Biomolecular and isotopic characterisation of lipid residues absorbed in Impressed Wares from the Early Neolithic village of Skorba, Malta

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Organic residue analysis (ORA) was used to characterise the lipid content of Impressed Ware vessels recovered from the Early Neolithic village of Skorba, Malta. ORA utilises both chromatographic and isotopic analytical techniques, and provides direct evidence for the function of ceramic vessels analysed. Lipid residues were interpreted against authentic reference fats of Mediterranean origin, and in light of the archaeological data available. The results showed that lipid yields were generally low, however direct evidence for the processing of an admixture comprising ruminant fat and marine oil was obtained in a vessel dated to the Early Neolithic period. This investigation also tests the feasibility of carrying out ORA on ceramic vessels recovered from Maltese archaeological contexts.

Background to the problem

The first archaeological evidence for human settlement in Malta dates to around 5000 BC, when Sicilian farmers are thought to have crossed the channel bringing with them various aspects of the Neolithic, including pottery and domesticates (Trump 2002). Good weather permitting, both islands can be easily sighted, with better visual contact being made from the island of Gozo (Pace 2004). Archaeological remains dating to this period have been retrieved from the caves of Għar Dalam (Malta) and Għajn Abdul (Gozo), and the Neolithic village of Skorba (Mġarr, Malta), which is the only Neolithic village uncovered on the islands to date. The remains of a wall were found, indicating a ditched enclosure, however extensive investigations have been limited due to the subsequent building of a megalithic temple structure above the village. Pace (2004) suggested that perhaps other such scenarios might have existed in Malta, hence by the Għar Dalam phase, the islands may have been populated by other such settlements, which in later centuries may or may not have been developed into megalithic monuments.

To date, archaeological and genetic studies support a Levantine origin for agrarian practices and domestication (e.g. Bruford and Townsend 2006; Luikart *et al.* 2006; Zeder 2011). Their subsequent spread across the Mediterranean has been widely disputed (Ammerman and Cavalli-Sforza 1984; Barker 1985; Zilhão 2001). The current model proposed to explain the expansion of farming in this region suggests a punctuated event, brought about by pioneer seafaring farmers, who spread rapidly westwards from the Levant area (Zilhão 2001). At this time, characteristic Impressed/Cardial pottery and domesticates appear simultaneously on Mediterranean coastal sites. Pottery has for a long time been perceived as an indicator of agrarian settlements, in particular because of the assumption that the earliest ceramic vessels were produced and used by early farmers, thus implying that the spread of pottery and farming was linked (Jordan and Zvelebil 2010b). Archaeological research has shown that this is not necessarily the case; for example, in Japan, China and the Russian Far East, pottery vessels were used millennia before the onset of plant and animal husbandry (e.g. Aikens 1995; Jordan and Zvelebil 2010a and references therein), and similarly in North Africa, agriculture appears two or three thousand years after the beginning of pottery production (Close 1995). On the other hand, evidence for agrarian practices in the Levant occurs well before the earliest indications for pottery production (Moore 1995). The Mediterranean makes an interesting comparison. Current data shows that during the transition to

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agriculture in the eastern Mediterranean, evidence that aceramic communities utilised domestic taxa has been found (e.g. Perlès 2001), while pottery has also been retrieved from contexts associated with purely wild taxa (e.g. Forenbaher and Miracle 2005). Yet excavations in the central and western Mediterranean more consistently show a simultaneous introduction of domesticates and pottery (e.g. Trump 1966, Muntoni, 2009), which appear to have spread rapidly westwards as a package (Zilhão, 2001).

Characterising the absorbed lipid content of ceramic vessels using organic residue analysis (ORA) provides an excellent opportunity to directly determine vessel use. ORA is a well-established technique which has been routinely used over the past two decades to characterise a wide range of animal products (e.g. animal adipose, ruminant dairy fats and marine oils), plant oils and epicuticular waxes, beeswax, bitumen, resins, and birch bark tar (see reviews Evershed 2008b; Debono Spiteri *et al.* 2011; Regert 2011). The premise for using ORA is that when animal and plant products are processed in unglazed ceramics, the heat generated causes fatty components to become absorbed within the walls of ceramic vessels. Lipid residues in charred visible crusts, which are sometimes found adhering to the surface of ceramic vessels, can also be similarly extracted and characterised (Craig *et al.* 2013). By quantifying lipid yields along the profile of the vessels tested, the cooking method can also be identified (Charters *et al.* 1993). Lipid extracts are analysed using Gas Chromatography (GC), which separates out and quantifies the lipid constituents present in the extracted residue, and GC-Mass Spectrometry (GC-MS), which provides structural information on the lipids present, allowing a preliminary identification of the original fatty source to be made. In the absence of specific biomarkers, it is difficult to distinguish between different types of fat, in particular ruminant and non-ruminant adipose, and ruminant dairy fats, because the lipid profiles of these degraded fats are very similar. Another technique, GC-combustion-Isotope Ratio MS (GC-c-IRMS), is applied. GC-c-IRMS measures the isotopic ratios of individual compounds within a mixture, in this case the $^{13}C/^{12}C$ of palmitic ($C_{16:0}$) and stearic ($C_{18:0}$) fatty acids, denoted as $\delta^{13}C$ values. The $\delta^{13}C$ measurements of these two fatty acids vary in different fatty products because of variation in the way they are biosynthesised and routed within the organism, which in turn, allows different fats to be categorically distinguished (Evershed *et al.* 2002). $C_{16:0}$ and $C_{18:0}$ fatty acids are present in all living organisms, their $\delta^{13}C$ values are not affected by diagenesis over archaeological timescales (Evershed *et al.* 1999), and they are readily extractable from prehistoric pottery, making these two fatty acids excellent compounds to target for GC-c-IRMS analysis.

The research presented here is part of a wider study in which 301 Impressed/Cardial Ware vessels recovered from 14 Early to Middle Neolithic sites in the Mediterranean were tested (Debono Spiteri 2012), primarily to re-evaluate the role of Impressed/Cardial Wares at the transition to agriculture. This paper focuses on the results obtained from the analysis of 16 Impressed Ware vessels collected from Early Neolithic contexts at Skorba, and (i) addresses: the feasibility of applying ORA to pottery vessels recovered from the Maltese archaeological record in terms of lipid preservation; (ii) investigates the content of the selected Early Neolithic vessels, and therefore obtain direct evidence for their function; (iii) informs on the culinary practices of the first settlers on the islands.

The site

The Neolithic village of Skorba is thought to have been established by 5000 BC (Trump 2008). The site lies on a hill overlooking Żebbiegħ, in the north-west of Malta (Fig. 1). Valleys located to the north and south of the site provided good arable soil, with a freshwater spring in the former and easy access to the coast. Revised calibrated radiocarbon dates suggest that the Neolithic took over a millennium to develop, and is subdivided into three phases: Għar Dalam (GħD) and Grey Skorba (GSk) which overlap (5500-4100 Cal. BC), and Red Skorba (RSk, 4350-3650 Cal. BC) (Fenech 2007). The structures associated with the Neolithic village include a hut and an 11m wall, founded firmly in the rock and dated to the first level of occupation on the islands (GħD), a GSk phase wall, and two huts built during the RSk phase. During the Temple Period, around 3500 BC, a standard early three-apsed temple was built on the site, but the extent to which this structure and later additions affected the village is still unknown (Trump 1966). Ploughing over the years has led to further destruction of the archaeological structures, however what remains provides evidence for a flourishing Neolithic village. The site was excavated between 1961 and 1963 (Trump 1966).

Materials and method

Sherds were selected from domestic contexts and comprised a selection of rims, bases and body fragments from ceramic vessels associated with cooking, serving and perhaps storage of food commodities (Table 1). Sampling was carried out using a Dremmel modelling drill with a tungsten bit. About 2g of ceramic powder was drilled from the internal surface of each sherd to a depth of around 4mm, discarding the first layer to remove possible contamination introduced by handling and contact with plastic. External surfaces were also sampled to test for exogenous contamination. The ceramic powder was accurately weighed and 1µg tetratricontane was added as an internal standard for quantification purposes. Lipids were extracted three times by sonicating in a mixture of dichloromethane and methanol (2:1; v:v) (HPLC grade; Fischer). Following centrifugation, the solvent was pipetted off into clean screw-capped vials, then evaporated under a gentle stream of nitrogen and mild heating to obtain the total lipid extract (TLE), which was then partitioned (50%). Prior to High Temperature-GC (HT-GC) and GC-MS analyses, one half of the partitioned lipid extracts was derivatised using N,O-bis(trimethylsilyl)trifluoracetamide (BSTFA) with 1% trimethylchlorosilane (TMCS) (4 drops; 70°C; 1 hour).

One sample (SKR-16I) contained sufficient $C_{16:0}$ and $C_{18:0}$ fatty acids for GC-c-IRMS analysis. To release esterified fatty acids, the remaining TLE was saponified by adding 0.5M sodium hydroxide solution made up in a methanol and water solution (9:1, v:v), and heating at 70°C for 1 hour. The samples were allowed to cool, then neutralised. The lipids were extracted into hexane (Fischer; HPLC Grade), and the solvent was gently evaporated. Saponification was also carried out on a selection of the extracted ceramic powder samples, to analyse the 'bound' lipid fraction not released by solvent extraction.

Fatty acid methyl esters (FAMEs) were prepared using 200µL of Boron trifluoride methanol solution (14%; Sigma Life Science) and heating for 1 hour at 70°C. The FAMEs were extracted into hexane and the solvent reduced. $C_{16:0}$ and $C_{18:0}$ fatty acid standards of known carbon isotopic composition were methylated alongside the sample, and were later used to correct the $\delta^{13}C$ values obtained for the carbon atom added during methylation. The sample was analysed using GC and GC-c-IRMS.

Results

Lipid yields were considered significant if more than 5µg of lipid per gram of sherd (µg g^{-1}) were obtained, which ensured that archaeological lipid profiles could be securely distinguished from background contamination (Evershed 2008a). All but one of the 16 samples analysed contained negligible (<5 µg g^{-1}) amounts of absorbed lipid residue (Table 1), and comprised mainly traces of fatty acids and alkanes, probably introduced from the burial environment as they are consistent, in lipid profile and quantification, with the five external sherds analysed. Saponification of the extracted ceramic powder to release the 'bound' lipid fraction also produced negligible results. Sample SKR-16I was obtained from the interior surface of a vessel dated to the GħD phase, whose shape could not be identified. The fabric was black, coarse and uneven, and had numerous large, black and white inclusions. GC-MS analysis of the lipid extracted from SKR-16I is consistent with a ruminant fat. It comprised $C_{16:0}$ and $C_{18:0}$ fatty acids, the latter being more abundant, indicating an animal fat. The presence of $C_{15:0}$ and $C_{17:0}$, produced by microorganisms in the rumen, suggest a ruminant fat, while the short chain fatty acids ($<C_{12:0}$) identified are indicative of a dairy fat (Fig. 2). A series of diacylglycerols was present, indicating hydrolysis of triacylglycerols which were absent when the sample was run on HT-GC. Two long-chain ketones, derived from the condensation of fatty acids during heating, were present and are indicative of cooking (criteria consistent with Raven *et al.* 1997).

GC-c-IRMS analysis of SKR-16I showed a $\Delta^{13}C$ value of -3.1‰, which is consistent with a ruminant fat, possibly, but not unequivocally a dairy residue (Fig. 3). The $\delta^{13}C$ value of the $C_{16:0}$ fatty acid is quite high, plotting within the isotopic range denoting marine oils. This can be due to contribution from C_4 vegetation (e.g. maize or sorghum), or marine oil to the pot content. C_4 plants had not yet been introduced in the Mediterranean during the Neolithic (Hunt *et al.* 2008), so in their absence this residue may potentially represent a mixture comprising ruminant fat and marine oil, possibly introduced during sequential cooking episodes. However, i) marine fish biomarkers were not present in the lipid profile, either because they did not survive, or because fish had not been processed in the pot in the first place, and ii) fish bones were not recorded in the archaeological deposit. Hence a marine input cannot be securely suggested.

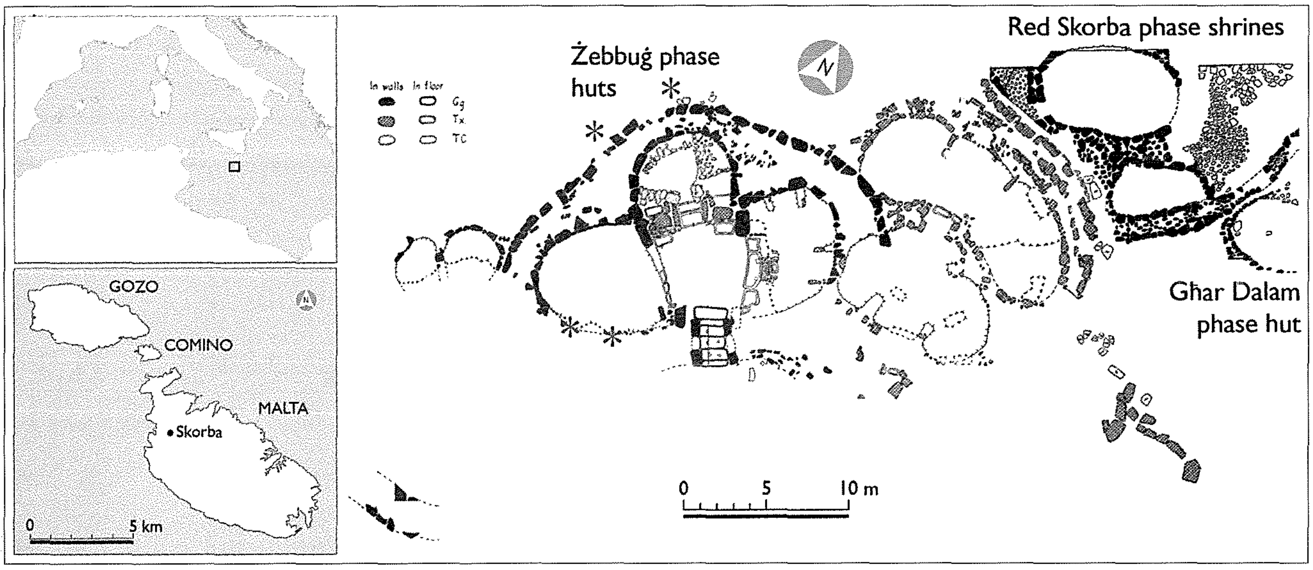

Figure 1. Plan of Skorba showing the main features pertaining to the Neolithic village and the later apsed temple (after Trump 2002: Figures 3 and 10; Map courtesy of Heritage Malta. [*: denotes the location where the pottery vessels sampled were recovered]. Redrawn by Maxine Anastasi).

Discussion

Organic residues are more likely to survive in waterlogged and desiccated environments, rather than in areas where seasonal variations alternate between heavy rainfall and hot dry spells, which is more consistent with the Mediterranean climate (complementing results obtained by Evershed *et al.* 2008; Gregg *et al.* 2009). Furthermore, the calcareous burial deposits, characteristic of the geology of the Mediterranean, do not favour the survival of lipid residues, mainly because they support a richer microbial population than acidic environments, therefore enhancing lipid degradation (Moucawi *et al.* 1981). Low lipid yields pose two major problems: (i) they preclude a secure characterisation of residues because of heightened signals from background contamination, and (ii) negligible residues do not necessarily represent advanced phases of lipid degradation, but could also be produced as a result of the function assigned to individual vessels, which need not have been conducive to the formation of an absorbed residue. Distinguishing between the two using ORA is difficult. This however, does not preclude the application of ORA to Mediterranean contexts, as significant lipid residues have been successfully extracted from pottery recovered from other sites (Debono Spiteri 2012). Increasing the number of vessels tested would, in this case, produce a more representative analysis.

One of the original aims of the study was to observe variation, if any, in pottery use over time (hence the selection of sherds dating from the Early Neolithic (GħD) to the beginning of the Temple Period (Żb)), as well as changes in vessel use with respect to fabric, vessel shape and decoration. Trump (1966) provides a detailed description of the ceramics uncovered at Skorba, which could have been used for a variety of purposes, including cooking, storage and serving. However, the low lipid yields extracted preclude further discussion, as a secure identification of vessel content could not be made in all but one vessel. Low lipid yields add complexity in interpreting the function of Impressed Wares from Skorba since several potential explanations can be put forward to explain the low quantities of lipid present. These include the use of vessels as storage containers for non-fatty products and/or processing of plant material. Simulation experiments have shown that low levels of plant oils become absorbed within the ceramic matrix during vessel use, which is not conducive to survival over archaeological timescales, and is therefore likely to yield negligible lipid quantities (Evershed *et al.* 1995). It is not surprising that plants should be found to play an important role in Early Neolithic cuisine. Their use as a key dietary component is widespread and pre-dates both farming and the production of pottery vessels, which are not essential when processing plant material (e.g. the use of domestic plants is well documented during the Pre-Pottery Neolithic A phase (c. 9000-8500 BC) in the Levant). Remains of barley, wheat

Lab Code	Fabric	Vessel Part	Shape	Decoration	Period	Phase	Date (Cal. BC)	Quantification (μg g^{-1})
SKR-01I	Coarse	Rim	Open vessel	Undecorated	TP	Żb	4350-3050	0.43
SKR-02I	Coarse	Rim	Jar (?)	Undecorated	TP	Żb	4350-3050	0.13
SKR-03E	Coarse	Base	Small bowl	Undecorated	TP	Żb	4350-3050	0.80
SKR-03I	Coarse	Base	Small bowl	Undecorated	TP	Żb	4350-3050	0.48
SKR-04I	Fine	Body	Bowl (?)	Burnished	TP	Żb	4350-3050	0.30
SKR-05E	Fine	Rim	Jar	Burnished	TP	Żb	4350-3050	0.74
SKR-05I	Fine	Rim	Jar	Burnished	TP	Żb	4350-3050	1.27
SKR-06E	Fine	Body	Large vessel with a rounded body	Red slip	EN	RSk	4350-3650	1.73
SKR-06I	Fine	Body	Large vessel with a rounded body	Red slip	EN	RSk	4350-3650	2.36
SKR-07I	Fine	Body	Large vessel with a rounded body	Burnished; red slip	EN	RSk	4350-3650	0.13
SKR-08I	Fine	Rim	Open vessel	Burnished; red slip	EN	RSk	4350-3650	0.10
SKR-09I	Coarse	Body	Large vessel	Red slip	EN	RSk	4350-3650	0.04
SKR-10I	Fine	Body	Large vessel with a rounded body	Undecorated	EN	GSk	5500-4100	1.54
SKR-11I	Fine	Rim	Large vessel	Burnished	EN	GSk	5500-4100	0.21
SKR-12E	Fine	Base	Large open vessel (?)	Burnished	EN	GSk	5500-4100	1.68
SKR-12I	Fine	Base	Large open vessel (?)	Burnished	EN	GSk	5500-4100	0.50
SKR-13I	Fine	Body	Small bowl (?)	Burnished	EN	GħD	5500-4100	1.36
SKR-14E	Medium	Body	Large deep vessel	Undecorated	EN	GħD	5500-4100	3.12
SKR-14I	Medium	Body	Large deep vessel	Undecorated	EN	GħD	5500-4100	4.40
SKR-15I	Medium	Body	Large vessel	Undecorated	EN	GħD	5500-4100	0.56
SKR-16E	Coarse	Body	Bowl (?)	Undecorated	EN	GħD	5500-4100	0.23
SKR-16I	Coarse	Body	Bowl (?)	Undecorated	EN	GħD	5500-4100	5.44

Table 1. Table listing the details of the samples analysed [I: Internal; E: External; TP: Temple Period; EN: Early Neolithic; Żb: Żebbuġ Phase; RSk: Red Skorba Phase; GSk: Grey Skorba Phase; GħD: Għar Dalam Phase].

and lentils were collected from Skorba (Trump 1966, Appendix IV), although low quantities were recorded, which could have been due to floatation methods not having been used in Trump's case. If Impressed Wares at Skorba were being used to process plant remains, the lipid profiles and low lipid yields obtained could be attributed to plant use.

Faunal remains at Skorba show evidence for domestic animals, primarily ovicaprids, but remains of pigs and cows were also retrieved (Trump 1966, Appendix III; Borg 2008). Of interest, Borg (2008) noted the absence of wild species (e.g. deer), despite their earlier attestations in the archaeological record. He suggests that the number of wild animals present in Malta during the Neolithic could have seriously declined, or they could possibly have already become extinct. Hence, the faunal record complements a domestic ruminant fat interpretation to the residue extracted from vessel SKR-16I, in particular since the $\Delta^{13}C$ measurement for this sample plots within an area of overlap between ruminant dairy residues and wild ruminant (deer) adipose, more recently investigated by Craig *et al.* (2012) (Fig. 3). However, since the $\Delta^{13}C$ signal borders on both the ruminant adipose and ruminant dairy fat categories, it cannot be unequivocally interpreted as either, and further analyses are needed to securely attest to the presence of milk. A dairy result would be of significant importance within the Mediterranean framework as it would corroborate evidence from other studies which demonstrate that dairy products were consumed from the initial phases of the Neolithic (Vigne and Hemler 2007; Evershed *et al.* 2008), rather than later (Sherratt 1983). Although the allele responsible for lactose tolerance, 13.910*T, was not yet widespread during the European Neolithic to support a diet rich in dairy products, small quantities of milk can still be consumed by lactose intolerant people without ill-effects, while removing lactose from milk by processing into cheese or yoghurt, makes dairy products more widely available (Ingram *et al.* 2009). Dairy products may indeed have played a crucial role in the survival of early seafaring farming communities spreading throughout the western Mediterranean, in particular, since surplus dairy products can be processed and stored, hence made available all year round (Rowley-Conwy 2011).

Although not representative of the pottery assemblage analysed, the absence of marine biomarkers in sample SKR-16, which could not securely confirm a potential marine input obtained through isotopic analysis, is consistent with the apparent dietary shift from a predominantly marine to a terrestrial diet during the Mesolithic-Neolithic transition in Europe and the Mediterranean (Richards *et al.* 2001, 2003; Le Bras-Gaude and Claustre 2009; Lightfoot *et al.* 2011). It is difficult to perceive why people would turn their backs on a freely available resource, especially when considering that the site is located within walking distance of the Mediterranean coast. Although pots are not necessary to cook fish, the absence of fish bones provides no indication that perhaps different methods had been used to prepare marine products. This is suggestive of a conscious decision to avoid marine food products, in favour of terrestrial produce.

Conclusion

The application of ORA to Early Neolithic vessels from Skorba is the first of its kind to be carried out on pottery recovered from Maltese archaeological contexts. Unfortunately, the lipid content of the vessels analysed was generally low, with only one sample providing sufficient lipid residue for a secure characterisation. This emphasises the necessity to increase the sample size analysed when applying ORA, and in no way does it diminish the potential of this technique to inform on the use of ancient pots, and culinary preferences of the communities that produced them. The combined biomarker and isotopic approach applied identified a potential admixture of a ruminant fat and a marine oil in one vessel dated to the earliest Neolithic phase on the islands (GħD) Although low lipid yields precluded an in depth

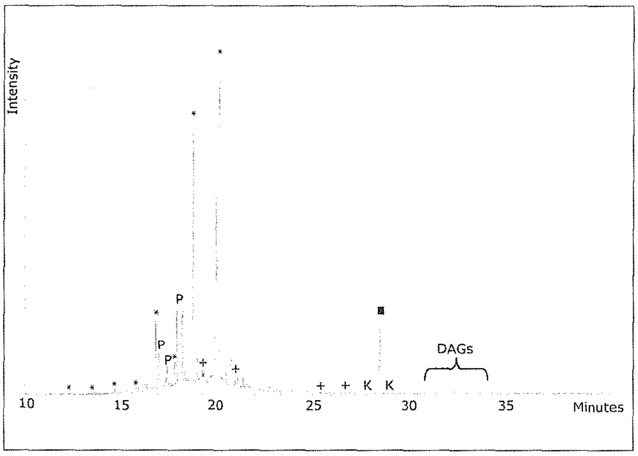

Figure 2. Total Ion Chromatogram of sample SKR-16I. [*: Fatty acids ($C_{10:0}$-$C_{18:0}$; +: Alkanes; K: Ketones; DAGs: Diacylglycerols; P: Phthalates; ◉: Internal standard (C_{34} *n*-alkane)].

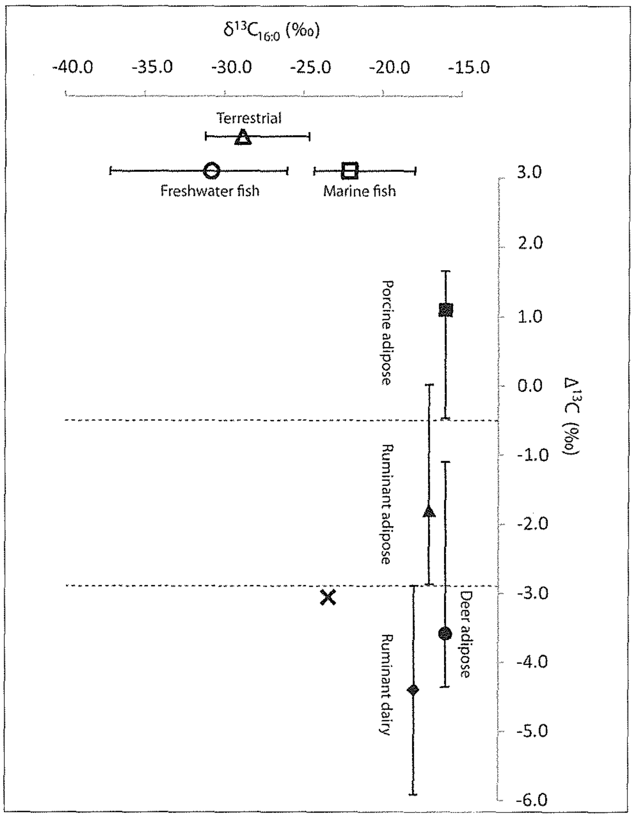

Figure 3. Plot of $\delta^{13}C_{16:0}$ against $\Delta^{13}C$ ($\delta^{13}C_{16:0}$-$\delta^{13}C_{18:0}$). Reference points plot the median values obtained from authentic modern animal fats in published literature (Dudd 1999; Craig *et al.* 2005, 2012; Gregg *et al.* 2009) and have been supplemented with data from blood samples of pigs (*n*=2), milk and blood samples of sheep (*n*=2), goats (*n*=2) and cows (*n*=2), all raised on C_3 Mediterranean grown plants, and fish tissue (*n*=6) caught from Mediterranean waters. All modern values have been corrected for post-industrial carbon (1.2‰; Friedli *et al.* 1986). The error bars denote the range at 95% confidence intervals.

discussion on culinary preferences, further testing may yet produce important results, in particular with regard to the potential presence of an agro-pastoral community at Skorba dating to the initial phases of the Neolithic, which would feed into current research on the origin and spread of dairying.

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