A Neolithic without dairy? Chemical evidence from the content of ceramics from the Pendimoun rockshelter (Castellar, France, 5750-5150 BCE)

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A Neolithic without dairy? Chemical evidence from the content of ceramics from the Pendimoun rock-shelter (Castellar, France, 5750-5150 BCE)

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Abstract

The early phases of Neolithic expansion in the Central and Western Mediterranean are relatively poorly understood with regards to the diversity in the subsistence economy and the degree of interaction with indigenous hunter-gatherers. Recent analysis of pottery manufacturing techniques also points to a surprisingly diverse range of practices across the region. Here, we explore the use of pottery during the early phases of the Neolithic in the North-western Mediterranean, through analysis of organic residues absorbed in the pots of the Pendimoun rock-shelter (Impresso-Cardial complex) in South-eastern France. Using molecular and single-compounds stable carbon isotopes analyses, our study reveals that the majority of pots were used for processing wild or domesticated ruminant carcase fats, although lipids derived from cereals and wild non-ruminant fats, such as hares, cannot be excluded. In addition, a few of the earlier Impressa vessels showed the presence of beeswax and porcine fats. Correlations between the contents of the vessels and their volume were found, suggesting that vessels were manufactured for specific uses. Only one vessel from the Cardial phase showed evidence of dairy fats strengthening the notion that milk was not heavily or systematically exploited by the earliest Neolithic populations of the Mediterranean. Overall, however, our study calls for more detailed regional investigations to fully understand the transition to farming according to the local landscape and environmental context.

Keywords

Organic residues analysis; Neolithic; pottery use; dairy products; ruminant adipose fat; beeswax

1. Introduction

The arrival of farming in the Mediterranean and the nature of the earliest farming communities is a key theme in European prehistory. In the Central and Western Mediterranean, many prehistorians now dismiss simple models of demic diffusion (Ammerman & Cavalli-Sforza, 1973) in favour of a more complex mosaic where farming dispersed at a non-uniform rate (e.g. Binder et al., 2017; Guilaine and Manen, 2007; Manen et al., 2019). At the heart of understanding of the dispersal of farming is the need to characterise variation in the nature of the earliest Neolithic activity and subsistence practices, as well as interactions, if any, with indigenous hunter-gatherers. Recent analysis of Impresso-Cardial material culture in the Mediterranean reveals great diversity in this respect, for example, in the variety of ceramic decoration (Binder, 2013; Manen et al., 2010), shaping methods (Gomart et al., 2017), in the variability of domestic animal exploitation (Debono Spiteri et al., 2016; Rowley-Conwy et al., 2013) and harvesting technology (Mazzucco et al., 2020). The first Neolithic communities in the North-western Mediterranean settled as pioneer groups across the Liguro-Provencal arc and in Languedoc at the beginning of the 6th millennium BC (Figure 1). While animal husbandry and cereal agriculture are well attested, exploitation of molluscs and wild fruits also appear to have been a part of the economy. Analyses of faunal assemblages obtained from the earliest well-dated sites of this region (Arene Candide, Pont-de-Roque-Haute), also show that domesticated animals were initially unlikely to have been heavily exploited for their milk (Debono Spiteri et al., 2016), in contrast to pioneer Neolithic settlers in other parts of Europe (Copley et al., 2005; Ethier et al., 2017). Such diversity of practice could be the consequence of multiple interactions between Neolithic populations originating from the Aegean and indigenous hunter-gatherers, but also between different Neolithic groups, as shown by the ancient DNA analysis (aDNA) of these populations (Rivollat et al., 2020).

Many questions are still outstanding. For example, to what degree was early Neolithic Mediterranean economy dependent on wild resources? Does this reflect strong interactions with the surrounding Mesolithic communities, or an adaptation of the pioneer Neolithic groups to the local environment? What was the nature of early animal husbandry practices? When was a fully agrarian economy established in this region?

Analyses of organic residues preserved in ceramics following their use provides an approach to address some of these issues. Organic residue analysis has revealed a strong association of early Neolithic pottery use for processing milk especially in Northern Europe (e.g. Copley et al., 2005; Cramp et al., 2014; Salque et al., 2013), a product that can be unambiguously assigned to animal domestication. More recently, this type of approach has contributed to deconstructing the overly simplistic model of sedentary European Neolithic societies relying solely on the economy of production. Recent work has shown that regional variation in the degree to which dairy products were processed in early Neolithic pottery (Cubas et al., 2020) and that other products that had no relation to the productive economy, such as fish, were also processed (Craig et al., 2011; Cramp et al., 2019). However, very little work has been carried on early pottery of the North-western Mediterranean.

Figure 1: Map of the earliest Neolithic sites in the North-western Mediterranean. In red, the site studied in this paper. Source: Google Maps

The Pendimoun rock-shelter offers a unique opportunity to study the Neolithisation mechanisms in the North-western Mediterranean through the reconstruction of pottery use by organic residue analysis. Pendimoun is one of the few stratified sites that has yielded Impressa and Cardial material, witnessing both pioneering Neolithic incursions and the permanent establishment of farming communities. A new set of recent dates has provided a reliable and detailed sequence of these occupations (Binder et al., 2017). The Impresso-Cardial pottery assemblage, composed of more than 200 identified vessels, has recently been subjected to an in-depth interdisciplinary study leading to a very detailed understanding of the assemblage in terms of clay pastes, manufacturing method and decoration (Gabriele, 2014; Gomart et al., 2017). Finally, the site is conveniently situated to study the interactions between local Mesolithic populations, occupying the highlands, and Neolithic groups settling along the coast. Organic remains mainly support an agropastoral Neolithic economy, with the predominance of domestic sheep and domesticated cereals (Binder et al., 2020; Delhon et al., 2020; Rowley-Conwy et al., 2013). Excavation of the site also revealed the presence of rocky marine shellfish, fruits and nuts (e.g. acorns, hazelnuts) and some exploitation of wild animals; evidence that collection and hunting activities continued in parallel with the productive economy (Binder et al., 2020).

Since the Mediterranean area is known for poor preservation of organic matter associated with ceramic vessels (e.g. Debono-Spiteri, 2012), the first aim of these investigations was to assess the degree of lipid preservation in the Impressa and Cardial pottery at the Pendimoun rock-shelter. Following this objective, we aimed to determine whether hunted, gathered or fished resources (mainly marine products, which are easily identified by organic residue analysis) were used in ceramics, or whether pottery was exclusively dedicated to processing produced foods (i.e. domesticated animal carcass fats, dairy products and cereals).

2. Material and methods

Archaeological site and pottery assemblage

The Pendimoun rock-shelter is located in the municipality of Castellar in the Alpes-Maritimes (France) at an altitude of about 690 m on a limestone massif, less than one kilometre from the Italian border (Barral, 1958; Binder et al., 1993). Its sedimentary sequence extends from the Epipalaeolithic to the historical periods, with a particularly well-developed stratigraphic sequence between the 6th and 3rd millennia BC.

The site was first excavated by L. Barral in 1955 and 1956 (Barral, 1958), then by D. Binder from 1985 to 1992, and from 1997 and 2006 (Binder et al., 1993, 2020). Part of the archaeological remains are located under the roof of the shelter (about 15 m^2), but the layers of occupation extend beyond the shelter itself. Based on pottery and AMS dating of short-lived samples, the two first phases of Neolithic occupation were during the 6th millennium BC. The Impressa phase, with finger and instrumental impressed pottery decorations dated to between 5730 and 5430 BCE. The Cardial phase, mainly with cardiidae shell decorated pottery, dated to between 5550 and 5150 BCE (Binder et al., 2020).

The economy of the site was based on the cultivation of cereals (naked barley, emmer and einkorn) and the rearing of domestic caprines, mainly sheep (Binder et al., 2020; Rowley-Conwy et al., 2013). Due to the small amount of faunal remains, it is not possible to study the livestock management for the Impressa phase. During the Cardial phase, production is mainly centred on meat. The succession of dung layers indicates the use of the shelter for penning small ruminants, probably on a seasonal basis (Binder et al., 1993, 2020). The hunted fauna is very restricted and mainly represented by hare, deer and wild cats. The wild or domestic status of the suids found at the site remains unclear. The gathering of wild fruits and marine molluscs completes the available resources: acorns, hazelnuts, and shells from rocky environments (mainly limpets, but also *Trochidae spp.*) have been discovered at the site (Binder et al., 1993, 2020).

Pottery is present in relatively large densities for this period: 126 individual vessels have been identified in the Impressa horizon and 83 in the Cardial horizon. Three groups of pastes were distinguished by mineralogical and petrographic analyses: local glauconitic earths, granitic earths from the Argentera-Mercantour massif (about 30 km as the crow flies from the site) and pastes made from mixtures of the two previous ones, in varying proportions (Binder and Sénépart, 2010; Gabriele, 2014). The Impressa and Cardial pottery of Pendimoun rock-shelter was made using the spiral patchwork manufacturing method, specific to the Tyrrhenian area (Gomart et al., 2017). The vessels are characterised by a great variability in morphology and volume, suggesting a diversity of function (Figure 2).

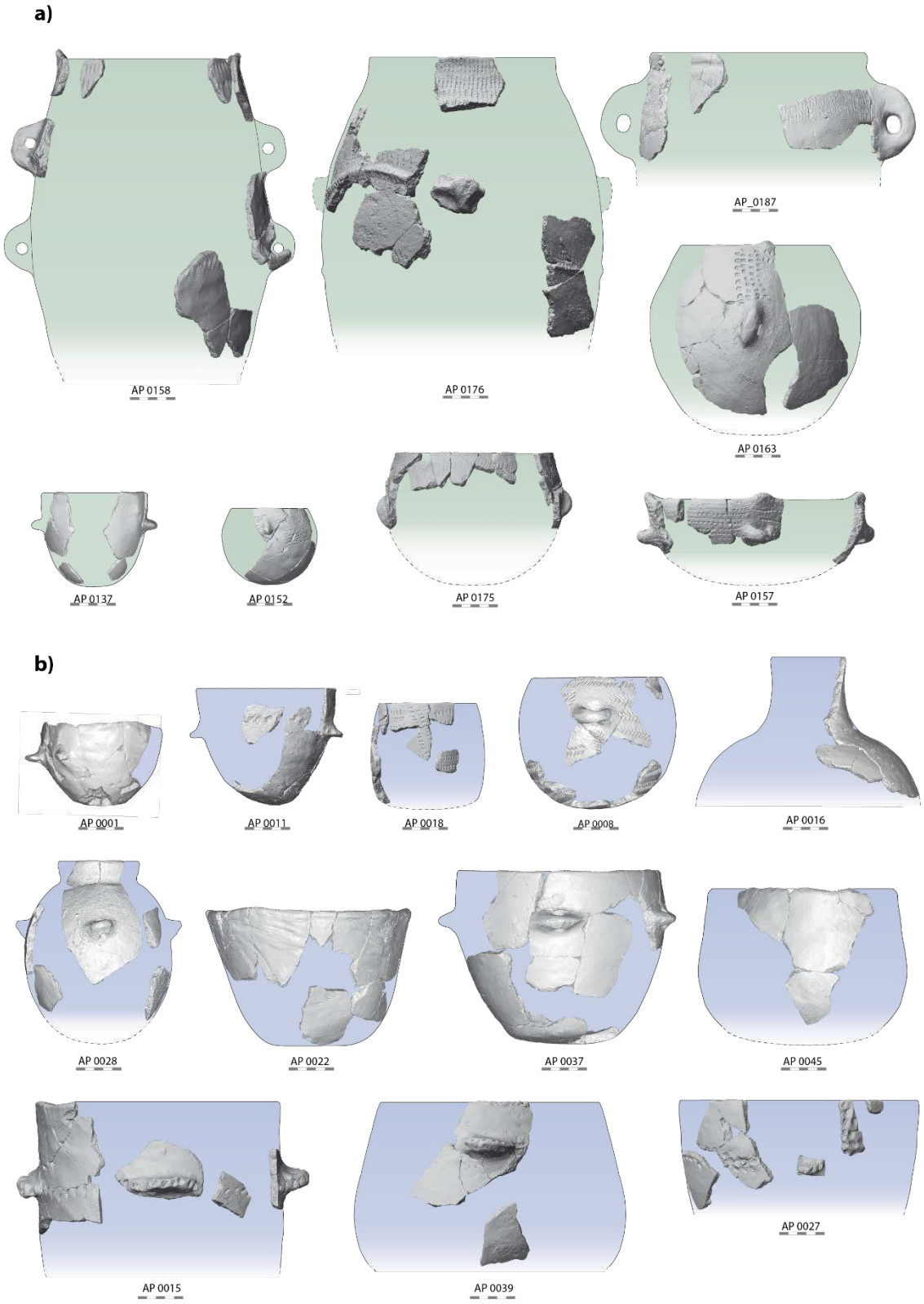

Figure 2: Examples of pottery shapes from Pendimoun rock-shelter. a) Cardial phase; b) Impressa phase. Reconstructions obtained by 3D scan.

Archaeological pottery sampling and capacity measurement

Ceramic samples obtained from fifty-two vessels and one foodcrust, adhering to the inside of a vessel, were investigated. Where possible, potsherds selected for analysis were taken from the upper part of the body of each vessel, which generally accumulates the most amount of lipids during cooking (Charters et al., 1993). Since all the reconstructed pots and significant isolated sherds were 3D scanned for technological analysis, the destructive sampling for chemical analysis did not cause any loss of information regarding the form and decoration of the vessels.

The ceramics were scanned with a portable 3D scanner Artec Spider, and 3D model without texture was obtained for each potsherd. The reconstructions of the vessels - virtual reshaping and creation of a form model - were carried out with the Cyclone 3DR software. In order to calculate the capacity, the shape of the ceramic was reconstructed by revolution of the profile. The capacity of the vessel was calculated between the inner walls and a maximum capacity level, positioned on the rim. An automatic capacity calculation function was applied, and the result given in litres. Where the entire lower part of the vessels was missing, only the minimum volume was calculated.

Modern products

Single-compounds stable carbon isotopes analyses have been performed on modern cereals (wheat, barley and einkorn) and animal products (goat adipose fat and goat milk). These products, originating from the South of France and Sicily, were purchased in organic supermarkets or collected from producers (Table S2). The diet of the animals was strictly composed of C_3 plants (outdoor grazing, barley and alfalfa fodder), to be as close as possible to Neolithic animal diet (Roffet-Salque et al., 2016).

Organic residue analysis

Only one sample had charred residues on the surface, which was collected using a clean scalpel. The surfaces of the potsherds were removed using a mini-drill to eliminate any exogenous lipids and the potsherds were drilled to collect around 1g of ceramic powder. Part of the charred residue (around 200 mg) was crushed with a mortar and pestle. An internal standard (*n*-alkane C_{34}, 10 µg, 1mg mL^{-1} in *n*-hexane) was then added to each sample.

The extraction of the samples was performed using 4 mL of methanol to which 800 µL of sulphuric acid was added drop by drop. After sonication for 15 min, the samples were heated for 4 hours at 70°C. The samples were then centrifugated and extracted three times in hexane. The hexane solution containing the lipids were then evaporated and re-dissolved in 100 µL of hexane. A second internal standard (*n*-alkane C_{36}, 10 µg, 1 mg mL^{-1} in *n*-hexane) was added to each sample. An aliquot of this solution was kept for GC-C-IRMS analyses and another was derivatised with *N,O*-bis(trimethylsilyl)trifluoroacetamide (BSTFA, 1% trimethylchlorosilane; 70°C, 1h) for GC and GC/MS analyses.

Dichloromethane / methanol (DCM/MeOH) extractions were performed on specific samples to check for the presence of triacylglycerols (TAGs), wax esters and alkylresorcinols. A second sample (1 g) was obtained for this purpose. After drilling, and the addition of 10 µg of *n*-alkane C_{34}, the samples were extracted in 10 mL of a DCM/MeOH solution (2:1, v/v) with ultrasonication. The samples were centrifuged (10 minutes at 3000 rpm) and the supernatants were collected. These operations were repeated twice more. The combined lipid extracts were

evaporated under nitrogen flow and dissolved in 100 µL of DCM/MeOH, before derivatisation with BSTFA. The lipid extracts were analysed on GC-FID and GC/MS after the addition of 10 µg of *n*-C_{36}.

GC-FID analyses were performed on an Agilent 7890B chromatograph equipped with a flame ionization detector (FID). After injection via split/splitless injector in splitless mode, the molecular compounds were separated into a 15 m DB-1HT (Agilent) apolar capillary column with an internal diameter of 0.32 mm and a phase thickness of 0.1 µm. The temperature program was as follow: 2 min isothermal at 50°C followed by a gradient increase to 375°C at 10°C/min, followed by 10 min at 375°C.

GC/MS analysis were performed on an Agilent Technologies 7890A device chromatograph coupled to an Agilent 5977B mass spectrometer. The separation was performed through a high temperature non-polar column (DB5-HT, 30 m x 0.25 mm i.d., 0.1 µm film thickness, Agilent J&W), after injection with a splitless injector. The temperature was set at 50°C for 2 min before an increase to 325°C at 10°C min^{-1}, and a final isothermal hold for 15 min. The mass spectrometer was running in electron ionization mode (EI, 70 eV) and mass spectra acquired over the range *m/z* 50–1000. Additional analyses were performed at high temperature on specific solvent extract (DCM/MeOH extractions) using the same column but with a temperature program set as follow: 50°C for 2 min before an increase to 375°C at 10°C min^{-1}, and a final isothermal hold for 12.5 min.

Additionally, samples were analysed on a DB-23 (50%-cyanopropyl)-methylpolysiloxane column (60m × 0.250mm × 0.25mm; J&WScientific) in Selected Ion Monitoring (SIM) mode to identify aquatic markers (isoprenoid fatty acids and *ω*-(*o*-alkylphenyl)alkanoic acids; Cramp and Evershed, 2014; Lucquin et al., 2016). The temperature program was set as follow: 50°C for 2 min and increase to 100°C at 10°C min^{-1}, increase to 140°C at 4°C min^{-1}, increase to 160°C at 0.5°C min^{-1}, increase to 250°C at 20°C min^{-1}, and a final isothermal hold for 10 min. The SIM mode was set to detect *4,8,12*-trimethyltridecanoic acid (*4,8,12*-TMTD) fragmentation (ions *m/z* 74, 87, 213, 270), pristanic acid (ions *m/z* 74, 88, 101, 312), phytanic acid (ions *m/z* 74, 101, 171, 326), and C_{16}- to C_{22}-ω-(*o*-alkylphenyl)alkanoic acids (ions *m/z* 74, 105, 262, 290, 318, 346). Helium was used as the carrier gas with a flow rate of 2.4 mL/min. The percentage of phytanic acid diastereoisomer SSR was obtained using the area of the ion *m/z* 101.

The well-preserved samples (more than 10 µg/g lipid) were analysed by GC-C-IRMS, after removal of sulphur, where present (Blumer, 1957). These analyses were performed using a Trace Ultra gas chromatograph (Thermo Fisher) with a GC Isolink II interface (Cu/Ni combustion reactor held at 1000°C; Thermo Fisher) coupled to a Delta V Advantage isotope ratio mass spectrometer (Thermo Fisher, Bremen, Germany). The carrier gas was ultrahigh-purity-grade helium with a flow rate of 2mL/min. 1 µL of each sample was injected into a DB-5MS fused-silica column (60m × 0.25mm× 0.25 µm; J&W Scientific). The temperature program was as follow: 0.5 min isothermal at 50°C followed by a gradient increase to 175°C at 25°C min^{-1}, then to 325°C at 8°C min^{-1}. Temperature was finally held for 20 min at 325°C. After ionization of the eluted compounds by electron impact (70°C), the $^{13}C/^{12}C$ ratio of each peak was automatically obtained by measuring the ionic intensities of *m/z* 44, 45 and 46. Computations were based on comparisons with repeated measurements a standard reference gas (CO_2). The results of the analysis were reported in per mile (‰) relative to the Vienna Pee

Dee belemnite (VPDB) international standard. For each batch of samples, a curve of expected vs. measured $\delta^{13}C$ values (international standards, Indiana A6 and F8-3 mixture) was used for calibration (average R^2 = 0.999 ± 0.001 in 6 batches). Control of the accuracy of the instrument was based on *n*-alkanoic acid ester standards of known isotopic composition and the precision on a laboratory standard mixture that was injected regularly between samples. Stable carbon isotope measurements were performed on the two main fatty acids in archaeological extracts: palmitic ($C_{16:0}$) and stearic ($C_{18:0}$) acid methyl esters. Values were corrected subsequent to analysis to account for the methylation of the carboxyl group using a mass balance formula. The $\Delta^{13}C$ value ($\delta^{13}C_{18:0}$ - $\delta^{13}C_{16:0}$) was calculated to overcome the influence of animal feed and the environment on stable carbon isotope values and to facilitate the identification of dairy products, ruminant and non-ruminant adipose fats (Evershed et al., 2002). The archaeological data were compared with the values obtained for modern animal fats published in the literature (Cramp and Evershed, 2014; Debono Spiteri, 2012; Dudd, 1999; Outram et al., 2009; Pääkkönen et al., 2020; Taché and Craig, 2015) and the data obtained in this study (cereals and animal fats, Table S2). The ^{13}C change in the atmosphere due to post-industrial carbon was mitigated by correcting the measured values on modern samples base on the estimated harvest and death date (Hellevang & Aagaard, 2015).

3. Results

Forty-six of the 52 pottery samples (88%) yielded enough lipids (> 10 µg/g) to be interpreted. Fatty acids (palmitic, $C_{16:0}$ and stearic, $C_{18:0}$), probably from animal fats were the main compounds extracted, generally in similar amounts ($C_{16:0}/C_{18:0}$ was between 1 and 1.5 in more than half of the samples; Figure 3). Small amounts of $C_{16:1}$ and $C_{18:1}$ were also present in many samples. Eight samples (AP_0002, AP_0011, AP_0015, AP_0029, AP_0037, AP_0039, AP_0053 and AP_0164) differed from the others in their high amounts of $C_{16:0}$ and sometimes $C_{18:1}$ ($C_{16:0}/C_{18:0}$ >2.0 and 2.5> $C_{18:1}/C_{18:0}$>0.2; Figure 3b), which suggest the presence of a plant oils rather than animal fats. Shorter fatty acids such as $C_{12:0}$ and $C_{14:0}$ were also sometimes detected. Linear and branched isomers of odd-carbon-numbered fatty acids ($C_{13:0}$ to $C_{19:0}$), derived from bacteria, potentially from the rumen of ruminants (Christie, 1978), were present in 15 samples (Figure 3a). Long chain fatty acids ($C_{20:0}$ to $C_{32:0}$), related to plant and insect waxes (Dunne et al., 2016; Eglinton & Hamilton, 1967; Roffet-Salque et al., 2015), and some of their unsaturated homologues ($C_{20:1}$ and $C_{22:1}$) were detected. AP_0011 displayed large amounts of $C_{24:0}$ (major long chain fatty acid), $C_{26:0}$, and $C_{28:0}$ (Figure 3d) and the long chain fatty acids of AP_0158 were dominated by $C_{26:0}$ (Figure 3c). Cholesterol (mainly occurring in animal products) and its derivatives (methyl cholesterol, cholesta-3,5-diene, cholest-2-ene) were present in some of the samples.

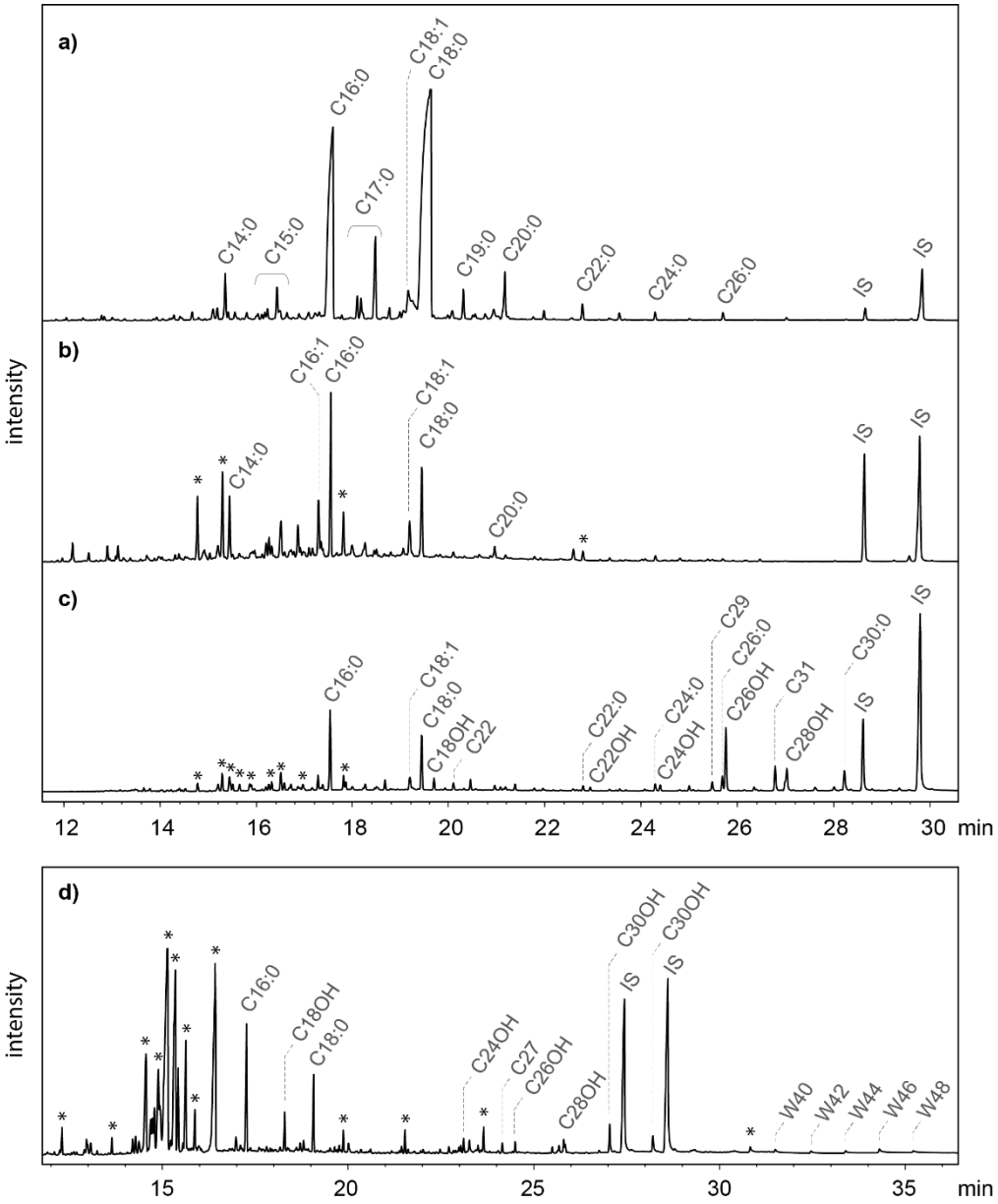

*Figure 3: Chromatograms representative of the substances identified in the pottery of the Pendimoun rock-shelter. a) ruminant adipose fat (AP_0175), b) plant oil (AP_0037) ; c) plant wax (AP_0158) ; d) beeswax and unidentified fat (AP_0011, solvent extraction). Cxx:x: fatty acids; CxxOH: linear alcohols; Cxx: linear alkanes; Wxx: wax esters; IS: internal standard; *: modern contamination.*

Isoprenoid fatty acids, from phytol transformation in ruminants and aquatic organisms (Lucquin et al., 2016), were identified in 19 samples. *4,8,12*-TMTD is detected in 15 samples and phytanic and pristanic acid in four samples (AP_0008, AP_0036, AP_0157, and AP_0175; Figure 4). The ratio of the two isomers of phytanic acid varied from 45 to 76% of SSR diastereoisomer. Markers of thermal transformation of animal fats (Bondetti et al., 2020; Hansel et al., 2004; Raven et al., 1997) were also detected occasionally. Traces of ω-(*o*-alkylphenyl)alkanoic acids (APAAs) with 18 carbon atoms were detected in 3 samples (AP_0008, AP_0027, and AP_0036; Figure 4) but no C_{20}-APAAS. Three mid-chain ketones (K_{31}, K_{33} and K_{35}) were detected in the foodcrust (AP_0176r).

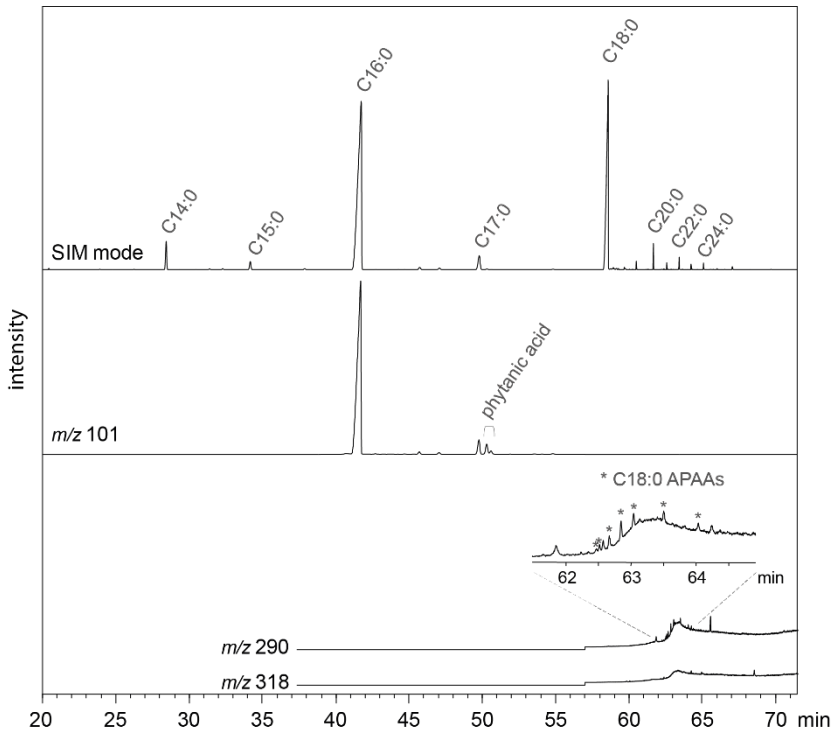

Figure 4: Example of a chromatogram obtained in SIM mode for the search for aquatic and ruminant biomarkers (AP_0008). The m/z 101 ion is used to indicate the presence of phytanic acid and the m/z 290 and 318 ions, C18- and C20-APAAs respectively.

Alkanes were generally present in small amounts, as a homologous series of even and odd-numbered chains of carbon atoms (centred around C_{22}). This profile remains difficult to interpret and contamination cannot be excluded. However, four samples had different alkane profiles: AP_0001, AP_0036, and AP_0158 had odd-numbered carbon-dominated alkane profiles dominated by C_{31}, and AP_0011 by C_{27} (Figure 3c and d), indicating the presence of waxy products (Dunne et al., 2016; Eglinton & Hamilton, 1967; Roffet-Salque et al., 2015). AP_0011 also yielded linear alcohols in significant amounts, dominated by even-carbon-numbered long chains (max $C_{30}OH$). The other samples exhibited low amounts of a homologous series of linear alcohols with even- and odd-carbon-numbered chains centred on $C_{18}OH$, possibly resulting from environmental contamination. High temperature chromatographic analyses (GC and GC-MS) revealed the presence of wax esters (palmitic acid wax esters, W_{40} to W_{48}) in AP_0011 (Figure 3d) and small amounts of palmitoleic and myristic acid wax esters in several other samples. Diterpenes (methyl-dehydroabietic, methyl-didehydroabietic and methyl 7-oxo-dehydroabietic acids and phenanthrene), originating from conifer exudates (Mills & White, 1977), were also present in very small amounts (0.1 to 1.1 µg/g) in 16 samples. Due to their low amount and the absence of pine in the archaeological record (Battentier et al., 2015), this presence is at the moment difficult to explain and these compounds will not be further discussed.

Many samples yielded significant amounts of phthalates (mass spectrum characterised by the *m/z* 149 ion) and benzoic acid derivatives (*m/z* 105 and 123 ions), revealing contamination by plastic products, and sometimes small amounts of squalene from the handling of the potsherds. Molecular sulphur was detected in variable amounts in 25 samples.

Stable carbon isotope measurements of $C_{16:0}$ and $C_{18:0}$ gave values mostly in the range of -30 to -28‰ ($C_{16:0}$) and -31 to -29‰ ($C_{18:0}$) and an offset around -1‰ (Figure 5). Three samples (AP_0001, AP_0022 and AP_0028) had fatty acids more enriched in ^{13}C (offset between 0 and 1‰) and one sample has a large $\Delta^{13}C$ offset (-5‰; AP_0157).

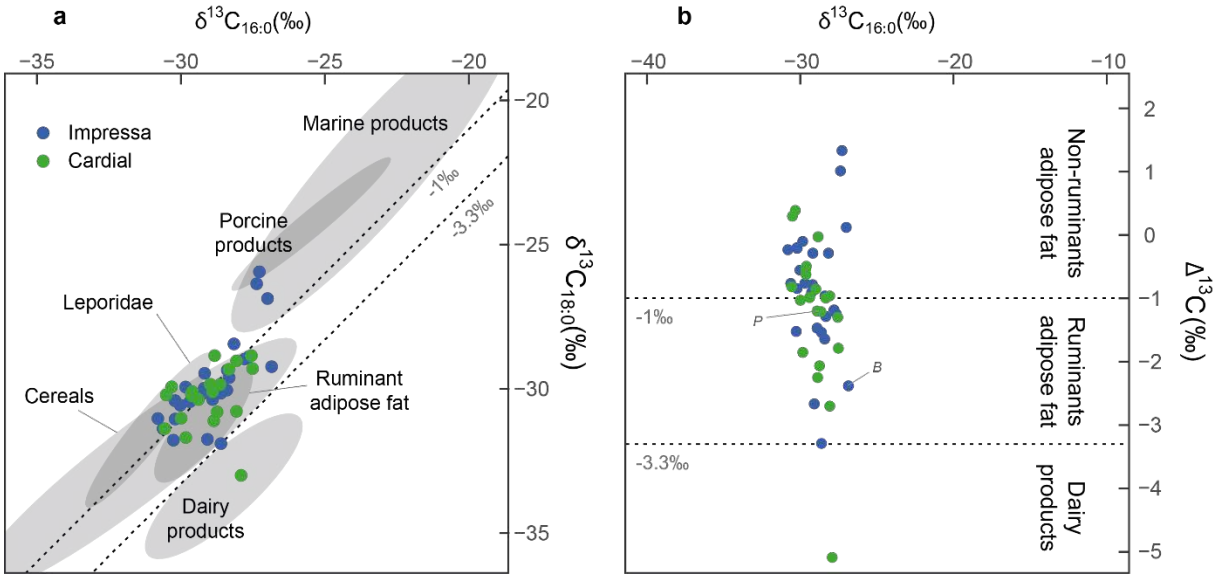

Figure 5: a) $\delta^{13}C$ values of fatty acids ($C_{16:0}$ and $C_{18:0}$) in pottery from the Pendimoun rock-shelter. Ellipses (95% confidence) are calculated from modern reference substances from the UK, Finland, Russia, Canada, Kazakhstan, Italy and Southern France (Cramp and Evershed, 2014; Debono Spiteri, 2012; Dudd, 1999; Outram et al., 2009; Pääkkönen et al., 2020; Taché and Craig, 2015; and this study). b) Plot of the $\Delta^{13}C$ (difference between the $\delta^{13}C$ values of the $C_{16:0}$ and $C_{18:0}$ fatty acids) and $\delta^{13}C_{16:0}$. B: beeswax; P: plant epicuticular wax.

4. Discussion

4.1. Identification of products contained in Pendimoun rock-shelter ceramics

Molecular analyses of the content of Pendimoun pottery show a predominance of animal fats, in particular from ruminants (largely predominant saturated fatty acids, linear and branched odd carbon-numbered fatty acids, cholesterol and derivatives, %SSR<76%; Figure 3a; Lucquin et al., 2016; Regert, 2011). These results are consistent with analyses of faunal assemblages that revealed the predominance of domestic sheep (Binder et al., 2020; Rowley-Conwy et al., 2013). Isotopic values also support the identification of ruminant adipose fats in many samples (-3.3‰ < $\Delta^{13}C$ < -1‰). The ^{13}C values of many other samples are close or higher to the limit between ruminant and non-ruminant adipose fat, but too depleted to be interpreted as porcine fats ($\Delta^{13}C$ > -1‰; $\delta^{13}C_{16:0}$ around -30‰; Figure 5). This shift could be due to the presence of freshwater fats, but neither molecular data nor faunal remains support this hypothesis. The presence of isoprenoids is not sufficient to formally identify fats of aquatic origin, as these lipids also exist in ruminant fats (Lucquin et al., 2016). When phytanic acid is detected in Pendimoun samples, the % of the SSR isomer corresponds to ruminant fat values (Figure 4; Lucquin et al., 2016). In addition, there is no evidence of C_{20}-APAAs that are characteristic of aquatic products (Cramp and Evershed, 2014). Another potential source are wild non-ruminants (e.g. hare) or cereals, both well attested at the site (Binder et al., 2020).

The molecular profile, mainly composed of $C_{16:0}$ and $C_{18:0}$ in similar amounts, is consistent with both animal fats or degraded cereal lipids (Debono Spiteri, 2012, p. 168). The absence of alkylresorcinols in all samples extracted with DCM/MeOH is not a criterion to rule out the presence of cereals because these molecular markers have shown that they were very poorly preserved in aerobic contexts (Hammann and Cramp, 2018).

Isotopic data indicated the presence of dairy products in a single vessel (AP_0157, $\Delta^{13}C < -3.3$ ‰; Figure 5; Regert, 2011). Three containers (AP_0001, AP_0022 and AP_0028) had ^{13}C-enriched fatty acids, which indicates the presence of either porcine fats or marine products ($\Delta^{13}C < -3.3$‰, $\delta^{13}C_{16:0} > -28$‰; Figure 5). The absence of marine molecular markers in these samples (the three isoprenoids together and APAAs; Cramp and Evershed, 2014) suggests the exploitation of porcine fats. This finding correlates with the faunal assemblage that contained either wild or domesticated pigs (Binder et al., 2020; Rowley-Conwy et al., 2013).

The presence of large amounts of $C_{16:0}$ and $C_{18:1}$ in 8 samples suggests the presence of plant oils (Figure 3b). Hazelnuts or acorns, identified in carpological assemblages (Binder et al., 2020), are potential candidates to explain such molecular signal. The presence of these oils may be due to a deliberate oil extraction or to cooking episodes, with or without other products. As solvent extraction of some of these samples also revealed the presence of $C_{14:0}$ and $C_{16:1}$ wax esters (AP_0002, AP_0029, AP_0037), used in the cosmetics industry (e.g. Takekoshi et al., 2002), modern contamination of these samples cannot be ruled out. One vessel (AP_0158, Figure 3c) shows the exploitation of leafy plants during the Cardial, through the presence of C_{29}, C_{31} and C_{33} alkanes and long-chain fatty acids dominated by $C_{26:0}$ (Dunne et al., 2016; Eglinton & Hamilton, 1967).

Sample AP_0011 shows a very clear beeswax profile ($C_{16:0}$ wax esters, long-chain alkanes dominated by C_{27} and long-chain alcohols dominated by $C_{30}OH$, large amount of $C_{16:0}$; Figure 3d; Roffet-Salque et al., 2015). The presence of beeswax in an Impressa pot demonstrates the long history of the exploitation of beehive products in the North-western Mediterranean, which had not been identified until now (Roffet-Salque et al., 2015). The presence of $C_{18:0}$ in the same sample suggests that the pots have contained a fatty product, mixed with the beeswax or used prior to or after the beeswax.

4.2. Correspondence between vessel shape and contents

Comparison of vessel capacity with lipid extraction yield (Figure 6b) shows a distinction between small capacity pots (between 1 and 2 L) with high lipid yields (> 200 µg/g) and very large vessels (V > 6 L) with very low lipid yields (between 8 and 22 µg/g). Differential preservation does not seem to be the cause of such a difference, as the lipid concentration shows no correlation with the period (Impressa or Cardial), the location of the potsherds within the site, or the type of ceramic paste. These two groups could correspond respectively to cooking pots and storage/serving vessels. However, AP_0028, one of the small lipid-rich vessels with a closed a shape would seem to be unsuitable for use on a fire (e.g. Rice, 1987, p. 237-240). The difference in lipid yields may be due to the different contents of these vessels. The molecular profile and isotope values of the small lipid-rich vessels clearly point to animal fats (ruminant, porcine or mixtures, depending on the pots; Figure 6a). In contrast, one of the largest vessel (AP_0158) had a clear signal of plant epicuticular waxes and the molecular and/or isotopic data from the other large pots also suggests a plant origin (large dominance of

$C_{16:0}$ over $C_{18:0}$ and/or isotope values in the cereal range; Figure 6a). Previous experiments demonstrate that the accumulation of animal fats from boiling meat in a single episode is over 150 times that from repeated boiling of leafy vegetables (Evershed, 2008). The difference observed in concentrations may therefore reflect their use, although a difference in the intensity of use of the pots cannot be ruled out.

Small vessels may have served as cooking pots for a single, or limited number of individuals (e.g. household or family group; Smith, 1988; Henrickson and McDonald, 1983; Tsirtsoni, 2001) or may reflect specialised use for processing and storage of animal fats (such as lard or tallow). The latter is perhaps most relevant for AP_0028 that also contained red ochre (Pradeau, 2015, p. 270-274). Although reuse cannot be excluded, it is possible that the pot was used to store a mixture of porcine adipose fat and ochre. The co-presence of ochre and animal fat (ruminant adipose fat) has also been identified in another vessel at the site (AP_0163; Pradeau, 2015, p. 270-274). Animal fats could be added to the ochre to facilitate its pliability allowing manipulation for technical, decorative or symbolic activities. Such a preparation could be used, for example, for the finishing treatment or maintenance of leathers, for body care and decoration (Audouin & Plisson, 1982; Rifkin, 2011), or for colouring some objects (such as the sculpted and coloured face discovered at the site; Binder et al., 2014). In comparison, the very large volume of some of the vessels that contained plant products (> 9 L) suggests use for a larger group than the household. The diversity of shapes indicates that these vessels do not constitute a single functional group but may have been used for cooking and storage of plant substances, especially the bottle (AP_0016) that might have been used to store or serve a plant- or cereal-based liquid. In addition, it is interesting to note that the only pot that unambiguously yielded dairy products is also the shallowest and most open of the vessels tested (AP_0157; Figure 6a). Such a form, also associated with dairy products during the Middle Neolithic period in the Jura (Drieu et al., 2020), does not evoke a use as a cooking pot. This vessel could have been employed for milk processing (e.g. for recovering filtered whey during cheesemaking, as has been suggested for Neolithic bowls in Poland; Salque et al., 2013), or for serving and consuming dairy products.

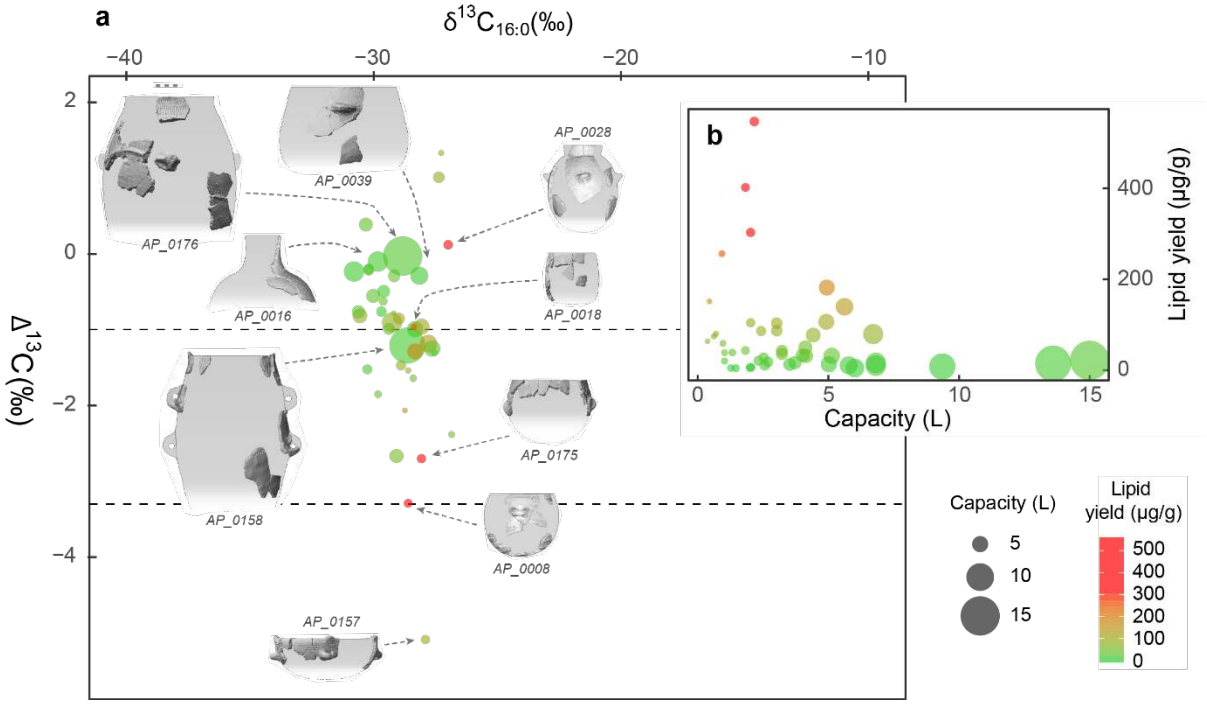

Figure 6: a) Plot of the $\Delta^{13}C$ and $\delta^{13}C_{16:0}$ representing samples as a function of vessel capacity and lipid yield. The shape of the vessels discussed in the text is displayed; b) Plot of vessel capacity as a function of the amount of lipids extracted. Included are drawings of vessels with specific volume/form/organic content relationships discussed in the text.

4.3. A distinctive subsistence economy?

The absence of aquatic products in the ceramics of Pendimoun is in-keeping with a terrestrial subsistence economy, similar to other Neolithic sites of the North-western Mediterranean (Debono Spiteri et al., 2016; Tarifa-Mateo et al., 2019). This suggests in particular that the shellfish found at the site were not collected to be processed in ceramic vessels and that terrestrial resources, particularly animal fats, constituted the major part of the everyday food prepared in pottery. This pattern, supported by analyses of bone collagen (Goude et al., 2011; Le Bras-Goude et al., 2010), shows that fish, shellfish and marine molluscs were not important dietary components although we cannot rule out low-level consumption and preparation techniques that did not involve ceramics.

Even if domestic ruminants were an important part of the diet, it should be noted that the exploitation of wild ruminants in ceramics, such as deer, identified in small quantities at the site (Binder et al., 2020; Rowley-Conwy et al., 2013), cannot be completely excluded from use in pottery as these cannot be formally identified using molecular markers or single compound stable carbon isotopes (Craig et al., 2012). Similarly, the status of pigs, domestic or wild, found in faunal assemblages is still undetermined (Binder et al., 2020; Rowley-Conwy et al., 2013). The potential contribution of hunting activities, a possible mark of Mesolithic way of life or interaction with Mesolithic groups, cannot currently be discussed in more detail.

Although the use of perishable containers to process and consume the milk cannot be excluded, the lack of pottery with a clear dairy product signal in Pendimoun (only 1 sample,

accounting for 2% of the interpretable samples, and dated from the end of the Impresso-Cardial phase) is in agreement with the archaeozoological data which suggests that the management of the domestic animals (mainly sheep) was oriented towards the exploitation of the meat (Rowley-Conwy et al., 2013). A different pattern seems to emerge from the analysis of pottery from other early Impresso-Cardial sites that delivered enough interpretable results (i.e. more than a few pots with enough lipids to be interpreted). These sites, located in Central and Southern Italy, on the eastern side of the Apennines, yielded a higher proportion of vessels dedicated to dairying (44% in Fondo Azzollini and 23% in Colle Santo Stefano; Debono Spiteri et al., 2016; Salque et al., 2012). The different extraction method that was used in these studies (DCM/MeOH extraction, which generally results in lower extraction yields; Correa-Ascencio & Evershed, 2013) may account for the difference in results, as dairy might yield higher amounts of lipids and therefore have a better chance of being detected using these approaches. However, archaeozoological analyses at Colle Santo Stefano also suggest the exploitation of dairy products (Debono-Spiteri et al., 2016).

Archaeobotanical and sediments analyses demonstrate that the Pendimoun rock-shelter has been used for penning small domestic ruminants (Battentier et al., 2015; Binder et al., 1993; Delhon et al., 2020). The scarcity of pottery specifically dedicated to the processing of dairy products at the site might be related to a complex system of land use, where the rock-shelter was occupied when the animals were not in lactation. Analysis of the phytoliths reveals that Pendimoun was mainly occupied from late spring to early autumn (Delhon et al., 2020). Stable oxygen isotope composition of enamel of sheep teeth suggests that, during the Neolithic, sheep birth generally occurred throughout the spring in Western and South-eastern Europe (Balasse et al., 2017). Because the lactation period is of a few months (at least 4 months; Haenlein, 2007), fresh milk would potentially have been available from late winter/early spring until the end of summer. Assuming that the scarcity of dairy products in the Pendimoun pottery is due to a specific occupation season, the combination of archaeobotanical, archaeozoological and organic residue analyses would suggest occupation of the Pendimoun rock-shelter only in early autumn. It cannot be excluded that, in addition to spring lambing, the Pendimoun breeders achieved autumn lambing, as at other Cardial and Epicardial sites in the south of France (Tornero et al., 2020). According to this hypothesis, the occupation window of the rock-shelter would have been very short, between the end of summer and the first autumn births. Assuming that the milk was exploited at other sites, as part of a complex seasonal exploitation of the territory involving transhumance, the vessels used to store and process the milk can be supposed to have been left on site. Such a hypothesis does not preclude possible transport and consumption of dairy products in Pendimoun (in the form of cheeses, for example, which are easier to transport and preserve). On the contrary, Fondo Azzollini and Colle Santo Stefano are open-air sites, which could have been occupied for a longer duration.

The distinction in dairy product exploitation between Impresso-Cardial sites has an interesting parallel in that pottery manufacturing methods also differ between the Pendimoun rock-shelter and Colle Santo Stefano (Gomart et al., 2017), perhaps indicating that culinary and cultural traditions were closely linked, and each associated with a different pioneer colonisation process. Possible avenues for studying Neolithic expansion routes from the culinary traditions can be found in the comparison with the first phases of the Neolithic in Northern Greece and on the Atlantic coast of the Iberian Peninsula. These two regions, for which a large amount of data on the contents of pottery is available, also appear to be centred on the exploitation of meat rather than dairy products (Cubas et al., 2020; Whelton et al., 2017).

Clearer evidence, both molecular and archaeozoological, for the exploitation of milk is seen during the Late Impresso-Cardial in Southern France and North-eastern Spain (Debono-Spiteri et al., 2016), representing a potential shift in the subsistence economy towards intensification in the exploitation of dairy products. Interestingly, while goats are absent from the early phases of occupation in Pendimoun, their appearance during the Cardial period is concomitant with the only ceramic vessel providing clear evidence of the exploitation of dairy products. The hypothesis that goats were exploited for their milk from the Cardial period is tempting, but studies on a larger corpus will have to be set up to verify whether milk exploitation really constitutes a turning point starting from the Cardial period.

5. Conclusion

Analysis of the contents of the Early Neolithic ceramics from the Pendimoun rock-shelter demonstrated, on the one hand, that the pottery was mainly used to process and consume livestock products, particularly ruminants. The fish and shellfish do not appear to have been prepared in pottery, but a contribution of hunting game, such as wild boar, hare or deer, cannot be ruled out. The lack of pottery with a clear dairy signal during the earliest occupation phase of the site contrasts with findings from Southern Italy at the origin of the Western Mediterranean farming dispersal. Therefore, the absence, or scarcity, of dairy at the first step of this spread may suggest the loss of some aspects of the original farming package. However, the seasonal occupation of the Pendimoun rock-shelter during non-lactation periods could also explain this absence, as opposed to open-air sites, which are potentially occupied year-round. This study highlights that the Neolithic "package" varies from one site to another, calling for future investigations to be undertaken at a regional scale in order to more accurately reflect the true diversity of culinary practices by Europe's first farming communities (Binder et al., 1991). With regard to these questions and their consequences for our perception of the economic and social changes induced by the spread of agriculture in Europe, biomolecular studies should be undertaken on a wider geographical scale, in the light of the detailed chronology recently published (Binder et al., 2017; Manen et al., 2019).

Acknowledgement

This work is carried out within the framework of the ANR CIMO (ANR-14-CE31-009). We thank Pierre Pradier and Egidio Gonzales for providing us with the modern samples of cereals and animal fats. We are grateful to Marie Balasse and Juliette Knockaert for the fruitful discussion on the periods of birth of caprines and lactation season during the Neolithic. Finally, we would like to thank the two reviewers for their constructive suggestions which helped to improve the quality of this article.

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Supplementary information

Table S1: List of analysed vessels, period, type of earth and main information from the organic residue analysis. FA: fatty acids; 4,8,12-TMTD: 4,8,12-Trimethyltridecanoic acid.

Table S2: List of modern cereals and stable carbon isotope values measured on palmitic ($C_{16:0}$) and stearic ($C_{18:0}$) acids.

Sample name	Period	Earth type	Minimum volume (L)	Lipid yield ($\mu g\ g^{-1}$)	C16:0/C18:0	C18:1/C18:0	other biomarkers	$\delta^{13}C_{16:0}$ (‰)	$\delta^{13}C_{18:0}$ (‰)	$\Delta^{13}C$ (‰) ($\delta^{13}C_{18:0}$-$\delta^{13}C_{16:0}$)	Interpretation of vessel contents
AP_0001	Impressa	Granitic earth	0,6	75,04	0,96	0,18	long chain FA (C20:0-C25:0)	-27,27	-25,94	1,33	Porcine adipose fat
AP_0002	Impressa	Mixed earth	3,2	36,27	2,12	1,04	long chain FA (C20:0-C26:0), cholesterol	-29,18	-29,46	-0,29	Plant product?
AP_0003	Impressa	Glauconitic earth	6,8	10,59	0,94	0,18	long chain FA (C20:0-C26:0)	-30,80	-31,03	-0,23	Undetermine fat (wild non ruminant or cereals). See Discussion part
AP_0004	Impressa	Glauconitic earth	2,4	86,77	1,22	0,23	long chain FA (C20:0-C26:0)	-28,90	-30,37	-1,47	Ruminant adipose fat
AP_0007	Impressa	Glauconitic earth	0,4	64,26	1,15	0,20	long chain FA (C20:0-C24:0), cholesterol	-29,19	-29,98	-0,78	Undetermine fat (wild non ruminant or cereals). See Discussion part
AP_0008	Impressa	Granitic earth	1,8	402,12	0,84	0,13	long chain FA (C20:0-C24:0), linear and branched odd-numbered FA (C15:0 and C17:0), C18:0 APAAs, 4,8,12-TMTD, pristanic and phytanic acid (76% SSR)	-28,61	-31,90	-3,29	Ruminant adipose fat
AP_0011	Impressa	Granitic earth	0,9	59,78	4,51	0,46	long chain FA (C20:0-C32:0), odd-numbered alkanes (C27-C33), linear alcohols (C24OH-C34OH), wax esters (W40-W48)	-26,85	-29,23	-2,38	Beeswax and animal adipose fat
AP_0014	Impressa	Mixed earth	6,0	5,48	1,73	0,27		n.d.	n.d.	n.d.	Low amount of lipid
AP_0015	Impressa	Mixed earth	9,4	8,95	2,67	0,37		n.d.	n.d.	n.d.	Plant product?
AP_0016	Impressa	Mixed earth	1,9	17,03	1,13	0,13	long chain FA (C20:0)	-29,84	-29,94	-0,10	Undetermine fat (wild non ruminant or cereals). See Discussion part
AP_0018	Impressa	Granitic earth	0,9	256,65	0,25	0,09	long chain FA (C20:0-C26:0), linear and branched odd-numbered FA (C15:0 and C17:0)	-28,39	-29,35	-0,96	Undetermine fat (wild non ruminant or cereals). See Discussion part
AP_0019	Impressa	Glauconitic earth	2,5	28,15	1,09	0,09	4,8,12-TMTD (tr.)	-30,20	-30,41	-0,21	Undetermine fat (wild non ruminant or cereals). See Discussion part
AP_0020	Impressa	Glauconitic earth	2,7	18,94	0,82	0,16	long chain FA (C20:0)	-30,21	-30,41	-0,20	Undetermine fat (wild non ruminant or cereals). See Discussion part
AP_0022	Impressa	Granitic earth	3,0	87,62	1,10	0,18	long chain FA (C20:0-C24:0)	-27,37	-26,36	1,01	Porcine adipose fat
AP_0027	Impressa	Mixed earth	5,6	140,14	0,91	0,17	long chain FA (C20:0-C22:0), C18:0 APAAs, 4,8,12-TMTD (tr.) and	-27,79	-28,97	-1,18	Ruminant adipose fat
AP_0028	Impressa	Granitic earth	2,1	546,88	1,25	0,43	long chain FA (C20:0-C26:0)	-26,99	-26,87	0,12	Porcine adipose fat
AP_0029	Impressa	Mixed earth with chamotte?	1,0	39,22	3,93	2,40	long chain FA (C20:0-C24:0)	-28,41	-30,05	-1,64	Ruminant adipose fat, plant product?
AP_0030	Impressa	Glauconitic earth	4,1	31,94	0,59	0,10	long chain FA (C20:0-C28:0), phytanic acid (tr.)	-30,02	-30,57	-0,55	Undetermine fat (wild non ruminant or cereals). See Discussion part
AP_0036	Impressa	Mixed earth	4,1	50,59	0,99	0,13	long chain FA (C20:0-C26:0), C18:0 APAAs, 4,8,12-TMTD, pristanic	-29,08	-31,75	-2,67	Ruminant adipose fat
AP_0037	Impressa	Glauconitic earth	5,1	32,22	2,06	0,51	long chain FA (C20:0-C24:0), 4,8,12-TMTD (tr.), cholesterol	-27,64	-28,89	-1,25	Ruminant adipose fat, plant product?
AP_0039	Impressa	Granitic earth	5,8	11,45	2,22	0,44		-28,16	-28,44	-0,29	Plant product?
AP_0042	Impressa	Glauconitic earth	3,7	17,38	1,48	0,16	long chain FA (C20:0-C26:0), cholesterol	-30,62	-31,38	-0,76	Undetermine fat (wild non ruminant or cereals). See Discussion part
AP_0045	Impressa	Granitic earth	4,6	182,17	0,57	0,17	long chain FA (C20:0-C24:0), linear and branched odd-numbered FA	-28,33	-29,61	-1,29	Ruminant adipose fat
AP_0046	Impressa	Glauconitic earth	1,2	5,60	1,70	0,26		n.d.	n.d.	n.d.	Low amount of lipid
AP_0052	Impressa	Glauconitic earth	2,3	21,57	1,49	0,47	long chain FA (C20:0-C26:0), cholesterol	-30,26	-31,78	-1,52	Ruminant adipose fat
AP_0053	Impressa	Glauconitic earth	2,5	11,52	2,40	1,01	long chain FA (C20:0-C30:0), 4,8,12-TMTD (tr.)	-29,69	-30,45	-0,76	Plant product?
AP_0081	Impressa	Granitic earth	n.d.	19,29	1,12	0,26	long chain FA (C20:0), 4,8,12-TMTD	-30,20	-31,05	-0,85	Undetermine fat (wild non ruminant or cereals). See Discussion part
AP_0137	Impressa	Mixed earth	0,7	80,68	1,37	0,49	long chain FA (C20:0-C26:0), 4,8,12-TMTD	-28,61	-30,15	-1,54	Ruminant adipose fat
AP_0038	Cardial (transition Impressa/Cardial)	Glauconitic earth	6,7	80,08	1,19	0,47	long chain FA (C20:0-C26:0), cholesterol	-29,26	-30,17	-0,91	Undetermine fat (wild non ruminant or cereals). See Discussion part
AP_0146	Cardial (transition Impressa/Cardial)	Glauconitic earth	4,4	77,82	1,10	0,25	long chain FA (C20:0-C26:0), 4,8,12-TMTD	-30,56	-31,38	-0,82	Undetermine fat (wild non ruminant or cereals). See Discussion part
AP_0040	Cardial	Glauconitic earth	1,4	5,21	2,79	0,00		n.d.	n.d.	n.d.	Low amount of lipid
AP_0148	Cardial	Glauconitic earth	2,0	7,00	1,23	0,11		n.d.	n.d.	n.d.	Low amount of lipid
AP_0152	Cardial	Glauconitic earth	0,4	151,70	1,85	0,24	long chain FA (C20:0-C26:0), 4,8,12-TMTD, cholesterol	-28,74	-30,80	-2,06	Ruminant adipose fat
AP_0157	Cardial	Glauconitic earth	2,0	104,91	1,03	0,08	long chain FA (C20:0-C26:0), linear and branched odd-numbered FA (C15:0 and C17:0), pristanic and phytanic acid (61% SSR)	-27,91	-33,00	-5,09	Dairy product
AP_0158	Cardial	Glauconitic earth	13,6	15,30	1,44	0,39	long chain FA (C20:0-C30:0), 4,8,12-TMTD, odd-numbered alkanes	-28,64	-29,85	-1,21	Ruminant adipose fat
AP_0163	Cardial	Glauconitic earth	4,9	176,17	0,55	0,03	long chain FA (C20:0-C26:0)	-28,07	-29,03	-0,96	Undetermine fat (wild non ruminant or cereals). See Discussion part
AP_0164	Cardial	Glauconitic earth	1,0	20,87	3,11	0,21		-27,55	-28,85	-1,30	Ruminant adipose fat, plant product?
AP_0165	Cardial	Glauconitic earth	3,2	43,24	1,26	0,19	long chain FA (C20:0-C22:0), 4,8,12-TMTD, cholesterol	-29,39	-30,37	-0,99	Undetermine fat (wild non ruminant or cereals). See Discussion part
AP_0166	Cardial	Glauconitic earth	3,5	13,25	1,33	0,29	long chain FA (C20:0-C26:0)	-29,60	-30,10	-0,49	Undetermine fat (wild non ruminant or cereals). See Discussion part
AP_0170	Cardial	Glauconitic earth	1,8	43,94	1,09	0,07	long chain FA (C20:0-C25:0)	-29,62	-30,24	-0,63	Undetermine fat (wild non ruminant or cereals). See Discussion part
AP_0171	Cardial	Glauconitic earth	n.d.	9,12	1,58	0,19	long chain FA (C20:0-C26:0), 4,8,12-TMTD	-28,86	-31,11	-2,25	Ruminant adipose fat
AP_0174	Cardial	Glauconitic earth	n.d.	34,14	1,08	0,03	long chain FA (C20:0-C22:0)	-29,64	-30,18	-0,54	Undetermine fat (wild non ruminant or cereals). See Discussion part
AP_0175	Cardial	Glauconitic earth	2,0	358,38	0,45	0,09	long chain FA (C20:0-C28:0), linear and branched odd-numbered FA (C15:0 and C17:0), pristanic and phytanic acid (46% SSR)	-28,07	-30,78	-2,70	Ruminant adipose fat
AP_0176	Cardial	Glauconitic earth	15,0	22,41	0,91	0,16	long chain FA (C20:0-C26:0)	-28,83	-28,85	-0,03	Undetermine fat (wild non ruminant or cereals). See Discussion part
AP_0176r	Cardial	Glauconitic earth	-	58,65	1,10	0,33	long chain FA (C20:0-C28:0), ketones (K31, K33, K35)	-27,51	-29,30	-1,79	Heated ruminant adipose fat
AP_0178	Cardial	Glauconitic earth	2,0	6,48	1,42	0,18	long chain FA (C20:0-C28:0)	n.d.	n.d.	n.d.	Low amount of lipid
AP_0180	Cardial	Glauconitic earth	1,3	39,57	1,63	0,26	long chain FA (C20:0-C28:0)	-29,83	-31,69	-1,85	Ruminant adipose fat
AP_0181	Cardial	Glauconitic earth	5,0	13,86	1,32	0,00		-28,33	-29,32	-0,99	Undetermine fat (wild non ruminant or cereals). See Discussion part
AP_0182	Cardial	Glauconitic earth	3,0	103,98	0,47	0,05	long chain FA (C20:0-C24:0)	-28,98	-29,84	-0,85	Undetermine fat (wild non ruminant or cereals). See Discussion part
AP_0187	Cardial	Glauconitic earth	4,0	33,75	1,36	0,12	long chain FA (C20:0-C24:6), 4,8,12-TMTD (tr.) and phytanic acid (tr.)	-30,32	-29,93	0,39	Undetermine fat (wild non ruminant or cereals). See Discussion part
AP_0206	Cardial	Granitic earth	n.d.	14,90	0,95	0,10	long chain FA (C20:0-C22:0)	-30,51	-30,21	0,30	Undetermine fat (wild non ruminant or cereals). See Discussion part
AP_0207	Cardial	Glauconitic earth	n.d.	66,23	0,44	0,12	long chain FA (C20:0-C26:0)	-29,99	-31,02	-1,03	Ruminant adipose fat
AP_0212	Cardial	Glauconitic earth	n.d.	11,37	1,34	0,33	4,8,12-TMTD, cholesterol	-28,90	-30,10	-1,20	Ruminant adipose fat

Sample name	Cereal	Provenance	Geographical origin	Date collected	$\delta^{13}C_{16:0}$ (‰)	$\delta^{13}C_{18:0}$ (‰)	$\Delta^{13}C$ (‰) ($\delta^{13}C_{18:0}$-$\delta^{13}C_{16:0}$)
YM0858	Wheat	Local producer (Azienda Agricola biologica San Luca)	Castronovo di Sicilia, Italy	2018	-29,6	-31,0	-1,4
YM0861	Barley	Local producer (Azienda Agricola biologica San Luca, Castronovo di Sicilia, Italy)	Castronovo di Sicilia, Italy	2018	-30,6	-31,2	-0,6
YM0868	Einkorn	Local organic supermarket	Provence, France	2019	-31,4	-32,9	-1,5
YM0869	Einkorn	Local organic supermarket	Provence, France	2019	-32,0	-33,00	-1,0
YM0870	Einkorn	Local organic supermarket	Provence, France	2019	-32,2	-33,9	-1,7
YM0871	Barley	Local organic supermarket	Provence, France	2019	-36,4	-35,6	0,8
YM0863	Goat	Local producer (Pierre Pradier)	Simiane-la-Rotonde, France	2016	-28,7	-31,1	-2,4
YM0867	Goat milk	Local producer (Pierre Pradier)	Simiane-la-Rotonde, France	2016	-26,8	-31,9	-5,1