition of parental magma (Barone et al., 2010; Leterrier et al., 1982). Indeed, crystal-chemistry of  
177 clinopyroxene is related to different geochemical and petrological magma affinities (Cellai et al., 1994; Cundari and Salviulo,  
178 1987; Gentili et al., 2014). Finally, to discern possible petrographic and geochemical sources, data available in scientific literature  
179 were used.

180 The bulk pottery compositions were obtained by Inductively Plasma Atomic Emission Spectrometry (ICP-AES) and Inductively  
181 Coupled Mass Spectrometry (ICP-MS) for major and trace elements respectively at the Geochemical and Petrographical Research  
182 Center in Nancy (SARM laboratory, CNRS-CRPG; Supplementary dataset) following the procedure described in Carignan et al.  
183 (2001).

184 Chemical analysis by environmental scanning electron microscope (FEI PHILIPS XL30 ESEM) equipped with an Energy  
185 Dispersive Spectroscopy (EDS) system for X-ray microanalysis (Quantax XFLASH6/30 silicon drift 10mm<sup>2</sup>) was applied on 74  
186 clinopyroxene and 53 K-feldspar (sanidine) selected single crystal grain minerals found as inclusions in polished thin sections and

187 using mapping of scanner images. The analyses were carried out at the laboratory of the Centre for Material Forming (CEMEF,  
188 Ecoles des Mines de Paris, CNRS, Sophia Antipolis, France).

189 Laser Ablation Inductively Coupled Plasma Mass Spectrometry (LA-ICP-MS) examinations for estimating major and trace  
190 elements were applied on clinopyroxene single crystals found as inclusions in the epoxy impregnated ceramic samples left from  
191 the processing of thin sections, by stereomicroscopy observations and using mapping of scanner images. LA-ICP-MS analysis  
192 were undertaken on 78 selected clinopyroxene grain minerals, other than those of the SEM-EDS investigations (Supplementary  
193 dataset). The largest clinopyroxenes were selected in order to avoid possible contamination by other mineral species or clay paste  
194 from the ceramic during the ablation process. LA-ICP-MS analysis was conducted at the IRAMAT laboratory, Centre Ernest-  
195 Babelon (Université d'Orléans, CNRS, France).

196 The analytical protocol developed for obsidian inclusion analysis (Palumbi et al., 2014) was adapted to the analysis of  
197 clinopyroxenes improving detection limits of elements such as rare earths. As encountered with the analysis of obsidian  
198 inclusions, one of the critical parameters of this type of analysis is the thickness of the analyzed clinopyroxene grains, specifically  
199 because they were inserted in a ceramic paste and thus may contain other mineral species in their structure.

200 Consequently, in order to avoid overshooting the inclusions and to maintain a high signal level, a 10 Hz laser pulse frequency was  
201 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  
202 mass scans from lithium to uranium.

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

207 However, the contribution or the modification brought by another mineral species to the signal measured for a perturbing  
208 mineral species in the whole signal is weak. It is thus only when the chemical contrast between both species is significant that the  
209 correction of the signal is possible, as illustrated by the presence of a zircon grain in one of the recorded spectra or by a transition  
210 between a clinopyroxene and a feldspar. For most of the other cases the presence of another mineral species may not be detected  
211 and will just increase the dispersion or the variability of the calculated compositions. To avoid clay contaminations, the analyses  
212 were carried out in the middle of the clinopyroxene grains. When possible the largest grains were selected for the analysis,  
213 however, analyses of very small grains were also carried out by adapting the laser beam diameter.

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

219

### 220 **3 Results and discussion of the comparative study of the non-local volcanic pottery**

#### 221 *3.1 Petrographic analysis of pottery pastes*

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

225 At thin section optical-microscopic scale, porosity is characterized by meso- and macro planes and few macro vughs. The porosity  
226 distribution and orientation is well expressed in the GLS samples, where concentric features and parallel, inclined and bow-like  
227 bands of oriented planes were recognized. A-plastic inclusions are common, mostly sub-rounded and rounded, moderately sorted,  
228 with fine-medium sand size. Grain distribution and orientations are weakly-moderately expressed, up to single- and double-spaced  
229 relative distance. Clinopyroxene and K-feldspar (sanidine) are the most common minerals (Fig. 3C, D). Clinopyroxene is

230 frequently rounded, coloured with green pleochroism and twinned (Fig. 3C-F). Sanidine is generally less rounded and larger (up to  
231 very coarse sand size) than clinopyroxene, fresh and Carlsbad twinned (Fig. 3C-F). Other mineral grains are identified in different  
232 proportions and size, within which there are oxides, quartz, plagioclase, and black and white micas. Lithic inclusions are generally  
233 rounded, heterometric, up to very coarse sand and very fine gravel size. Lithoclasts are identified as alkaline volcanic rocks (Fig.  
234 3G, H), sandstone, siliceous sedimentary rocks, and quartz-metamorphic rocks. The matrix is optical active in GLS samples with  
235 stippled-speckled b-fabric and striated b-fabric. The colour is heterogeneous, linked to Fe reduction on the margins and Fe  
236 oxidation on the core of the fragments.

237

### 238 *3.1.1 The importance of petrography for identifying potential volcanic source areas*

239 On the basis of petrographic and archaeological considerations, the most likely source of raw materials are the Italian Miocene-  
240 Quaternary potassic and ultrapotassic volcanic rocks from the so-called Volcanic Provinces (hereinafter referred to as VP) part of  
241 the Magmatic Provinces of the Tyrrhenian region (Fig. 1) (Conticelli et al., 2004; Peccerillo, 2017). Furthermore, petrographic  
242 pottery data, namely characterized by the association of sanidine and green clinopyroxene minerals with minor amounts of  
243 volcanic, sedimentary and metamorphic lithoclasts, suggest considering volcanic formations that are hydrographically or  
244 geomorphologically linked with formations of different geological origins. In this perspective, more suitable volcanic centres are  
245 the Monte Amiata in the Tuscany VP (hereinafter referred to as TVP) (Conticelli et al., 2015; Cristiani and Mazzuoli, 2003), and  
246 Vulcini (Barton et al., 1982; Holm, 1982; Palladino et al., 2014), Vico (Barbieri et al., 1988; Palladino et al., 2014; Perini et al.,  
247 2004; Perini and Conticelli, 2002) and Sabatini (Conticelli et al., 1997; Del Bello et al., 2014; Palladino et al., 2014) districts in the  
248 Roman VP (hereinafter referred to as RVP). We cannot a priori exclude Roccamonfina (Ghiara et al., 1979), Phlegrean Fields  
249 (Armienti et al., 1983; Belkin et al., 2016; Civetta et al., 1997; Fedele et al., 2009; Mollo et al., 2016) and Somma-Vesuvius  
250 (Bertagnini et al., 1998) districts in the Campania VP (hereinafter referred to as CVP) (in this paper Roccamonfina volcanic  
251 district is considered part of the CVP, Fig. 1; moreover for the CVP we haven't considered data on eruptions younger than 8 ka  
252 BP).

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

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

267

### 268 *3.2 Major elements analysis of single mineral inclusions in pottery pastes*

269 Data from the SEM-EDS analysis shows that alkali-feldspar minerals are compositionally homogenous with Or<sub>67</sub> to Or<sub>85</sub>, and in  
270 only one case (GLS02 sample) with Or<sub>50</sub>. The alkali-feldspar classification is represented in a ternary diagram in supplementary  
271 Figure 1.



272 Clinopyroxenes are predominantly composed of diopside and Fe-rich diopside; augite to Mg-rich augite and CaFe-rich  
273 clinopyroxene are also present (Supplementary dataset). Also in the case of LA-ICP-MS analysis, clinopyroxenes are  
274 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  
275 ( $Wo_{44}En_{33}Fs_{23}$ ) and CaFe-rich diopside ( $Wo_{51}En_{38}Fs_{11}$  to  $Wo_{52}En_{28}Fs_{20}$ ) are also present (Supplementary dataset). The  
276 clinopyroxene classification is represented in the QUAD diagram referring to Morimoto (1988) in supplementary Figure 2. The  
277 major-element chemical composition of clinopyroxene available in the scientific literature allows us to differentiate amongst  
278 previously indicated possible petrographic sources in the TVP (Aulinas et al., 2011; Conticelli et al., 2015, 2013; Feldstein et al.,  
279 1994), RVP (Barton et al., 1982; Comodi et al., 2006; Conticelli et al., 1997; Cundari, 1975; Dal Negro et al., 1985; Del Bello et  
280 al., 2014; Gentili et al., 2014; Holm, 1982; Kamenetsky et al., 1995; Palladino et al., 2014; Perini, 2000; Perini et al., 2004; Perini  
281 and Conticelli, 2002), CVP (Armienti et al., 1983; Aulinas et al., 2008; Belkin et al., 2016; Civetta et al., 1997; Fedele et al., 2009;  
282 Ghiara et al., 1979; Mollo et al., 2016; Pappalardo et al., 2008), AVP (Bindi et al., 1999; Caggianelli et al., 1990), and SVP  
283 (Dostal et al., 1982; Guarino et al., 2011). The Quad diagrams show substantial correspondence between Mg-rich augite and  
284 diopside composition of clinopyroxenes in pottery (Fig. 4A) and volcanic rocks of RVP, CVP and AVP (Fig. 4B, C, E). Instead,  
285 there is partial correspondence with rocks of TVP and SVP, especially due to the lack of clinopyroxene with augite composition in  
286 pottery pastes (Fig. 4D, F). Moreover, pottery clinopyroxenes are characterized by limited compositional variations in major  
287 elements, considered as cationic values. In  $Ti_{tot}$  vs  $Al_{tot}$  binary diagrams (Supplementary Figure 3), the cluster of pottery  
288 clinopyroxene composition fits in the field of clinopyroxenes of the RVP, TVP and CVP (Supplementary Figure 3B, C, D), but  
289 only partially fits in the clinopyroxene compositional fields of the AVP and SVP (Supplementary Figure 3E, F).

290

### 291 *3.3 Trace element analysis of bulk pottery pastes*

292 We conducted ICP-MS trace element analysis on two bulk ceramic samples from the two archaeological sites. A soil sample from  
293 the PRH site (Sedimentary Pliocene deposits) was also analyzed. Results are reported in the Supplementary Dataset. In the spider  
294 diagram (Fig. 5A) PRH and GLS pottery are geochemically indistinguishable. Their spectra display the same Large Ion Lithophile  
295 Elements (LILE) enrichment, the same high negative Ta and Ti anomalies, and the same slight Sr anomaly. Furthermore, trace  
296 element contents of rocks from the Languedoc Volcanic Province (Agde volcano and lava at the PRH site) do not display Ta, Sr  
297 and Ti anomalies (Fig. 5A), suggesting that volcanic minerals of these ceramics are not derived from southern France. Indeed,  
298 Languedoc Volcanic Province corresponds to homogeneous alkali basaltic geochemistry (Dautria et al., 2010), which is different  
299 from typical calc-alkaline geochemistry of the subduction zones (Italian Volcanic Provinces) (Peccerillo, 2017; Gasperini et al.,  
300 2002).

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

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

312

### 313 *3.4 Trace element analysis of clinopyroxene inclusions in pottery pastes*

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

333

### 334 *3.5 A unique source area for long distance exogenous pottery*

335 Our study shows a clear correspondence between the three archaeological pottery samples at every level of each method of  
336 analysis. This petrographic and chemical evenness suggests the exact same provenance for the volcanic pottery from both PRH  
337 and GLS sites, confirming the non-local origin of the vessels. The basaltic volcanic formations near the site of PRH can be  
338 excluded, both through petrographic and geochemical analyses. Moreover, Giglio Island is not a suitable source due to the  
339 exclusive presence of granitic and metamorphic formations and the absence of volcanic formations (Capponi et al., 1997;  
340 Westerman et al., 2003). Petrographic investigations allow us to highlight the correspondence of mineralogical and roundness  
341 textural features of the a-plastic inclusions of pottery pastes of both sites. The diversity visible in the other textural features of the  
342 fabric elements (i.e. granulometry) may depend on the internal variability of the deposits used as raw material. The roundness of  
343 inclusions shows, indeed, that secondary sedimentary deposits were used for pottery production (Capelli et al., 2008; Convertini,  
344 2007; Gabriele, 2014).

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

353 These results effectively demonstrate the efficiency and reliability of our methodology which is based on a chain of successive and  
354 complementary stages of analysis. We have demonstrated that petrography (both macro and micro observations) is the first and  
355 essential step, and must be confirmed and detailed with subsequent chemical analysis, in order to circumscribe real source areas of  
356 raw materials.

#### 358 **4 Unravelling early farming dynamics in the Western Mediterranean**

359 This comparative and multi-analytical pottery sourcing study provides the first evidence for interregional relationships at distances  
360 of more than 1000 km in the Western Mediterranean early Neolithic, through the circulation of pottery. We were moreover able to  
361 precisely circumscribe the source area for this pottery production, between the Fiora and the Tiber river basins in Southern  
362 Tuscany and Northern Latium, Italy.

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

367 This model is suggested to be at the origin of the settlement of small Neolithic seafaring groups far from their place of origin.  
368 Through this process, Neolithic practices and know-how were progressively transferred to an extended region. The technical  
369 traditions newly implemented in the North-Western Mediterranean are therefore expected to be very similar to those of the origin  
370 area, a topic which remains controversial. However, PRH potters clearly belong to the community of practices developed west of  
371 the Apennine range in Italy and significantly differ from the Adriatic and Balkan tradition (Gomart et al., 2017). Similar  
372 connections between the North-Western and the Tyrrhenian sites are observed regarding cropping practices based on hulled  
373 wheats and barley, and animal husbandry, since PRH ewes exhibit the same morphology as most Tyrrhenian ewes (Guilaine et al.,  
374 2007).

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

378 Paradoxically, volcanic pastes and obsidian sources exploited during the earliest Impressa stages are situated in areas of Central  
379 Italy where dwelling sites are poorly identified; the closest and earliest sites are Settecannelle cave site in the Fiora Valley (Ucelli  
380 Gnesutta, 2002), and La Marmotta site on the banks of the Bracciano Lake (Fugazzola Delpino, 2002) in Latium region, and  
381 Panicarola site located on the banks of the Trasimene Lake in the Umbria region (De Angelis, 2003) (Fig. 8). A similar situation  
382 can be observed regarding Sardinian obsidian exploitation: despite the trade of Monte Arci glass towards Liguria (Arene Candide;  
383 Ammerman and Polglase, 1997; Maggi, 1997) and Languedoc (Pont-de-Roque-Haute and Peiro Signado; Briois et al., 2009; De  
384 Francesco and Crisci, 2007), from 5850-5750 BCE, only one early dwelling place has been identified on this island and is  
385 hesitantly suspected to belong to this first phase (Su Coloru; Lugliè, 2018; Sarti et al., 2012). Similarly, evidence for the earliest  
386 impressed wares is very rare in Corsica (Campu Stefanu, Cesari et al., 2014; Albertini rock-shelter, Binder and Nonza-Micaelli in  
387 press).

388 Considering this scarcity, one could suspect that the area where raw materials were collected was in some way *terra incognita* for  
389 Neolithic pioneer groups. But the same lack of data could also indicate that the territorial meshing of early farmers is severely  
390 underestimated today, due to various hazards such as littoral submersion, sedimentary filling, site destruction or research lacunae.  
391 Nonetheless, these pottery analyses reveal invisible parts of the original meshing of groups and territory, just as pollen has  
392 revealed very early cropping within areas where Neolithic sites are currently unknown (Branch et al., 2014; Guillon et al., 2010).  
393 This observation suggests a peopling discontinuity between Southern Italy and the Franco-Ligurian region and strongly  
394 encourages the reassessment of the leapfrog dispersal model (Zilhão, 2014).

395 These results bring up new issues, of which one of the more pressing concerns the nature and temporality of the processes for  
396 acquiring various raw materials and for transferring pots or other goods at long-distance. This questions both the mobility regimes  
397 and the social interactions at the beginning of the Neolithic transition in the Western Mediterranean.

398 The hypothesis of short-term voyaging episodes, connecting the Northern Latium, the Tuscan Archipelago and the Mediterranean  
399 Languedoc, is strongly supported by our data. Indeed, the chronological resolution of radiocarbon dating, as well as the vagueness

400 of stylistic comparisons, cannot allow us to link those three regions solely to pioneer events. In fact, recent literature evokes a long  
401 duration of the production and trade of pottery from RVP, for instance in northern Latium (Settecannelle) (Ucelli Gnesutta and  
402 Bertagnini, 1993), Tuscan archipelago (Cala Giovanna Piano, Pianosa island) (Gabriele and Boschian, 2009), and Liguria (Pian  
403 del Ciliégio) (Capelli et al., 2017, 2008). At the same time, throughout the 6<sup>th</sup> millennium BCE, in the whole Tyrrhenian area and  
404 Liguria, several networks were developed at a smaller range as highlighted for example by movements of wares with low pressure  
405 ophiolitic components (Capelli et al., 2017; Gabriele and Boschian, 2009; Martini et al., 1996). In the Provençal area, the site of  
406 Nice - Caucade is a good example of regional multidirectional exploitation (early Impresa stage) (Convertini, 2010; Manen et al.,  
407 2006).

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

412 Surprisingly, during the following stage of the ICC, after 5500 BCE, this extended network seems to have collapsed. This is  
413 highlighted for instance by the general disappearance of the obsidian trade throughout Provence and Languedoc (Binder et al.,  
414 2012), by the increasing polymorphism of pottery styles (Manen, 2002), and by the diversification of economic patterns leaving a  
415 wider place to hunting activities (Binder, 1991b). This break could be the result of an increasing admixture between farmers and  
416 local Hunter-Gatherers or of an economic and social reorganization of communities facing new environments and a specific  
417 declension of the Neolithic Paradigm (Guilaine, 2018). The results provided here demonstrate that the development and  
418 comparison of both petrographic and mineral chemistry pottery sourcing methods are an indispensable contribution to  
419 understanding long-distance human mobility through time and space, as they complement and confirm obsidian sourcing data,  
420 while at the same time rendering visible unknown interactions and/or subtleties within known interactions. Furthering comparative  
421 approaches in ceramic sourcing are needed as they have great potential to expand our understanding of the complex dynamics of  
422 the neolithisation process, both in the Mediterranean region and elsewhere.

423  
424

#### 425 **Acknowledgments**

426 The authors wish to thank Mario Brandaglia, Paola Rendini and the *Soprintendenza per i Beni Archeologici della Toscana* for  
427 allowing us to study the ceramic materials from Giglio Island-Le Secche. The authors are also very grateful to the reviewers for  
428 their helpful comments, to Lamya Khalidi for her suggestions and for the English revision of the manuscript, and to Michel Dubar  
429 for his advice.

430 This work was supported by a grant from the *Agence Nationale de la Recherche* for the CIMO research project ‘Western  
431 Mediterranean Impressed Wares. An interdisciplinary research on Early Neolithic pottery (6th millennium cal BCE): sourcing,  
432 production, uses and transfers’ (ANR-14-CE31-009, D. Binder dir.).

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772  
773  
774 **Figure captions**

775 Figure 1

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

778  
779 Figure 2

780 Studied archaeological pottery from (A) Pont de Roque-Haute (drawn by J. Coularou in Manen and Guilaine 2007, fig. 49) and  
781 from (B-C) Le Secche (B macrophotography and C stereo-microphotography).

782  
783 Figure 3

784 Microphotography comparison of pottery samples. Arrows point out the main mineral components of pottery pastes: rounded  
785 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  
786 Roque-Haute (PRH). A-B stereomicroscopic observations; C-H thin section microscopic observations.

787  
788 Figure 4

789 QUAD classification diagram of Wollastonite (Wo), Enstatite (En), Ferrosilite (Fs) for (A) clinopyroxenes from archaeological  
790 samples analysed by SEM-EDS and LA-ICP-MS and for (B-F) archaeological samples and selected Italian volcanic provinces  
791 (Armienti et al., 1983; Aulinas et al., 2008; Barton et al., 1982; Belkin et al., 2016; Bindi et al., 1999; Caggianelli et al., 1990;  
792 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  
793 al., 2009; Feldstein et al., 1994; Ghiara et al., 1979; Guarino et al., 2011; Holm, 1982; Mollo et al., 2016; Palladino et al., 2014;  
794 Pappalardo et al., 2008; Perini et al., 2004; Perini and Conticelli, 2002). A to F diagrams correspond to the enlarged part of the  
795 QUAD diagram (grey coloured area).

796  
797 Figure 5

798 Primitive mantle normalised trace-element spider diagram for (A) bulk archaeological samples, PRH soil and Languedoc volcanic  
799 formations (Dautria et al., 2010) and for (B) bulk archaeological samples and selected Italian volcanic formations (Gasparini et al.,  
800 2002; Peccerillo, 2017). Normalisation values from McDonough and Sun (1995).

801  
802 Figure 6

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

807  
808 Figure 7

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

812  
813 Figure 8

- 814 Map of north western Mediterranean study area showing the identified volcanic source area for pottery provenance and location of  
815 Neolithic archaeological sites. Tyrrhenian geological obsidian outcrops are also reported.



Pont-de-Roque-Haute

Giglio-Le Secche

0 50 km

Languedoc Volcanic Province

Tuscany Volcanic Province

Roman Volcanic Province

Campania Volcanic Province

Apulian Volcanic Province

Sardinia Volcanic Province



A

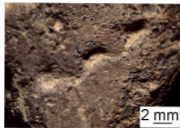


5 cm



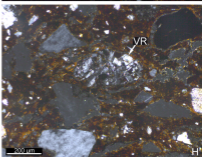
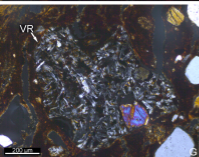
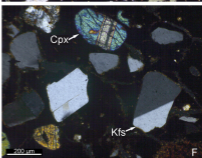
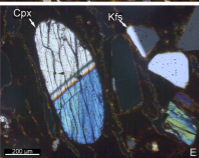
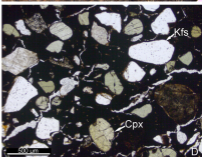
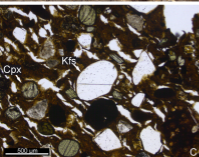
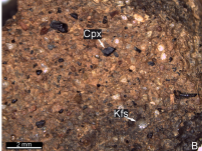
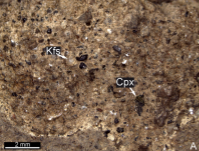
3 cm

B



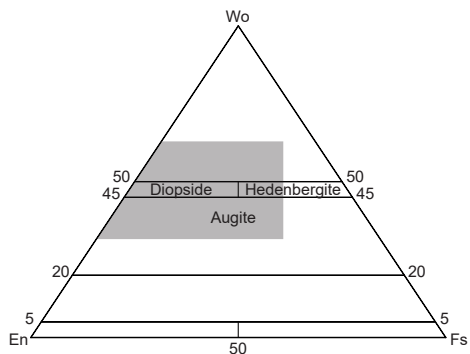
2 mm

C



GLS

PRH

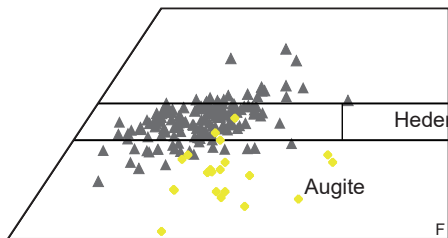
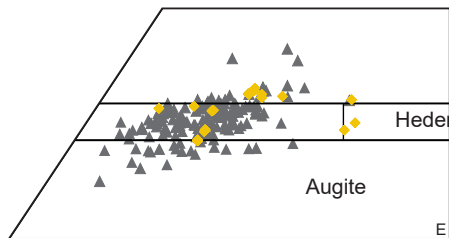
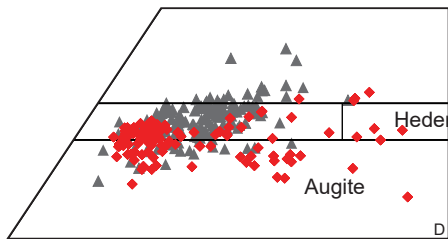
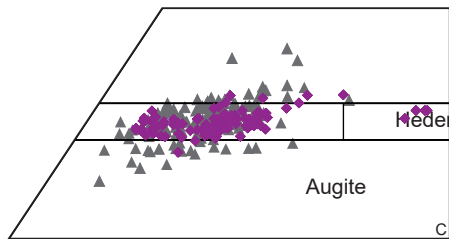
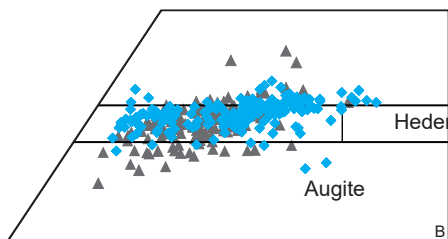
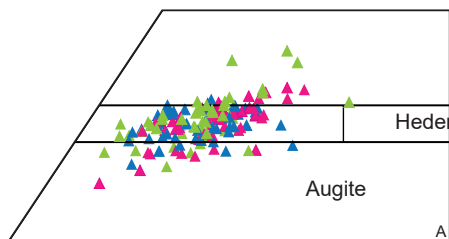


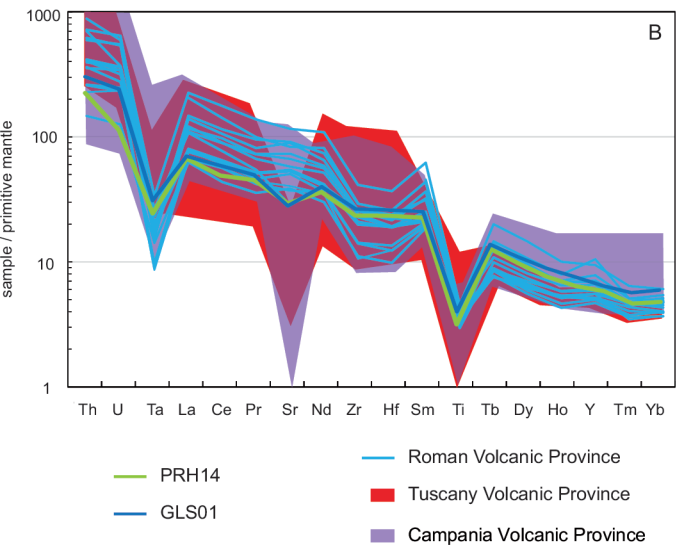
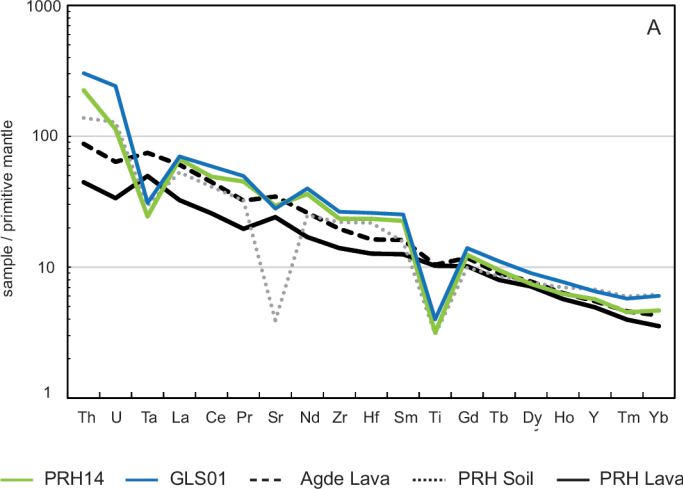
## Archaeological samples

- ▲ PRH14
- ▲ GLS01
- ▲ GLS02

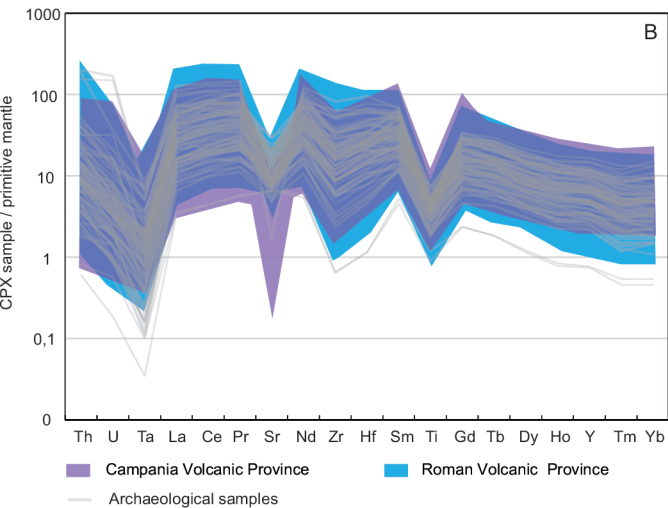
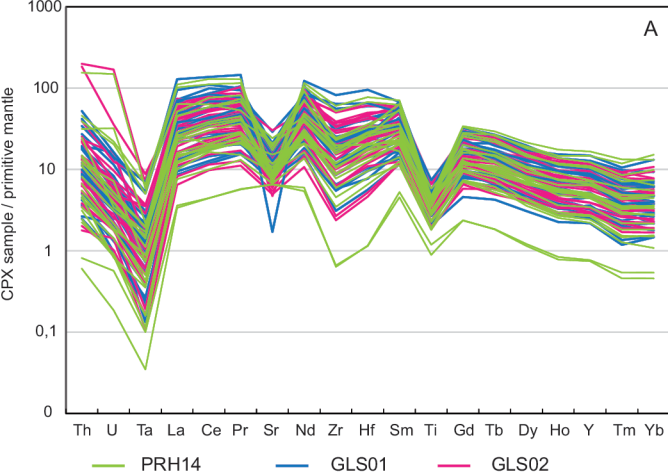
## Volcanic Provinces

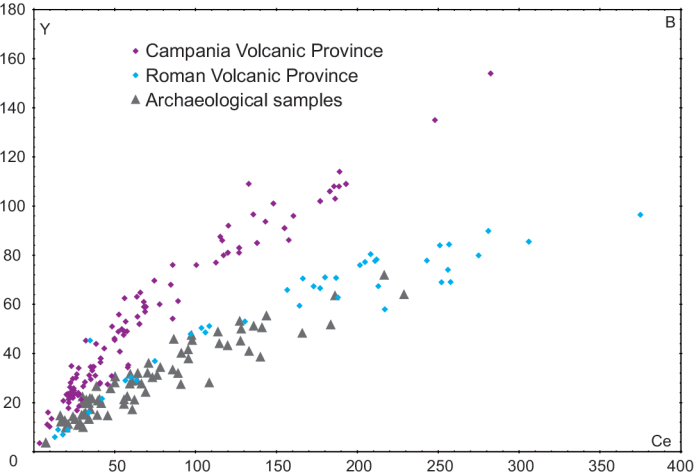
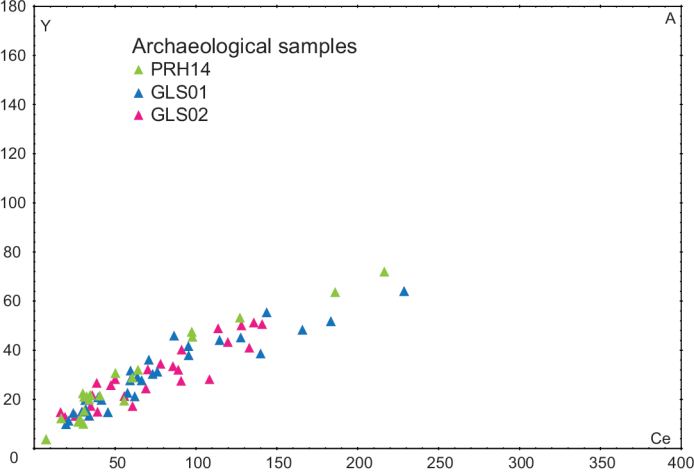
- ◆ Roman Volcanic Province
- ◆ Campanian Volcanic Province
- ◆ Tuscany Volcanic Province
- ◆ Apulian Volcanic Province
- ◆ Sardinia Volcanic Province
- ▲ Archaeological samples













■ Roman Volcanic Province

■ Associated river basins

Neolithic archaeological sites

1 - Peiro-Signado 2 - Caucade 3 - Pendimoun 4 - Arene Candide 5 - Pian del Ciliegio 6 - Panicarola 7 - Settecannelle

8 - La Marmotta 9 - Pianosa - Cala Giovanna 10 - Albertini 11 - Campu Stefanu 12 - Su Coloru

Obsidian geological outcrops

13 - Monte Arci 14 - Palmarola