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Domestic activities and pottery use in the Iron Age Corsican settlement of Cuciurpula revealed by organic residue analysis

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Abstract

The excavation of the protohistoric site of Cuciurpula (South Corsica, France) revealed a significant amount of potsherds, often bearing visible surface crusts, sometimes very thick. This exceptional case in the Mediterranean region, suggesting a good preservation of organic substances, provided a unique opportunity to address questions related to pottery function and natural organic substances exploited in Corsica during the first half of the 1st millennium BC. The molecular analysis (GC and GC/MS) of organic residues from three houses of the site, preserved in both pottery walls and charred surface crusts, highlighted the wide diversity and the various roles of substances contained and processed in ceramic vessels: animal fats, plant oils and waxes, beeswax, and conifer resin. These molecular data, considered together with the shapes of the vessels and their location into the habitation units, revealed the diversity of pottery function (culinary and technical) and spatial organisation of domestic activities between houses or in a house (distinction between storage and cooking areas).

Keywords

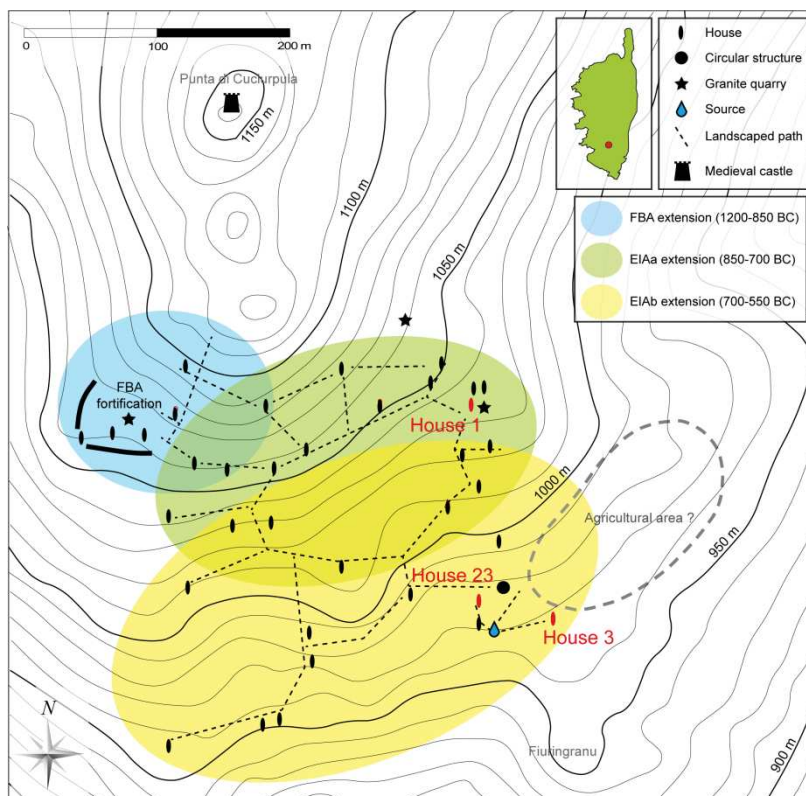
Organic residue analysis; pottery function; Iron Age; Corsica; spatial distribution; adhesive making

1. Introduction

For more than thirty years, organic residue analysis mainly focused on the study of the first pottery and the spread of Neolithic economy (e.g. Craig et al., 2011; Debono Spiteri et al., 2016; Evershed et al., 2008; Salque et al., 2013). Unlike the Neolithic period, largely studied in Europe and the Near East, Protohistoric sites attracted much less attention. For this period, organic residue analysis has been mainly performed on ceramic or wooden containers from sites located in the Alps (Carrer et al., 2016; Colonese et al., 2017; Evershed et al., 1995; Hayek et al., 1991; Raven et al., 1997), the British Islands (Copley et al., 2005a, 2005b; Craig et al., 2005; Cramp et al., 2014b, 2015; Dudd, 1999; Hayek et al., 1991), and Scandinavia (Cramp et al., 2014a; Hayek et al., 1991; Isaksson et al., 2010; McGovern et al., 2013). Only few data from other regions are available: Russia (Kostyukevich et al., 2016), Poland (Heron et al., 2016), France (for hafting residues analysis; Regert et al., 2003; Regert and Rolando, 2002) and Eastern Mediterranean (Decavallas, 2011; Roumpou et al., 2003; Steele and Stern, 2017). In particular, organic residue analysis data on protohistoric pottery content from north-western Mediterranean are very scarce (Faraco et al., 2016; Manzano et al., 2015), due to the lack of

45 studies in this region or to the generally poor preservation of organic substances in Mediterranean
46 contexts.

47 The protohistoric site of Cuciurpula is settled on the hillside of la Punta di Cuciurpula in south central
48 Corsica (Figure 1). The excavations carried out between 2010 and 2015 revealed a large settlement of
49 about 40 well-preserved structures occupied from the 12th to the 6th century BC (Late Bronze Age to
50 the beginning of the Second Iron Age; Peche-Quilichini et al., 2015). Based on exhaustive excavation
51 data of seven of the structures and further analysis of various artefacts, these structures have been
52 interpreted as habitation units. The presence of grinding stones together with scarce cereals and
53 domestic animal remains attests for agriculture and herding activities (Peche-Quilichini et al., 2014a).
54 The surrounding forest was also exploited as acorn and pine nuts were discovered at the site (Peche-
55 Quilichini et al., 2014a). This very limited picture of the exploitation of biological substances by
56 protohistoric societies at Cuciurpula has been partially completed thanks to organic residue analysis
57 focusing on adhesive and waterproofing substances (Rageot et al., 2016). In a complementary
58 approach, the present paper enlarges the scope of natural substances by studying the various fatty
59 substances contained and processed in ceramic vessels, in order to explore the whole diversity of
60 products exploited during Protohistory in Corsica. Secondly, by relating the content of pottery with
61 the shapes of the vessels and their location inside the habitation units, we aim at understanding how
62 these products were transformed, stored and consumed at the site. This will highlight the largely
63 unknown daily domestic activities of Corsican communities during the beginning of the 1st
64 millennium BC. With these aims, ceramic vessels from three different Iron Age houses of Cuciurpula
65 were selected. The lipids preserved in their walls were extracted and studied by gas chromatography
66 and mass spectrometry (GC and GC/MS).



67
68 *Figure 1: Location and map of the site. In red are the three habitation units considered in the present study.*

69 2. Materials and methods

70 2.1. Samples

71 For organic residues analysis, three different types of sample can be considered: free lumps
72 recovered in sediment, organic molecules trapped inside the pottery walls, and visible surface
73 residues (Regert, 2007, 2011). Among the latter, different categories could also be distinguished,
74 based on their location on the vessel, their adherence to ceramic surface, and their aspect (colour,
75 brightness, transparency, etc.). As described by Rageot *et al.* (2016), five classes of residues have
76 been identified based on simple observation at Cuciurpula: reparation residue along the edges of
77 ancient cracks (class A); thin residues on the inner surface interpreted as ceramic internal treatment,
78 maybe for waterproofing (class B); black residues on the external surface, maybe for decoration or
79 treatment of the exterior of the vessels (class C); free lumps, possibly adhesive storage before use or
80 manufacturing waste (class D); and thick visible remains on the interior surface, probably residues
81 related to the ceramic content (class E). The present study focuses on class E residues and on an
82 additional class, comprising organic molecules preserved in the porous ceramic matrix but invisible to
83 the naked eyes (class F) in order to investigate pottery use.

84 Three habitation units were selected for sampling. House 1 (850-600 BC) was chosen to complete the
85 data obtained during the adhesive substances study (Rageot *et al.*, 2016). Two supplementary
86 habitation units, House 3 and House 23 (700-550 BC), were selected to compare two
87 contemporaneous houses located close to each other. Two vessels from an additional house (house
88 38) were sampled because of their perforated walls suggesting a particular function. Due to the high
89 fragmentation of ceramics, only part of the sampled potsherds originated from ceramic vessels of
90 known shapes (deep vases, small pots, goblets, bowls and perforated shallow containers). A total of
91 39 potsherds and 20 visible residues was analysed (Table 1).

Sample name	Archaeo. number	House	US	Square	Morphology	Analysed residue
MR2696a and r	4891	1	135	K14	Unknown	Class F, E
MR2698a and r	4719	1	135	J14	Unknown	Class F, E
MR2699a and r	4506	1	135	J14	Unknown	Class F, E
MR2701a and r	5211	1	114	K9-10-11	Unknown	Class F, E
MR2702a and r	5178	1	114	J13	Unknown	Class F, E
MR2705a and r	5172	1	114	L13	Unknown	Class F, E
MR2706a and r	5243	1	114	DE13	Unknown	Class F, E
MR2707a and r	5211	1	114	K9-10-11	Unknown	Class F, E
MR2711a and r	5211	1	114	K9-10-11	Unknown	Class F, E
MR2709a and r	2794	1	114	C9	Unknown	Class F, E
MR2713a and r	2762	1	103	K12	Unknown	Class F, E
MR2714a and r	3053	1	103	H14	Unknown	Class F, E
MR2717a and r	2763	1	103	K13	Unknown	Class F, E
MR2727a and r	5235	1	122	AB456	Deep vase, large opening	Class F, E
LD10650a and r	964	1	105	E5	Big vase with thick walls	Class F, E
LD10651a and r	755	1	105	E5	Big vase with thick walls	Class F, E
LD10652a		38	5		Perforated shallow containers	Class F
LD10653a	5211	1	114	K9-10-11	Perforated shallow containers	Class F
LD10654a	2243	1	115	C10	Perforated shallow containers	Class F
LD10655a		38	1		Perforated shallow containers	Class F
LD10656a	1845	1	101	A12	Unknown	Class F
LD10659a and r	910	1	105	E3	Big vase with thick walls	Class F, E
LD10660a	110	23	13	E6	Deep vase, slightly restricted opening	Class F
LD10661a and r	121	23	16	E7	Unknown	Class F, E
LD10662a and r	155	23	16	E6	Unknown	Class F, E

LD10663a	255	23	16	E4	Deep and close vase with straight neck	Class F
LD10664a	204	23	16	E7	Bowl	Class F
LD10665a	96	23	13	E7	Deep and close vase	Class F
LD10666a	37	23	16	E4	Deep and close vase	Class F
LD10667	93	23	13	F5	Deep and close vase	Class F
LD10668a	459	3	405		Carinated bowl	Class F
LD10669a	528	3		Fosse 410	Carinated bowl	Class F
LD10670a	742	3	405	F8	Convex cup	Class F
LD10671a	600	3	402b	E7	Convex pot	Class F
LD10672a and r	266	3	402b		Convex neck pot	Class F, E
LD10673r	608	3	405	F8	S-profile pot	Class F
LD10674a		3	405		S-profile pot	Class F
LD10675a	349	3			Deep vase, restricted opening, thin bottom	Class F
LD10676a	648	3	425	D8	Deep vase, restricted opening	Class F

92 Table 1: List of samples analysed during the study and excavation data. r and E: visible residue related to the content of the
93 vessel; a and F: organic residue potentially absorbed inside the pottery matrix.

94 Surrounding sediments were also sampled at two different locations at the site to compare their lipid
95 composition with pottery sherds and surface visible residues. In order to study the effect of the
96 environmental context on lipid preservation, the acidity of these soil samples was also measured.

97 2.2. Lipid analysis

98 Sample treatment and analysis were carried out following Evershed et al. (1990), with some small
99 modifications. Visible surface residues were removed from sherds with a sterile scalpel. The surfaces
100 of the potsherds were then scraped using a clean scalpel to remove any exogenous lipids. Around 2 g
101 of potsherds and between 40 and 400 mg of visible carbonised residues were crushed using solvent-
102 washed mortar and pestle. An internal standard (20 µg of *n*-C₃₄, 1 mg.mL⁻¹ in *n*-hexane) was added
103 for quantitation. Solvent extraction was performed using dichloromethane/methanol solution
104 (DCM/MeOH; 2:1 v/v, 10 mL) and sonication. After centrifugation, the supernatant was evaporated
105 to dryness and dissolved in 500 µL of DCM/MeOH to obtain the total lipid extract (TLE). An aliquot of
106 the TLE (100 µL) was treated with *N,O*-bis(trimethylsilyl)trifluoroacetamide containing 1%
107 trimethylchlorosilane (BSTFA; 70°C, 1h). The excess BSTFA was evaporated under nitrogen and the
108 derivatised aliquot dissolved in hexane for molecular analyses.

109 Gas chromatographic (GC) analyses were performed on an Agilent Technologies 7890A device. 1 µL
110 of sample was introduced via an on-column injector into a 15 m x 0.32 mm i.d. fused silica capillary
111 (DB5-MS, 0.1 µL film thickness, Agilent J&W), with helium used as carrier gas. The GC temperature
112 programme was as follows: increased from 50°C to 100°C at 15°C min⁻¹, then from 100°C to 375°C at
113 10°C min⁻¹. For GC/MS analysis, the instrument was a Shimadzu GC 2010 PLUS chromatograph
114 coupled to a Shimadzu QP 2010 ULTRA mass spectrometer, fitted with a high temperature non-polar
115 column (DB5-HT, 15 m x 0.322 mm i.d., 0.1 µm film thickness, Agilent J&W). The injection was
116 performed using a splitless injector. The temperature programme consisted of a 1 min isothermal
117 hold at 50°C followed by an increase to 100°C at 15°C.min⁻¹, then to 240°C at 10°C.min⁻¹ and to
118 380°C.min⁻¹ and a final isothermal hold for 7 min. The GC-MS interface was maintained at a
119 temperature of 300°C and the mass spectrometer run in electron ionization mode (EI, 70 eV). Mass
120 spectra were acquired over the range *m/z* 50–950.

121 2.3. Complementary data on sediments

122 For pH measurements of the soils, 2 g of sample were sieved to 2 mm and dissolved in 25 mL distilled
123 water. The hand-held pH meter (Eutech PC 450, Thermo Scientific) was calibrated using 3 different

124 buffer solutions (pH 4.00, 7.00 and 9.81), and all pH measurements were made while stirring with the
 125 aid of a magnetic stirrer. Determination of carbonate content was performed using a Bernard
 126 Calcimeter calibrated with a known amount of pure CaCO₃. The amount of carbonate content was
 127 determined by quantifying the CO₂ released when the soil samples reacted with HCl (6M, 10 mL).

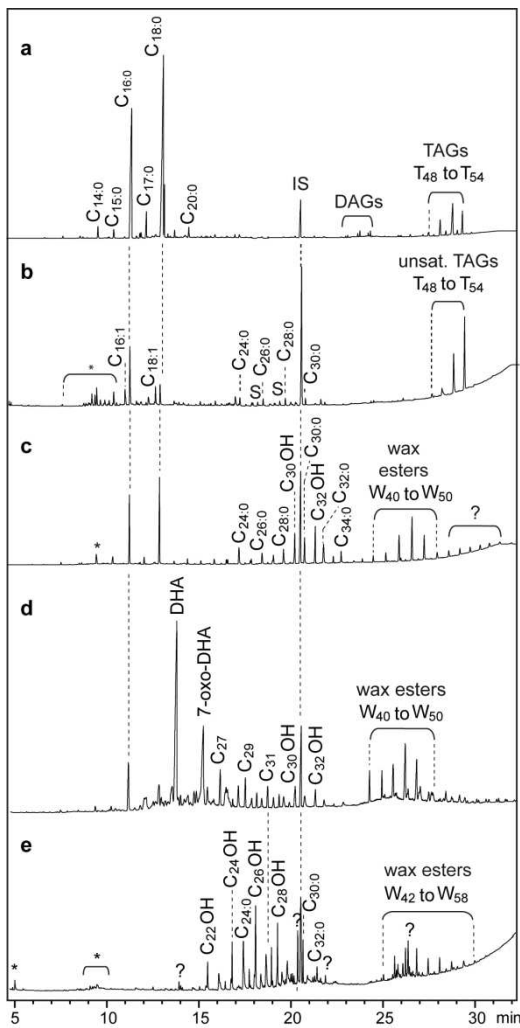
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129 3. Results and molecular interpretation

130 3.1. Extraction yields from ceramics

131 Lipid recovery from both pottery sherds and carbonised surface residues was surprisingly very high
 132 (66% of analysed potsherds contained more than 5 µg.g⁻¹, with a maximum of 3855 µg.g⁻¹) compared
 133 to other Mediterranean sites (Debono Spiteri et al., 2016; Evershed et al., 2008; Fanti et al.,
 134 submitted; Fanti, 2015; Šoberl et al., 2014). The exceptional preservation of organic substances at
 135 the site is also attested by the presence in some TLE of unsaturated triacylglycerols (TAGs), highly
 136 sensitive to both microbial and chemical degradation processes. To the best of our knowledge,
 137 Cuciurpula is one of the few archaeological contexts where they were preserved in pottery
 138 (Decavallas, 2011, pp. 176–177; Evershed et al., 2003; Fanti et al., submitted). More common
 139 molecules, such as saturated TAGs and their degradation products (di- and monoacylglycerols, and
 140 fatty acids), wax esters, *n*-alkanes, *n*-alcohols, and diterpenoid compounds have been preserved,
 141 highlighting the wide diversity of substances contained and processed in pottery vessels at
 142 Cuciurpula (Figure 2).

143

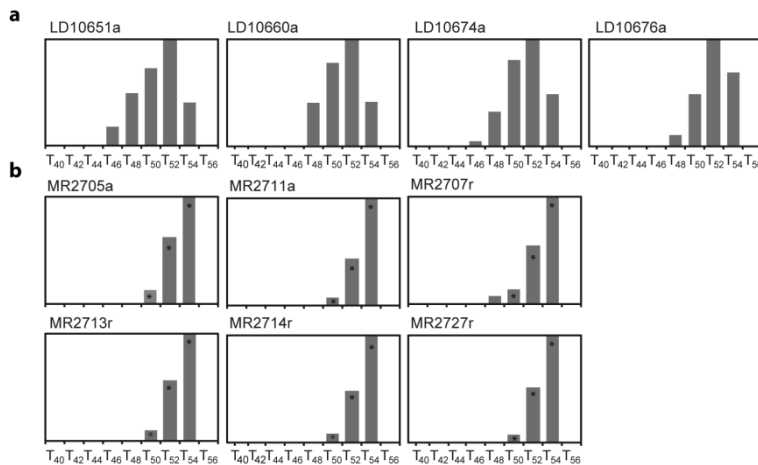


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145 Figure 2: Gas chromatograms of lipids from ceramic vessels (a to d) and of sediment (e). a) animal fat (LD10676a); b) plant
 146 oil (MR2707r); c) plant wax (LD10669a); d) beeswax and conifer resin (LD10662a). C_{xx}: fatty acids; DHA: dehydroabietic
 147 acid; C_{xx}OH: linear alcohols; C_{xx}: linear alkanes; S: saccharides; DAGs: diacylglycerols; W_{xx}: wax esters; TAGs: triacylglycerols;
 148 ?: unknown compound; IS: internal standard; *: modern contamination.

149 3.2. Fatty substances

150 TAGs were detected in 6 pottery sherds and 4 carbonised residues (LD10651a, LD10660a, LD10674a,
 151 LD10676a, MR2705a, MR2707r, MR2711a, MR2713r MR2714r, and MR2727r; Figure 3). The diversity
 152 of their profiles and the occasional presence of high quantity of unsaturated compounds suggest that
 153 both animal fats and plant oils were processed in pottery vessels.
 154



155 Figure 3: TAG profiles: a) mostly saturated; b) mostly unsaturated. *: unsaturated TAGs.
 156

157 When only saturated TAGs were detected (Figure 3a), their restricted profile (T₄₆ to T₄₈-T₅₄)
 158 dominated by T₅₂ suggested the presence of ruminant adipose fats (LD10651a, LD10660a, LD10674a
 159 and LD10676a; Dudd et al., 1999; Dudd and Evershed, 1998). In these samples, the ratio of palmitic
 160 to stearic acid was always dominated by the latter (P/S comprised between 0.52 and 0.71),
 161 supporting the animal origin of the organic substances. Absent from samples with TAG typical of
 162 plant oils, linear and branched C_{15:0} and C_{17:0} fatty acids were identified in some samples with
 163 saturated TAGs. Their presence strengthens the hypothesis of ruminant fat, as they can result from
 164 the activity of bacteria in the rumen (Dudd et al., 1999; Evershed, 1993). Based on molecular analysis,
 165 dairy products do not seem absorbed in the studied pottery but further isotopic analysis will be
 166 carried out to assess this point (Dudd and Evershed, 1998).

167 Unsaturated TAG profiles were clearly identified in 6 samples based on their retention time and their
 168 specific mass spectrum, different from their saturated counterpart (MR2705a, MR2707r, MR2711a,
 169 MR2713r, MR2714r, and MR2727r; Figure 3b). This particular profile is close to plant oils with the
 170 dominating T₅₄ (triolein) and the T₅₂ and T₅₀ peaks comprising oleic and palmitic acid moieties (Copley
 171 et al., 2005c; Garnier et al., 2009). Besides C_{16:0} and C_{18:0}, most of these samples also contained
 172 substantial amounts of unsaturated C_{16:1} and C_{18:1}, and odd and even-carbon numbered long-chain
 173 fatty acids (C_{20:0} to C_{34:0}), confirming a plant origin. A small amount of saccharides was also
 174 sometimes detected, thanks to ions *m/z* 204, 217 and 361 in mass spectra (Medeiros and Simoneit,
 175 2007).

176 The absence of other molecular markers except palmitic and stearic acids in thirteen more TLEs, did
 177 not allow identification of the source of the fatty substances present in samples LD10656a,
 178 LD10659a, LD10664a, LD10665a, LD10667a, LD10673a, LD10670a, LD10675, MR2696a, MR2699r,
 179 MR2702r, MR2706a and MR2727a.

180 Methylated fatty acids have been detected in three samples (LD10651a, LD10656a, and LD10659a).
 181 These compounds are known to be formed when fatty substances are exposed to very high

temperatures (Raven et al., 1997), but they can also result from low heating of animal fat or plant oil in ceramics at less than 100°C, probably when catalysed by metal salts in the pottery fabric, as revealed by recent experiments (Drieu et al., unpublished results). Methyl esters may also correspond to artefacts resulting from the reaction, possibly catalysed by clays, of glycerol esters or linear carboxylic acids with methanol during extraction of the samples. At this stage of the investigation the origin of these methylated esters is still not fully understood.

3.3. Waxes

Typical molecular signals of waxy substances were identified in several samples: wax esters and their degradation products (even-carbon numbered long-chain fatty acids and *n*-alcohols) and ranges of long-chain odd-carbon numbered *n*-alkanes. The diversity of profiles of the homologous series of these molecular compounds suggests various origins.

First, three samples displayed classical profiles of well-preserved beeswax (LD10656a, LD10661a and LD10662a) with palmitic wax esters in the range W₄₀–W₅₀ dominated by W₄₆, and the corresponding hydroxyesters (Figures 2d and 4a; Heron et al., 1994; Regert et al., 2001; Roffet-Salque et al., 2015; Tulloch, 1971). Typical beeswax distributions of *n*-alkanes (C₂₇ to C₃₁, maximizing at C₂₇) and long-chain fatty acids (C_{24:0} to C_{32:0}) were also detected. Partial hydrolysis of wax esters is attested by the presence of *n*-alcohols from C_{24OH} to C_{32OH} (Charters et al., 1995; Evershed et al., 1997). Degraded beeswax was detected in 5 more samples (LD10661r, LD10662r, MR2696r, MR2714a and r) by the characterisation of slightly different profiles, altered by heating or natural processes: reduction of shorter wax esters (W₄₀ to W₄₄) and changes in the *n*-alkanes profiles (Figure 4b; Heron et al., 1994; Regert et al., 2001).

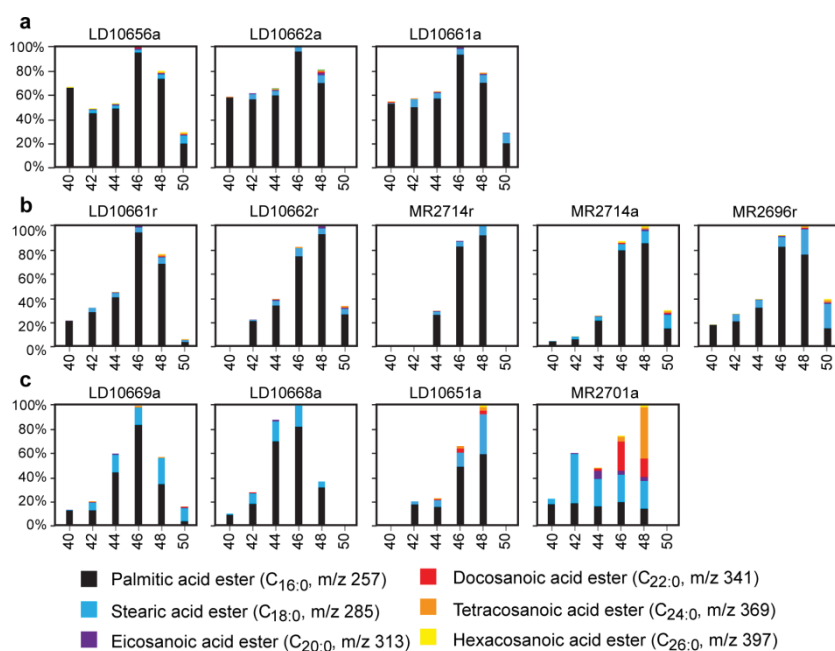


Figure 4: Wax esters composition in samples containing waxy substances. a) well-preserved beeswax profile; b) degraded beeswax profile; c) probably plant wax profile.

Waxy profiles were also attested in 4 other samples (LD10651a, LD10668a, LD10669a, and MR2701a) but with unusual distributions, suggesting a different natural origin. Mass spectrometric investigations of these samples revealed that palmitic wax esters were coeluted with several isomers (C_{18:0} to C_{26:0} wax esters; Figure 4c). Other components present included *n*-alkanes profiles, dominated by C₂₉ or C₃₁ and various long-chain fatty acids (Figure 2c). These molecular markers probably originate from plant epicuticular waxes (Ribechini et al., 2008) absorbed in pottery wall

212 during the cooking of leafy vegetables, for example. A mixing of epicuticular plant wax and beeswax
213 is not to be excluded for some samples (Ribechini et al., 2008).

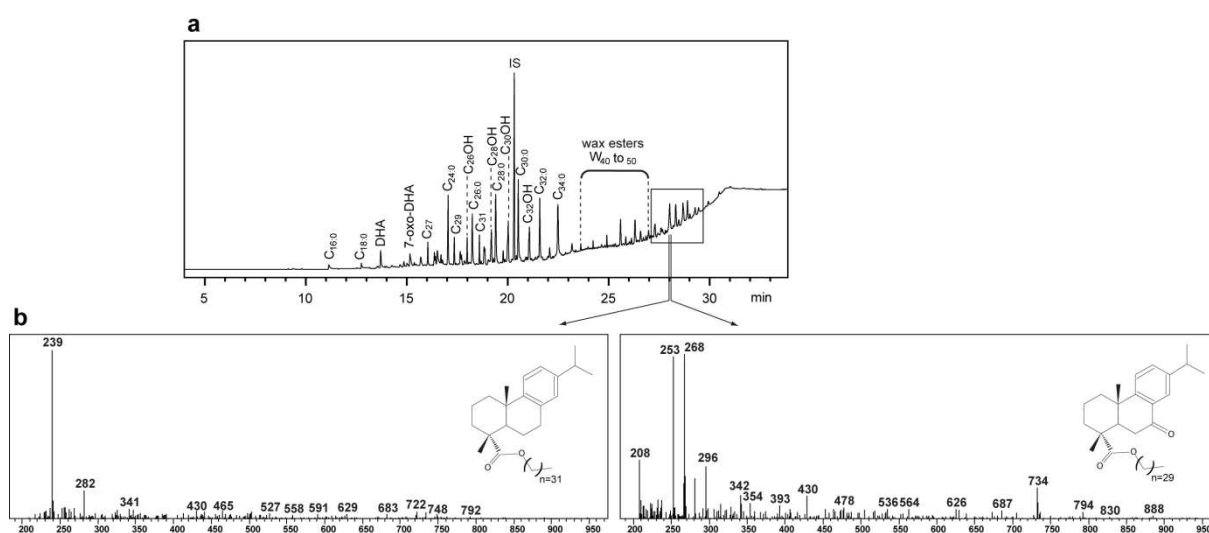
214 *N*-alkanes and *n*-alcohols also occurred in samples with wax esters existing as traces (LD10676a,
215 LD10672a, and MR2727r), making the origin of the waxy substance difficult to determine.

216 3.4. Diterpenoid compounds

217 Seven TLE from sherds and their associated visible crusts yielded substantial quantities of diterpenoid
218 constituents (LD10661a and r, LD10662a and r, LD10671a, LD10672a and r), mainly by-products of
219 the degradation of abietic acid: dehydroabietic acid (DHA), didehydroabietic acid, 7-oxo-
220 dehydroabietic (7-oxo-DHA) and 15-hydroxy-7-oxo-dehydroabietic acid (15-hydroxy-7-oxo-DHA).
221 Together with the presence of pimaric and isopimaric acids and unknown DHA derivatives (with mass
222 spectra dominated by the m/z 239 ion), these markers attested the presence of conifer resin
223 (Colombini et al., 2005; Regert and Rolando, 2002).

224 The presence of these biomarkers in carbonised surface residues related to the content of the vessels
225 (i.e. with appearance, localisation on the vessel and adherence to the wall not suggesting pottery
226 coating of repair) and the identification of retene in some samples indicate that conifer resin was
227 heated in ceramic vessels. Furthermore, conifer resin was identified together with beeswax
228 biomarkers in four samples (LD10661a and r, LD10662a and r), while beeswax is absent, or present as
229 traces in others (LD10671a, LD10672a and r). In four samples (LD10661a, LD10662a and r,
230 LD10672a), two homologous series of unusual molecular compounds eluted after wax esters
231 (between 27 and 30 min; Figure 5a). Their mass spectra, respectively dominated by ions m/z 239, and
232 both 253 and 268, suggested that they were esters of DHA or 7-oxo-DHA and long-chain linear
233 alcohols (Figure 5b). The length of these *n*-alcohols could have been partly unravelled since the
234 molecular peaks of part of these spectra fitted with esters of 7-oxo-DHA and $C_{26}OH$ to $C_{30}OH$ *n*-alcohols.
235 Esters of terpenic alcohols and fatty acids have already been determined in archaeological samples
236 (Charrié-Duhaut et al., 2007; Dudd and Evershed, 1999), but to the best of our knowledge, this is the
237 first identification of esters of diterpenoid acids and linear alcohols. Their formation should result from
238 an esterification reaction between diterpenoid compounds and free *n*-alcohols (also present as free
239 alcohols in the sample and probably resulting from the hydrolysis of wax esters), maybe during the
240 heating of beeswax and conifer resin.

241



242 Figure 5: Example of TLE containing unusual molecular compounds eluted after the wax esters (sample LD10662a). a) partial
243 gas chromatogram; b) mass spectra of the peaks eluted at 28 min and proposed molecular structures.
244

245 3.5. Sediments analysis

246 The sediments of Cuciurpula are clearly acidic, with pH values comprised between 4.9 and 5.0. The
247 acidic properties of the sediments are likely to be due to the granitic substrate of Corsica allowing the
248 development of soils rich in SiO₂ and depleted in Ca²⁺ ions and thus favourable to acidification

249 (Fabian et al., 2014). This hypothesis is supported by the very low total carbonate content we
250 measured in the sediment samples (1%).

251 The sediments yielded respectively 51.2 and 4.6 $\mu\text{g.g}^{-1}$ of lipids, with a clear predominance of *n*-
252 alcohols ($\text{C}_{22\text{OH}}$ to $\text{C}_{30\text{OH}}$, with a maximum at $\text{C}_{26\text{OH}}$), a typical pattern of plant waxes (Figure 2e;
253 Eglinton and Hamilton, 1967; Gülz, 1994; Kolattukudy, 1970). Sample LD10678 also revealed wax
254 esters (W_{42} to W_{58} , maximum W_{46} and W_{48}), long chain fatty acids from $\text{C}_{22:0}$ to $\text{C}_{30:0}$ and odd-
255 numbered *n*-alkanes (C_{27} - C_{33} , with maximum at C_{31}), confirming the plant origin of the lipids
256 contained in the soils (Eglinton and Hamilton, 1967; Evershed et al., 1994; Evershed and Lockheart,
257 2007; Gülz, 1994; Kolattukudy, 1970; Tulloch and Hoffman, 1973). Although long chain fatty acids, *n*-
258 alkanes and *n*-alcohols were also detected in some potsherds, their respective distributions in soils
259 and in pottery are clearly different (for example, $\text{C}_{26\text{OH}}$ is never dominant *n*-alcohols in potsherds).

260

261 4. Discussion

262 4.1. Preservation of the lipid signal

263 The Mediterranean climate being unfavourable to lipid preservation (Evershed, 2008a), the good
264 preservation of lipids at Cuciurpula is surprising. The molecular composition of the TLE from pottery
265 sherds and visible residues are very different from the lipids extracted from the sediments,
266 confirming that the detected lipids are due to pottery use and did not migrate from the surrounding
267 soils (Heron et al., 1991). The acidic properties of the sediments could however explain the
268 significant amount of lipids preserved at the site, because they are unfavourable to microorganisms
269 development (DeLaune et al., 1981; Moucawi et al., 1981). Furthermore, in acidic conditions fatty
270 acids are not present as soluble ions and are thus less easily eliminated by leaching (Evershed et al.,
271 1997).

272

273 4.2. Exploitation of natural substances at Cuciurpula during the 1st 274 Iron Age

275 The presence of animal fats in ceramic vessels is quite common in pre- and protohistoric pottery (e.g.
276 Copley et al., 2005a, 2005b; Dudd et al., 1999; Salque et al., 2012). The molecular assemblages and
277 particularly the TAGs profiles suggest that ruminant fats were mainly exploited in ceramic vessels at
278 Cuciurpula, probably cattle and ovicaprids. These results are coherent with the scarce faunal remains
279 discovered and studied at Cuciurpula (Peche-Quilichini et al., 2014a). Based on molecular profiles of
280 TAGs, it was not possible to detect any dairy product but this has now to be confirmed by isotopic
281 analysis (Dudd & Evershed, 1998).

282 More unusual is the large quantity of samples yielding plant substances: pottery seems to have been
283 used equally for animal (11 vessels) and plant products (9 vessels), sometimes mixed together (3
284 vessels). This uncommonly high percentage of pottery used to contain or process diverse plant oils
285 and waxes may be due to the good preservation of organic molecules at the site. Nevertheless, it also
286 gives evidence for significant exploitation of the plant kingdom by protohistoric societies of the first
287 Iron Age in Corsica. Plant substances seem to have been heavily contained and processed in pottery
288 during Protohistory, in particular in the Eastern Mediterranean region (Greece, Cyprus, and
289 Macedonia) and Britannic Islands (Copley et al., 2005a, 2005b; Decavallas, 2011; Evershed et al.,
290 2003; Steele and Stern, 2017). This is confirmed by archaeobotanical studies at Cuciurpula, which
291 highlighted the exploitation of acorn, pine nuts and barley (Peche-Quilichini et al., 2014a). Olive
292 exploitation is not attested at the site but at other Corsican settlements (Bronzini de Caraffa et al.,
293 2005; de Lanfranchi, 2005; Magdeleine and Ottaviani, 1983; Terral et al., 2005). The unsaturated TAG
294 profile extracted from archaeological sherds could originate from olive oil, acorn or pine nut fats
295 (Copley et al., 2005c; Garnier et al., 2009; León-Camacho et al., 2004; Lopes and Bernardo-Gil, 2005;
296 Nergiz and Dönmez, 2004), but saccharides and fatty acid distributions are different from these

297 substances (Al-Rousan et al., 2013; Buonasera, 2007; De Man, 1985; Nergiz and Dönmez, 2004).
298 Analysis of phytoliths or starch grains on potsherds and carbonised surface residues could provide
299 new data to help identifying these plant substances.

300 The presence of beeswax in several pottery walls and visible residues related to the content of the
301 vessels confirm the exploitation of beehive products at Cuciurpula, previously discussed by Rageot et
302 al. (2016). When detected together with animal fats or plant oils, beeswax can be interpreted as a
303 residue of honey mixed with other commodities for meal preparation or as waterproofing agent of
304 the vessel walls before its use to process edible commodities (Regert et al., 2001; Roffet-Salque et al.,
305 2015). The detection of beeswax in pottery from Cuciurpula enters within a global picture of beehive
306 products exploitation during Protohistory. Beeswax is attested in potsherds from an Etruscan
307 contemporaneous site in Italy (Garnier et al., 2002), but also in earlier settlements in Eastern
308 Mediterranean (Decavallas, 2011; Evershed et al., 1997; Roumpou et al., 2003; Steele and Stern,
309 2017). Whether the exploited beehives at Cuciurpula were domestic or wild is difficult to establish,
310 but domestic beehives are not to be excluded since they are attested contemporaneously in the Near
311 East (Bloch et al., 2010) and a few centuries later in Greece (Evershed et al., 2003) and in the Po plain
312 (Castellano et al., 2017).

313 The identification of diterpenoid compounds confirms the exploitation of conifer resin already
314 highlighted by Rageot et al. (2016). No molecular marker of birch bark tar has been detected during
315 the present study. Considering the good stability of such molecular compounds in archaeological
316 contexts, this absence suggests that birch bark tar was not contained or processed in the ceramics,
317 but used to repair and coat the external walls of some vessels (Rageot et al., 2016).

318

319 4.3. Pottery function

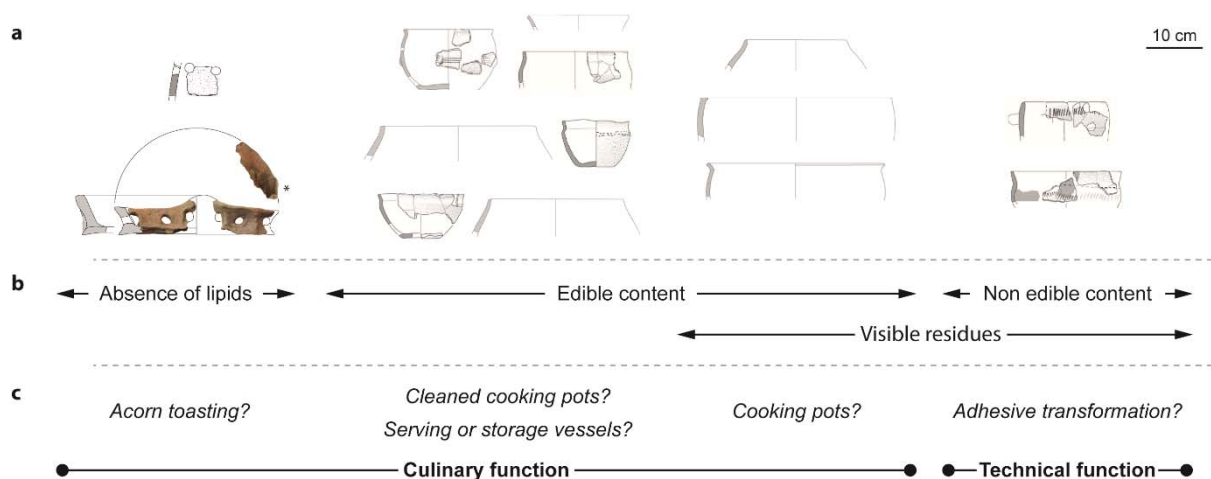
320 Based on the shape of the vessels, on the presence/absence of carbonised residues and on the fatty
321 substances identified, four different types of uses have been distinguished.

322 Considering the edible substances (animal fats, plant oils and waxes) identified in 25 ceramics, it is
323 clear that they were used, at least once, as culinary utensils (Figure 6). The presence of carbonised
324 residues on the surface of 15 of them indicates that their content was heated. The absence of
325 molecular markers of thermal transformation (such as ketones and ω -(*o*-alkylphenyl)alkanoic acids)
326 could be due to the lack of metallic salts catalysing their formation or to cooking processes at low
327 temperature, like simmering (Evershed, 2008b; Raven et al., 1997). The co-occurrence of various
328 products (animal fats, plant oils and waxes and / or beeswax) in several ceramic vessels from House 1
329 represents either intentional mixing resulting from culinary recipes or successive uses. For this
330 category of vessels, the diversity of molecular signals, especially when comparing the TLE from the
331 pottery walls (accumulation of the successive contents) and the corresponding food crusts (last use;
332 Oudemans and Boon, 1991), suggests that each ceramic vessel was used for various commodities.
333 The shape of these “cooking pots” was rarely possible to reconstruct due to their high fragmentation,
334 but the few known shapes are vases of large diameter. The thick wall of some vases (more than 8.5
335 mm for 1/3 of the pottery displaying food crusts) could seem contradictory with a use as cooking pot
336 (Bronitsky, 1986, p. 250; Rice, 1987, p. 229). However, thick walls could have been intentionally
337 chosen to strengthen the vessels against mechanical shocks potentially occurring when moving the
338 pots or stirring the content (Rice, 1987, p. 228).

339 Ten ceramics with edible substances preserved inside their walls do not display any visible residues
340 on their surface (Figure 6), maybe due to their cleaning or to the absence of contact with fire - as
341 serving or storage vessels.

342 The analysis of the four perforated containers revealed a complete absence of lipids (Figure 6).
343 Considering the excellent general preservation of lipids at the site, this absence seems incompatible
344 with potential use for straining substances such as dairy or beehive products, as already identified in

345 Neolithic sites (Regert et al., 2001; Salque et al., 2013). Considering the shallow shape suggested by a
 346 similar perforated sherd at the recently excavated site of Valchiria (Sartène, South Corsica; Peche-
 347 Quilichini et al., in press), another hypothesis of use is the toasting of acorns. Such function could be
 348 related to grilled acorn stocks revealed by excavation in a house of the site (Peche-Quilichini et al.,
 349 2014a). Furthermore, this use should not allow any release and absorption of lipids in the pottery
 350 wall nor the formation of foodcrusts and could be thus consistent with the results of our analysis.
 351 Finally, the analysis of four vessels highlighted the presence of exclusively non-comestible products
 352 (conifer resin, sometimes mixed with beeswax), suggesting a technical use probably related to
 353 adhesives (Figure 6). As already discussed by Rageot et al. (2016), the mixing of beeswax and conifer
 354 resin can be intentionally made in order to modify the physical properties of the adhesives. The
 355 systematic presence of thick carbonised surface residue on the corresponding potsherds, the
 356 identification of molecular markers of heating, and the degradation of the beeswax molecular profile
 357 in some carbonised residues related to the content of the vessels indicate that the adhesives were
 358 heated inside the pottery. The absence of methyl-dehydroabietate suggests that conifer resin was
 359 used instead of conifer pitch.
 360

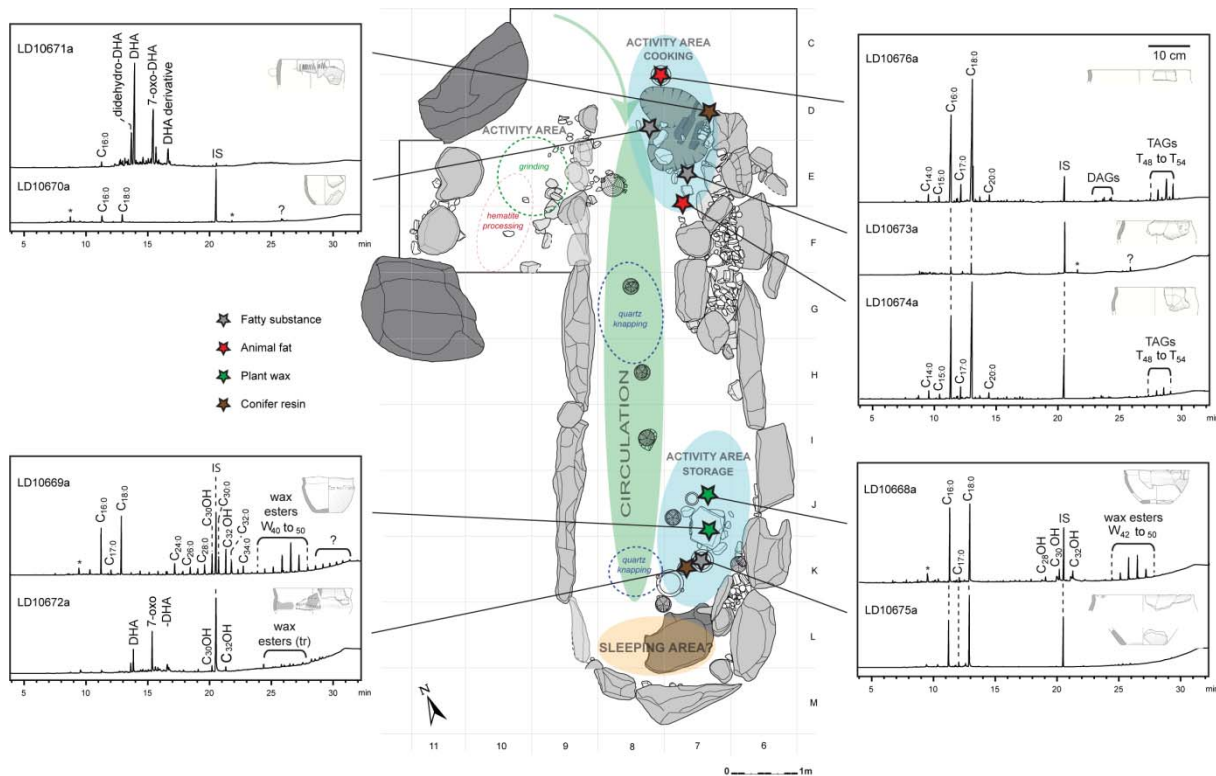


361
 362 *Figure 6: Pottery functional groups: shapes of the vessels (a) classified according to the edibility of lipid content and the*
 363 *presence of visible residues (b), and hypothesis of vessels use (c). * proposed shape based on similar perforated sherd*
 364 *excavated at the site of Valchiria (Sartène, South Corsica; Peche-Quilichini et al., in press). Vessels drawings: K. Peche-*
 365 *Quilichini and T. Lachenal.*

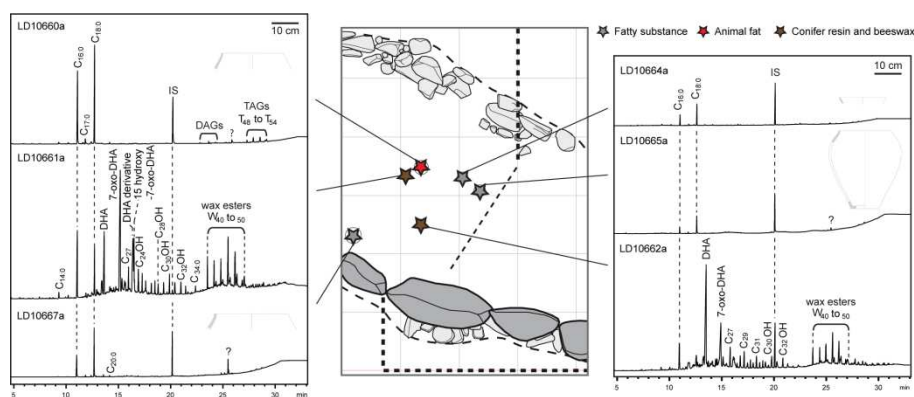
366

367 4.4. Spatial distribution

368 General spatial organisation in the habitation units at Cuciurpula has been unravelled for Houses 3
 369 and 23, based on the distribution of pottery and lithic tools (Peche-Quilichini et al., 2014b, 2015).
 370 Potsherds originate from a unique area in House 23 and two distinct ones in House 3 (probably
 371 respectively cooking and storage areas). The sampling in House 1 was mainly carried out in a waste
 372 area near the actual house.
 373



374
 375 Figure 7: Spatial distribution of pottery from House 3 and gas chromatograms of associated TLE. Cxx:x: fatty acids; DAGs:
 376 diacylglycerols; CxxOH: linear alcohols; Wxx: wax esters; TAGs: triacylglycerols; DHA: dehydroabiatic acid; ?: unidentified
 377 compound; IS: internal standard; * : modern contaminant. Computer-aided design of House 3 and vessels drawings: T.
 378 Lachenal.



379
 380 Figure 8: Spatial distribution of pottery from House 23 and gas chromatograms of associated TLE. Cxx:x: fatty acids; DAGs:
 381 diacylglycerols; CxxOH: linear alcohols; Wxx: wax esters; TAGs: triacylglycerols; DHA: dehydroabiatic acid; ?: unidentified
 382 compound; IS: internal standard; * : modern contaminant. Computer-aided design of House 23 and vessels drawings: K.
 383 Peche-Quilichini.

384 Organic residue analysis in pottery revealed some differences in pottery use inside each domestic
 385 unit. In Houses 3 and 23, pottery related to both culinary (animal and/or plant fats) and technical
 386 (conifer resin, with or without beeswax) activities do not seem spatially distinct inside the habitation
 387 unit (Figure 7 and Figure 8). “Adhesive recipes” seem however to be specific to each house: conifer
 388 resin was used alone in House 3, but was mixed with beeswax in House 23. In House 1, adhesive-
 389 related activities do not seem to have occurred: pottery usually contained animal fats, plant oils and
 390 plant waxes; when traces of diterpenoid compounds are detected, we suggest they correspond to
 391 residues of the surface treatment identified by Rageot et al. (2016). Plant products are also unequally
 392 distributed between houses. Plant oils are largely identified in pottery from House 1 and plant waxes
 393 in samples from House 3, but they both seem absent from House 23. No spatial data is available for

394 House 1 but in House 3, plant products occur only in vessels originating from the storage area (one of
395 them was identified in a pit closed by a granite slab), while animal fats were detected in pottery near
396 the hearth (Figure 7). Beeswax also seems to have been exploited differently from one house to
397 another. Absent from House 3, it was detected in potsherds from House 23 always mixed with
398 conifer resin – suggesting technical use. In samples from House 1, its co-occurrence with animal fat
399 or plant oil suggests that beeswax could be a residue of recipes assembling various commodities and
400 honey. As carbonised surface residues indicate that most of the samples from this house have been
401 heated, the use of beeswax as a sealant is very unlikely - beeswax melts at 60°C and is soluble in
402 animal fat (Charters et al., 1995; Regert et al., 2001).

403 The differential spatial distribution of substances contained and processed in pottery can be due to
404 the specificity of the sampling (waste area vs. inside of the habitation units), or to the differential
405 preservation of lipids between houses. These variations can also result from slight differences in the
406 organisation of domestic areas and activities from a house to another. However, these preliminary
407 results did not reveal any real specialisation in the activities, as already identified for Neolithic
408 settlements (Matlova et al., 2017; Vieugué, 2010).

409

410 5. Conclusion

411 The exceptional case of the site of Cuciurpula, located in the largely granitic island of Corsica,
412 provided an unique opportunity to explore the exploitation of natural substances during Protohistory
413 in the Mediterranean region, generally unfavourable to lipid preservation. The present study
414 revealed the diversity of substances consumed and used for both culinary and technical activities:
415 animal fats, plant oils and waxes, conifer resin, and beeswax. Furthermore, the careful recording of
416 the location of the potsherds inside domestic structures enabled us to tentatively reconstruct areas
417 of activity inside domestic units, and to the best of our knowledge, for the first time using organic
418 residue analysis. In particular, differences of pottery content between cooking and storage areas
419 were highlighted for one of the houses. Despite the globally homogenous pattern of exploitation of
420 natural substances at the site, we could also identify differential behaviours from one house to
421 another in cooking habits and adhesive making. These preliminary observations must be confirmed
422 by enlarging the sampling to include other houses with clear spatial distribution of pottery at the site
423 (e.g. House 1 and House 6; Peche-Quilichini et al., 2015) and in other sites from the first Iron Age in
424 Corsica.

425

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434

435

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