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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 (^{14}C) analyses. Here we studied the site of Bornais (Outer Hebrides, UK) where a local MRE, ΔR of -65 ± 45 yr 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 $\delta^{13}C$ values of $C_{16:0}$ and $C_{18:0}$ FAs revealed ruminant and marine product mixing for 71% of the vessels. Compound-specific ^{14}C analysis (CSRA) of FAs yielded intermediate ^{14}C ages between those of terrestrial and marine bones from the same contexts, confirming an MRE existed. A database containing $\delta^{13}C$ values for FAs from reference terrestrial and marine organisms provided endmembers for calculating the percentage marine-derived C ($\%_{marine}$) in FAs. We show that lipid ^{14}C dates can be corrected using determined $\%_{marine}$ and ΔR values, such that pottery vessels from coastal locations can be ^{14}C dated by CSRA of FAs.

KEYWORDS: compound-specific radiocarbon analysis, lipid residues, marine reservoir effect, mixed marine/terrestrial corrections, pottery vessels.

INTRODUCTION

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

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

Marine product processing in pots can be identified by the presence of specific aquatic biomarkers alongside the $C_{16:0}$ and $C_{18:0}$ FAs, namely: (i) long-chain dihydroxy fatty acids (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, $\delta^{13}C$ values determined for the $C_{16:0}$ and $C_{18:0}$ 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 $C_{16:0}$ and $C_{18:0}$ FAs differ between terrestrial and marine organisms and the relationship between their mixing proportions and the resulting $\delta^{13}C$ 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 $C_{16:0}$ and $C_{18:0}$ FAs in a potsherd are no longer consistent within 2-σ (Casanova et al. 2018, in press). It is certainly possible that $C_{16:0}$ and $C_{18:0}$ FAs in sherds arising from mixtures of terrestrial- and marine-derived food residues may yield different apparent ^{14}C 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 ($\%_{marine}$) present (Cook et al. 2015). The ΔR values can be obtained by ^{14}C dating historical marine specimens (of known date of collection), pairing ^{14}C measurements on terrestrial and marine organisms from secure contexts at the site of interest or by both dating tephra layers deposited at sea and on land (Ascough et al. 2005). The evaluation of the $\%_{marine}$, 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, $\delta^{13}C$ and $\delta^{15}N$ 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 ^{14}C 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 focused 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 of pottery vessels including compound-specific $\delta^{13}C$ 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) ^{14}C 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

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marine and terrestrial lipids, and (v) application of relevant marine reservoir corrections to the ^{14}C 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 four mounds with a long duration of occupation defined by 109 ^{14}C 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 (ca.18,000 bones, including those of small vertebrates, birds, fish and mollusks; 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 mollusk shells (limpets and winkles) were extremely abundant at the site (ca. 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 analyzed 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 δ^{13}C values of FAs were determined by GC-Combusted-Isotope ratio MS (GC-C-IRMS; supplementary material S2).

Pretreatment Methods for ^{14}C Analyses

Approximately 300 mg of coarse bone powder were weighed into a culture tube and pretreated 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, ~18 hr, 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 × 10 mL) in between each acid and base wash and centrifuged (3000 rpm, 5 min). The collagen was then gelatinized at pH 3 with HCl (0.001 M, 10 mL, 75 °C, 20 hr) and filtered through precombusted glass fiber 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 × 10 mL) and dried at 60 °C in a drying cabinet (Knowles et al. 2019).

Sherds containing lipid concentrations, typically above 500 $\mu g.g^{-1}$, were selected for ^{14}C determinations. Pieces of 2–10 g of the potsherd was sampled, depending on the lipid

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concentrations and size of the potsherds. The lipids were extracted in culture tubes using H_2SO_4/MeOH (4% *v*/*v*, 3 x 8 mL, 70°C, 1 hr). Samples were centrifuged after each extraction (2500 rpm, 10 min) and the three supernatants (methanolic fractions) combined into a second culture tube containing double-distilled water (5 mL). The lipids, including fatty acid methyl esters (FAMEs) formed from the reaction of methanol with the FAs during the first step, were extracted from the methanolic solution with *n*-hexane (4 × 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 hr). 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 ca. 5 $\mu g.\mu L^{-1}$ 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 GC (pcGC) instrument following the methods described in Casanova et al. (2017, 2018, in press).

^{14}C Determinations

Organic materials (FAMEs and collagen) were combusted to CO_2 using a Vario Microcube Elemental Analyser (EA, Elementar). The shells (carbonate-based) were digested in H_3PO_4 (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 CO_2. Resulting CO_2 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 CO_2 was reduced to graphite under H_2 (580°C, 2 hr, 420 mbar) on a preconditioned iron catalyst. A Pneumatic Sample Press (PSP, Ionplus) was used to press the graphitized samples into Al targets.

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

Corrections and Calibration of ^{14}C Measurements

^{14}C measurements 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-σ 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 v4.3 (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).

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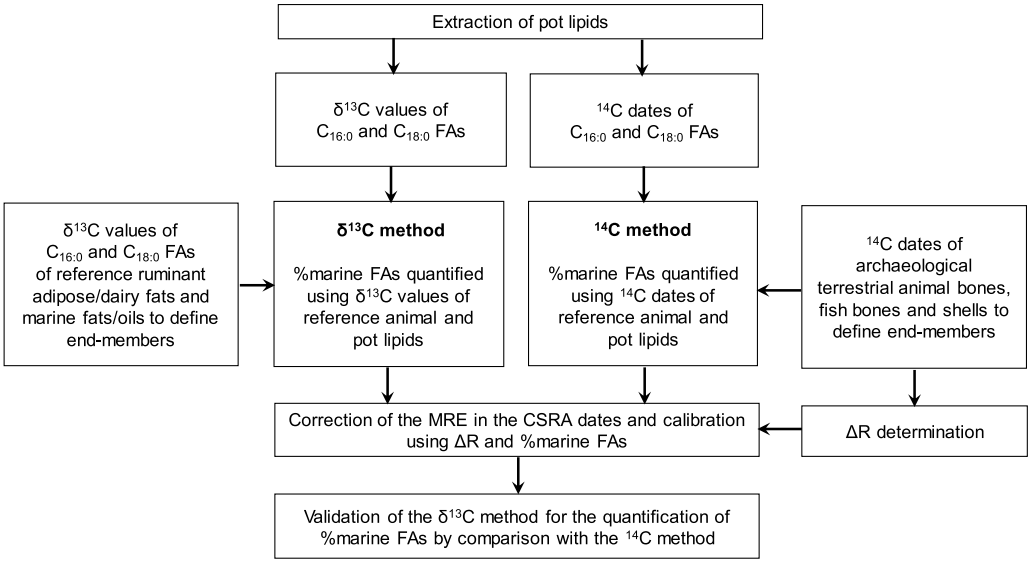

Figure 1 Flowchart showing the methods used to assess the validity of the $\delta^{13}C$ values method for the estimation of the $\%_{marine}$ products and correction of the CSRA dates on the FAs.

The mixing of marine/terrestrial commodities was quantified in each potsherd using two independent methods: ^{14}C dates and $\delta^{13}C$ 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 $\delta^{13}C$ 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 ^{14}C dates. This is 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 ^{14}C ages.

The first method of quantifying the $\%_{marine}$ is based on the weighted average of ^{14}C determinations on the short-lived terrestrial and marine organisms, from the same context/phase as the potsherds dated, as endmembers using Equation (1).

$$\%_{marine} = \frac{\left(Age_{pot} - Age_{terr}\right)}{\left(Age_{marine} - Age_{terr}\right)} \times 100 \quad (1)$$

Where $\%_{marine}$ is the percentage of aquatic C in the lipid residue and Age_{pot}, Age_{terr}, Age_{marine} are the combined ^{14}C ages on the individual FAs, for terrestrial animals and marine organisms, respectively.

The second method uses the $\delta^{13}C$ 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 hypothesized 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 $C_{16:0}$ and $C_{18:0}$ FAs and were found to be $\delta^{13}C_{16:0} = -30.0 \pm 0.6‰$ and $\delta^{13}C_{18:0} = -32.2 \pm 0.6‰$ for adipose fats and $\delta^{13}C_{16:0} = -29.2 \pm 1.0‰$ and $\delta^{13}C_{18:0} = -34.0 \pm 0.9‰$ for dairy fats. Both ruminant

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adipose and dairy values were used as endmembers to evaluate whether one should be used over the other. The marine endmembers were $\delta^{13}C_{16:0} = -22.7 \pm 2.2‰$ and $\delta^{13}C_{18:0} = -21.7 \pm 2.5‰$. The relationship between the relative proportions of marine and terrestrial fats and the $\delta^{13}C$ 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 $\%_{marine}$ contribution and associated uncertainty using the propagation of analytical errors. The $\%_{marine}$ 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 $\%_{marine}$ values must be constrained between 0 and 100%.

As a comparison, the $\%_{marine}$ 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 $\%_{marine}$ after Bayesian modeling was exported and implemented as a prior information file into the mixing marine/terrestrial tool in OxCal.

RESULTS AND DISCUSSION

Characterization 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 $\mu g.g^{-1}$ 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 $C_{16:0}$ and $C_{18:0}$ FAs 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 three classes of aquatic biomarkers, 21% ($n = 10$) contained two 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 $\delta^{13}C$ values of the palmitic and stearic acids were determined by GC-C-IRMS (Figure 2d). Significantly, the $C_{16:0}$ and $C_{18:0}$ FAs displayed $\delta^{13}C$ 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 conductive to the formation of thermally produced aquatic biomarkers (APAAs). The use of $\Delta^{13}C$ ($= \delta^{13}C_{18:0} - \delta^{13}C_{16:0}$) values allows the identification of sherds where FAs are predominantly of dairy product

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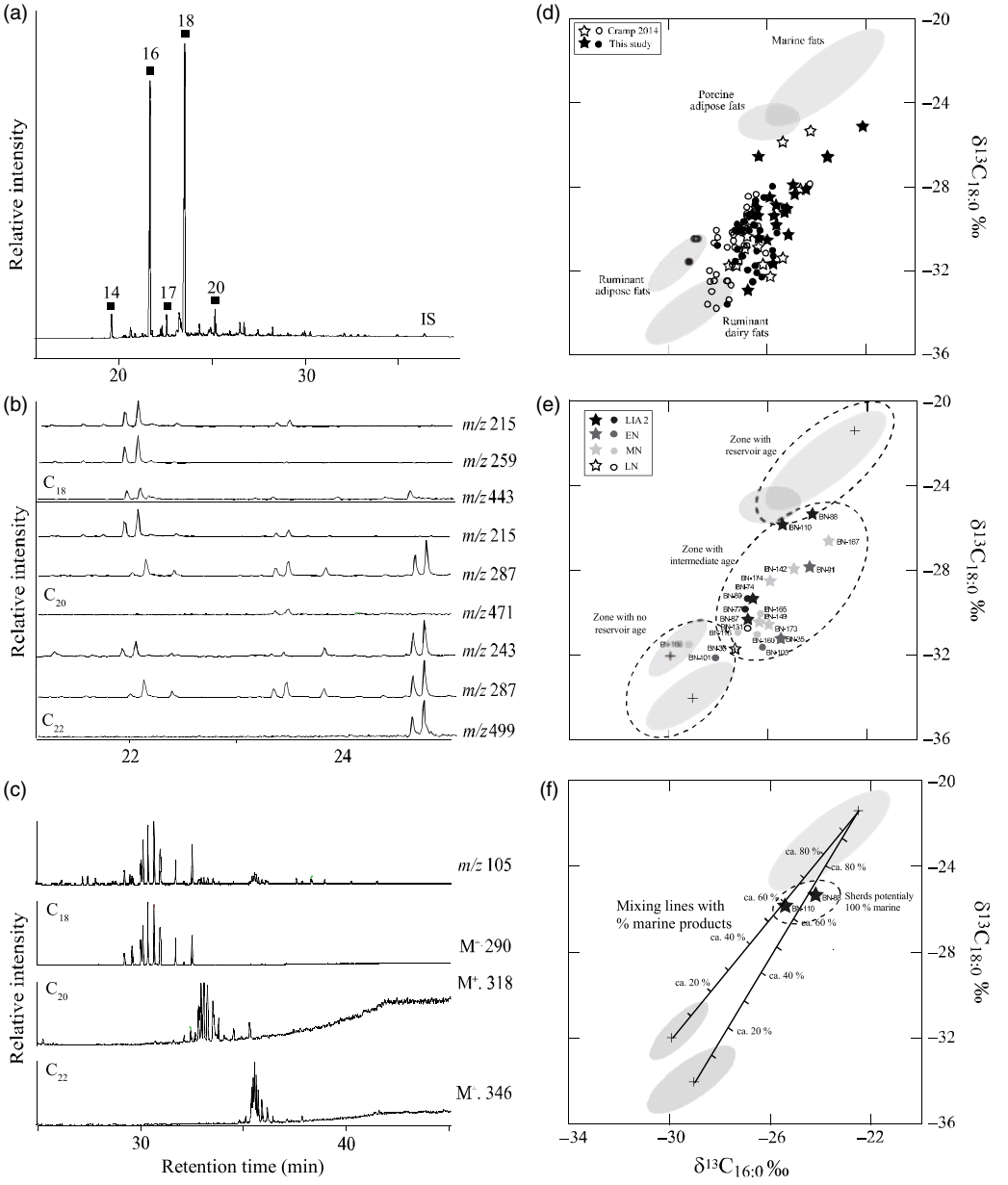

Figure 2 Partial gas chromatogram of the TLE (a), and GC/MS SIM mass chromatograms showing detection of DHYAs (b) and APAAs (c), for potsherd BN-173. Scatter plots of $\delta^{13}C_{16:0}$ plotted against $\delta^{13}C_{18:0}$ from lipid residues characteristic of animal fats at Bornais for all the TLEs (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 $\delta^{13}C$ values of modern animals and the crosses are the values used as endmembers. The dashed lines correspond to the areas where lipid residues are hypothesized to be affected to varying degrees by the MRE.

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origin (< –3.1‰; Copley et al. 2003). A total of 22 sherds yielded $\Delta^{13}C$ 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 $\Delta^{13}C$ 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 analyzed 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 LIA1 phase lacked aquatic biomarkers.

A total of 21 potsherds (from all phases) with sufficient lipid concentration 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 mollusk shells ($n = 14$) and terrestrial animal bones ($n = 8$) were ^{14}C dated (Table 1). These were assessed together with other available ^{14}C 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 ^{14}C 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 mollusk 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 for fish bones).

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 ΔRs failed the statistical identicality test, therefore layer BAG ($\Delta R = -214 \pm 26$), 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

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Table 1 ^{14}C determinations of terrestrial and marine organisms from Bornais and **ΔR**s calculated for the diverse contexts based on the multiple paired terrestrial/marine organisms. *refers to statistical outliers that have been excluded from **ΔR** calculation.

		Terrestrial organisms			Marine organisms			
Phase	Layer	Material	Lab nr	Conventional ^{14}C age	Material	Lab nr	Conventional ^{14}C age	ΔR
LIA2	BAC	Cattle	BRAMS-1710	1272 ± 25	Limpet-1	BRAMS-1727	1624 ± 26	–47 ± 23
		Caprine	BRAMS-1711	1298 ± 25	Limpet-2	BRAMS-1728	1627 ± 26	
		Unidentified	BRAMS-1713.1	1258 ± 25	Limpet-5	BRAMS-1731	1642 ± 26	
			BRAMS-1713.2	1264 ± 25	Limpet-6	BRAMS-1732	1616 ± 26	
	BAF	Cattle	BRAMS-1712	1348 ± 25	Fish-9	BRAMS-1725*	1651 ± 25	–165 ± 26
					Fish-10	BRAMS-1726*	1294 ± 25	
					Limpet-3	BRAMS-1729	1565 ± 26	
					Limpet-4	BRAMS-1730	1584 ± 26	
	BAG	Cattle	BRAMS-1708	1320 ± 25	Fish-1	BRAMS-1717	1509 ± 25	–214 ± 26
		Cattle	BRAMS-1709	1320 ± 25				
EN	BBD	Cattle	BRAMS-1715*	1082 ± 25	—			—
		Cattle	BRAMS-1719*	945 ± 25				
	BBA	—			Limpet-7	BRAMS-1733	1622 ± 26	—
MN	BCC	Cattle	SUERC-2684	925 ± 35	Fish-11	BRAMS-2049 (c)	1306 ± 25	–102 ± 35 (a)
		Red deer	SUERC-22894*	875 ± 30	Fish-12	BRAMS-2050 (c)	1318 ± 25	248 ± 37 (b)
		Pig	SUERC-22890*	1035 ± 30	Fish-13	BRAMS-2051 (c)	1323 ± 25	–26 ± 40 (c)
		Seed	GU-18290	—	Fish-14	BRAMS-2052 (c)	1365 ± 25	35 ± 150
		Cattle	SUERC-22896	970 ± 30	Fish-15	BRAMS-2053 (c)	1308 ± 25	(all)
		Cattle	SUERC-22897	975 ± 25	Limpet-8	BRAMS-2041*	1380 ± 24	
		Sheep	OxA-15420	903 ± 27	Limpet-9	BRAMS-2042.1 (a)	1263 ± 25	
		Cattle	OxA-15522	985 ± 26		BRAMS-2042.2 (a)	1236 ± 24	
					Limpet-10	BRAMS-2043(b)*	1575 ± 24	
					Limpet-11	BRAMS-2044 (b)*	1593 ± 25	

Table 1 (*Continued*)

		Terrestrial organisms			Marine organisms			
Phase	Layer	Material	Lab nr	Conventional ^{14}C age	Material	Lab nr	Conventional ^{14}C age	ΔR
					Winkle-1	BRAMS-2045 (a)	1257 ± 24	
					Winkle-2	BRAMS-2046 (b)*	1614 ± 25	
					Winkle-3	BRAMS-2047 (a)	1243 ± 24	
					Winkle-4	BRAMS-2048 (b)*	1613 ± 25	
	AD	Cattle	BRAMS-1716	956 ± 25	Fish-4	BRAMS-1720	1167 ± 25	–84 ± 34
					Fish-5	BRAMS-1721	1261 ± 25	
					Fish-6	BRAMS-1722	1257 ± 25	
LN	AG	Sheep	BRAMS-1714	930 ± 25	Fish-7	BRAMS-1723.1*	1268 ± 25	—
						BRAMS-1723.2*	1237 ± 25	
					Fish-8	BRAMS-1724*	1183 ± 25	
Overall site								–65 ± 46

statistically identical (T' = 69.3, T'(5%) = 71.0, υ = 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) Hebridian Islands of –47 ± 52 for the period 3500 BC-1450 cal AD (Reimer et al. 2002; Ascough et al. 2004, 2005, 2006, 2007, 2009, 2017; Russell et al. 2010, 2015; see supplementary material S4).

^{14}C Dating of Pottery Vessels

$C_{16:0}$ and $C_{18:0}$ 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 ^{14}C dates of the $C_{16:0}$ and $C_{18:0}$ 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 δ^{13}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 and the internal quality control criterion is still applicable.

The four sherds from the BAC and BAF contexts of phase LIA2, the three from the EN phase and the three from the BCC context of the MN phase were shown to have ^{14}C ages between the age of the terrestrial organisms and their contemporaneous marine analogues (Tables 1 and 2). These include the five sherds (BN-89, BN-77, BN-105, BN160 and BN-165) which did not exhibit aquatic biomarkers. These dates suggest, therefore, mixing of terrestrial and marine resources in all the pots from which the sherds derived.

The sherd BN-88 from the BAG context LIA2 phase exhibited not only the most enriched δ^{13}C values but also the oldest age obtained in this investigation. This date is older than the marine reference fish bone from this context and, indeed the reference fish bones from other LIA2 contexts. The second dating of the potsherd confirmed the accuracy of the compound-specific ^{14}C measurement, suggesting the FAs likely derived from a pure marine fat/oil residue and that the MRE (based on only one pair) was underestimated in this case, unless

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Table 2 Summary of ^{14}C dated potsherds from Bornais, including the presence of aquatic biomarkers, $\delta^{13}C$ values of individual FAs, combined ^{14}C determinations of $C_{16:0}$ and $C_{18:0}$ FAs (which passed the internal quality control 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 $\delta^{13}C$ values on ruminant adipose products ($\%_{marine}$ $\delta^{13}C_{adipose}$) and $\delta^{13}C$ values on ruminant dairy products ($\%_{marine}$ $\delta^{13}C_{milk}$) as endmembers and finally using $\delta^{13}C$ values implemented in FRUITS ($\%_{marine}$ $\delta^{13}C$ FRUITS). *refers to the preferred endmembers for the terrestrial fats based on the $\Delta^{13}C$ value (i.e. milk if $\Delta^{13}C < -3.1‰$, 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 distributions are in supplementary material S6).

Phase	Layer	Pot#	Aquatic biomarkers	$\delta^{13}C_{16:0}$ (‰)	$\delta^{13}C_{18:0}$ (‰)	Lab nr	Age ± 1 σ (BP)	$\%_{marine}$ ^{14}C	$\%_{marine}$ $\delta^{13}C_{adipose}$	$\%_{marine}$ $\delta^{13}C_{milk}$	$\%_{marine}$ $\delta^{13}C$ FRUITS
LIA2	BAC	BN89 (1)	—	–26.8	–29.4	BRAMS-1549.1	1368 ± 25	27 ± 12	30 ± 22*	37 ± 22	35 ± 10
		BN89 (2)				BRAMS-1549.2	1365 ± 25	26 ± 12			
		BN74 (1)	APAAs	–26.6	–29.4	BRAMS-1551.1	1383 ± 30	31 ± 12	31 ± 23*	39 ± 16	36 ± 10
		BN74 (2)				BRAMS-1551.2	1286 ± 25	4 ± 11			
	BAF	BN77	—	–26.9	–29.9	BRAMS-1605	1370 ± 24	—	26 ± 23*	34 ± 14	32 ± 10
		BN87	APAAs	–26.8	–30.4	BRAMS-1604	1304 ± 24	—	21 ± 25	33 ± 15*	32 ± 12
	BAG	BN88 (1)	APAAs,	–24.2	–25.4	BRAMS-1548.1	1757 ± 25	—	69 ± 31*	73 ± 27	71 ± 12
		BN88 (2)	DHYAs			BRAMS-1548.2	1762 ± 25				
EN	BBD	BN35	APAAs, DHYAs	–25.4	–31.4	BRAMS-1552	1156 ± 27	—	12 ± 12	31 ± 31*	33 ± 10
		BN105 (1)	—	–26.2	–31.7	BRAMS-1547.1	1268 ± 25	—	8 ± 36	28 ± 24*	27 ± 10
		BN105 (2)				BRAMS-1547.2	1327 ± 27	—			
	BBA	BN110	APAAs	–25.4	–25.9	BRAMS-1608	1326 ± 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	1201 ± 25	74 ± 16	16 ± 31	32 ± 20*	29 ± 10
		BN165	—	–26.3	–30.1	BRAMS-2063	1060 ± 25	32 ± 14	25 ± 28	38 ± 18*	36 ± 10
		BN174	APAAs	–25.9	–28.5	BRAMS-2062	1115 ± 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

the potsherd was residual and corresponds to the LIA1 phase, although no aquatic biomarkers were detected in potsherd extracts from this particular phase.

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

For the LN phase, the FA date on pot BN-36 (786 ± 25 BP) is younger than that of the terrestrial organism (BN-MB-7: 930 ± 25 BP). Based on the $\delta^{13}C$ values, the sherd plots close to the reference dairy fat ellipses despite containing aquatic biomarkers. This result is surprising and suggests that the dating of this phase, based on only one terrestrial organism could be erroneous. Younger ages from other LN contexts from Bornais were obtained in the range 900 to 650 ^{14}C years BP (uncalibrated), which would support this 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 $\delta^{13}C$ values and ^{14}C dates of FAs and this does not appear to have an adverse effect on the internal quality control criterion. 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 $\delta^{13}C$ values away from the ruminant products ellipses.

Correction of the MRE and Calibration

The $\%_{marine}$ 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 $\delta^{13}C$ 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 $\delta^{13}C$ 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 $\%_{marine}$ were noted in the use of $\delta^{13}C$ 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 $\Delta^{13}C < -3.1‰$, ruminant adipose if $\Delta^{13}C > -3.1‰$).

The range of calibrated terrestrial dates on mammals for BRAMS-1710, BRAMS-1711, BRAMS-1713, in the LIA2 phase, BAC context, were 672–773 cal AD, 662–769 cal AD, and 685–772 cal AD, respectively (95% probability, Figure 3a). The $\%_{marine}$ within the FAs in pot BN-89 was determined to be 27 ± 12% using ^{14}C dates, 30 ± 22% using $\delta^{13}C$ values of adipose endmembers in the simple linear mixing and 35 ± 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).

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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 $\delta^{13}C$ values of adipose FAs as endmembers for the mixing lines and FRUITS, respectively. The calibrated age from pot BN-74 (first extract) agrees with the age of terrestrial organisms (Figure 3a). Nonetheless, the second extract of the pot BN-74, which yielded results statistically different to the first 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 $\delta^{13}C$ values in this case. As mentioned previously, this potsherd is likely affected by inhomogeneous deposition of the marine fats in certain areas of the vessel, implying that determination of $\delta^{13}C$ values and ^{14}C dates on the same TLE is required for satisfactory quantification of the $\%_{marine}$ using $\delta^{13}C$ values.

The range of calibrated terrestrial dates (excluding outliers, Table 1) varies from 993–1052 cal AD (55% probability) and 1081–1152 cal AD (OxA-15522; 41% probability) to 1039–1206 cal AD (OxA-1540, 95% probability) for the MN phase, BCC context (Figure 3b). The results for potsherds BN-165 using $\delta^{13}C$ values of milk FAs and BN174 using $\delta^{13}C$ values of adipose FAs showed, similarly to BN-89 and BN-74 (first extract), a good agreement with the age of reference terrestrial animals using the different methods (Figure 3b).

The $\%_{marine}$ in the pot BN-160 is, however, estimated to be 74 ± 16% based on ^{14}C dates, 26 ± 34% and 29 ± 10% based on $\delta^{13}C$ values milk endmembers in the linear mixing curve and FRUITS, respectively (Table 2). These results are not identical within a 1-σ error but are within 2-σ. The potsherd BN-160 was calibrated to 908–1212 cal AD by ^{14}C estimates, 730–1044 cal AD and to 780–1016 cal AD (95% probability for all) using the dairy endmember in the linear mixing model 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 dominant in the TLE whereas the $\delta^{13}C$ 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 $\delta^{13}C$ values recorded on the initial TLE are not representative of the second TLE used for ^{14}C dating.

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

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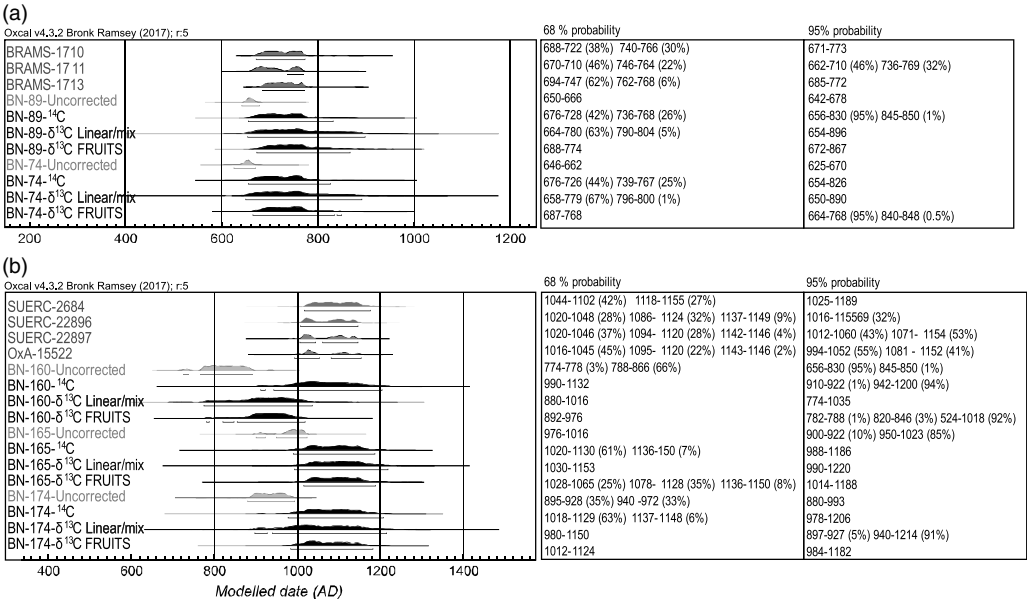

Figure 3 Corrections and calibrations for potsherds of the (a) LIA phase, (b) MN phase in OxCal v4.3 against the IntCal13 calibration curve (Bronk Ramsey 2009; Reimer et al. 2013). The distributions plotted in dark grey correspond to the reference age of terrestrial animal bones, in light grey the uncorrected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

the same lipid extract to avoid potential inconsistencies associated with inhomogeneous deposition of the lipids within vessels and use more reliable δ^{13}C values for the quantification of marine products.

One limitation of the δ^{13}C approach is the estimation of $\%_{marine}$ 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 δ^{13}C values plotting at the edge of the reference marine ellipse can be purely marine but, the $\%_{marine}$ 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 δ^{13}C values plotted just outside the marine reference ellipse and marine-derived C was quantified to be 69 ± 31% with the adipose endmember, and the $\%_{marine}$ appeared to be underestimated in this case. Potsherd BN-110 could also contain a dominance of marine products based on FA δ^{13}C values (i.e. 63 ± 28% with adipose FAs 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 $\%_{marine}$ products would lead to some underestimation when applying MRE corrections in the case of a dominance of marine products. This would be overcome using the full probability distribution calculated in FRUITS as prior information on the percentage marine.

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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 $\%_{marine}$ 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 ^{14}C dates of associated marine and terrestrial samples also enabled the detection of marine product processing in cases were 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 product 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 $\delta^{13}C$ values of 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 reference materials which confirmed the efficacy of using FA $\delta^{13}C$ values to estimate the $\%_{marine}$, 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 calibrations of lipids dates, we recommend:

(i) Calculating a ΔR for the site using a paired terrestrial/marine reference materials approach or using a previously published ΔR relevant for the spatiotemporal area.
(ii) Recording $\delta^{13}C_{16:0}$ and $\delta^{13}C_{18:0}$ values from the same TLE as that used for ^{14}C dating to determine the $\%_{marine}$, 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 $\delta^{13}C$ 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 $\Delta^{13}C < -3.1‰$ or using endmembers from the reference ruminant adipose fats values for potsherds with $\Delta^{13}C > -3.1‰$, to determine $\%_{marine}$ in the TLEs.
(v) Employing FRUITS or other Bayesian approaches (if available) to quantify $\%_{marine}$ in the TLEs using a probability density function.
(vi) Correcting CSRA dates for the MRE using mixed atmospheric and marine calibration curves (e.g. in OxCal).

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SUPPLEMENTARY MATERIAL

To view supplementary material for this article, please visit https://doi.org/10.1017/RDC.2020.11

REFERENCES

Ascough PL, Cook GT, Dugmore AJ. 2005. Methodological approaches to determining the marine radiocarbon reservoir effect. Progress in Physical Geography 29(4):532–547.

Ascough PL, Dugmore AJ, Cook GT, Higney E, Barber J, Scott EM. 2004. Holocene variations in the Scottish marine radiocarbon reservoir effect. Radiocarbon 46(2):611–620.

Ascough PL, Cook GT, Church MJ, Dugmore AJ, Arge SV, McGovern TH. 2006. Variability in North Atlantic marine radiocarbon reservoir effects at c. AD 1000. The Holocene 16(1):131–136.

Ascough PL, Cook GT, Dugmore AJ, Scott EM. 2007. The North Atlantic marine reservoir effect in the Early Holocene: Implications for defining and understanding MRE values. Nuclear Instruments and Methods in Physics Research B 259(1):438–447.

Ascough PL, Cook GT, Dugmore AJ. 2009. North Atlantic marine ^{14}C reservoir effects: implications for late-Holocene chronological studies. Quaternary Geochronology 4(3):171–180.

Ascough PL, Church MJ, Cook GT. 2017. Marine radiocarbon reservoir effects for the Mesolithic and Medieval periods in the Western Isles of Scotland. Radiocarbon 59(1):17–31.

Berstan R, Stott AW, Minnitt S, Bronk Ramsey C, Hedges REM, Evershed RP. 2008. Direct dating of pottery from its organic residues: new precision using compound-specific carbon isotopes. Antiquity 82(317):702–713.

Bronk Ramsey C. 2009. Dealing with outliers and offsets in radiocarbon dating. Radiocarbon 51(3):1025–1045.

Casanova E, Knowles TDJ, Williams, C, Crump MP, Evershed RP. 2017. Use of a 700 MHz NMR microcryoprobe for the identification and quantification of exogenous carbon in compounds purified by preparative capillary gas chromatography for radiocarbon determinations. Analytical Chemistry 89(13):7090–7098.

Casanova E, Knowles TDJ, Williams C, Crump MP, Evershed RP. 2018. Practical considerations in high precision compound-specific radiocarbon dating: Eliminating the effects of solvent and sample cross-contamination on accuracy and precision. Analytical Chemistry 90(18):11025–11032.

Casanova E, Knowles TDJ, Bayliss A, Dunne J, Barański M.Z, Denaire A, Lefranc P, di Lernia S, Roffet-Salque M, Smyth J, Barclay AJ, Gillard T, Claßen E, Coles BJ, Illet M, Jeunesse C, Krueger M, Marciniak A, Minnitt, Rotunno R, Van De Velde P, van Wijk I, Cotton J, Daykin A, Evershed RP. In press. Accurate compound-specific ^{14}C dating of archaeological pottery vessels. Nature.

Cook GT, Ascough PL, Bonsall C, Hamilton WD, Russell N, Sayle KL, Scott EM, Bownes JM. 2015. Best practice methodology for ^{14}C calibration of marine and mixed terrestrial/marine samples. Quaternary Geochronology 27:164–171.

Copley MS, Berstan R, Dudd SN, Docherty G, Mukherjee AJ, Straker V, Payne S, Evershed RP 2003. Direct chemical evidence for widespread dairying in prehistoric Britain. Proceedings of the National Academy of Sciences 100(4):1524–1529.

Copley MS, Hansel FA, Sadr K, Evershed RP. 2004. Organic residue evidence for the processing of marine animal products in pottery vessels from the pre-colonial archaeological site of Kasteelberg D east, South Africa. South African Journal of Science 100(5–6):279–283.

Correa-Ascencio M, Evershed RP. 2014. High throughput screening of organic residues in archaeological potsherds using direct acidified methanol extraction. Analytical Methods 6(5):1330–1340.

Cramp LJE, Evershed RP. 2014. "Reconstructing aquatic resource exploitation in human prehistory using lipid biomarkers and stable

isotope". In Treatise on geochemistry: Archaeology and anthropology. Amsterdam: Elsevier Oxford 12:319–339.

Cramp LJE, Evershed RP, Lavento M, Halinen P, Mannermaa K, Oinonen M, Kettunen J, Perola M, Onkamo P. Heyd V. 2014a. Neolithic dairy farming at the extreme of agriculture in northern Europe. Proceedings of the Royal Society B: Biological Sciences 281(1791):20140819.

Cramp LJE, Jones J, Sheridan A, Smyth J, Whelton H, Mulville J, Sharples N, Evershed RP. 2014b. Immediate replacement of fishing with dairying by the earliest farmers of the northeast Atlantic archipelagos. Proceedings of the Royal Society of London B: Biological Sciences 281(1780):20132372.

Cramp LJE, Casanova E, Evershed RP. Forthcoming. "Organic residues from Late Iron Age and Norse pottery. A Norse settlement in the Outer Hebrides". Excavation on Mounds 2 and 2A, Bornais, South Uist. Oxford: Oxbow Books.

Dunne, J, Evershed RP, Salque, M, Cramp LJE, Bruni S, Ryan K, Biagetti S, di Lernia S. 2012. First dairying in green Saharan Africa in the fifth millennium BC. Nature 486(7403):390–394.

Evershed RP, Heron C, Goad LJ. 1990. Analysis of organic residues of archaeological origin by high-temperature gas chromatography and gas chromatography-mass spectrometry. Analyst 115(10):1339–1342.

Evershed RP, Copley MS, Dickson L, Hansel FA. 2008. Experimental evidence for the processing of marine animal products and other commodities containing polyunsaturated fatty acids in pottery vessels. Archaeometry 50(1):101–113.

Fernandes R, Millard AR, Brabec M, Nadeau M-J, Grootes P. 2014. Food Reconstruction Using Isotopic Transferred Signals (FRUITS): A Bayesian model for diet reconstruction. PLOS ONE 9(2):e87436.

Hansel FA, Copley MS, Madureira LAS, Evershed RP. 2004. Thermally produced ω-(*o*-alkylphenyl) alkanoic acids provide evidence for the processing of marine products in archaeological pottery vessels. Tetrahedron Letters 45(14): 2999–3002.

Hansel FA, Evershed RP. 2009. Formation of dihydroxy acids from *Z*-monounsaturated alkenoic acids and their use as biomarkers for the processing of marine commodities in archaeological pottery vessels. Tetrahedron Letters 50(40):5562–5564.

Hedges RM, Tiemei C, Housley RA. 1992. Results and methods in the radiocarbon dating of pottery. Radiocarbon 34(3):906–915.

Heron C, Craig OE. 2015. Aquatic resources in foodcrusts: Identification and implication. Radiocarbon 57(4):707–719.

Knowles TDJ, Monaghan PS, Evershed RP. 2019. Radiocarbon sample preparation procedures and the first status report from the Bristol radiocarbon AMS (BRAMS) facility. Radiocarbon 61(5):1541–1550.

Longin R. 1971. New method of collagen extraction for radiocarbon dating. Nature 230(5291):241.

Marshall P. 2005. "The Chronology". In A Norse farmstead in the Outer Hebrides: Excavations at Mound 3, Bornais, South Uist. Oxbow Books(8):153–156.

Marshall P, Bronk Ramsey C, Cook G. 2016. "Chronology". In A late Iron Age farmstead in the Outer Hebrides: Excavations at Mound 1, Bornais, South Uist. Oxbow Books: 211–223.

Marshall P, Bronk Ramsey C, Cook G. Forthcoming. "The chronology". In: A Norse settlement in the Outer Hebrides. Excavation on Mounds 2 and 2A, Bornais, South Uist. Oxford: Oxbow Books.

Mukherjee AJ, Copley MS, Berstan R, Clark KA, Evershed RP. 2005. Interpretation of $\delta^{13}C$ values of fatty acids in relation to animal husbandry, food processing and consumption in prehistory. 9th ICAZ Conference. Durham: Oxbow Books. p. 77–93

Reimer PJ, McCormac FG, Moore J, McCormick F, Murray EV. 2002. Marine radiocarbon reservoir corrections for the mid to late Holocene in the eastern subpolar North Atlantic. The Holocene 12(2):129–135.

Reimer PJ, Bard E, Bayliss A, Beck JW, Blackwell PG, Bronk Ramsey C, Buck C, Cheng H, Edwards RL, Friedrich M, Grootes PM, Guilderson TP, Haflidason H, Hajdas I, Hatté C, Heaton TJ, Hoffmann DL, Hogg AG, Hughen KA, Kaiser KF, Kromer B, Manning SW, Niu M, Reimer RW, Richards DA, Scott EM, Southon JR, Staff RA, Turney CSM, van der Plicht J. 2013. IntCal13 and Marine13 radiocarbon age calibration curves 0–50,000 years cal BP. Radiocarbon 55(4):1869–1887.

Reimer RW, Reimer PJ. 2016. An online application for ΔR calculation. Radiocarbon 59:1623–1627.

Russell N, Cook GT, Ascough PL, Dugmore AJ. 2010. Spatial variation in the marine radiocarbon reservoir effect throughout the Scottish post-Roman to late Medieval period: North Sea values (500–1350 BP). Radiocarbon 52(3):1166–1181.

Russell N, Cook GT, Ascough P, Barrett JH, Dugmore A. 2011. Species specific marine radiocarbon reservoir effect: a comparison of ΔR values between Patella vulgate (limpet) shell carbonate and Gadus morhua (Atlantic cod) bone collagen. Journal of Archaeological Science 38(5):1008–1015.

Russell N, Cook GT, Ascough PL, Scott EM. 2015. A period of calm in Scottish seas: A comprehensive study of ΔR values for the northern British Isles coast and the consequent implications for archaeology and oceanography. Quaternary Geochronology 30: 34–41.

Sharples N, Ingrem C, Marshall P, Mulville J, Powell A, Reed K. 2016. "The Viking occupation of the Herbides: evidence from the excavations at Bornais, South Uist". In: Barrett J, Gibbon S-J,

https://doi.org/10.1017/RDC.2020.11 Published online by Cambridge University Press

editors. Maritime societies of the Viking and medieval world. Monograph 37, chapter 17. Leeds: Maney. p. 247–268.

Sharples N. Forthcoming. "A Norse settlement in the Outer Hebrides". In Excavation on Mounds 2 and 2A, Bornais, South Uist. Oxford: Oxbow Books.

Sharples N. and Davis O. Forthcoming. "The Middle Norse house on Mound 2(BC)". In Excavation on Mounds 2 and 2A, Bornais, South Uist. Oxford: Oxbow Books.

Stott AW, Berstan R, Evershed P, Hedges, REM, Bronk Ramsey C, Humm MJ. 2001. Radiocarbon dating of single compounds isolated from pottery cooking vessel residues. Radiocarbon 43(2A):191–197.

Stott AW, Berstan, R, Evershed RP, Bronk-Ramsey C, Hedges REM, Humm MJ. 2003. Direct dating of archaeological pottery by compound-specific ^{14}C analysis of preserved lipids. Analytical Chemistry 75(19):5037–5045.

Synal H-A, Stocker M, Suter M. 2007. MICADAS: A new compact radiocarbon AMS system. Nuclear Instruments and Methods in Physics Research B 259(1):7–13.

Wacker L, Němec M, Bourquin J. 2010. A revolutionary graphitisation system: Fully automated, compact and simple. Nuclear Instruments and Methods in Physics Research B 268(7–8):931–934.

Wacker L, Fülöp RH, Hajdas I, Molnár M, Rethemeyer J. 2013. A novel approach to process carbonate samples for radiocarbon measurements with helium carrier gas. Nuclear Instruments and Methods in Physics Research B 294:214–217.

https://doi.org/10.1017/RDC.2020.11 Published online by Cambridge University Press