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Compound-specific radiocarbon, stable carbon isotope and biomarker analysis of mixed marine/terrestrial lipids preserved in archaeological pottery vessels

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ABSTRACT

At archaeological sites located on islands or near the coast, the potential exists for lipid extracts of potsherds to contain fatty acids (FA) from both aquatic and terrestrial organisms, meaning that consideration must be given to marine reservoir effects (MRE) in radiocarbon analyses. Here we studied the site of Bornais (Outer Hebrides, UK) where a local MRE, ΔR of -65 ± 45 y was determined through the paired ^{14}C determinations of terrestrial and marine faunal bones. Lipid analysis of 49 potsherds, revealed aquatic biomarkers in 45% of the vessels, and $\delta^{13}\text{C}$ values of $\text{C}_{16:0}$ and $\text{C}_{18:0}$ FAs revealed ruminant and marine product mixing for 71% of vessels. Compound-specific radiocarbon analysis (CSRA) of FAs yielded intermediate radiocarbon ages between those of terrestrial and marine bones from the same contexts, confirming an MRE existed. A database containing $\delta^{13}\text{C}$ values for FAs from reference terrestrial and marine organisms provided endmembers for calculating the percentage marine-derived C ($\%_{\text{marine}}$) in FAs. We show that lipid ^{14}C dates can be corrected using determined $\%_{\text{marine}}$ and ΔR values, such that pottery vessels from coastal locations can be radiocarbon dated by CSRA of FAs.

KEYWORDS: Pottery vessels, Lipid residues, Compound-specific radiocarbon analysis, Marine reservoir effect, Mixed marine/terrestrial corrections.

35

36 INTRODUCTION

37 Due to their central importance to, and survival in the archaeological record, accurate direct
38 radiocarbon dating of pottery vessels has been one of the “Holy Grails” of archaeology.
39 Compound-specific radiocarbon dating of lipids preserved within the clay matrix of
40 archaeological potsherds is technically extremely challenging, with previous attempts failing
41 to achieve the accuracy and precision required (e.g. Hedges *et al.* 1992, Stott *et al.* 2001,
42 2003b; Berstan *et al.* 2008). Recently however, we have reported the first accurate dates
43 achieved for such residues based on compound-specific ^{14}C analyses of $\text{C}_{16:0}$ and $\text{C}_{18:0}$ fatty
44 acids (FAs) isolated from the clay walls of Neolithic pottery vessels (Casanova *et al.*, in
45 press). The preparative-capillary gas chromatography (pcGC) isolation technique required
46 two major advances, namely a new trap design allowing the solvent-less recovery of the
47 trapped analytes and a heat-based cleaning method to prevent cross-contamination (Casanova
48 *et al.* 2018). These methodological improvements have enabled reliable and accurate dating
49 of the two FAs characteristic of degraded animal fats. Furthermore, the two independent
50 radiocarbon dates obtained provide an important internal quality control; the radiocarbon age
51 of the FAs should agree at the 2σ error level (Casanova *et al.* 2018). The samples used thus
52 far in the validation of the compound-specific pot lipid dating method, outlined in Casanova
53 *et al.* (in press) have originated from archaeological sites located inland where human dietary
54 subsistence was dominated by domesticated terrestrial animals, such that the target FAs
55 derived from dairy or carcass fats of ruminant and non-ruminant animals.

56 None of the pottery dated thus far has originated from coastal areas where the exploitation of
57 marine products may have occurred. At such locations, FAs preserved in pottery vessels
58 would likely be affected by a reservoir effect (Heron and Craig 2015), requiring marine
59 reservoir correction in order to obtain reliable calibrated dates (Cook *et al.* 2015). Particularly
60 problematic would be potsherds containing mixed marine- and terrestrial-derived FAs
61 (Cramp and Evershed 2014; Cramp *et al.* 2014a), as this would increase the complexity of
62 marine reservoir corrections.

63 Marine product processing in pots can be identified by the presence of specific aquatic
64 biomarkers alongside the $\text{C}_{16:0}$ and $\text{C}_{18:0}$ FA, namely: (i) long-chain dihydroxy fatty acids
65 (DHYAs), (ii) isoprenoid fatty acids (IFAs) and (iii) long-chain ω -(o-alkylphenyl)alkanoic

acids (APAAs); (Hansel *et al.* 2004; Evershed *et al.* 2008; Hansel and Evershed 2009; Cramp and Evershed 2014). Furthermore, $\delta^{13}\text{C}$ values determined for the $\text{C}_{16:0}$ and $\text{C}_{18:0}$ FAs can reveal the mixing of both terrestrial and marine commodities in the same vessel (Copley *et al.* 2004, Cramp *et al.* 2014a, 2014b). It is known, however, that the relative abundances of $\text{C}_{16:0}$ and $\text{C}_{18:0}$ FAs differ between terrestrial and marine organisms and the relationship between their mixing proportions and the resulting $\delta^{13}\text{C}$ values is not necessarily linear (Mukherjee *et al.* 2005). It is unclear whether this effect will adversely affect the validity of the internal quality control criteria, such that the ^{14}C dates obtained for $\text{C}_{16:0}$ and $\text{C}_{18:0}$ FAs in a potsherd are no longer consistent within 2σ (Casanova *et al.* 2018, in press). It is certainly possible that $\text{C}_{16:0}$ and $\text{C}_{18:0}$ FAs in sherds arising from mixtures of terrestrial- and marine-derived food residues may yield different apparent radiocarbon ages.

Generally, MRE corrections require generation of terrestrial/marine mixing curves using dedicated software (e.g. OxCal, CALIB). This requires an understanding of the local deviation (ΔR) from the global marine calibration curve for a specific time period as well as the percentage of marine-derived C ($\%_{\text{marine}}$) present (Cook *et al.* 2015). The ΔR values can be obtained by radiocarbon dating historical marine specimens (of known date of collection) from museum collections, pairing ^{14}C measurements on terrestrial and marine organisms from secure contexts at the site of interest or by dating tephra layers deposited at sea and on land (Ascough *et al.* 2005). The evaluation of the $\%_{\text{marine}}$, however, is more challenging. Such considerations are often applied to bone collagen from omnivores which can feed on both terrestrial and marine resources (Cook *et al.* 2015). Typically, $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ values are recorded on bulk collagen to understand the local diet and the percentage of marine resources consumed. Preferably, endmembers for pure terrestrial and pure marine organisms are recorded for samples local to the site, but in the majority of cases, more general (non-local) reference values for endmembers are used (Cook *et al.* 2015).

Herein, we evaluate whether the approach commonly applied to bone collagen to estimate the contribution of aquatic resources could be applied to FAs extracted from pottery vessels for MRE correction of pot lipids ^{14}C dates. The approach was to undertake radiocarbon dating in order to determine the influence of aquatic resources on CSRA of lipids from potsherds and establish appropriate methods to correct for the MRE. We focussed on lipids preserved in pottery vessels with a clearly mixed marine/terrestrial signal from the site of Bornais (South Uist, UK). Our approach involved: (i) lipid residue analyses on pottery vessels including compound-specific $\delta^{13}\text{C}$ determinations on FAs, (ii) calculation of the local deviation from

the global marine calibration curve at the site using paired marine and terrestrial animal remains, (iii) radiocarbon dating of FAs from a range of pottery vessels, (iv) a multiproxy investigation (i.e. biomarkers, stable isotopes and ^{14}C analyses) to evaluate the proportion of mixing of marine and terrestrial lipids and (v) application of relevant marine reservoir corrections to the radiocarbon dates obtained from pot lipids.

METHODS

Site description

The site of Bornais is located on the island of South Uist, in the Outer Hebrides, UK (Supplementary material S1). The site comprises 4 mounds with a long duration of occupation defined by radiocarbon dates on seeds and bone collagen, from the late Iron Age (LIA 1 and LIA 2; 5th-6th century AD) to the Early, Middle and Late Norse (EN, MN, LN, respectively; mid-9th-14th century AD) period (Marshall 2005, 2016, forthcoming; Sharples forthcoming). The recovery of plant macrofossils indicates the cultivation of rye and barley, while the faunal assemblage displays a particularly rich diversity of terrestrial animals (*c.* 18,000 bones), small vertebrates, birds, fish and molluscs; Sharples and Davis forthcoming, Sharples, *et al.* 2016). Domesticated animals dominate (~ 95 %) the terrestrial faunal assemblage, which comprised cattle (*ca.* 40 %), sheep (*ca.* 45 %) and pigs (*ca.* 10%). The mortality profiles derived from the cattle suggest they were exploited for their milk (Sharples, *et al.* 2016). Fish bones (eel, saithe, cod, haddock, ray, turbot, mackerel etc.) and mollusc shells (limpets and winkles) were extremely abundant at the site (*c.* 17,000 identified specimens), while marine mammal bones, e.g. seal, were rare (Sharples, *et al.* 2016).

Lipid residue analysis

Lipid residue analyses of samples of pottery were performed using a methanolic sulphuric acid extraction procedure (Correa-Ascencio and Evershed 2014). The total lipid extracts (TLEs) were analysed by gas chromatography (GC) and GC-Mass spectrometry (GC-MS) for the identification and quantification of biomarkers, including aquatic biomarkers following established procedures (Evershed *et al.* 1990, Cramp and Evershed 2014). Compound-specific $\delta^{13}\text{C}$ values of fatty acids were determined by GC-Combusted-Isotope ratio MS (GC-C-IRMS; supplementary material S2).

Pretreatment methods for radiocarbon analyses

Approximately 300 mg of coarse bone powder were weighed into a culture tube and pre-treated using a modified Login procedure (Longin 1971) as described in Knowles, *et al.* (2019). Briefly, bone powder was demineralized in HCl (0.5 M, 10 mL, ~ 18h, room temperature (RT)) followed by a wash with NaOH (0.1 M, 10 mL, 30 min, RT) and a second acid wash with HCl (0.5 M, 10 mL, 30 min, RT). The extracted collagen was rinsed with ultrapure MilliQ-water (MQ-water; 3 x 10 mL) in between each acid and base wash and centrifuged (3000 rpm, 5 min). The collagen was then gelatinised at pH 3 with HCl (0.001 M, 10 mL, 75 °C, 20 h) and filtered through pre-combusted glass fibre before freeze drying (Knowles, *et al.* 2019).

Surface cleaned shells were ultrasonically agitated in MQ-water (5 mL, 5 min) before drying at 60°C. When dried, the shells (~ 30 mg) were crushed roughly before the surface was acid etched (~20 %) with HCl (0.2 M, 10 mL). Samples were rinsed with MQ-water (3 x 10 mL) and dried at 60°C in a drying cabinet (Knowles, *et al.* 2019).

Sherds containing lipid concentrations, typically above 500 µg.g⁻¹, were selected for radiocarbon determinations. Pieces of 2 to 10 g of the potsherd was sampled, depending on the lipid concentrations and size of the potsherds. The lipids were extracted in culture tubes using H₂SO₄/MeOH (4 % v/v, 3 x 8 mL, 70°C, 1 h). Samples were centrifuged after each extraction (2500 rpm, 10 min) and the 3 supernatants (methanolic fractions) combined into a second culture tube containing double-distilled water (5 mL). The lipids, including fatty acid methyl esters (FAMES) created from the reaction of methanol with the FAs during the first step, were extracted from the methanolic solution with *n*-hexane (4 x 5 mL) and blown down to dryness at room temperature under a gentle nitrogen stream. The TLEs were derivatized with BSTFA (20 µL, 70 °C, 1 h). Excess BSTFA (*N,O*-bis(trimethylsilyl)trifluoroacetamide) was removed under a nitrogen stream, then ~180 µL of *n*-hexane was added to obtain a solution containing C_{16:0} and C_{18:0} FAMES at a concentration at *c.* 5 µg.µL⁻¹ of carbon. The solution was transferred to an autosampler vial for isolation of C_{16:0} and C_{18:0} into individual traps using a preparative capillary gas chromatography (pcGC) instrument following the methods described in Casanova *et al.* (2017, 2018, in press).

Radiocarbon determinations

Organic materials (FAMES and collagen) were combusted to CO₂ using a Vario Microcube Elemental Analyser (EA, Elementar). The shells (carbonate-based) were digested in H₃PO₄ (1

mL, 85 % v/v, 70 °C) under a He headspace using a Carbonate Handling System (CHS, Ionplus; Wacker, *et al.* 2013; Knowles, *et al.* 2019) to generate CO₂. Resulting CO₂ was transferred to the Automated Graphitisation Equipment (AGE 3, Ionplus; Wacker *et al.* 2010; Knowles, *et al.* 2019) under a He stream and adsorbed on Zeolite traps before being released into reaction tubes. The CO₂ was reduced to graphite under H₂ (580 °C, 2 h, 420 mbar) on a pre-conditioned iron catalyst. A Pneumatic Sample Press (PSP, Ionplus) was used to press the graphitised samples into Al targets.

All ¹⁴C determinations were performed at the BRAMS (Bristol Accelerator Mass Spectrometer) facility which is equipped with a mini radiocarbon dating system (BRIS-MICADAS) instrument (ETH Zurich, Zurich, Switzerland; Synal *et al.* 2007). Samples were analysed alongside size-matched processing standards and blanks (Casanova *et al.* 2018; Knowles *et al.* 2019).

Corrections and calibration of ¹⁴C measurements

Radiocarbon measurement on FAs from single pottery vessels were corrected for the presence of the methyl derivative C (Casanova *et al.* 2017, 2018) and subjected to a 2-sigma equivalency test and, if successful, combined as described in Casanova *et al.* (in press) before testing the validity of calibration on mixed marine/terrestrial resources. Reservoir correction and calibration of the mixed resources was performed in OxCal v 4.3 (Bronk Ramsey 2009) using the 'Marine/mixed curve' tool using the IntCal13 and Marine13 curves (Reimer *et al.* 2013). This incorporates the percentage of marine derived resources present in the TLEs and the ΔR value for the site (see results section).

The local reservoir effect was calculated for every pair combination of terrestrial/marine organisms in each context using the online ΔR calculation tool (Reimer and Reimer, 2016). These individual ΔR values were subjected to a χ^2 test at the 5 % level (both for each context and all together) to detect potential outliers before calculation of their weighted average, with error calculation as recommended by Russell *et al.* (2010).

The mixing of marine/terrestrial commodities was quantified in each potsherd using two independent methods: ¹⁴C dates and $\delta^{13}\text{C}$ values of C_{16:0} and C_{18:0} FAs (Figure 1). By comparing the mixing ratios obtained by the two methods, it is possible to evaluate whether

the FA $\delta^{13}\text{C}$ values (determined by GC-C-IRMS) can be used to estimate the proportion of marine-derived C in the FAs for use in MRE corrections of their radiocarbon dates. This constitutes an important consideration, especially for sites where terrestrial and marine remains are absent from the archaeological record and so cannot be used to provide reference radiocarbon ages.

Figure 1

The first method of quantifying the $\%_{\text{marine}}$ is based on the weighted average of radiocarbon determinations on the short-lived terrestrial and marine organisms, from the same context/phase as the potsherds dated, as endmembers using Equation (1).

$$(1) \quad \%_{\text{marine}} = \frac{(Age_{\text{pot}} - Age_{\text{terr}})}{(Age_{\text{marine}} - Age_{\text{terr}})} * 100$$

Where $\%_{\text{marine}}$ is the percentage of aquatic C in the lipid residue and Age_{pot} , Age_{terr} , Age_{marine} are the combined radiocarbon ages on the individual FAs, for terrestrial animals and marine organisms, respectively.

The second method uses the $\delta^{13}\text{C}$ values of the individual FAs of UK reference animals (cattle and sheep raised on a pure C_3 diet; Copley, *et al.* 2003) as the terrestrial endmembers (pigs were hypothesised not to have been processed in potsherds; Sharples, *et al.* 2016, and fish, winkles and limpets captured from UK waters (corrected for the Suess effect; Cramp and Evershed, 2014) to serve as the marine endmembers. The terrestrial endmembers correspond to the average values for both $\text{C}_{16:0}$ and $\text{C}_{18:0}$ FAs and were found to be

$\delta^{13}\text{C}_{16:0} = -30.0 \pm 0.6 \text{ ‰}$ and $\delta^{13}\text{C}_{18:0} = -32.2 \pm 0.6 \text{ ‰}$ for adipose fats and $\delta^{13}\text{C}_{16:0} = -29.2 \pm 1.0 \text{ ‰}$ and $\delta^{13}\text{C}_{18:0} = -34.0 \pm 0.9 \text{ ‰}$ for dairy fats. Both ruminant adipose and dairy values were used as endmembers to evaluate whether one should be used over the another. The marine endmembers were $\delta^{13}\text{C}_{16:0} = -22.7 \pm 2.2 \text{ ‰}$ and $\delta^{13}\text{C}_{18:0} = -21.7 \pm 2.5 \text{ ‰}$. The relationship between the relative proportions of marine and terrestrial fats and the $\delta^{13}\text{C}$ values is theoretically non-linear, due to the differing relative abundances of the of FAs in the different foodstuffs (Mukherjee, *et al.* 2005), however, the success of the internal quality control on the CSRA dates (see results section) suggests a linear relationship within analytical uncertainty. A linear mixing curve could therefore be employed to estimate the $\%_{\text{marine}}$ contribution and associated uncertainty using the propagation of analytical errors. The $\%_{\text{marine}}$ values obtained on both FAs were then combined as a weighted average with uncertainties calculated according to Russell *et al.* (2010). Such a model is a conservative approach and probably overestimates the uncertainties. Furthermore, it cannot take into account the fact that the true $\%_{\text{marine}}$ values must be constrain between 0 and 100 %.

As a comparison, the $\%_{\text{marine}}$ values of the TLEs were also estimated (using the same endmembers) using the software package FRUITS (v2.1). This software employs a Bayesian approach to quantify the contribution of different food sources using isotopic data (Fernandes *et al.* 2014). The output of this software is given both as means and standard deviations (represented by box-and whiskers plots) or as probability distributions constrained to between 0 and 100 %. The full range of data points for the probability distributions of the $\%_{\text{marine}}$ after Bayesian modelling was exported and implemented as a prior information file into the mixing marine/terrestrial tool in OxCal.

RESULTS AND DISCUSSION

Characterisation of lipid residues in pottery vessels

Forty-nine pottery vessels from layer BCC, Mound 2, MN period were subjected to lipid residue analyses (S2). TLEs with concentrations $>5 \text{ } \mu\text{g.g}^{-1}$ were recovered from 96 % ($n = 47$) of the potsherds, at an average lipid concentration of 1.2 mg.g^{-1} (supplementary material S3). A total of 80 % of the TLEs with residues ($n = 39$) were dominated by the $\text{C}_{16:0}$ and $\text{C}_{18:0}$ fatty acids characteristic of degraded animal fats (Figure 2a). Many of the TLEs (47 % of the sherds with residues; $n = 22$) exhibited marine biomarkers. The long-chain DHYAs (C_{18} , C_{20} and C_{22}) were detectable in 30 % ($n = 14$; Figure 2b), long-chain APAAs (C_{18} , C_{20} and C_{22}) in

23 % ($n = 11$; Figure 2c) and the IFAs (phytanic acid and 4,8,12-trimethyltridecanoic acid (TMTD)) in 30 % ($n = 14$) of the sherds with residues. In total, only 4 % ($n = 2$; BN-140, BN-173) of the potsherds with lipid residues contained all 3 classes of aquatic biomarkers, 21 % ($n = 10$) contained 2 aquatic biomarkers and 26 % ($n = 12$) showed one aquatic biomarker. No aquatic biomarkers were detected in the remainder of the TLEs ($n = 25$, 53 % of the sherds with organic residues). The $\delta^{13}\text{C}$ values of the palmitic and stearic acids were determined by GC-C-IRMS (Figure 2d). Significantly, the $\text{C}_{16:0}$ and $\text{C}_{18:0}$ fatty acids displayed $\delta^{13}\text{C}$ values characteristic of mixtures between ruminant and marine or porcine products (Cramp *et al.* 2014a, 2014b). The extracts yielding the most enriched stable carbon isotope values also contained aquatic biomarkers, strongly suggesting the processing of marine products rather than porcine. Several TLEs were relatively more enriched in ^{13}C , but show no detectable aquatic biomarkers suggesting they did not survive or could denote the processing of marine commodities under conditions not conducive to the formation of thermally-produced aquatic biomarkers (APAAs). The use of $\Delta^{13}\text{C}$ ($= \delta^{13}\text{C}_{18:0} - \delta^{13}\text{C}_{16:0}$) values allows the identification of sherds where FAs are predominantly of dairy product origin (< -3.1 ‰; Copley *et al.* 2003). A total of 22 sherds yielded $\Delta^{13}\text{C}$ values below -3.1 ‰, with aquatic biomarker identification for half of them supports the hypothesis of some mixing of dairy products with marine products. The other sherds with higher $\Delta^{13}\text{C}$ values of > -3.1 ‰ could result from the mixing of ruminant carcass and marine products.

Additionally, 131 potsherds from all the phases and mounds at the site were previously analysed by lipid residue analysis (Cramp *et al.* 2014b, forthcoming). The results suggested a dominance of dairy and ruminant carcass product processing, as well as some mixing of non-ruminant and marine fat/oil (Figure 1d). Only the pottery from the LIA 1 phase lacked aquatic biomarkers.

Figure 2

A total of 21 potsherds (from all phases) with sufficient lipid concentrations and containing either none or at least one aquatic biomarker, were subjected to CSRA (Figure 1e).

ΔR value determination

In order to determine the age of the structures associated with the pots, and the local reservoir effect at the site of Bornais a range of fish bones ($n = 13$), marine mollusc shells ($n = 14$) and terrestrial animal bones ($n = 8$) were radiocarbon dated (Table 1). These were assessed together with other available radiocarbon measurements on terrestrial animal bones and

grains (layer BCC, $n = 7$; Marshall *et al.* forthcoming-b). All these materials derive from the LIA2, EN, MN and LN settlement structures. On a context-by-context basis, marine and terrestrial organisms were subjected to χ^2 statistical testing to detect outliers for exclusion (Table 1). Two marine samples from context BAF and two terrestrial animal bones from context BCC were, therefore, excluded from ΔR determination. The ΔR values calculated using all the pairs of terrestrial/marine organism (80 in total) per context are reported in Table 1. No ΔR was calculated for contexts BBA and BBD as they were dated based on only one material type, and for AG, the two marine organisms from this context failed the χ^2 test.

With the exception of context BCC, all the contexts demonstrated a negative ΔR , varying from -214 ± 26 to -45 ± 21 . Interestingly, layer BCC (MN phase) shows a ΔR of 28 ± 150 ; the large uncertainty associated with this value results from high variability in the radiocarbon ages of the marine organisms, which could be classified into three distinct groups. Group (a) gave a ΔR of -107 ± 54 , Group (b) 242 ± 55 and Group (c) -31 ± 56 . The MRE of Group (c), comprised only fish bones and likely reflects the mobility of the fish species (Russell, *et al.* 2011). Groups (a) and (b) comprise both winkles and limpets and their MREs do not appear to be species dependent. The grouping could, therefore, either correspond to two different collection points of the mollusc shells (likely collection points nearby are either completely coastal, or sea lochs with the potential for substantial terrestrial runoff) or simply the introduction of older material into a later context (although, this offset was only observed for some limpet and winkle shells, but not fish bones).

Table 1: Radiocarbon determinations on terrestrial and marine organisms at Bornais and ΔR calculated for the diverse contexts based on the multiple paired terrestrial/marine organisms. *refer to statistical outliers that have been excluded from ΔR calculation.

Phase	Layer	Terrestrial organisms			Marine organisms			ΔR
		Material	Laboratory#	Conventional ^{14}C age	Material	Laboratory#	Conventional ^{14}C age	
LIA 2	BAC	Cattle	BRAMS-1710	$1,272 \pm 25$	Limpet-1	BRAMS-1727	$1,624 \pm 26$	-47 ± 23
		Caprine	BRAMS-1711	$1,298 \pm 25$	Limpet-2	BRAMS-1728	$1,627 \pm 26$	
		Unidentified	BRAMS-1713.1	$1,258 \pm 25$	Limpet-5	BRAMS-1731	$1,642 \pm 26$	
			BRAMS-1713.2	$1,264 \pm 25$	Limpet-6	BRAMS-1732	$1,616 \pm 26$	
	BAF	Cattle	BRAMS-1712	$1,348 \pm 25$	Fish-9	BRAMS-1725*	$1,651 \pm 25$	-165 ± 26
					Fish-10	BRAMS-1726*	$1,294 \pm 25$	
					Limpet-3	BRAMS-1729	$1,565 \pm 26$	
					Limpet-4	BRAMS-1730	$1,584 \pm 26$	
EN	BAG	Cattle	BRAMS-1708	$1,320 \pm 25$	Fish-1	BRAMS-1717	$1,509 \pm 25$	-214 ± 26
		Cattle	BRAMS-1709	$1,320 \pm 25$				
	BBD	Cattle	BRAMS-1715*	$1,082 \pm 25$	-			-
		Cattle	BRAMS-1719*	945 ± 25				
	BBA	-			Limpet-7	BRAMS-1733	$1,622 \pm 26$	-

MN	BCC	Cattle	SUERC-2684	925 ± 35	Fish-11	BRAMS-2049 (c)	1,306 ± 25	-102 ± 35 (a) 248 ± 37 (b) -26± 40 (c) 35 ± 150 (all)
		Red deer	SUERC-22894*	875 ± 30	Fish-12	BRAMS-2050 (c)	1,318 ± 25	
		Pig	SUERC-22890*	1,035 ± 30	Fish-13	BRAMS-205.1 (c)	1,323 ± 25	
		Seed	GU-18290	-	Fish-14	BRAMS-2052 (c)	1,365 ± 25	
		Cattle	SUERC-22896	970 ± 30	Fish-15	BRAMS-2053 (c)	1,308 ± 25	
		Cattle	SUERC-22897	975 ± 25	Limpet-8	BRAMS-2041*	1,380 ± 24	
		Sheep	OxA-15420	903 ± 27	Limpet-9	BRAMS-2042.1 (a)	1,263 ± 25	
		Cattle	OxA-15522	985 ± 26		BRAMS-2042.2 (a)	1,236 ± 24	
					Limpet-10	BRAMS-2043(b)*	1,575 ± 24	
					Limpet-11	BRAMS-2044 (b)*	1,593 ± 25	
					Winkle-1	BRAMS-2045 (a)	1,257 ± 24	
					Winkle-2	BRAMS-2046 (b)*	1,614 ± 25	
					Winkle-3	BRAMS-2047 (a)	1,243 ± 24	
					Winkle-4	BRAMS-2048 (b)*	1,613 ± 25	
	AD	Cattle	BRAMS-1716	956 ± 25	Fish-4	BRAMS-1720	1,167 ± 25	-84 ± 34
					Fish-5	BRAMS-1721	1,261 ± 25	
					Fish-6	BRAMS-1722	1,257 ± 25	
LN	AG	Sheep	BRAMS-1714	930 ± 25	Fish-7	BRAMS-1723.1*	1,268 ± 25	-
						BRAMS-1723.2*	1,237 ± 25	
					Fish-8	BRAMS-1724*	1,183 ± 25	
Overall site							-65 ± 46	

The MREs calculated from Groups (a) and (c) gave statistically indistinguishable ^{14}C determinations and are in good agreement with ΔR values calculated for the other contexts. Only shells from Group (b) were excluded from the overall ΔR determination due to uncertainty in the security of the context in light of its high ΔR value. The remaining 56 ΔR s failed the statistical identity test, therefore layer BAG ($\Delta\text{R} = -214 \pm 26$), showing the lowest ΔR and the pairs BN-F-14/SUERC-2684, BN-F-14/OxA15420 which showed the highest ΔR values were excluded from the calculation. With the removal of the organisms and terrestrial/marine pairs identified as outliers the remaining 53 ΔR values are statistically identical ($T' = 69.3$, $T'(5\%) = 71.0$, $v = 53$) and average to -65 ± 46 . These data suggest there is no significant difference in the reservoir effect from the LIA2 to LN period at the site. This ΔR value of -65 ± 46 is also consistent with the previously reported ΔR values for the North Atlantic, including the (Inner, and Outer) Hebrides Island of -47 ± 52 for the period 3500 BC-1450 cal AD (Reimer, *et al.* 2002; Ascough, *et al.* 2004, 2005, 2006, 2007, 2009, 2017; Russel, *et al.* 2010, 2015; see supplementary material S4).

Radiocarbon dating of pottery vessels

$\text{C}_{16:0}$ and $\text{C}_{18:0}$ FAs from 21 sherds were dated, of which six were dated in duplicate. This includes potsherds from all phases present at Bornais, both with and without aquatic biomarkers present. Of these, 17 sherds successfully passed the internal quality control criterion, whereby the radiocarbon dates of the $\text{C}_{16:0}$ and $\text{C}_{18:0}$ FAs must agree within 95 % confidence. Three failed, and seven did not yield sufficient C for both FAs to be dated

independently. These last ten sherds were therefore not further considered as no internal control on the C_{16:0} and C_{18:0} FAs was present to ensure the security of the dates (Supplementary materials S5).

Two of the pottery vessels dated in duplicate failed the internal quality control the first time, but either passed it the second time (BN-35) or yielded insufficient C for two targets (BN-101). Three pot dates that were duplicated gave indistinguishable dates for both extracts. The duplicate analysis of potsherd BN-74 produced statistically non-identical results between the two extractions. The CSRA dates successfully passed the internal criterion for both extractions and as the C_{16:0} and C_{18:0} dates are essentially independent, it is unlikely that both FAs in one extraction could be contaminated to the same degree (giving rise to identical, but inaccurate, dates; Casanova, *et al.* 2018). This difference could, therefore, reflect an inhomogeneous partitioning of the marine and terrestrial products in the same potsherd (due to different filling levels during cooking for example), and could potentially be monitored and corrected for in the future by recording $\delta^{13}\text{C}$ values on the two different TLEs (not performed in this case). Table 2 reports the combined measurements on the potsherds which passed the internal control.

These results suggest that the internal quality control is valid in this case of mixed marine/terrestrial resources and can be used as evidence for the reliability of the CSRA measurements. The error introduced by mixing the FAs of different abundances is likely below the AMS error, justifying the hold of the internal criteria.

Table 2: Summary of radiocarbon dated potsherds from Bornais, including the presence of aquatic biomarkers, $\delta^{13}\text{C}$ values of individual FAs, combined radiocarbon determinations of C_{16:0} and C_{18:0} FAs (which passed the internal criterion) and the percentage of marine fat/oil within the TLEs. The %_{marine} were calculated using reference ^{14}C measurements on marine-terrestrial samples (%_{marine} ^{14}C), using a linear mixing with $\delta^{13}\text{C}$ values on ruminant adipose products (%_{marine} $\delta^{13}\text{C}_{\text{adipose}}$) and $\delta^{13}\text{C}$ values on ruminant dairy products (%_{marine} $\delta^{13}\text{C}_{\text{milk}}$) as endmembers and finally using $\delta^{13}\text{C}$ values implemented in FRUITS (%_{marine} $\delta^{13}\text{C}_{\text{FRUITS}}$). *refers to the preferred endmembers for the terrestrial fats based on the $\Delta^{13}\text{C}$ value (i.e. milk if $\Delta^{13}\text{C} < -3.1\text{‰}$, ruminant adipose otherwise) and used for the %_{marine} calculation using FRUITS (v2.1; here the mean and standard deviation are presented and the full probability distribution are in supplementary material S6).

Phase	Layer	Pot#	Aquatic biomarkers	$\delta^{13}\text{C}_{16:0}$ (‰)	$\delta^{13}\text{C}_{18:0}$ (‰)	Laboratory#	Age $\pm 1 \sigma$ (BP)	% _{marine} ^{14}C	% _{marine} $\delta^{13}\text{C}_{\text{adipose}}$	% _{marine} $\delta^{13}\text{C}_{\text{milk}}$	% _{marine} $\delta^{13}\text{C}_{\text{FRUITS}}$
LIA 2	BAC	BN89 (1)	-	-26.8	-29.4	BRAMS-1549.1	1,368 $\pm$ 25	27 $\pm$ 12	30 $\pm$ 22*	37 $\pm$ 22	35 $\pm$ 10
		BN89 (2)	-	-	-	BRAMS-1549.2	1,365 $\pm$ 25	26 $\pm$ 12	-	-	-
		BN74 (1)	APAAs	-26.6	-29.4	BRAMS-1551.1	1,383 $\pm$ 30	31 $\pm$ 12	31 $\pm$ 23*	39 $\pm$ 16	36 $\pm$ 10
		BN74 (2)	-	-	-	BRAMS-1551.2	1,286 $\pm$ 25	4 $\pm$ 11	-	-	-
	BAF	BN77	-	-26.9	-29.9	BRAMS-1605	1,370 $\pm$ 24	-	26 $\pm$ 23*	34 $\pm$ 14	32 $\pm$ 10
		BN87	APAAs	-26.8	-30.4	BRAMS-1604	1,304 $\pm$ 24	-	21 $\pm$ 25	33 $\pm$ 15*	32 $\pm$ 12
	BAG	BN88 (1)	APAAs,	-24.2	-25.4	BRAMS-1548	1,757 $\pm$ 25	-	69 $\pm$ 31*	73 $\pm$ 27	71 $\pm$ 12

		BN88 (2)	DHYAs			BRAMS-1548	1,762 ± 25	-			
EN	BBD	BN35	APAAs, DHYAs	-25.4	-31.4	BRAMS-1552	1,156 ± 27	-	12 ± 12	31 ± 31*	33 ± 10
		BN105 (1)	-	-26.2	-31.7	BRAMS-1547.1	1,268 ± 25	-	8 ± 36	28 ± 24*	27 ± 10
		BN105 (2)	-			BRAMS-1547.2	1,327 ± 27	-			
	BBA	BN110	APAAs	-25.4	-25.9	BRAMS-1608	1,326 ± 25	-	64 ± 28*	63 ± 24	63 ± 13
MN	BCA	BN115	-	-27.2	-31.0	BRAMS-1609	987 ± 24	-	15 ± 24	28 ± 13*	27 ± 9
	BCC	BN160	-	-26.4	-31.0	BRAMS-2066	1,201 ± 25	74 ± 16	16 ± 31	32 ± 20*	29 ± 10
		BN165	-	-26.3	-30.1	BRAMS-2063	1,060 ± 25	32 ± 14	25 ± 28	38 ± 18*	36 ± 10
		BN174	APAAs	-25.9	-28.5	BRAMS-2062	1,115 ± 26	49 ± 15	40 ± 26*	48 ± 20	46 ± 12
LN	AG	BN36	APAAs, DHYAs	-27.3	-31.8	BRAMS-1607	786 ± 25	-	7 ± 26	24 ± 14*	21 ± 8

350

351 The four sherds from the BAC and BAF contexts of phase LIA2, the three from the EN phase
352 and the three from the BCC context of the MN phase were shown to have radiocarbon ages
353 between the age of the terrestrial organisms and their contemporaneous marine analogues
354 (Table 1 and 2). These include the five sherds (BN-89, BN-77, BN-105, BN160 and BN-165)
355 which did not exhibit aquatic biomarkers. These dates suggest, therefore, mixing of terrestrial
356 and marine resources in all the sherds.

357 The sherd BN-88 from the BAG context LIA2 phase exhibited not only the most enriched
358 $\delta^{13}\text{C}$ values but also the oldest age obtained in this investigation. This date is older than the
359 marine reference fish bone from this context and, indeed the reference fish bones from other
360 LIA 2 contexts. The second dating of the potsherd confirmed the accuracy of the compound-
361 specific ^{14}C measurement, suggesting the FAs likely derived from a pure marine fat/oil
362 residue and that the MRE (based on only one pair) was underestimated in this case, unless the
363 potsherd was residual and corresponds to the LIA1 phase, although no aquatic biomarkers
364 were detected in potsherds extracts from this particular phase.

365 The potsherd BN-115 (987 ± 24 BP) from context BCA (not dated) of the MN phase, which
366 lacked aquatic biomarkers exhibited an age consistent with the MN phase, and thus is likely
367 to be entirely composed of terrestrial animal fats.

368 For the LN phase, the FA date on the pot BN-36 (786 ± 25 BP) is younger than that of the
369 terrestrial organisms (BN-MB-7: 930 ± 25 BP). Based on the $\delta^{13}\text{C}$ values, the sherd plots
370 close to the reference dairy fat ellipses despite containing aquatic biomarkers. This result is
371 surprising and suggests that the dating of this phase, based on only one terrestrial organism
372 could be erroneous. Younger ages from other LN contexts from Bornais were obtained in a
373 range of 900 to 650 radiocarbon years BP (uncalibrated), which would support this
374 hypothesis (Marshall *et al.* 2016, forthcoming).

These measurements clearly confirm that lipid dates can be affected by the marine reservoir effect and such dates will therefore require calibration using relevant ΔR values and proportionately mixed terrestrial/marine curves. The mixing of marine and terrestrial products influences the determined $\delta^{13}\text{C}$ values and radiocarbon dates of FAs and this does not appear to have an adverse effect on the internal quality control criteria. Interestingly, MREs are evident in TLEs from potsherds lacking detectable aquatic biomarkers. The results therefore suggest that ^{14}C dates could be used to detect a (low-)level of marine organism processing in pots where aquatic biomarkers are undetectable. This would be especially relevant for sites where potential exists for processing of non-ruminant products or where aridity effects are possible, shifting the $\delta^{13}\text{C}$ values away from the ruminant products ellipses.

Correction of the MRE and calibration

The $\%_{\text{marine}}$ in the lipid residues was quantified for the sherds which passed the internal quality control criterion (Table 2; Supplementary material S5, S6). To ensure a fair evaluation of the use of FA $\delta^{13}\text{C}$ values for determination of the degree of marine/terrestrial product mixing, only potsherds from contexts which were securely dated using more than one marine/terrestrial organism were used for validating the correction and calibration (BAC and BCC). The validity of using $\delta^{13}\text{C}$ values of FAs for the quantification of marine-derived C was evaluated by comparison with reference values obtained by ^{14}C dates.

Overall, no significant differences in $\%_{\text{marine}}$ were noted in the use of $\delta^{13}\text{C}$ values from ruminant adipose or milk fats as terrestrial end members in a simple linear mixing model (Table 2, Figure 2f). Therefore, only the one most representative of the terrestrial endmembers was used for MRE corrections (i.e. milk if $\Delta^{13}\text{C} < -3.1\text{‰}$, ruminant adipose if $\Delta^{13}\text{C} > -3.1\text{‰}$).

The range of calibrated terrestrial dates on mammals for BRAMS-1710, BRAMS-1711, BRAMS-1713, in the LIA 2 phase, BAC context, were 672 - 773 cal AD, 662 - 769 cal AD and 685 - 772 cal AD, respectively (95% probability, Figure 3a). The $\%_{\text{marine}}$ within the FAs in pot BN-89 was determined to be $27 \pm 12\%$ using ^{14}C dates, $30 \pm 22\%$ using $\delta^{13}\text{C}$ values of adipose endmembers in the simple linear mixing and $35 \pm 10\%$ (mean and standard deviation) when implemented in FRUITS (Table 2). All these estimates are statistically

indistinguishable and the calibrated ages after MRE correction agrees with the reference age of the terrestrial organisms (Figure 3a).

Turning to potsherd BN-74, the first extract yielded estimates of 31 ± 12 % marine fat/oil using ^{14}C dates, and 31 ± 23 % and 36 ± 10 % using $\delta^{13}\text{C}$ values of adipose FAs endmembers on the mixing lines and FRUITS, respectively. The calibrated age from pot BN-74 (1st extract) agrees with the age of terrestrial organisms (Figure 3a). Nonetheless, the 2nd extract of the pot BN-74, which yielded results statistically different to the 1st extract, showed a %_{marine} of 4 ± 11 % using ^{14}C as end-members, suggesting an underestimation of the proportion of marine products in the TLE based on the $\delta^{13}\text{C}$ values in this case. As mentioned previously, this potsherd is likely affected by a differential deposition of the marine fats in certain areas of the vessel, implying that determination of $\delta^{13}\text{C}$ values and ^{14}C dates on the same TLE is required for a satisfactory quantification of the %_{marine} using $\delta^{13}\text{C}$ values.

Figure 3

The range of calibrated terrestrial dates (excluding outliers, Table 1) varies from 993 – 1,052 cal AD (55 % probability) and 1,081 – 1,152 cal AD (OxA-15522; 41% probability) to 1,039 – 1,206 cal AD (OxA-1540, 95 % probability) for the MN phase, BCC context (Figure 3b). The results for potsherds BN-165 using $\delta^{13}\text{C}$ values of milk fatty acids and BN174 using $\delta^{13}\text{C}$ values of adipose fatty acids showed, similarly to BN-89 and BN-74 (1st extraction), a good

agreement with the age of reference terrestrial animals using the different methods (Figure 3b).

The $\%_{\text{marine}}$ in the pot BN-160 is, however, estimated to be $74 \pm 16 \%$ with the use of ^{14}C dates, $26 \pm 34 \%$ and $29 \pm 10 \%$ with the use of $\delta^{13}\text{C}$ values milk endmembers in the linear mixing curve and FRUITS, respectively (Table 2). These results are not identical within a 1σ error but are within 2σ . The potsherd BN-160 was calibrated to $908 - 1,212 \text{ cal AD}$ by radiocarbon estimates, $730 - 1,044 \text{ cal AD}$ and to $780 - 1,016 \text{ cal AD}$ (95 % probability for all) by dairy endmember in linear mixing and FRUITS, respectively. The end of the last two distributions overlap only at the start of the calibration of the reference terrestrial organisms (Figure 3b). It should be noted that for potsherd BN-160, the ^{14}C dates suggest that marine fat/oil are dominate in the TLE whereas the $\delta^{13}\text{C}$ values suggest a dominance of dairy products. Unless the CSRA date is inaccurate, this implies that this potsherd, like BN-74, could be affected by a differential partitioning of the marine products and that $\delta^{13}\text{C}$ values recorded on another TLE are not representative of the TLE used for ^{14}C dating.

Overall, MRE corrections of lipid residues, using $\delta^{13}\text{C}$ calculations using the simple linear mixing model showed a wider probability distribution than those obtained using radiocarbon dates and $\delta^{13}\text{C}$ values used in the software FRUITS. However, the calibrated range of the corrected CSRA determinations on pot lipids using both methods clearly overlaps the calibrated range of the reference terrestrial organisms. The precision of the calibrated ages depends almost entirely on the uncertainties associated with the calculated $\%_{\text{marine}}$, as illustrated with reduced errors obtained using FRUITS software instead of the simple linear mixing curve. The results demonstrate no significant difference in the use of ruminant adipose or dairy $\delta^{13}\text{C}$ values as end members for the quantification of the $\%_{\text{marine}}$. In practice, however, one should be chosen over the other based on the $\Delta^{13}\text{C}$ values to ensure that the terrestrial endmember is representative of the animal products processed in the vessels at the time (i.e. dairy if $\Delta^{13}\text{C} < -3.1\%$ or adipose if $\Delta^{13}\text{C} > -3.1\%$). On the other hand, the ^{14}C dates provide an accurate estimate of the $\%_{\text{marine}}$ present in the FAs and could be used for quantification of marine products in TLEs instead of a calendar age. The $\%_{\text{marine}}$ in potsherds BN-74 (2nd extraction) and BN-160 were underestimated, leading to inappropriate corrections. However, this could be accounted for in the future if the ^{14}C measurements and $\delta^{13}\text{C}$ values are recorded on the same lipid extract to avoid potential inconsistencies

associated with inhomogeneous deposition of the lipids within vessels and use more reliable $\delta^{13}\text{C}$ values for the quantification of marine products.

One limitation of the $\delta^{13}\text{C}$ approach is the estimation of $\%_{\text{marine}}$ due to the wide range of reference values (from ca. -26 ‰ to -20 ‰) observed in modern marine organisms. The reference ellipses commonly plotted comprise only 68 % of the reference values (i.e. 1- σ). The average values used to generate an endmember here are not centred in the ellipses (Figure 2f). Therefore, potsherds with individual FAs $\delta^{13}\text{C}$ values plotting at the edge of the reference marine ellipse can be purely marine but, the $\%_{\text{marine}}$ deposited in the sherd can be underestimated using the linear mixing curves (Table 2, Figure 2f). This phenomenon is illustrated in the case of potsherd BN-88 which is likely to contain predominantly marine fats based on the CSRA dates. The $\delta^{13}\text{C}$ values plotted just outside the marine reference ellipse and marine-derived C was quantified to be 69 ± 31 % with adipose endmember, and the $\%_{\text{marine}}$ appeared to be underestimated in this case. Potsherd BN-110 could also contain a dominance of marine products based on fatty acid $\delta^{13}\text{C}$ values (i.e. 63 ± 28 % with adipose used as endmembers; Table 2, Figure 2f). This suggests that the linear mixing does not account particularly well for the dominance of marine products at the boundaries. Therefore, the use of the mean and standard deviation for the reported $\%_{\text{marine}}$ products would lead to some underestimation when applying MRE corrections in the case of dominance of marine products. This would be overcome using the full probability distribution calculated in FRUITS as prior information on the percentage marine.

We suggest that a linear mixing curve can give valid corrections if marine products are not dominant in the TLE, however, the FRUITS software would deal more adequately with the boundaries (if probability distribution are used) and should be used preferably to access the $\%_{\text{marine}}$ in the TLEs.

CONCLUSION

The processing of mixed terrestrial/marine fats in pottery vessels at the site of Bornais was revealed through lipid biomarker and CSRA analyses. CSRA and comparison with the radiocarbon dates of associated marine and terrestrial samples also enabled the detection of marine product processing in cases where no aquatic biomarkers were detected. We therefore suggest that in such circumstances, ^{14}C measurements could be used as a tracer for the

detection and quantification of marine products processing in pots. Compound-specific dates from potsherds from Bornais were successfully subjected to MRE correction, and assessed against independent ages determined for contemporaneous terrestrial organisms using:

- (i) An appropriate ΔR (-65 ± 45) for the site and time period.
- (ii) An estimate of the proportion of marine resource processed in the pots calculated using $\delta^{13}\text{C}$ values on individual pot FAs and from a modern reference database (linear mixing or implementation in FRUITS).
- (iii) A mixed calibration approach in OxCal software.

These corrected ages agreed well with the calibrated age of terrestrial samples which confirmed the efficacy of using FA $\delta^{13}\text{C}$ values to estimate the $\%_{\text{marine}}$, meaning that an approach similar to that commonly adopted for bone collagen can be used to correct for MRE present in lipids. For future MRE corrections and calibration on lipids dates, we recommend:

- (i) Calculating a ΔR for the site using a paired terrestrial/marine sample approach or using a previously published ΔR relevant for the spatiotemporal area.
- (ii) Recording $\delta^{13}\text{C}_{16:0}$ and $\delta^{13}\text{C}_{18:0}$ values from the same TLE as that used for ^{14}C dating to determine the $\%_{\text{marine}}$, avoiding the negative impact of potential inhomogeneity of lipid distribution in vessels.
- (iii) Using mixing model endmembers calculated from modern reference values valid for the location of interest, by using either those of the database for UK animals (excluding the species not present at the site; Copley, *et al.* 2003; Cramp and Evershed 2014) or $\delta^{13}\text{C}$ values recorded from reference animals, representative of other locations and environmental conditions (e.g. arid environments, Dunne *et al.* 2012).
- (iv) Using endmembers from dairy reference fats in the case of potsherds with $\Delta^{13}\text{C} < -3.1$ ‰ or using endmembers from the reference ruminant adipose fats values for potsherds with $\Delta^{13}\text{C} > -3.1$ ‰, to determine $\%_{\text{marine}}$ in the TLEs.

- (v) Employ FRUITS or other Bayesian approaches (if available) to quantify %_{marine} in the TLEs using a probability density function.
- (vi) Correct CSRA dates for the MRE using mixed atmospheric and marine calibration curves (e.g. in OxCal).

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663

Figure captions:

Figure 1: Flowchart showing the method used to assess the validity of the $\delta^{13}\text{C}$ values method for the estimation of the $\%_{\text{marine}}$ products and correction of the CSRA dates on the FAs.

Figure 2: Partial gas chromatogram of the TLE (a), and GC/MS SIM mass chromatograms showing detection of DHYAs (b) and APAAs (c), for potsherd BN-173. Scatter plots of $\delta^{13}\text{C}_{16:0}$ plotted against $\delta^{13}\text{C}_{18:0}$ from lipid residues characteristic of animal fats at Bornais for all the TLEs (Cramp *et al.*, 2014b, forthcoming and this study), (d) for the 22 potsherd extracts selected for ^{14}C dating by CSRA and position of the average reference values (crosses) (e), and the theoretical mixing lines of terrestrial and marine end-members with the approximate percentage of marine fat/oil marked on the lines (f). Stars denote the detection of aquatic biomarkers. Shaded areas indicate the reference ellipses for $\delta^{13}\text{C}$ values on modern animals and the crosses are the values used as endmembers. The dashed lines correspond to the areas where lipid residues are hypothesised to be affected to varying degrees by the MRE.

Figure 3: Corrections and calibration for potsherds of the (a) LIA phase, (b) MN phase in OxCal v4.3 against the IntCal13 calibration curve (Bronk Ramsey 2009, Reimer *et al.* 2013). The distributions plotted in dark grey correspond to the reference age of terrestrial animal bones, in light grey the uncorrected determinations on pot lipids and in black the corrected determinations on pot lipids using either the ^{14}C or $\delta^{13}\text{C}$ methods using adipose or milk as endmembers.

Supporting Materials

Compound-specific radiocarbon, stable carbon isotope and biomarker analysis of mixed marine/terrestrial lipids preserved in archaeological pottery vessels

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S1- Site location and stratigraphic information of Mound 2

Figure S1: Location of the site of Bornais and the 4 mounds excavated. From Sharples *et al.* (2016), Fig.17.1.

Figure S2: Schematic diagram showing the stratigraphic information for the Mound 2. Green contexts correspond to the ones studied in this paper.

S2- Detailed method for lipid residue analyses

To remove surface contaminants, a small part of the potsherd (~ 2 - 7 g) was cleaned with a modelling drill then sampled using hammer and chisel and ground to fine powder using mortar and pestle. Approximately 1 to 2 g of ground potsherd were weighed into a clean culture tube (I) with stopper and 20 µL of internal standard (IS; *n*-tetratriacontane) at 1 mg.mL⁻¹ was added. The lipids were extracted using a solution of H₂SO₄/MeOH (4% v/v, 5 mL, 70 °C, 1 h.). The supernatant of culture tube I was then centrifuged (2500 rpm, 10 min) and transferred to a clean culture tube (II) before adding double distilled water (2 mL). *N*-hexane was added (2 x 3 mL) to culture tube I and the supernatant transferred to culture tube II. Following this, 2 x 2 mL *n*-hexane was added directly to the H₂SO₄/MeOH solution in culture tube II and whirlimixed to extract the remaining residues, then transferred to the 3.5 mL vials and blown down until a full vial of *n*-hexane remained. A procedural blank was prepared and analysed alongside every batch of archaeological materials to assess whether contamination was introduced during the protocol. Before analysis, an aliquot of the total lipid extract (TLE; 1/4) was derivatised by the addition of BSTFA (*N,O*-bis(trimethylsilyl)trifluoroacetamide; 20 µL, 70 °C, 1 h). Excess BSTFA was blown down at 40 °C under a gentle stream of nitrogen, and an appropriate amount of *n*-hexane was added, prior to analysis with GC, GC-MS and GC-C-IRMS (Correa-Ascencio and Evershed 2014).

GC analysis of TLEs (Section 2.3.2.1) for quantification was performed on a Hewlett Packard 5890 series II gas chromatograph or an Agilent Technologies 7890A GC. Helium was used as carrier gas at constant flow (2 mL.min⁻¹), and a flame ionisation detector (FID) used to monitor column effluent. Lipids extracts (1 µL) were injected into a non-polar fused silica capillary column (50 m x 0.32 mm i.d., DB1 stationary phase (100 % dimethylpolysiloxane), 0.17 µm film thickness, Agilent technologies). The oven temperature program started with an isothermal hold at 50 °C for 2 min, then the temperature was increased at 10 °C.min⁻¹ to 300 °C and held for 10 min (Evershed *et al.* 1990).

GC-MS analysis of TLEs for molecular identification was performed on a Finnigan Trace MS quadrupole instrument coupled to a Trace GC, or on a Thermo Scientific ISQ LT single quadrupole GC-MS coupled to a Trace 1300, with manual or auto-sampling injections. The lipid extracts (1 µL) were introduced into a non-polar fused silica capillary column (50 m x 0.32 mm i.d., DB1 stationary phase, 0.17 µm film thickness, Agilent Technologies). For TLEs analysis the oven temperature program started with an isothermal hold at 50 °C during 2 min, then the temperature increased at 10 °C.min⁻¹ to 300 °C and held for 10 min. The MS used electron ionization (EI) mode operating at 70 eV with a GC interface temperature of 300 °C and a source temperature of 200 °C. Acquisition used the total ion current (TIC) mode over the range *m/z* 50-650 Daltons at 8.3 scans.s⁻¹ (Evershed *et al.* 1990). Screening for di-hydroxy fatty acid methyl esters (DHYAs, aquatic biomarkers) used selected ion monitoring

(SIM) mode, monitoring m/z 159, 187, 215, 243, 259, 287, 315, 443, 459, 471, 487, 499 and 515 (for -COOMe derivatives instead of -COOTMS as published; Cramp and Evershed 2014).

In order to determine the presence of other aquatic biomarkers (APAAs and isoprenoid acids), TLEs were run on a polar column (60 m x 0.32 mm i.d., VF-23ms stationary phase (polydimethylsiloxane highly substituted with cyanopropyl groups), 0.15 μm film thickness, Agilent Technologies). The temperature program started with an isothermal hold at 70 °C for 2 min, followed by a ramp at 10 °C.min⁻¹ to 220 °C, then a ramp at 4 °C.min⁻¹ to 300 °C and finally an isothermal hold for 10 min. Full scan mode m/z 50-650 and SIM mode, screening for the masses m/z 105, 262, 290, 318 and 346, were performed for the detection of APAAs (Cramp and Evershed 2014).

The GC-C-IRMS analyses on C_{16:0} and C_{18:0} FAs (for identification of the source of animal fats) was performed on an Agilent Technologies 7890A, coupled via an IsoPrime GC5 combustion interface (CuO and silver reactor, 850 °C) to an IsoPrime 100 mass spectrometer. The FAME extracts (1 μL) were injected into a non-polar column (50 m x 0.32 mm i.d., DB1 stationary phase, 0.17 μm film thickness, Agilent technologies). The GC oven temperature was held for 2 min at 40 °C and increased to 300 °C at 10 °C.min⁻¹ and held for 10 min. The MS used EI at 70 eV and had three Faraday cups collecting for the masses m/z 44, 45 and 46. Data were acquired and processed by the IonVantage software (Copley *et al.* 2003).

S3- Results of lipid residue analysis

Table S1: Results of lipid residue analysis of potsherds from the site of Bornais Mound 2, House 2. P corresponds to the Phytanic acid and TMDT to the 4,8,12-trimethyltridecanoic acid. Compounds in brackets corresponds to trace amounts (not clearly identified).

Sherd #	Block	Code	C ($\mu\text{g}\cdot\text{g}^{-1}$)	FAs	DHYAs	APAAs	IFAs	$\delta^{13}\text{C}_{16:0}$ (‰)	$\delta^{13}\text{C}_{18:0}$ (‰)	$\Delta^{13}\text{C}$ (‰)	Assignment (before CSRA dating)
BN-132	BCA	1259/3790	849	C ₁₄ -C ₁₈	-	-	(TMTD), P	-25.8	-31.0	-5.2	Mixture dairy, non-ruminant fats
BN-133	BCA	1280/9448/3/3887	137	C ₁₄ -C ₁₈	C ₁₈	-	-	-	-	-	nd
BN-134	BCB	6/8656	190	C ₁₄ -C ₂₂	-	C ₁₈ , C ₂₀ , C ₂₂	TMTD, P	-	-	-	nd
BN-135	BCB	1089/8654	1444	C ₁₄ -C ₂₂	C ₁₈ , C ₂₀ , C ₂₂	-	(TMTD), P	-26.3	-26.6	-0.2	Mixture ruminant adipose, marine fats
BN-136	BCB/C	1074/2/3529	207	C ₁₄ -C ₂₂	C ₁₈ , C ₂₀ , C ₂₂	C ₁₈	-	-26.7	-32.9	-6.2	Mixture dairy, marine fats
BN-137	BCC	182/1/8659	676	C ₁₄ -C ₂₂	-	C ₁₈ , C ₂₀ , C ₂₂	TMTD, P	-24.9	-28.4	-3.5	Mixture dairy, marine fats
BN-138	BCC	528/2/199	19	C ₁₄ -C ₂₂	-	-	-	-	-	-	nd
BN-139	BCC	549/2264	3135	C ₁₄ -C ₂₀	C ₁₈ , C ₂₂	C ₁₈	-	-26.6	-29.3	-2.7	Ruminant adipose fats
BN-140	BCC	550/5/2458	216	C ₁₄ -C ₂₂	C ₁₈ , C ₂₀ , C ₂₂	C ₁₈ , C ₂₀ , C ₂₂	TMTD, P	-22.1	-25.2	-3.1	Marine fats
BN-141	BCC	550/5/2458	28	C ₁₆ -C ₂₂	(C ₁₈)	-	-	-26.6	-32.5	-5.9	Mixture dairy, non-ruminant fats
BN-142	BCC	557/5/2341	2220	C ₁₄ -C ₂₂	C ₁₈ , C ₂₀ , C ₂₂	C ₁₈ , (C ₂₀)	(TMTD), P	-24.9	-27.9	-3.0	-
BN-143	BCC	557/5/8660	5630	C ₁₄ -C ₂₀	C ₁₈	C ₁₈	P	-25.8	-27.9	-2.2	Mixture ruminant adipose, marine fats
BN-144	BCC	558/5/8670	2483	C ₁₄ -C ₂₂	C ₁₈ , C ₂₀ , C ₂₂	C ₁₈	(TMTD), P	-25.6	-28.9	-3.3	Mixture dairy, marine fats
BN-145	BCC	565/8655	1425	C ₁₄ -C ₂₂	C ₁₈	-	P	-27.0	-31.3	-4.3	Mixture dairy, non-ruminant fats
BN-146	BCC	921/2/992	1349	C ₁₄ -C ₂₂	C ₁₈	C ₁₈	P	-25.6	-30.2	-4.6	Mixture dairy, non-ruminant fats
BN-147	BCC	1008/3063	56	C ₁₆ -C ₂₂	(C ₁₈)	-	-	-	-	-	nd
BN-148	BCC	1008/9452/8657	60	C ₁₄ -C ₂₂	C ₁₈ , C ₂₀ , C ₂₂	C ₁₈ , (C ₂₀ , C ₂₂)	TMTD, P	-25.3	-29.2	-4.0	Mixture dairy, marine fats
BN-149	BCC	1010/9685/2/3279	3984	C ₁₄ -C ₂₂	C ₁₈ , C ₂₀ , C ₂₂	C ₁₈	(TMTD), P	-26.3	-30.4	-4.1	Mixture dairy, marine fats
BN-150	BCC	1010/2/3266	1460	C ₁₄ -C ₂₂	C ₁₈ , (C ₂₀ , C ₂₂)	C ₁₈ , C ₂₀ , C ₂₂	TMTD, P	-25.2	-29.0	-3.8	Mixture dairy, marine fats
BN-151	BCC	1049/9809/8661	2141	C ₁₄ -C ₂₂	C ₁₈ , (C ₂₀ , C ₂₂)	C ₁₈	P	-26.9	-29.6	-2.7	Mixture ruminant, non-ruminant adipose fats
BN-152	BCC	1057/9895/8656	0	-	-	-	-	-	-	-	-
BN-153	BCC	1057/9894/3522	1409	C ₁₄ -C ₂₂	(C ₁₈)	C ₁₈	-	-26.5	-29.8	-3.3	Mixture dairy, non-ruminant fats
BN-154	BCC	1057/9893/3461	1153	C ₁₄ -C ₂₀	C ₁₈ , C ₂₀ , C ₂₂	C ₁₈ , (C ₂₀ , C ₂₂)	TMTD, P	-27.0	-30.1	-3.0	Mixture ruminant adipose, marine fats
BN-155	BCC	1057/9894/3535	4198	C ₁₄ -C ₂₂	-	-	-	-26.5	-28.6	-2.2	Mixture ruminant, non-ruminant adipose fats
BN-156	BCC	1057/9894/8666	276	C ₁₄ -C ₁₈	-	-	-	-25.3	-28.9	-3.6	Mixture dairy, non-ruminant fats
BN-157	BCC	1079/2/8671	11	C ₁₆ -C ₂₂	C ₁₈	-	TMTD, P	-	-	-	nd
BN-158	BCC	1220/9991/3662	12	-	(C ₁₈)	-	-	-	-	-	nd
BN-159	BCC	1220/9991/8664	1178	C ₁₄ -C ₂₀	C ₁₈	C ₁₈	P	-26.4	-32.0	-5.6	Mixture dairy, non-ruminant fats
BN-160	BCC	1234/9492/8665	4559	C ₁₄ -C ₂₂	C ₁₈	C ₁₈	(TMTD), P	-26.4	-31.0	-4.6	Mixture dairy, non-ruminant fats
BN-161	BCC	1260/9467/3779	546	C ₁₄ -C ₁₈	(C ₁₈ , C ₂₀ , C ₂₂)	-	-	-25.7	-31.2	-5.5	Mixture dairy, non-ruminant fats
BN-162	BCC	1260/9465/8672	1574	C ₁₆ , C ₁₈	(C ₁₈)	-	-	-26.7	-29.3	-2.6	Mixture ruminant, non-ruminant adipose fats
BN-163	BCC	2192/11902/10/2192	452	C ₁₄ -C ₁₈	C ₁₈	C ₁₈	-	-27.3	-31.5	-4.2	Mixture dairy, non-ruminant fats
BN-164	BCC	2225/11943/6230	129	C ₁₆ -C ₂₂	C ₁₈ , C ₂₀ , C ₂₂	C ₁₈	TMTD, P	-25.8	-31.6	-5.8	Mixture dairy, marine fats

Sherd #	Block	Code	C ($\mu\text{g}\cdot\text{g}^{-1}$)	FAs	DHYAs	APAAs	IFAs	$\delta^{13}\text{C}_{16:0}$ (‰)	$\delta^{13}\text{C}_{18:0}$ (‰)	$\Delta^{13}\text{C}$ (‰)	Assignment (before CSRA dating)
BN-165	BCC	2231/11968/14/6214	1823	C ₁₄ -C ₁₈	(C ₁₈)	-	-	-26.3	-30.1	-3.7	Mixture dairy, non-ruminant fats
BN-166	BCC	2258/11287/15/8663	246	C ₁₆ , C ₁₈	C ₁₈	C ₁₈ , C ₂₀	TMTD, P	-25.8	-31.7	-5.9	Mixture dairy, marine fats
BN-167	BCC	2264/6314	946	C ₁₄ -C ₂₂	(C ₁₈ , C ₂₀ , C ₂₂)	C ₁₈ , C ₂₀ , C ₂₂	TMTD, P	-23.5	-26.6	-3.1	Marine fats
BN-168	BCC	2264/14/6312	1105	C ₁₆ , C ₁₈	C ₁₈	C ₁₈	-	-29.2	-31.5	-2.3	Ruminant adipose fats
BN-169	BCC	2264/14/8667	2	C ₁₈ , C ₂₂	-	-	-	-	-	-	-
BN-170	BCC	2285/11278/19/8662	4231	C ₁₄ -C ₂₂	-	C ₁₈ , C ₂₀ , C ₂₂	TMTD, P	nd	nd	nd	Marine fats
BN-171	BCC	2297/11318/19/8658	872	C ₁₄ -C ₂₂	C ₁₈	C ₁₈	P	-26.2	-32.2	-6.0	Mixture dairy, non-ruminant fats
BN-172	BCC	2613/9/6398	834	C ₁₆ -C ₂₀	(C ₁₈ , C ₂₀)	C ₁₈	P	-26.1	-28.5	-2.4	Mixture ruminant, non-ruminant adipose fats
BN-173	BCC	2637/11398/6500	2376	C ₁₄ -C ₂₂	C ₁₈ , C ₂₀ , C ₂₂	C ₁₈ , C ₂₀ , C ₂₂	TMTD, P	-26.0	-30.5	-4.5	Mixture dairy, marine fats
BN-174	BCC	2657/11464/16/6523	1176	C ₁₄ -C ₂₂	C ₁₈ , C ₂₀ , C ₂₂	C ₁₈ , (C ₂₀ , C ₂₂)	(TMTD), P	-25.9	-28.5	-2.6	Mixture ruminant adipose, marine fats
BN-175	BCC	2673/11489/13/8669	41	C ₁₆ -C ₂₂	C ₁₈ , (C ₂₂)	C ₁₈ , C ₂₀ , C ₂₂	(TMTD), P	-25.6	-29.8	-4.2	Mixture dairy, marine fats
BN-176	BCC	2692/12041/7300	924	C ₁₆ -C ₂₀	C ₁₈ , (C ₂₀ , C ₂₂)	C ₁₈ , (C ₂₀ , C ₂₂)	P	-26.4	-29.0	-2.6	Mixture ruminant adipose, marine fats
BN-177	BCC	2700/12057/6631	497	C ₁₆ -C ₂₂	C ₁₈ , C ₂₀ , C ₂₂	C ₁₈ , (C ₂₀)	TMTD, P	-25.1	-30.3	-5.2	Mixture dairy, marine fats
BN-178	BCC	2715/12090/20/6637	280	C ₁₆ -C ₂₂	C ₁₈ , C ₂₀ , C ₂₂	C ₁₈ , C ₂₀ , (C ₂₂)	TMTD, P	-	-	-	-
BN-179	BCC	2731/1204/20/6686	1083	C ₁₄ -C ₂₀	(C ₁₈)	C ₁₈	P	-26.5	-29.8	-3.2	Mixture dairy, non-ruminant fats
BN-180	BCC	545+39/2501	536	C ₁₆ -C ₂₀	(C ₁₈)	C ₁₈ , C ₂₀ , C ₂₂	P	-24.4	-28.1	-3.7	Mixture dairy, marine fats

S4- Summary of published Δ determinations in the Hebrides Islands

Table S2: Summary of ΔR values published for the Hebridian Islands

Location	Site	$\Delta\text{R} \pm 1\sigma$	Time period	Reference
Outer Hebrides Lewis and Harris	Guinnessro	-130 $\pm$ 36	1,460 - 1,630 AD	Ascough <i>et al.</i> 2017
	Bostadh	-56 $\pm$ 14	893 - 984 AD	Ascough <i>et al.</i> 2009
	Garenin	-85 $\pm$ 17	887 - 995 AD	Ascough <i>et al.</i> 2009
	Traigh na Beirigh	-126 $\pm$ 39	4,540 - 4,240 BC	Ascough <i>et al.</i> 2017
	Northton	64 $\pm$ 41 79 $\pm$ 32	6,390 - 6,290 BC 6,390 - 6,230 BC	Ascough <i>et al.</i> 2017 Ascough <i>et al.</i> 2007
Outer Hebrides North Uist	Baleshare	-79 $\pm$ 17 68 $\pm$ 95	252 BC - 149 AD 77 BC - 111 AD	Ascough <i>et al.</i> 2004 Reimer <i>et al.</i> 2002
Outer Hebrides South Uist	Hornish point	-79 $\pm$ 17 -184 $\pm$ 122 -146 $\pm$ 71	252 BC - 149 AD 394 BC - 24 AD	Ascough <i>et al.</i> 2004 Reimer <i>et al.</i> 2002
Inner Hebrides (& Mainland)	Carding Mill Bay	150 $\pm$ 28	3,641 - 3,521 BC	Russell <i>et al.</i> 2015
		86 $\pm$ 67	3,942 - 3,653 BC	Reimer <i>et al.</i> 2002
		-44 $\pm$ 91	3,965 - 3,714 BC	Reimer <i>et al.</i> 2002
	Sand	64 $\pm$ 41 64 $\pm$ 19	6,480 - 6,420 BC 6,480 - 6,420 BC	Ascough <i>et al.</i> 2017 Ascough <i>et al.</i> 2007

S5- Details on CSRA dates

Table S3: Details of CSRA dates on pottery vessels and aquatic biomarker detection

Sample	Phase	Layer	BRAMS #	mCO ₂ (µg)	Age ± 1σ (BP)	σ range	Comments
BN89-C _{16:0}	LIA 2	BAC	1549.1.1	199	1396 ± 29	••	No aquatic biomarkers
BN89-C _{18:0}	LIA 2	BAC	1549.1.2	312	1336 ± 27		
BN89-C _{16:0}	LIA 2	BAC	1549.2.1	230	1404 ± 30		
BN89-C _{18:0}	LIA 2	BAC	1549.2.2	373	1322 ± 29		
BN74-C _{16:0}	LIA 2	BAC	1551.1.1	388	1394 ± 27	••	APAAs
BN74-C _{18:0}	LIA 2	BAC	1551.1.2	534	1369 ± 26		
BN74-C _{16:0}	LIA 2	BAC	1551.2.1	379	1299 ± 29		
BN74-C _{18:0}	LIA 2	BAC	1551.2.2	448	1273 ± 29		
BN77-C _{16:0}	LIA 2	BAF	1605.1.1	234	1364 ± 29	•	No aquatic biomarkers
BN77-C _{18:0}	LIA 2	BAF	1605.1.2	293	1375 ± 28		
BN87-C _{16:0}	LIA 2	BAF	1604.1.1	175	1292 ± 29	•	APAAs
BN87-C _{18:0}	LIA 2	BAF	1604.1.2	246	1315 ± 28		
BN88-C _{16:0}	LIA 2	BAG	1548.1.1	216	1720 ± 28	•	APAAs, DHYAs
BN88-C _{18:0}	LIA 2	BAG	1548.1.2	141	1729 ± 30		
BN88-C _{16:0}	LIA 2	BAG	1548.2.1	228	1726 ± 30		
BN88-C _{18:0}	LIA 2	BAG	1548.2.2	165	1728 ± 32		
BN35-C _{16:0}	EN	BBD	1552.1.1	112	1389 ± 33	X	APAAs, DHYAs
BN35-C _{18:0}	EN	BBD	1552.1.2	142	1255 ± 30		
BN35-C _{16:0}	EN	BBD	1552.2.1	136	1151 ± 33		
BN35-C _{18:0}	EN	BBD	1552.2.2	125	1161 ± 34		
BN91-C _{16:0}	EN	BBD	1603.1.1	207	1372 ± 29	-	APAAs, DHYAs - No internal control
BN91-C _{18:0}	EN	BBD	1603.1.2	79	-		
BN101-C _{16:0}	EN	BBD	1550.1.1	110	1033 ± 27	X	No aquatic biomarkers
BN101-C _{18:0}	EN	BBD	1550.1.2	105	1420 ± 31		
BN101-C _{16:0}	EN	BBD	1550.2.1	110	1469 ± 33		
BN101-C _{18:0}	EN	BBD	1550.2.2	70	-		
BN105-C _{16:0}	EN	BBD	1547.1.1	164	1288 ± 30	•	No aquatic biomarkers
BN105-C _{18:0}	EN	BBD	1547.1.2	128	1247 ± 31		
BN105-C _{16:0}	EN	BBD	1547.2.1	120	1370 ± 34		
BN105-C _{18:0}	EN	BBD	1547.2.2	117	1280 ± 34		
BN110-C _{16:0}	EN	BBA	1608.1.1	184	1360 ± 29	••	APAAs
BN110-C _{18:0}	EN	BBA	1608.1.2	153	1288 ± 30		
BN115-C _{16:0}	MN	BCA	1609.1.1	219	989 ± 28	•	No aquatic biomarkers
BN115-C _{18:0}	MN	BCA	1609.1.2	214	984 ± 29		
BN142-C _{16:0} C _{18:0}	MN	BCC	2069.1.1	238	1007 ± 29	-	DHYAs - No internal control
BN149-C _{16:0} C _{18:0}	MN	BCC	2064.1.1	145	1768 ± 34	-	DHYAs - No internal control
BN160-C _{16:0}	MN	BCC	2066.1.1	-	1230 ± 29	•	No aquatic biomarkers
BN160-C _{18:0}	MN	BCC	2066.1.2	216	1165 ± 31		
BN165-C _{16:0}	MN	BCC	2063.1.1	311	1080 ± 29	•	No aquatic biomarkers
BN165-C _{18:0}	MN	BCC	2063.1.2	285	1040 ± 29		
BN167-C _{16:0} C _{18:0}	MN	BCC	2068.1.1	225	1295 ± 31	-	APAAs, TMTD - No internal control
BN168-C _{16:0}	MN	BCC	2061.1.1	123	1211 ± 34	X	No aquatic biomarkers
BN168-C _{18:0}	MN	BCC	2061.1.2	253	1062 ± 34		
BN173-C _{16:0} C _{18:0}	MN	BCC	2067.1.1	167	1234 ± 32	-	APAAs, DHYAs, TMTD - No internal control
BN174-C _{16:0}	MN	BCC	2062.1.1	219	1152 ± 31	••	APAAs
BN174-C _{18:0}	MN	BCC	2062.1.2	296	1065 ± 33		
BN38-C _{16:0}	MN	AD	1606.1.1	121	871 ± 31	-	No aquatic biomarkers - No internal control
BN36-C _{16:0}	LN	AG	1607.1.1	201	767 ± 29	••	APAAs, DHYAs
BN36-C _{18:0}	LN	AG	1607.1.2	115	816 ± 31		

S6- Quantification of marine derived-C using FRUITS (v2.1)

Table S4: Determination of the percentage marine products in pottery vessels CSRA dated using FRUITS.

Target/ Consumer	Source/Food	Mean	sd	2.5pc	median	97.5pc
BN89	ruminant adipose	0.646	0.1001	0.4117	0.6601	0.8014
	marine	0.354	0.1001	0.1986	0.3399	0.5884
BN74	ruminant adipose	0.6361	0.09734	0.4126	0.6486	0.7901
	marine	0.3639	0.09734	0.21	0.3514	0.5875
BN174	ruminant adipose	0.537	0.1158	0.2521	0.5545	0.7138
	marine	0.463	0.1158	0.2862	0.4455	0.7491
BN160	milk	0.7067	0.09375	0.5054	0.7157	0.8641
	marine	0.2933	0.09375	0.136	0.2843	0.4951
BN165	milk	0.6372	0.1027	0.389	0.6489	0.7997
	marine	0.3628	0.1027	0.2004	0.3511	0.6111
BN77	ruminant adipose	0.6823	0.09332	0.4724	0.6928	0.8323
	marine	0.3177	0.09332	0.1678	0.3072	0.5276
BN88	ruminant adipose	0.2925	0.1237	0.03621	0.3041	0.505
	marine	0.7075	0.1237	0.4951	0.696	0.9638
BN110	ruminant adipose	0.3656	0.1253	0.08533	0.3807	0.5757
	marine	0.6344	0.1253	0.4243	0.6193	0.9148
BN87	milk	0.6823	0.08895	0.4844	0.6914	0.8288
	marine	0.3177	0.08895	0.1713	0.3086	0.5156
BN35	milk	0.6692	0.1059	0.4316	0.681	0.8465
	marine	0.3308	0.1059	0.1536	0.319	0.5685
BN105	milk	0.7343	0.094	0.5228	0.7439	0.8934
	marine	0.2657	0.094	0.1067	0.2561	0.4774
BN115	milk	0.4731	0.1099	0.2199	0.4864	0.6534
	marine	0.5269	0.1099	0.3468	0.5136	0.7803
BN36	milk	0.4835	0.1136	0.2177	0.4982	0.6682
	marine	0.5165	0.1136	0.3323	0.5018	0.7824

Figure S3: Box and whisker plot for the marine contribution in potsherds dominated by (a) ruminant adipose fats and (b) dairy fats for the potsherds CSRA dated and corrected in the main paper (from FRUITS v2.1).

Figure S4: Probability distribution for the proportion of marine (green) and terrestrial (red) resources for the potsherds CSRA dated and corrected in the main paper (from FRUITS v2.1)

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