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

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

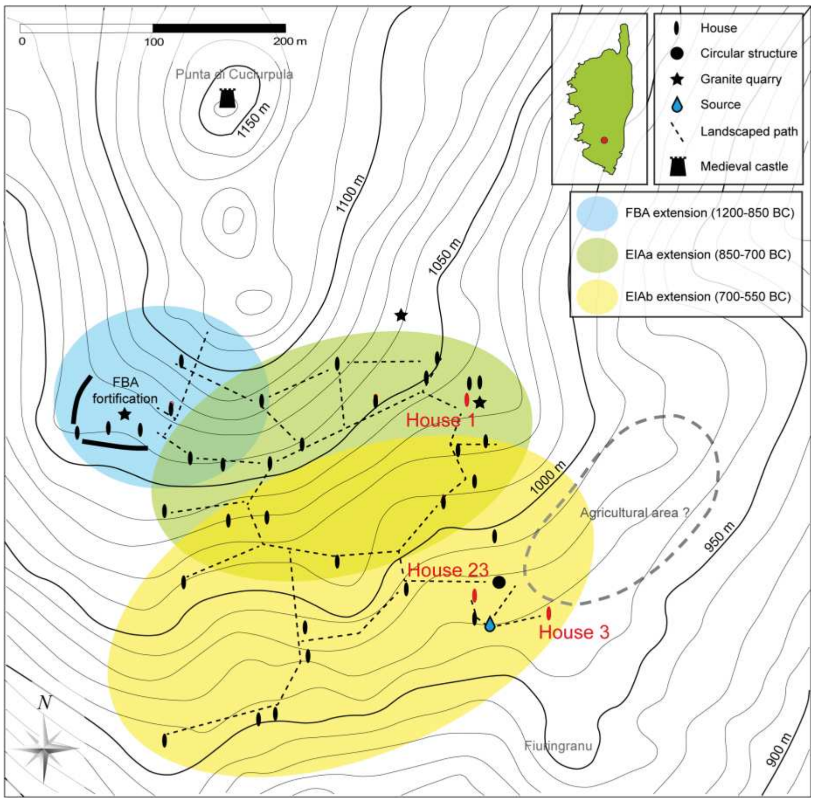

Figure 1: Location and map of the site. In red are the three habitation units considered in the present study.

2. Materials and methods

2.1. Samples

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

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

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

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

2.2. Lipid analysis

Sample treatment and analysis were carried out following Evershed et al. (1990), with some small modifications. Visible surface residues were removed from sherds with a sterile scalpel. The surfaces of the potsherds were then scraped using a clean scalpel to remove any exogenous lipids. Around 2 g of potsherds and between 40 and 400 mg of visible carbonised residues were crushed using solvent-washed mortar and pestle. An internal standard (20 µg of n-C_{34}, 1 mg.mL^{-1} in n-hexane) was added for quantitation. Solvent extraction was performed using dichloromethane/methanol solution (DCM/MeOH; 2:1 v/v, 10 mL) and sonication. After centrifugation, the supernatant was evaporated to dryness and dissolved in 500 µL of DCM/MeOH to obtain the total lipid extract (TLE). An aliquot of the TLE (100 µL) was treated with *N*,*O*-bis(trimethylsilyl)trifluoroacetamide containing 1% trimethylchlorosilane (BSTFA; 70°C, 1h). The excess BSTFA was evaporated under nitrogen and the derivatised aliquot dissolved in hexane for molecular analyses.

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

2.3. Complementary data on sediments

For pH measurements of the soils, 2 g of sample were sieved to 2 mm and dissolved in 25 mL distilled water. The hand-held pH meter (Eutech PC 450, Thermo Scientific) was calibrated using 3 different buffer solutions (pH 4.00, 7.00 and 9.81), and all pH measurements were made while stirring with the aid of a magnetic stirrer. Determination of carbonate content was performed using a Bernard Calcimeter calibrated with a known amount of pure $CaCO_3$. The amount of carbonate content was determined by quantifying the CO_2 released when the soil samples reacted with HCl (6M, 10 mL).

3. Results and molecular interpretation

3.1. Extraction yields from ceramics

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

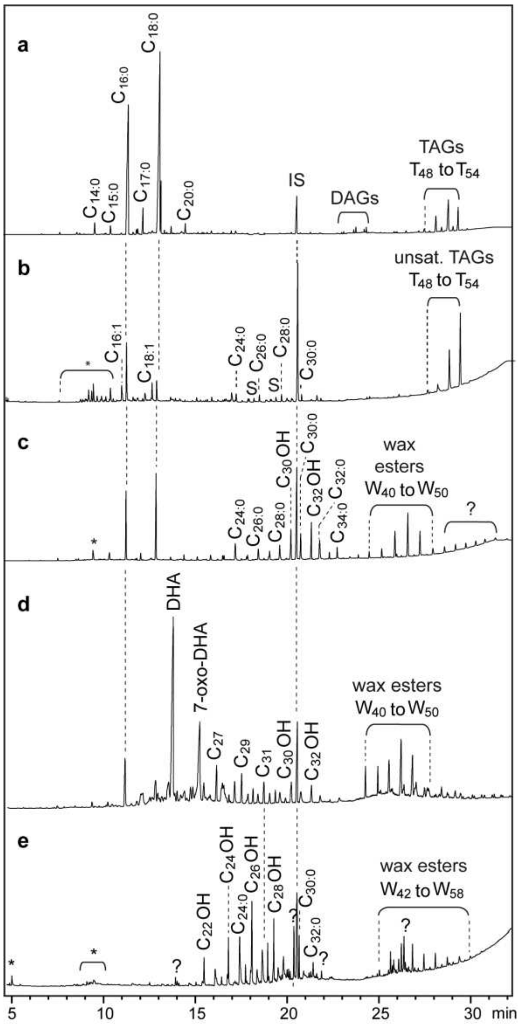

*Figure 2: Gas chromatograms of lipids from ceramic vessels (a to d) and of sediment (e). a) animal fat (LD10676a); b) plant oil (MR2707r); c) plant wax (LD10669a); d) beeswax and conifer resin (LD10662a). $C_{xx:x}$: fatty acids; DHA: dehydroabietic acid; C_{xx}OH: linear alcohols; C_{xx}: linear alkanes;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

3.2. Fatty substances

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

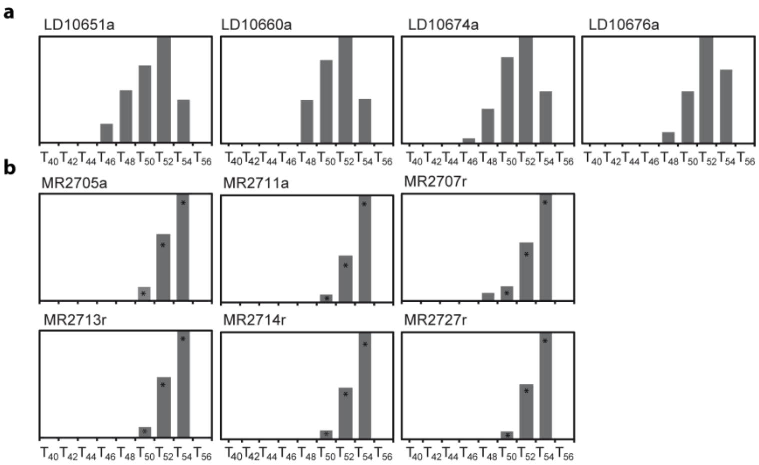

*Figure 3: TAG profiles: a) mostly saturated; b) mostly unsaturated. *: unsaturated TAGs.*

When only saturated TAGs were detected (Figure 3a), their restricted profile (T_{46} to T_{48}-T_{54}) dominated by T_{52} suggested the presence of ruminant adipose fats (LD10651a, LD10660a, LD10674a and LD10676a; Dudd et al., 1999; Dudd and Evershed, 1998). In these samples, the ratio of palmitic to stearic acid was always dominated by the latter (P/S comprised between 0.52 and 0.71), supporting the animal origin of the organic substances. Absent from samples with TAG typical of plant oils, linear and branched $C_{15:0}$ and $C_{17:0}$ fatty acids were identified in some samples with saturated TAGs. Their presence strengthens the hypothesis of ruminant fat, as they can result from the activity of bacteria in the rumen (Dudd et al., 1999; Evershed, 1993). Based on molecular analysis, dairy products do not seem absorbed in the studied pottery but further isotopic analysis will be carried out to assess this point (Dudd and Evershed, 1998).

Unsaturated TAG profiles were clearly identified in 6 samples based on their retention time and their specific mass spectrum, different from their saturated counterpart (MR2705a, MR2707r, MR2711a, MR2713r, MR2714r, and MR2727r; Figure 3b). This particular profile is close to plant oils with the dominating T_{54} (triolein) and the T_{52} and T_{50} peaks comprising oleic and palmitic acid moieties (Copley et al., 2005c; Garnier et al., 2009). Besides $C_{16:0}$ and $C_{18:0}$, most of these samples also contained substantial amounts of unsaturated $C_{16:1}$ and $C_{18:1}$, and odd and even-carbon numbered long-chain fatty acids ($C_{20:0}$ to $C_{34:0}$), confirming a plant origin. A small amount of saccharides was also sometimes detected, thanks to ions *m/z* 204, 217 and 361 in mass spectra (Medeiros and Simoneit, 2007).

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

Methylated fatty acids have been detected in three samples (LD10651a, LD10656a, and LD10659a). 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 no 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 W_{40}–W_{50} dominated by W_{46}, 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 (C_{27} to C_{31}, maximizing at C_{27}) 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 (W_{40} to W_{44}) and changes in the *n*-alkanes profiles (Figure 4b; Heron et al., 1994; Regert et al., 2001).

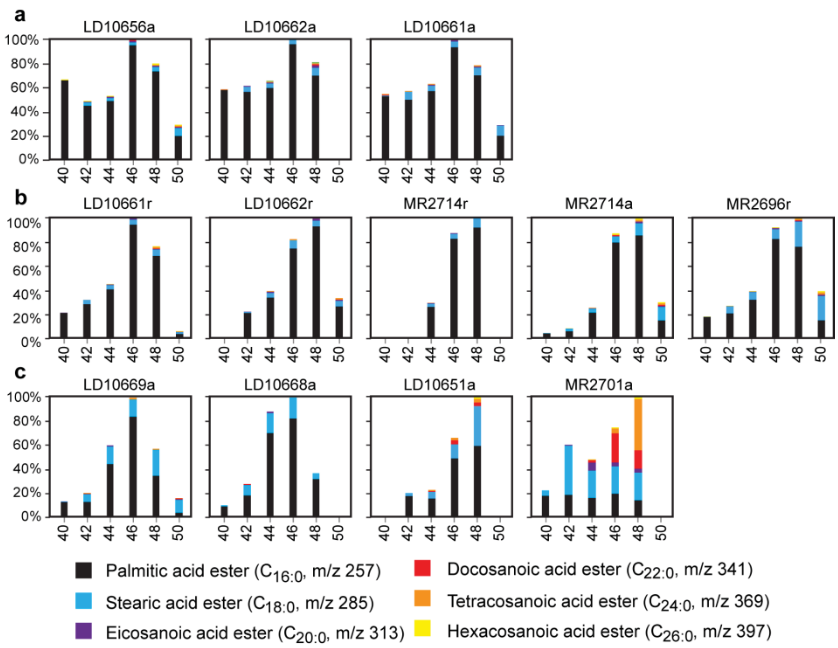

Figure 4: Wax esters composition in samples containing waxy substances. a) well-preserved beeswax profile; b) degraded beeswax profile; c) probably plant wax profile.

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 C_{29} or C_{31} 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 during the cooking of leafy vegetables, for example. A mixing of epicuticular plant wax and beeswax is not to be excluded for some samples (Ribechini et al., 2008).

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

3.4. Diterpenoid compounds

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

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

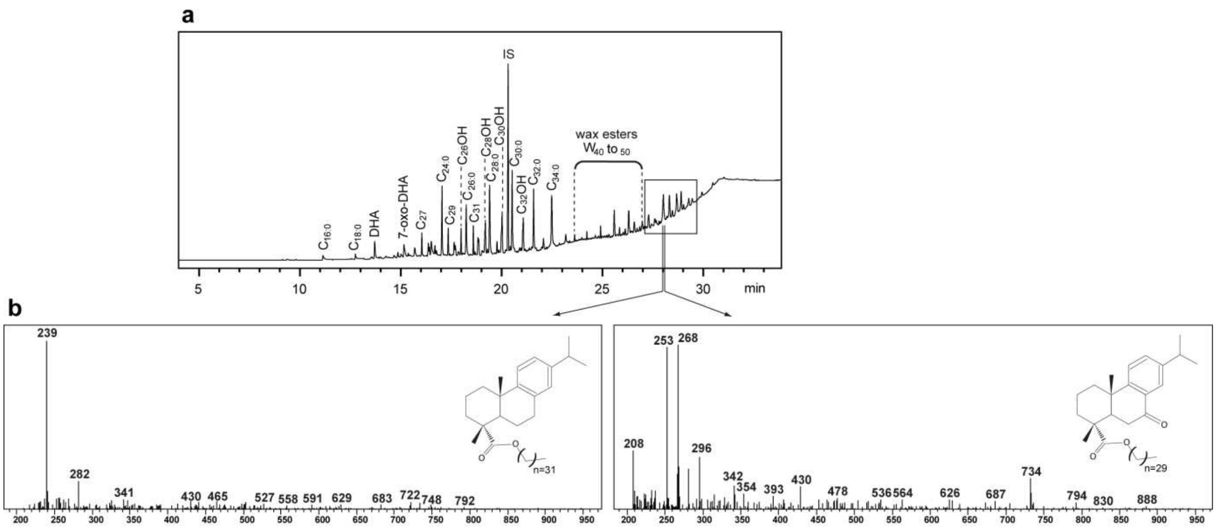

Figure 5: Example of TLE containing unusual molecular compounds eluted after the wax esters (sample LD10662a). a) partial gas chromatogram; b) mass spectra of the peaks eluted at 28 min and proposed molecular structures.

3.5. Sediments analysis

The sediments of Cuciurpula are clearly acidic, with pH values comprised between 4.9 and 5.0. The acidic properties of the sediments are likely to be due to the granitic substrate of Corsica allowing the development of soils rich in SiO_2 and depleted in Ca^{2+} ions and thus favourable to acidification

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

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

4. Discussion

4.1. Preservation of the lipid signal

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

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

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

More unusual is the large quantity of samples yielding plant substances: pottery seems to have been used equally for animal (11 vessels) and plant products (9 vessels), sometimes mixed together (3 vessels). This uncommonly high percentage of pottery used to contain or process diverse plant oils and waxes may be due to the good preservation of organic molecules at the site. Nevertheless, it also gives evidence for significant exploitation of the plant kingdom by protohistoric societies of the first Iron Age in Corsica. Plant substances seem to have been heavily contained and processed in pottery during Protohistory, in particular in the Eastern Mediterranean region (Greece, Cyprus, and Macedonia) and Britannic Islands (Copley et al., 2005a, 2005b; Decavallas, 2011; Evershed et al., 2003; Steele and Stern, 2017). This is confirmed by archaeobotanical studies at Cuciurpula, which highlighted the exploitation of acorn, pine nuts and barley (Peche-Quilichini et al., 2014a). Olive exploitation is not attested at the site but at other Corsican settlements (Bronzini de Caraffa et al., 2005; de Lanfranchi, 2005; Magdeleine and Ottaviani, 1983; Terral et al., 2005). The unsaturated TAG profile extracted from archaeological sherds could originate from olive oil, acorn or pine nut fats (Copley et al., 2005c; Garnier et al., 2009; León-Camacho et al., 2004; Lopes and Bernardo-Gil, 2005; Nergiz and Dönmez, 2004), but saccharides and fatty acid distributions are different from these substances (Al-Rousan et al., 2013; Buonasera, 2007; De Man, 1985; Nergiz and Dönmez, 2004). Analysis of phytoliths or starch grains on potsherds and carbonised surface residues could provide new data to help identifying these plant substances.

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

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

4.3. Pottery function

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

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

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

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

Neolithic sites (Regert et al., 2001; Salque et al., 2013). Considering the shallow shape suggested by a similar perforated sherd at the recently excavated site of Valchiria (Sartène, South Corsica; Peche-Quilichini et al., in press), another hypothesis of use is the toasting of acorns. Such function could be related to grilled acorn stocks revealed by excavation in a house of the site (Peche-Quilichini et al., 2014a). Furthermore, this use should not allow any release and absorption of lipids in the pottery wall nor the formation of foodcrusts and could be thus consistent with the results of our analysis.

Finally, the analysis of four vessels highlighted the presence of exclusively non-comestible products (conifer resin, sometimes mixed with beeswax), suggesting a technical use probably related to adhesives (Figure 6). As already discussed by Rageot et al. (2016), the mixing of beeswax and conifer resin can be intentionally made in order to modify the physical properties of the adhesives. The systematic presence of thick carbonised surface residue on the corresponding potsherds, the identification of molecular markers of heating, and the degradation of the beeswax molecular profile in some carbonised residues related to the content of the vessels indicate that the adhesives were heated inside the pottery. The absence of methyl-dehydroabietate suggests that conifer resin was used instead of conifer pitch.

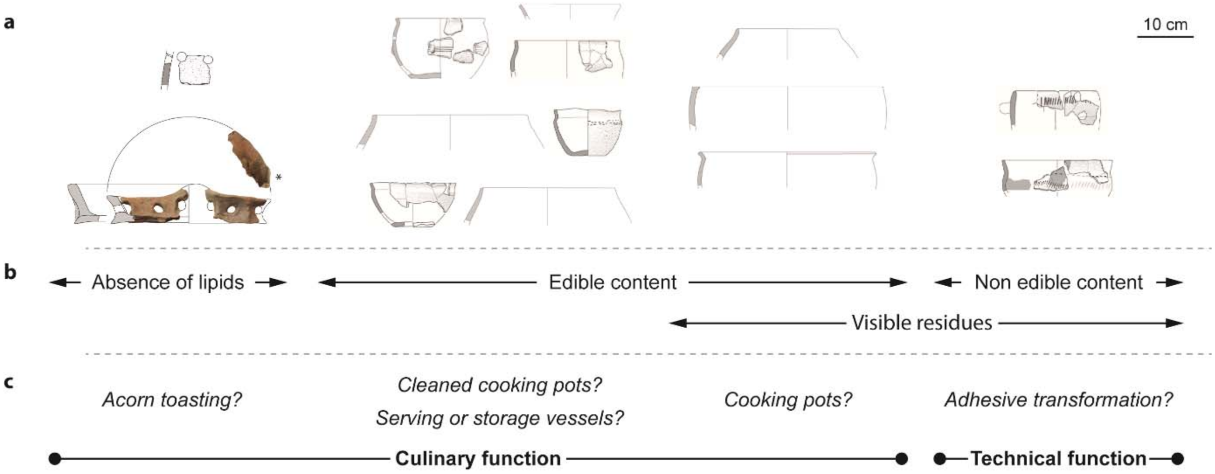

*Figure 6: Pottery functional groups: shapes of the vessels (a) classified according to the edibility of lipid content and the presence of visible residues (b), and hypothesis of vessels use (c). * proposed shape based on similar perforated sherd excavated at the site of Valchiria (Sartène, South Corsica;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

4.4. Spatial distribution

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

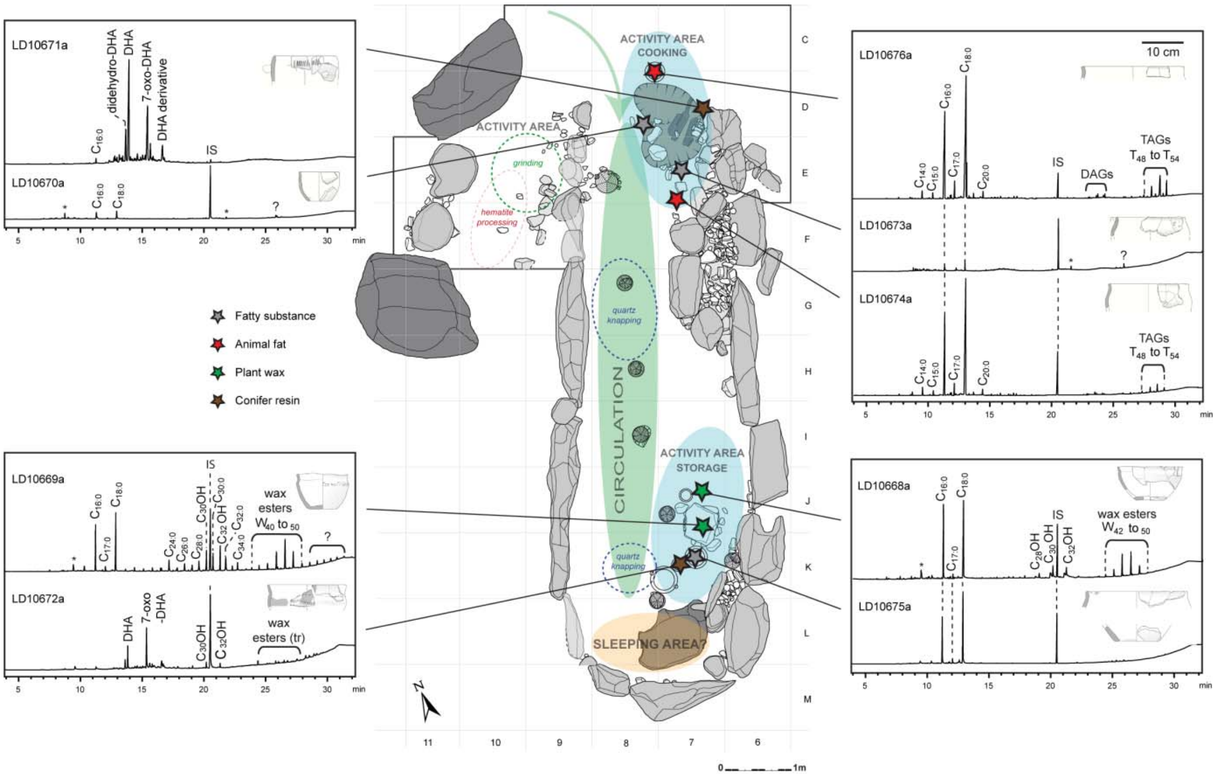

*Figure 7: Spatial distribution of pottery from House 3 and gas chromatograms of associated TLE. Cxx:x: fatty acids; DAGs: diacylglycerols; CxxOH: linear alcohols; Wxx: wax esters; TAGs: triacylglycerols; DHA: dehydroabietic acid; ?: unidentified compound; IS: internal standard; * : modern contaminant.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

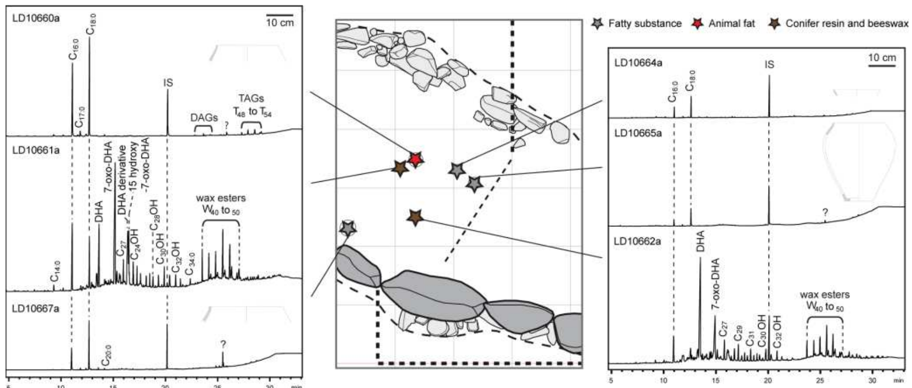

*Figure 8: Spatial distribution of pottery from House 23 and gas chromatograms of associated TLE. Cxx:x: fatty acids; DAGs: diacylglycerols; CxxOH: linear alcohols; Wxx: wax esters; TAGs: triacylglycerols; DHA: dehydroabietic acid; ?: unidentified compound; IS: internal standard; * : modern contaminant.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

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

House 1 but in House 3, plant products occur only in vessels originating from the storage area (one of them was identified in a pit closed by a granite slab), while animal fats were detected in pottery near the hearth (Figure 7). Beeswax also seems to have been exploited differently from one house to another. Absent from House 3, it was detected in potsherds from House 23 always mixed with conifer resin – suggesting technical use. In samples from House 1, its co-occurrence with animal fat or plant oil suggests that beeswax could be a residue of recipes assembling various commodities and honey. As carbonised surface residues indicate that most of the samples from this house have been heated, the use of beeswax as a sealant is very unlikely - beeswax melts at 60°C and is soluble in animal fat (Charters et al., 1995; Regert et al., 2001).
The differential spatial distribution of substances contained and processed in pottery can be due to the specificity of the sampling (waste area vs. inside of the habitation units), or to the differential preservation of lipids between houses. These variations can also result from slight differences in the organisation of domestic areas and activities from a house to another. However, these preliminary results did not reveal any real specialisation in the activities, as already identified for Neolithic settlements (Matlova et al., 2017; Vieugué, 2010).

5. Conclusion

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

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