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ABSTRACT

Carbon isotope measurements of individual fatty acids ($C_{16:0}$ and $C_{18:0}$) recovered from archaeological pottery vessels are widely used in archaeology to investigate past culinary and economic practices. Typically, such isotope measurements are matched with reference to food sources for straightforward source identification, or simple linear models are used to investigate mixing of contents. However, in cases where multiple food sources were processed in the same vessel, these approaches result in equivocal solutions. To address this issue, we tested the use of a Bayesian mixing model to determine the proportional contribution of different food sources to a series of different mixed food compositions, using data generated both by simulation and by experiment. The model was then applied to previously published fatty acid isotope datasets from pottery from two prehistoric sites: Durrington Walls, near Stonehenge in southern Britain and Neustadt in northern Germany. We show that the Bayesian approach to the reconstruction of pottery use offers a reliable probabilistic interpretation of source contributions although the analysis also highlights the relatively low precision achievable in quantifying pottery contents from datasets of this nature. We suggest that, with some refinement, the approach outlined should become standard practice in organic residue analysis, and also has potential application to a wide range of geological and geochemical investigations.

Keywords: Fatty acids; carbon isotopes; pottery use; Bayesian mixing models; FRUITS

1. Introduction

Organic residue analysis is a well-established method for determining the contents of archaeological pottery. This approach has been particularly important for establishing major

changes in prehistoric economic (e.g., Evershed, 2008; Cramp et al., 2014) and culinary practices (Craig et al., 2011), as well as understanding the origins of ceramic technology itself (Craig et al., 2013). Many of these studies have relied on measurements of the stable carbon isotope ratios (δ^{13}C) of saturated fatty (*n*-alkanoic) acids (e.g., $C_{16:0}$ and $C_{18:0}$) to distinguish different products (Regert, 2011; Craig et al., 2012). These fatty acids are commonly preserved in archaeological pottery and their isotope ratios have been well characterised in a range of authentic modern food products. Occasionally, vessels dedicated for specific uses can be discerned using this approach due to their clear and distinctive isotope composition (Salque et al., 2013). Yet for most situations in the past, it is likely that pots were used for preparing a range of foodstuffs, either as a result of these items being cooked together, or through sequential use of the pot over time. These processes add complexity when making inferences about the relative proportions of food types processed in the vessel.

Determining the ratio of different foods contributing to mixtures is difficult, since different foodstuffs not only vary isotopically, but also contain different amounts of $C_{16:0}$ and $C_{18:0}$ acids. Attempts have been made to resolve mixtures of foods in pottery quantitatively, using simple two end-member models which consider the concentration of each fatty acid determined from authentic reference samples (Mukherjee et al., 2008; Craig et al., 2011). However, in cases where multiple foods were potentially processed, this approach can result in equivocal solutions. A more robust method for resolving mixtures in archaeological pottery is therefore needed, both to confirm product identification, but also to identify ancient culinary practices that may involve the combination or separation of foods.

The primary goal of this study was to employ a Bayesian approach to quantify the proportions of different foodstuffs in archaeological pottery based on previously published

carbon isotope analysis of fatty acids from two prehistoric sites; the inland Late Neolithic henge monument of Durrington Walls, near Stonehenge in southern Britain, (Craig et al., 2015) and the coastal Late Mesolithic/Early Neolithic site of Neustadt on the Baltic coast of Germany (Craig et al., 2011). The performance of this Bayesian approach was first tested using both simulated examples and isotopic data from experimental pots, where known mixtures of three different foods were cooked in a controlled experiment.

2. Model specification

A model instance, represented by Equation 1, is defined here with the following characteristics: (i) Lipid groups are defined by the $\delta^{13}C$ measurements of multiple fatty acids (for the present study these are $C_{16:0}$ and $C_{18:0}$) in modern authentic foodstuffs (i.e. reference samples); (ii) Non-weighted model: excluding taphonomic effects it is assumed that the source of carbon for a particular fatty acid extracted from the ceramic matrix can only be the same fatty acid found in the lipid sources; (iii) Offset model: since modern reference isotopic values are employed it is necessary to include an offset quantifying the difference between modern and past stable carbon isotopes ratios due to fluctuations in atmospheric $\delta^{13}C$ values; (iv) Concentration-dependent model: the concentration of each fatty acid within each lipid group is included in the model.

The model for the observed value of the k-th isotope signal:

$$H_k \sim N(\mu_{H,k}, \sigma_{H,k}^2) \qquad \text{(Equation 1)}$$

where:

$$\mu_{H,k} = \frac{\sum_{i=1}^{N_k} \alpha_i C_{ik}(T_{ik} + I_{ik})}{\sum_{i=1}^{N_k} \alpha_i C_{ik}}$$

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and: H_k represents the k-th isotopic signal measured in the pottery lipid extracts. This corresponds to $\delta^{13}C$ measurements on fatty acids (in the present study $C_{16:0}$ and $C_{18:0}$) extracted from the archaeological ceramic extracts; α_i represents the contribution from the i-th lipid group. The α_i's are unknown and their estimation, together with estimation of their uncertainties, represents the ultimate analytical goal. Physical restrictions apply: $0 \leq \alpha_i \leq 1$ for $i = 1, \dots, n$ and $\sum_{i=1}^{n} \alpha_i = 1$ where n represents the number of lipid groups; I_{ik} is the isotopic signal (e.g., $\delta^{13}C_{16:0}$ and $\delta^{13}C_{18:0}$) measured for the i-th lipid group contributing to the k-th isotopic signal measured in the pot. Due to the presence of measurement errors (and inter-individual heterogeneity), it is assumed to behave as a random variable which is modelled by a multivariate normal distribution, $I_i \sim N(\mu_I, \Sigma_I)$ with an average vector μ_I and a Σ_I variance-covariance matrix; T_{ik} is the offset for the k-th isotopic signal in the i-th lipid group due to fluctuations in atmospheric $\delta^{13}C$ values. This is modelled as a normal variable, $T_{ik} \sim N(\mu_{T,ik}, \sigma_{T,ik}^2)$; C_{ik} is the concentration of the k-th fatty acid in the i-th lipid group. This is modelled by a multivariate normal distribution, $C_i \sim N(\mu_C, \Sigma_C)$ with an average vector μ_C and a Σ_C variance-covariance matrix.

2.1. Adding prior information

A simple approach was developed for incorporating a priori constraints of non-standard types into the expanded version of the model in Equation 1. Prior expert opinion is incorporated through user-defined algebraic expressions $y(\alpha_i, I_{ik}, C_{ik})$ that serve to express relationships of equality or inequality between model parameters (e.g., when prior knowledge allows imposing that certain lipid groups contribute more than others).

To link a relationship of equality into the model a parameter p (Equation 2) is assigned a normal distribution with a mean given by $y(\alpha_i, I_{ik}, C_{ik})$ and a user-defined uncertainty, r. The equality constraint is imposed by having an 'observed' value of zero for p.

$p \sim N(y(\alpha_i, I_{ik}, C_{ik}), r^2)$ (Equation 2)

To link an inequality relationship, a parameter l (Equation 3) is assigned a Bernoulli distribution $Bernoulli(k)$ where k is a Heaviside function, $H(y(\alpha_i, I_{ik}, C_{ik}))$, which provides a value of one or zero depending on whether $y(\alpha_i, I_{ik}, C_{ik})$ is positive or negative. The parameter l may also include an additional error term ε modelled as a normal distribution, $\varepsilon \sim N(0, r^2)$ with 0 average and a user-defined uncertainty, r. The inequality constraint is then imposed by having the 'observed' value of one for l.

$l \sim Bernoulli(H(y(\alpha_i, I_{ik}, C_{ik}) + N(0, r^2))$ (Equation 3)

2.2. Bayesian inference

Modelling was carried out using the 3.0 Beta version (available at http://sourceforge.net/projects/fruits/) of the Bayesian mixing model FRUITS (Fernandes et al., 2014a). Although FRUITS has mainly been used for the reconstruction of ancient human diets (e.g., Fernandes et al., 2012), the model is also applicable to any problems that aim at estimating the contributions from different sources to a given mixture, using quantitative signals (e.g., elemental or isotopic profiles) as input data. The FRUITS generic model (Fernandes et al., 2014a) includes a weight parameter that allows building model instances in which different food fractions (e.g., food nutrients, single compounds) contribute in varying proportions to a single target signal (here, $\delta^{13}C$ measured in fatty acids extracted from archaeological potsherds). However, since it is assumed that there is a one-to-one correspondence between pot and food

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fatty acids the weight parameter has a value of one when target and source fatty acid match and zero otherwise resulting in the simplified model representation (Equation 1).

Numerical Bayesian inference was performed using the BUGS software, a Markov chain Monte Carlo (MCMC) method that employs Gibbs sampling and the Metropolis-Hastings algorithm (Gilks et al., 1996). The first 5,000 iterations of the MCMC chains were discarded (burn-in steps) and these were then run for an additional 10,000 iterations. Model convergence for the different α_i's was checked by inspecting if the trace plots of the respective posterior chains exhibited an asymptotic behaviour. Trace autocorrelation plots were also inspected to assess convergence.

3. Model implementation

The first stage of model building is to identify the lipid groups (i.e. foodstuffs) that potentially contributed to the ceramic organic residue. This is usually based on locally available archaeological and historical evidence. For example, at Durrington Walls (see Section 5.1), the composition of the faunal assemblage and the near absence of plant remains (Craig et al., 2015), indicates that pottery was likely only used for the processing of cattle (meat and/or milk) and porcine products. At Neustadt (see Section 5.1) a broader range of foods was available including marine fish and mammals and wild ruminants, both of which made up a significant proportion of the faunal assemblage (Glykou, 2014). Freshwater fish were much sparser and consequently were not considered. Reference $\delta^{13}C_{16:0}$ and $\delta^{13}C_{18:0}$ values for these products (Table 1) were obtained from modern fats obtained from southern Britain (Copley et al., 2003) Denmark (Craig et al., 2011) and Poland (Craig et al., 2012).

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An additional source of uncertainty is the so-called Suess effect that describes the ^{13}C depletion of atmospheric CO_2 as a result of the burning of fossil fuels during the 19th and 20th centuries (Friedli et al., 1986). Under the defined model (Equation 1), through the offset parameter T_{ik}, it is assumed that each lipid group has a specific δ^{13}C offset between modern and ancient values. This is expected since modern references were likely sampled at different times. However, since collection times are not exactly known an approximation was made and the same offset value was used for all terrestrial lipids. Modern terrestrial lipid references were sampled during the 1990s and 2000s and an atmospheric δ^{13}C value of −8.1‰ ± 0.2‰ was taken as reference for this time period (Hellevang and Aagaard, 2015). During the second half of the Holocence, which includes the archaeological periods under study, atmospheric δ^{13}C values were ca. −6.35‰ ± 0.1‰ (Schmitt et al., 2012). Thus, the offset inlcuded in model (T_{ik}) to account for the differences in δ^{13}C$_{16:0}$ and δ^{13}C$_{18:0}$ between modern and archaelogical values was conservatively estimated to be 1.75‰ ± 0.2‰. For lipid references from modern marine organisms, the extent of mixing of CO_2 between the atmosphere and the Baltic Sea needs to be considered. Global estimates for the full oceanic ^{13}C Suess effect since pre-industrial times are estimated to be as high as 40% (±10%) of the atmospheric, depending on the depth and extent of mixing (Eide et al., 2017). The marine fish and mammals used as references here (Craig et al., 2011), and those available to prehistoric fisher-hunter-gathererers were likely to have fed both in the shallow Baltic and deeper North Atlantic waters (Craig et al., 2006). For the modelling purposes here, we used an estimate of 50% of the atmospheric Suess effect (i.e. 0.875‰ ± 0.2‰).

The concentration of C$_{16:0}$ and C$_{18:0}$ fatty acids (Table 1) as a proportion of total fatty acids in the modern authentic fats were either measured and reported with the δ^{13}C values or, where these were unavailable, obtained from previously published values. Whilst the proportion of C$_{16:0}$

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and $C_{18:0}$ in total fatty acids are generally well reported for different food classes (Table 1), the amount of fat by weight varies greatly between species and between tissues. This represents the greatest source of uncertainty in reconstructing pottery use in terms of relative weights of different foods. In the majority of cases, the outputs are given in weight percentage of total fatty acid. Fatty acids account for < 90% of the fat present in most animal tissues (Weihrauch et al., 1977), so the output values are a reasonable approximation for the relative weight of different animal fats and oils processed in the pottery vessels.

For the different lipid groups, the $\delta^{13}C_{16:0}$ and $\delta^{13}C_{18:0}$ values are highly correlated (Fig. 1); this is the motivation for allowing the correlations between isotopic values. Therefore, isotopic, and also concentration, values of fatty acids from the lipid groups are described within the model by multivariate normal distributions defined by a mean vector (Table 1) and a variance-covariance matrix. The observed isotopic distributions are naturally randomly distributed around a central value. Thus, with longer periods of pottery use it becomes more likely that a wide range of isotopic values would have been sampled and the overall combined value would tend towards the mean. In this case, as an approximation, the standard error of the mean (SEM) of multivariate normal distributions would be an appropriate reference. As with many prehistoric pots, the vessels from both Durrington Walls and Neustadt had built up accumulations of soot on their exteriors and had other signs of use-wear consistent with repeated use. In a study of pottery use from a Danish Early Neolithic site, a low median value (6 months) was taken as the reference for the usage of ceramic pots although compiled ethnographic data for the same study showed that the duration of use could actually be as high as several years (Madsen and Jensen, 1982). Nonetheless, the number of samples employed to define isotopic and concentration values of lipid groups is still relatively small (Table 1). Thus, we have examined two models to estimate

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source contribution to the archaeological pottery: a conservative model with, and a non-conservative model without, elements of covariance matrices divided by number of samples.

Where several potential lipid groups exhibit considerable overlap in their distribution of $\delta^{13}C_{16:0}$ and $\delta^{13}C_{18:0}$, it may not be possible to generate unambiguous estimates of proportions. For these cases, individual lipid groups may be aggregated into a new, enlarged, group. This is illustrated in Table 1 where the lipid group "combined ruminant" corresponds to the aggregation of the lipid groups "wild ruminant adipose", "domesticated ruminant adipose", and "dairy fats". However, when data are combined in this manner the assumption about the natural randomness and symmetry of isotopic values is no longer valid.

For our experimental material, replicate measurements corresponding to extracts of different portions of the same vessel showed a reproducibility equal or better than 0.2‰ for $\delta^{13}C_{16:0}$ and typically equal or better than 0.4‰ for $\delta^{13}C_{18:0}$. However, besides measurement uncertainties there are also other sources of uncertainty. These include potentially unknown contributing lipid groups that were not taken into account, degradation processes, cooking and lipid absorption processes. For instance, previous cooking experiments have shown that different cooking methods could result in isotopic differences between cooked and raw food which are larger than the measurement uncertainty (Fernandes et al., 2014b). In contrast, there is good evidence that fatty acids absorbed to pottery retain their $\delta^{13}C$ values despite extensive degradation under oxic and anoxic conditions (Evershed, 2008). Here, an uncertainty of 0.6‰ was taken as reference for measurements of $\delta^{13}C_{16:0}$ and $\delta^{13}C_{18:0}$ from ceramic containers. We believe this to be a conservative uncertainty since it represents the double of the value (0.3‰) commonly used for measurement reproducibility.

4. Model evaluation

4.1. Testing the model using simulated data

To test model performance we first employed simulated scenarios. A large variety of scenarios could have been selected but we chose four similar to those that are likely to occur in actual archaeological case studies. These were defined as: scenario 1 consisting of two lipid groups ("wild ruminant adipose" and "marine oils"); scenario 2 consisting of three lipid groups ("dairy fats", "domesticated ruminant adipose", and "porcine adipose"); scenario 3 in which the different ruminant fats are combined into a single group ("combined ruminant") and "porcine adipose" as a separate lipid group; scenario 4, as with scenario 2, but with additional prior information added to the model. For each scenario of available lipid groups, three different cases were simulated. Fig. 2 shows expected values and model outputs for the different scenarios. Expected mixed isotopic values were calculated using the average isotope and concentration values of $C_{16:0}$ and $C_{18:0}$ fatty acids given in Table 1 and represented in Fig. 1.

For scenario 1, consisting of two well-separated food groups, model outputs were in excellent agreement with simulated values (Fig. 2). For scenario 2, simulated contributions are contained within the 68% credible intervals. However, credible intervals for scenario 2 are larger than those observed for scenario 1. The ambiguity in model estimates is a consequence of the distribution of isotopic values for the three lipid groups (Fig. 1). The "domestic ruminant adipose" values are relatively close to the mixing line between "porcine adipose" and "dairy fat". Thus, contributions from "domestic ruminant adipose" may also be interpreted as the mixing of "porcine adipose" and "dairy fat".

In order to reduce the uncertainties different strategies may be adopted: (1) adding other isotopic or elemental proxies (e.g., bulk $\delta^{13}C$ or $\delta^{15}N$ or %C or %N, if available); (2) including

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additional prior information in the model; or, (3) grouping ranges of lipids that have similar values. In scenario 3, "combined ruminant" corresponds to the aggregation of the lipid groups "domesticated ruminant adipose", "dairy fats", and also "wild ruminant adipose" that was not included in scenario 2. The estimates of the contributions from "combined ruminant" vs "porcine" show that for the former the credible intervals are narrower than those observed for "wild ruminant adipose" or "dairy fats" in scenario 2. Scenario 4 is similar to scenario 2, however prior information was added to constrain model estimates (see Section 2.1). Introduced constraints impose relationships of inequality on the contributions from the different lipids groups or of equality with an uncertainty of 20% (Fig. 2). These types of relationships may be employed, for example, when palaeodietary evidence can be independently and safely assessed from the archaeological evidence. This could rely on the study of well-preserved faunal or botanical assemblages, but may also require the taphonomic analysis of assemblages to determine employed cooking methods (Roberts et al., 2002; Koon et al., 2003). In cases where robust archaeological information is lacking, and taphonomic effects may be unaccounted for, different scenarios should be tested to verify the sensitivity of model estimates. The presence of lipid biomarkers for specific products could also be employed to impose threshold values on the contribution from certain foods, although experimental work is still required to establish these values. One has to be aware of the fact that while imposing correct relationships can help efficiency, imposing wrong relationships can be entirely detrimental to the whole model (results can be easily biased heavily).

4.2. Reconstructing mixtures from pots with known contents

Eleven replica ceramic vessels, of approximately 9 cm diameter and fired at 750 °C, were used to prepare ten different mixtures of whole wild salmon, ground chestnut flour and wild red deer, plus a blank control containing only distilled water. These were chosen due to their expected distinct isotopic values. They are also products typically exploited by northern Eurasian Holocene hunter-gatherers. None of the animals are protected and they were not killed for the purpose of this research. All animals were pre-killed by license holders using the appropriate methods of killing as outlined in Appendix D of the Animals (Scientific Procedures) Act 1986. The salmon and deer tissue samples were homogenised in a food blender, and each product was weighed in preparation for addition to the pots in proportions shown in Table 2.

The total amount of each of the individual wet tissue mixtures was added to the experimental pots, which were then boiled continuously for 4 h over an open wood-fuelled fire in the York Experimental Archaeology (YEAR) Centre. Distilled water was added to each pot throughout the experiment to maintain a level up to the rim. Once completed, the remaining cooked foodstuffs were removed from each pot. The pots were placed in a drying oven at 40 °C for 48 h until dry. All pots were then wrapped in aluminium foil and stored in a cool, dry environment until needed for further analysis.

Approximately 1 g portions of ceramic powder were obtained from each pot by drilling to a depth of 2–5 mm from the interior surface. Separate portions of each pot were extracted at least in duplicate and methylated in one-step with acidified methanol as described previously (Craig et al., 2013; Papakosta et al., 2015). Briefly, HPLC grade methanol was added to each sample (4 mL/g). Each sample was sonicated for 15 min, and then acidified with concentrated sulphuric acid (800 μl). The acidified suspension was heated in sealed tubes for 4 h at 70 °C before being

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allowed to cool. Lipids were extracted with *n*-hexane (3×2 ml), and quantified using a GC-FID

equipped with a DB23 column (60 m × 0.25 mm × 0.25 μm; J&W Scientific, Folsom, CA, USA).

Untreated samples of the three foodstuffs (ca. 300–900 mg) used in this experiment, chestnut

flour, deer and salmon, were freeze dried and then extracted in the same manner as described for

the ceramic powder.

Carbon isotope measurements were determined on two fatty acid methyl esters (FAMEs),

$C_{16:0}$ and $C_{18:0}$ from sample aliquots that had been cleaned using $AgNO_3$-impregnated silica gel

columns. The fraction containing saturated fatty acids, eluted from the silica gel columns using *n*-

hexane:DCM (1:1, v:v), were dried under N_2, re-dissolved in *n*-hexane and directly analysed by

GC–C–IRMS using standard conditions and protocols (Craig et al., 2007, 2012). Analysis was

performed on an Isoprime 100 (Isoprime, Cheadle, UK) linked to a Hewlett Packard 7890B series

gas chromatograph (Agilent Technologies, Santa Clara, CA, USA) with an Isoprime GC5

interface (Isoprime, Cheadle, UK). The gases eluting from the GC were directed through the GC5

furnace held at 850 °C to oxidise all carbon species to CO_2. All $\delta^{13}C$ values are expressed relative

to Vienna Pee Dee Belemnite (VPDB), based on in-house reference gases (CO_2, BOC) adjusted

daily using an international standard mixture of *n*-C_{16} to *n*-C_{30} alkanes (the isotope ratios of

which were measured offline by A. Schimmelmann, Biogeochemical Laboratories, Indiana

University). Sample values were also corrected to account for the methylation of the carboxyl

group that occurs during acid extraction, based on comparisons with an in-house standard

containing $C_{16:0}$ and $C_{18:0}$ fatty acids of known isotopic composition processed in the same way as

the experimental pottery samples.

The $\delta^{13}C$ values of fatty acids from each pot are shown in Table 2. No lipids could be

identified in the blank control containing only water. Modelling was carried out using these as

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target values and the lipid group isotope information and the concentration data (Table 3). No additional prior information was added to the model.

The model output is shown in Fig. 3 as the contribution by wet weight of each product to each pot. Results from the experimental vessels show that the model can predict the salmon component of the mixture with relatively small credible intervals (Fig. 3). However, the credible intervals for the chestnut and deer are often considerably larger given the proximity in $\delta^{13}C_{16:0}$ and $\delta^{13}C_{18:0}$ values for these two foodstuffs. The known foodstuff contributions are contained within the 95% credible intervals except in two instances where they are just outside of the 95% interval (pots 2 and 3), while for pot 6 the model output for deer contribution is clearly distant from the known contribution.

In a separate model, deer and chestnut were aggregated as a single 'terrestrial' lipid group. The $\delta^{13}C$ values of the aggregated lipid group were calculated to reflect the different fatty acid concentrations of each single food group. The outputs for a two-source model are shown in Fig. 4 and predictably, the credible intervals for the terrestrial aggregated lipid group are narrower (Fig. 4) than those observed in the previous model output for deer and chestnut lipid groups (Fig. 3). In the new model instance the 95% credible intervals always contain the known foodstuff contribution. The model performance suggests that the uncertainty adopted for pot isotopic values is suitable (i.e. 0.6‰). In addition, the measured values of the pots containing single sources (pots 1–3) correspond very well with measurements of the raw foodstuffs (Tables 2 and 3), suggesting that the process of cooking and absorption does not cause any isotopic fractionation of these compounds. In contrast, the observed discrepancies between the actual contents and the content predicated by the model may be attributable to differential rates of absorption of lipids from different foodstuffs when cooked together.

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5. Archaeological case studies

5.1. Sites and materials available

Durrington Walls is a Late Neolithic henge monument in Southern Britain located near the site of Stonehenge. The main occupation of the site dates to 2,535–2,475 cal BCE and includes large amounts of animal bones, broken 'Grooved Ware' ceramics and other food-related debris, and has been interpreted as a place of feasting (Craig et al., 2015). Faunal remains at the site are dominated by domesticated pigs and, to a lesser extent, cattle. Other wild and domesticated animals and plants are extremely sparse. Lipids extracted from 151 potsherds were previously classified into three food categories (dairy, ruminant adipose, and non-ruminant) based on the difference stable carbon isotope ratios (Δ^{13}C) of their $C_{16:0}$ and $C_{18:0}$ n-alkanoic acids (Craig et al., 2015, Fig. 5A), using the criteria of Copley et al. (2003), although substantial mixing, particularly of ruminant adipose (presumably beef) and non-ruminant (presumably pork) was thought to have occurred (Craig et al., 2015).

Neustadt is a submerged coastal site on the Baltic coast of northern Germany that was occupied from ca. 4,600 to 3,800 cal BCE. The pottery at the sites includes both Late Mesolithic Ertebølle (EBK) and Early Neolithic Funnel Beaker (TRB) typologies (Saul et al., 2013). Exploitation of marine resources was very important at this site, indicated by thousands of fish bones and a particularly high frequency of marine mammals (Glykou, 2014). Terrestrial fauna are also well represented, including a small number of domesticates in the Early Neolithic layers. Organic residue analysis of 46 vessels has identified the presence of marine foods throughout the sequence, but also the presence of dairy and other ruminant products (Craig et al., 2011), which could also include wild ruminants such as deer, aurochs or moose. Five vessels have been

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interpreted as oil lamps thought to have been used exclusively for burning marine oil (Heron et al., 2013). The presence of aquatic biomarkers (Hansel et al., 2004), including ω-(o-alkylphenyl) alkanoic acids and isoprenoid fatty acids, on a number of vessels shows that fish or marine mammal oils made a contribution to the residue and may be used to constrain the model based on $C_{16:0}$ and $C_{18:0}$ measurements.

5.2. Model estimates for pottery residues from Durrington Walls

Source estimates were obtained for 121 sherds from Durrington Walls using the reference ranges for 'dairy fat', 'porcine fat' and 'domesticated ruminant adipose' as defined in Table 1. Credible intervals (68%) for the contributions from food sources to each vessel were estimated using conservative and non-conservative (standard error of the mean) models. The posterior distributions generated by the model are represented by ternary plots of the median values with marginal 68% credible intervals represented by error bars (Fig. 5B and 5C) allowing large number of samples to be visually compared. It should be noted, however, that the intervals in the three directions are not independent, and therefore the bars do not correspond to the true 68% credible area around the point, but they give a simple representation of the scale of the uncertainty. Overall, estimates provided by the model shows that for about two-thirds of pots, the main contributions are either from dairy or domesticated ruminant adipose. Under a conservative model credible intervals for the two lipid groups are relatively broad and in most cases it is not possible to exclude a significant contribution (ca. 20%) of either of these products in each pot. This ambiguity is slightly ameliorated when a non-conservative model is employed and the estimates typically indicate higher contributions from 'dairy fat'. There are also several pots (ca. 35%) showing large contributions from 'porcine fat' with more confidence.

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The overlap in credible intervals for different products is attributable to the isotopic proximity of fatty acids from ruminant adipose and dairy. Previous studies (e.g. Evershed, 2008; Cramp et al., 2014) have used the difference between the $\delta^{13}C$ values of $C_{16:0}$ and $C_{18:0}$ fatty acids ($\Delta^{13}C$) to distinguish these products. It has been demonstrated that the $C_{18:0}$ acid is depleted in ^{13}C compared to the $C_{16:0}$ component in ruminant tissues and to an even greater extent in ruminant milk, due to physiological differences in the biosynthesis of these lipids (Copley et al., 2003). The established $\Delta^{13}C$ ranges for non-ruminants, ruminants and dairy products are approximately 1‰ to −1‰, −1‰ to −3.3‰ and −3.3‰ to −7‰ respectively (Fig. 5A). To evaluate the utility of this approach, the mean contributions of each of these products to the 121 vessels, as estimated through the conservative model, are compared with the $\Delta^{13}C$ values for each vessel in Fig. 5D–F.

Vessels interpreted as dairy and porcine based on their $\Delta^{13}C$ values were estimated to have contained at least 50% of fatty acids from these sources, showing the value of this proxy. However, many vessels with $\Delta^{13}C$ values in the ruminant adipose fat range were estimated to have contained less than half their fatty acids from this source (Fig. 5E). This discrepancy serves as a note of caution when interpreting data of this nature. Mixing of sources with distinct isotope end-members, in this case dairy and porcine products, produces similar isotope values to sources with intermediate values, i.e. ruminant adipose. It is difficult to distinguish, for example, an absence of dairy fats when there is a possibility that this product was mixed with other non-ruminant sources. Furthermore, even pots that fall clearly within the $\Delta^{13}C$ ranges for dairy and porcine fats could credibly also contain substantial amounts of other products. The issue of equifinality is often overlooked in residue analysis studies, but becomes clear when more refined estimations including uncertainties are provided using the Bayesian approach we describe here.

5.3. Model estimates for pottery residues from Neustadt

Source estimates were obtained for previously reported data from 46 sherds from the site of Neustadt (Craig et al., 2011). The vessels were typologically classified as belonging to the Late Mesolithic Ertebølle culture (EBK) or Early Neolithic Funnel Beaker culture (TRB). Ertebølle vessels were sub-divided into two distinct forms; cooking pots (EBK), or lamps (EBK lamps). The defined model contained three sources: combined ruminant (including dairy and all adipose tissues), porcine and marine as described in Section 4.1. In this case, the model estimates have relatively narrow credible intervals (Fig. 6). Furthermore, the credible intervals are considerably reduced when the non-conservative model is used (Fig. 6b).

Biomarkers for aquatic products, including isoprenoid fatty acids and long-chain (C_{18}–C_{22}) ω-(*o*-alkylphenyl) alkanoic acids (APAAs: Hansel et al., 2004; Craig et al., 2011), were observed in 13 of the samples analysed (Fig. 6). These are only formed by protracted or repeated heating of the fish, shellfish or marine mammal oils. Seven of these samples were estimated to have lipids derived predominantly from a marine source (Fig. 6), but four samples were estimated to only have contained at most low amounts of marine-derived lipid. Unfortunately, without further information regarding the conditions required for their formation, the presence or absence of such aquatic biomarkers is unable to provide quantitative information on source contributions and therefore constrain the model. Experiments to ascertain such quantified limits would be useful for future studies. In addition, the use of other quantitative measures of aquatic product content, such as the ratio of diastereomers of phytanic acid (Lucquin et al., 2016) or elemental analysis-IRMS of the nitrogen in carbonised deposits (Heron and Craig, 2015) could be easily incorporated as additional proxies or priors into the model.

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Even without incorporating further proxies or prior information, the analysis of mixtures shows some interesting patterning in pottery use that are difficult to infer from plotting isotope data alone. Firstly, the model estimations reveal that marine fats were extensively mixed with ruminant products in the Neolithic (TRB), but less so in the Mesolithic pots (EBK). Conversely, porcine and ruminant products were mixed in late Mesolithic vessels, whilst porcine products were much less common in the Neolithic vessels (Fig. 6). These data contribute to the debate regarding the changing values and culinary roles of specific foods across the transition to farming (Saul et al., 2013). At Neustadt, pottery use does not reflect the proportion of different animals exploited, as determined by the well preserved faunal assemblage, indicating that very deliberate culinary choices were made on what to include or exclude from pottery, and perhaps what foods were allowed to be mixed together, and that the value of these foods changed following the introduction of domesticated species. Secondly, it is estimated that the six Ertebølle lamps, believed to be used for burning marine oils, may have had a more variable use than previously thought (Heron et al., 2013). The estimates show that the isotope data supports substantial use of porcine fat in addition to marine oils (Fig. 7). For one vessel (N1009) there is also a discrepancy between lipids absorbed into the ceramic (N1009i) and those recovered from a carbonised surface deposit (N1009s), suggesting that terrestrial fats and marine oils were burnt during different episodes in this lamp.

5.4. Future developments and applications

As a next step, we suggest incorporating additional datasets to improve model performance. This includes the use of independent prior information, such as data from the presence of lipid biomarkers. These data could be used to broadly constrain the model parameters but require

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understanding of the bounding conditions (e.g., minimum relative source concentrations) necessary for their formation. Further quantitative information on relative source contributions can also be obtained by employing additional proxies. These could include ratios of different stable isotopes (e.g., δ^2H or δ^{15}N), isotopic measurement of a wider range of compounds and the relative concentrations of different individual compounds, provided these are preserved during diagenesis (e.g., phytanic acid diastereomers). Furthermore, it is necessary to establish the parameter values of food sources with greater confidence through measurement of a greater number of authentic reference samples. Notably, the uncertainty due to geographical effects, Suess effect and other temporal effects could be reduced by establishing in situ reference ranges directly from archaeological bones of known species that are contemporary with the artefacts to be analysed (Colonese et al., 2015).

Finally, we note that this approach may have applications beyond archaeological science and could be used to more accurately quantify source contributions to mixtures in a range of environmental and geological settings using different forms of geochemical data. Such applications might include the sourcing of sediment inputs in fluvial systems using, for example, a combination of organic molecular (e.g., Chen et al., 2017) and inorganic elemental data (Haddadchi et al., 2014), alongside compound-specific radiocarbon measurements (Pearson and Eglinton, 2000; Feng et al., 2015; Fernandes et al., 2017). Other applications, include estimating the contributions of different end members (plants, microorganisms) to modern (e.g., Eley et al., 2012) or ancient sediments, based on the molecular and isotopic properties of their biomarkers (Newman et al., 2016).

6. Conclusions

In this study, we provide a new approach for assessing the contribution of different foodstuffs to archaeological pottery vessels based on fatty acid stable carbon isotope measurements, using a Bayesian mixing model. These estimates, expressed as posterior probability distributions, were determined by taking into account both the uncertainties associated with the measurement of isotope values in fatty acids extracted from potsherds and the uncertainties associated with the concentration and carbon isotope composition of reference food groups. We contend that this approach provides a more nuanced interpretation of vessel use than has been achieved before. By testing the model under different simulated scenarios and by applying it to data from pots used in controlled cooking experiments, we are also able assess the degree of ambiguity when estimating contents based solely on fatty acid carbon isotope values. In particular, we highlight the problem of equifinality, i.e. where many possible solutions account for the observed the data, which is due to the proximity of isotope signals from different food reference groups, and co-linearity among these values.

In an attempt to circumvent this problem, we improved the model's performance by using the standard error of the mean (SEM) to represent the error describing the distribution of the source data or by aggregating reference food groups, albeit at the loss of source resolution. When applied to archaeological data, we show that the mixing model could identify patterns in pottery use that were hitherto unknown, although the precision of the estimations was generally low with generally broad credibility intervals for each product. However, these data highlight the real precision achievable with this approach, a fact that needs to be considered when assigning pottery use to a single or mixture of source(s) and when making archaeological interpretations.

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Figure captions

Fig. 1. Distribution of stable carbon isotopic values (δ^{13}C) of fatty acids from different lipid groups. The data have not been corrected for Suess effect (see text).

Fig. 2. Comparison of simulated contributions and model estimates. Box plots show model output, while filled circles show simulated contribution. The boxes represent a 68% credible interval while the whiskers represent a 95% credible interval. The horizontal continuous line indicates the mean while the horizontal discontinuous line indicates the median.

Fig. 3. Modelled versus actual percentage of each foodstuff added to the experimental pots. Box plots show model output, while filled circles show actual content. The boxes represent a 68% credible interval while the whiskers represent a 95% credible interval. The horizontal continuous line indicates the mean while the horizontal discontinuous line indicates the median. All estimations are reported as percentage contribution by wet weight to each pot.

Fig. 4. Modelled versus actual percentage of aggregated terrestrial and salmon input to the experimental pots. Box plots show FRUITS output, while filled circles show actual content. The boxes represent a 68% credible interval while the whiskers represent a 95% credible interval. The horizontal continuous line indicates the mean while the horizontal discontinuous line indicates the median. All estimations are reported as percentage contribution by wet weight to each pot.

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Fig. 5. (:); Plot of $\delta^{13}C_{16:0}$ and $\Delta^{13}C$ values of fatty acids extracted from individual vessels at Durrington Walls against the ranges (median, max, min) in $\Delta^{13}C$ from authentic reference fats. (B and C): Ternary plots with points representing the median contributions from each source using conservative (B) and non-conservative (C) model parameters. Whiskers represent the 68% credible intervals of marginal distributions. For each lipid group whiskers are parallel to a bisector axis of the triangular plot (lipid group identified at the vertices of the triangular plot) and constrained by the length of the bisector they lie parallel to. As such, whiskers may extend beyond the boundaries of the triangular plot. (D–F): Plots showing medians and 68% credible intervals representing the contributions of each source to each potsherd, as estimated from non-conservative mixing model, against their $\Delta^{13}C$ values.

Fig. 6. Ternary plots with points representing the median source contributions to vessels from Neustadt, Germany using conservative (A) and non-conservative model parameters (B). Whiskers represent the 68% credible intervals of marginal distributions. For each lipid group whiskers are parallel to a bisector axis of the triangular plot (lipid group identified at the vertices of the triangular plot) and constrained by the length of the bisector they lie parallel to. As such, whiskers may extend beyond the boundaries of the triangular plot. All estimations are reported as % fatty acid of each source in total fatty acid. All estimations are reported as % fatty acid of each source in total fatty acid. The presence of aquatic biomarkers (isoprenoid fatty acids and C_{20}, C_{18} APAAs) is marked with a star.

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Fig. 7. Estimates for contributions of different sources to samples of Late Mesolithic 'lamps' from Neustadt using conservative (A) and non-conservative (B) model parameters. The boxes represent a 68% credible interval while the whiskers represent a 95% credible interval. The horizontal continuous line indicates the mean while the horizontal discontinuous line indicates the median. All estimations are reported as % fatty acid of each source in total fatty acid. Pot sample number are shown. i indicates interior drilled residue, s and f are carbonised surface deposits.

Figure 1

Figure 2

Figure 3

Figure 4

Figure 5

Figure 6

A

B
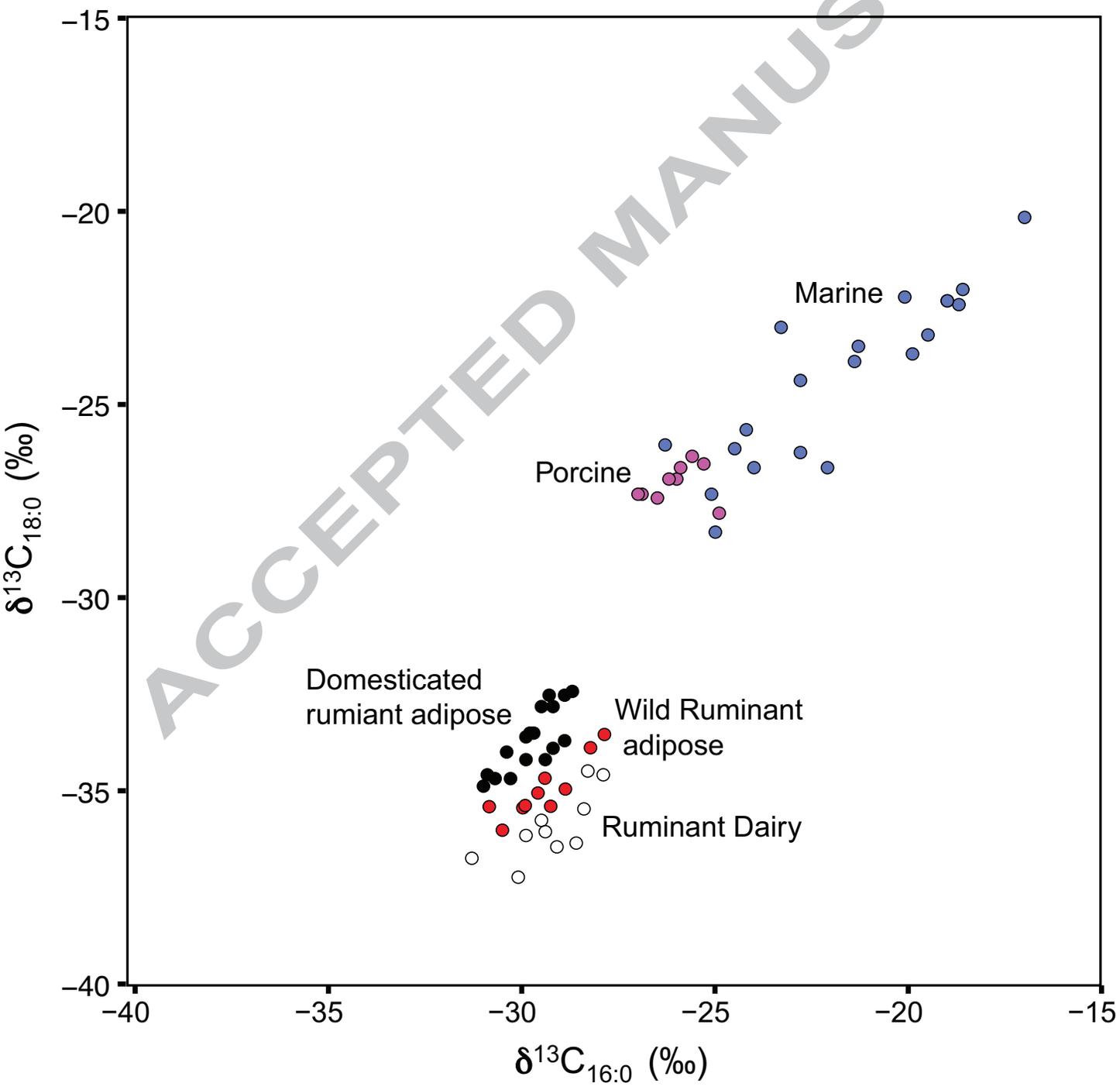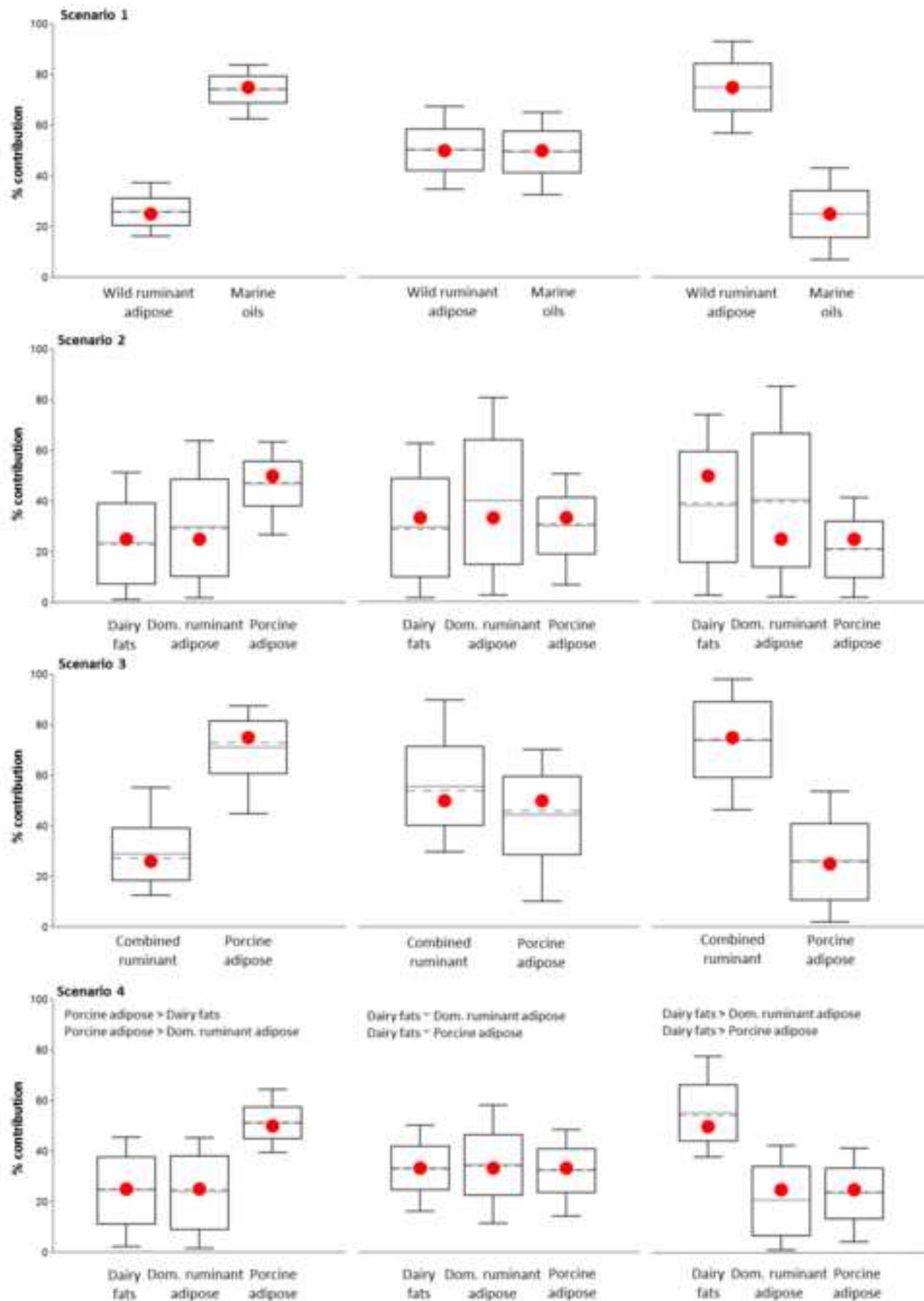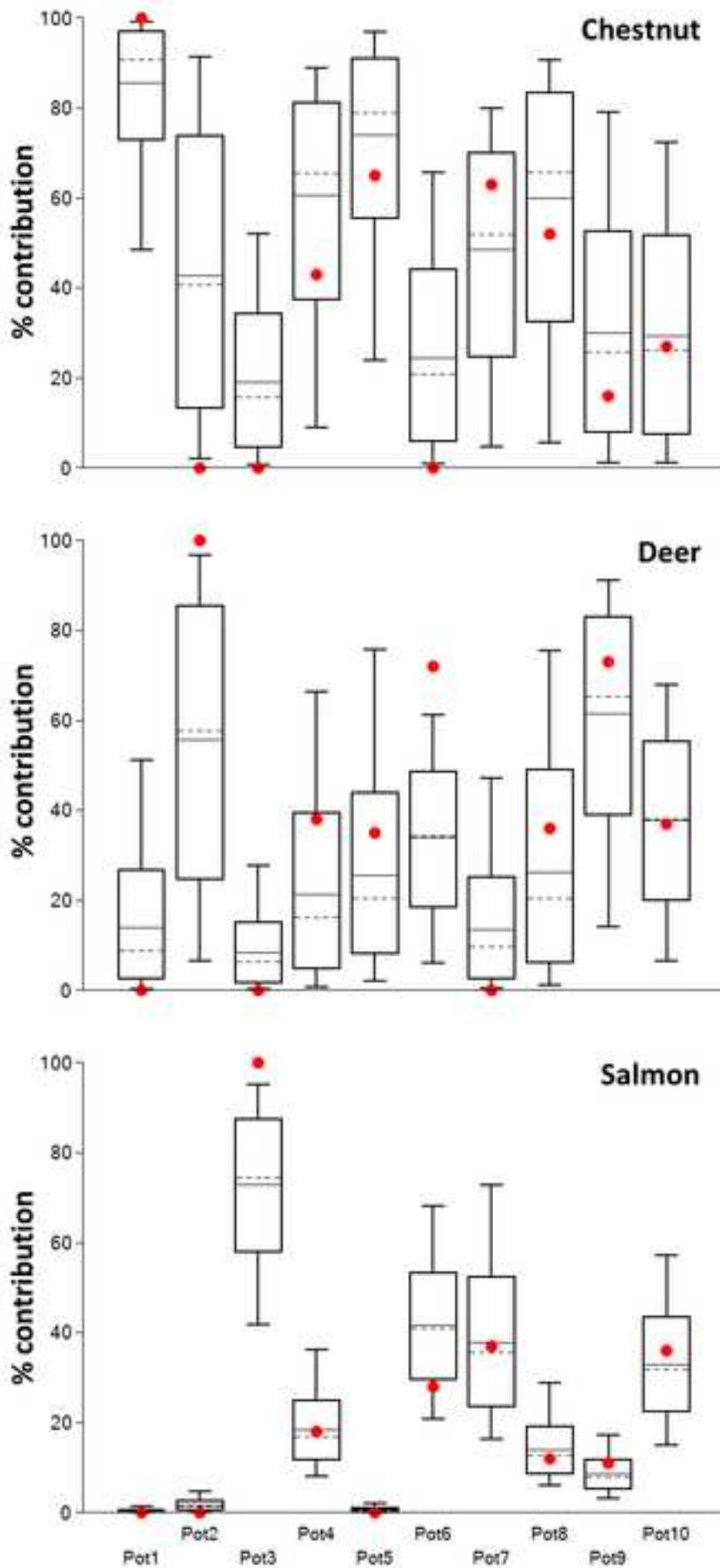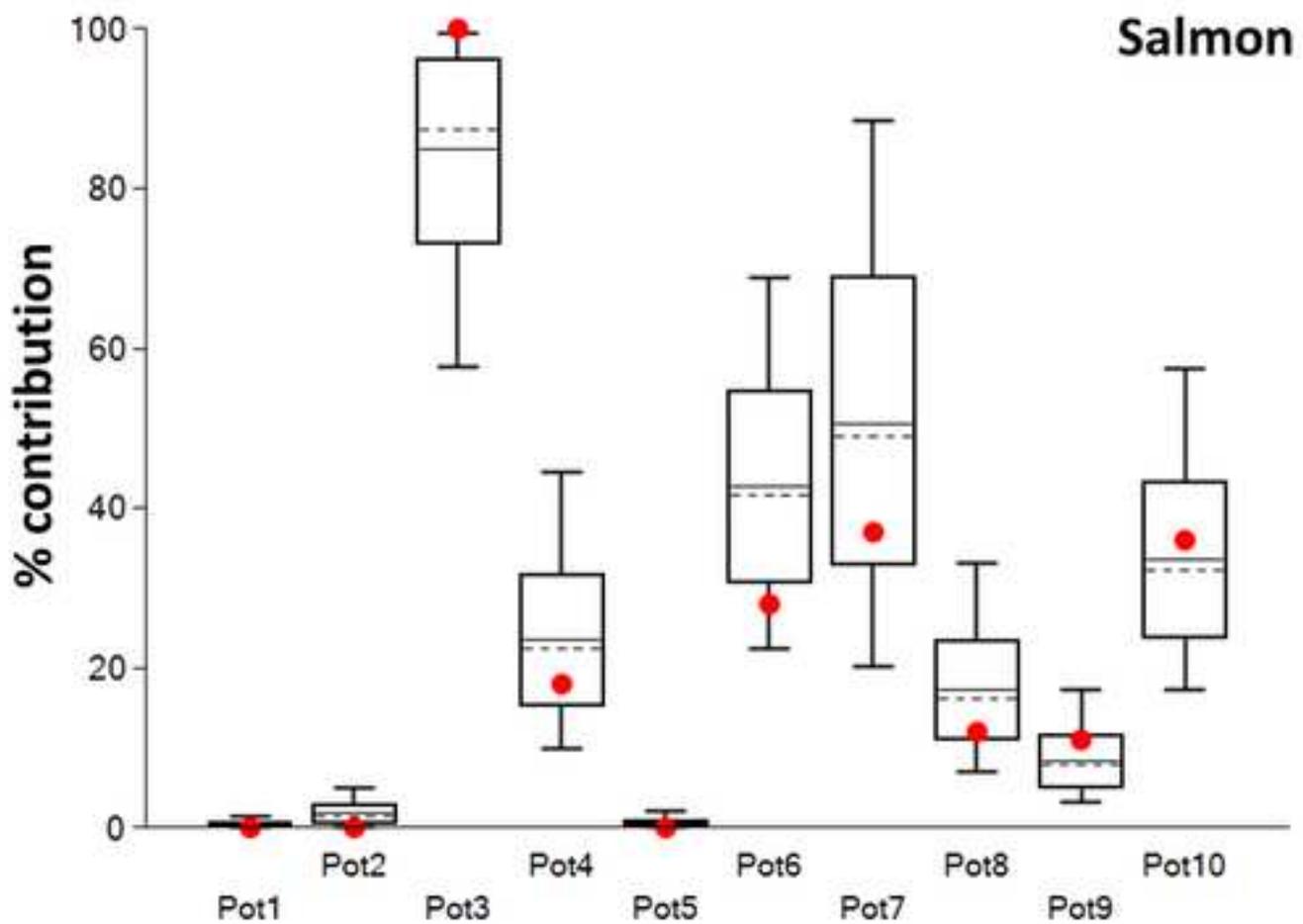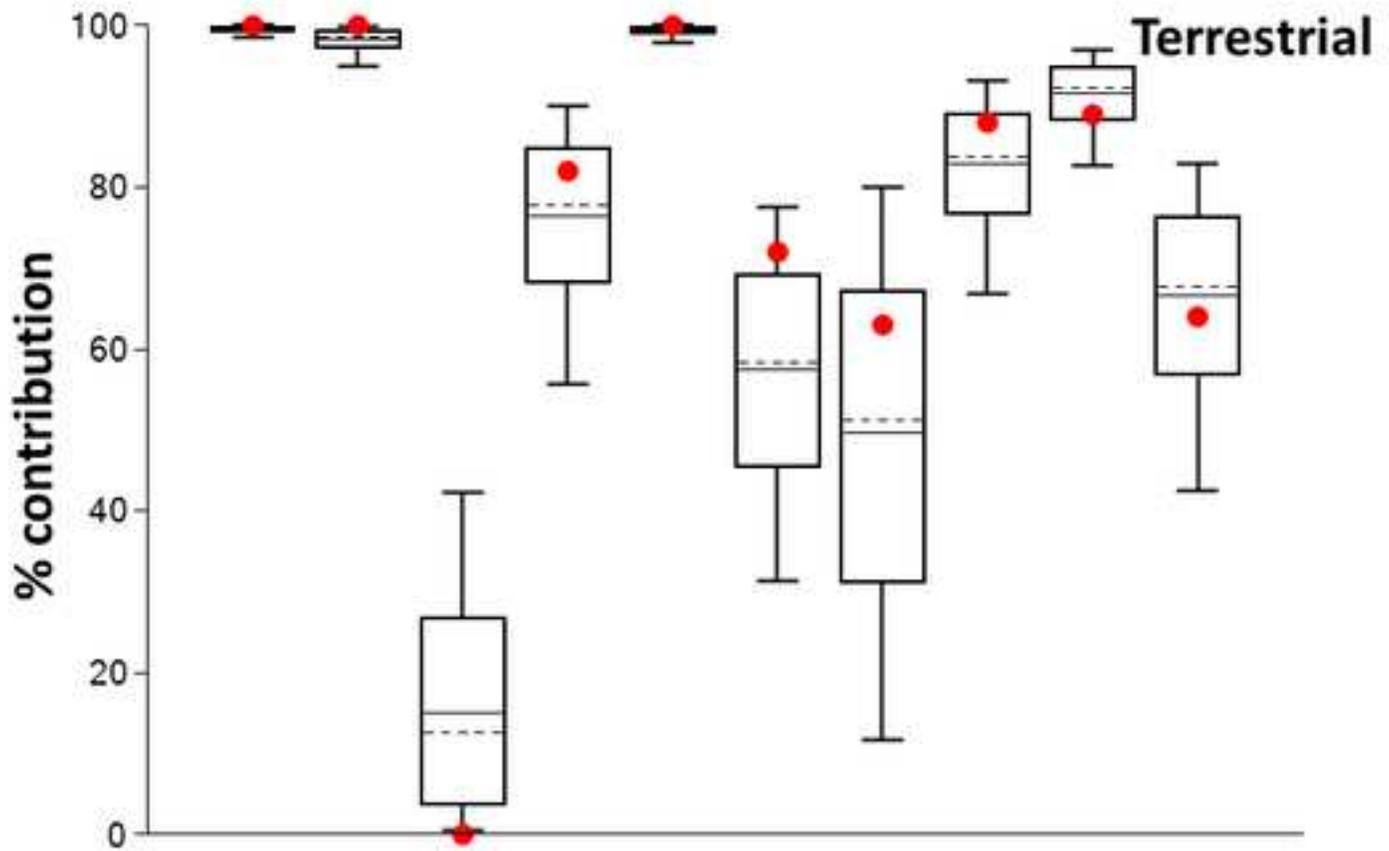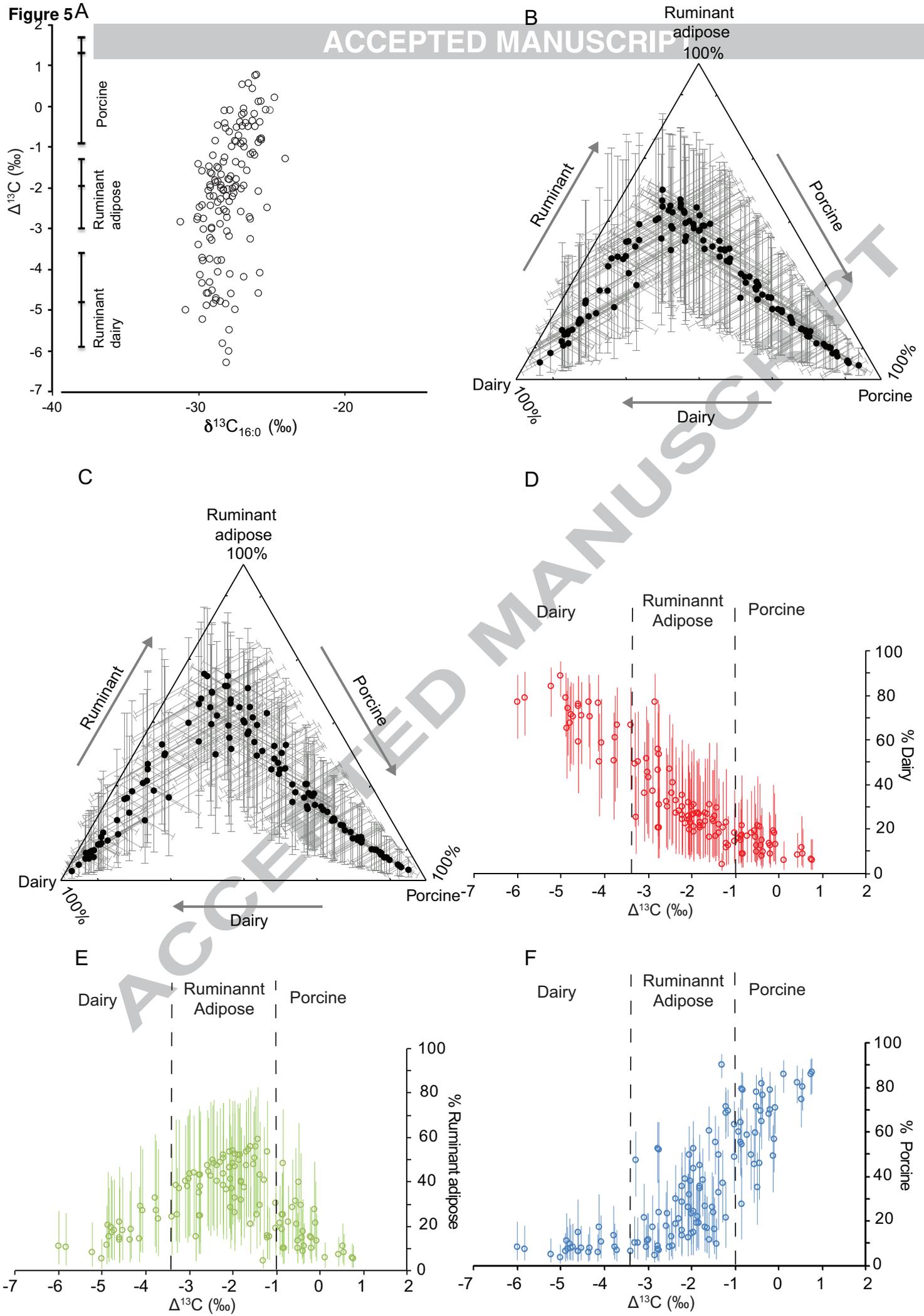
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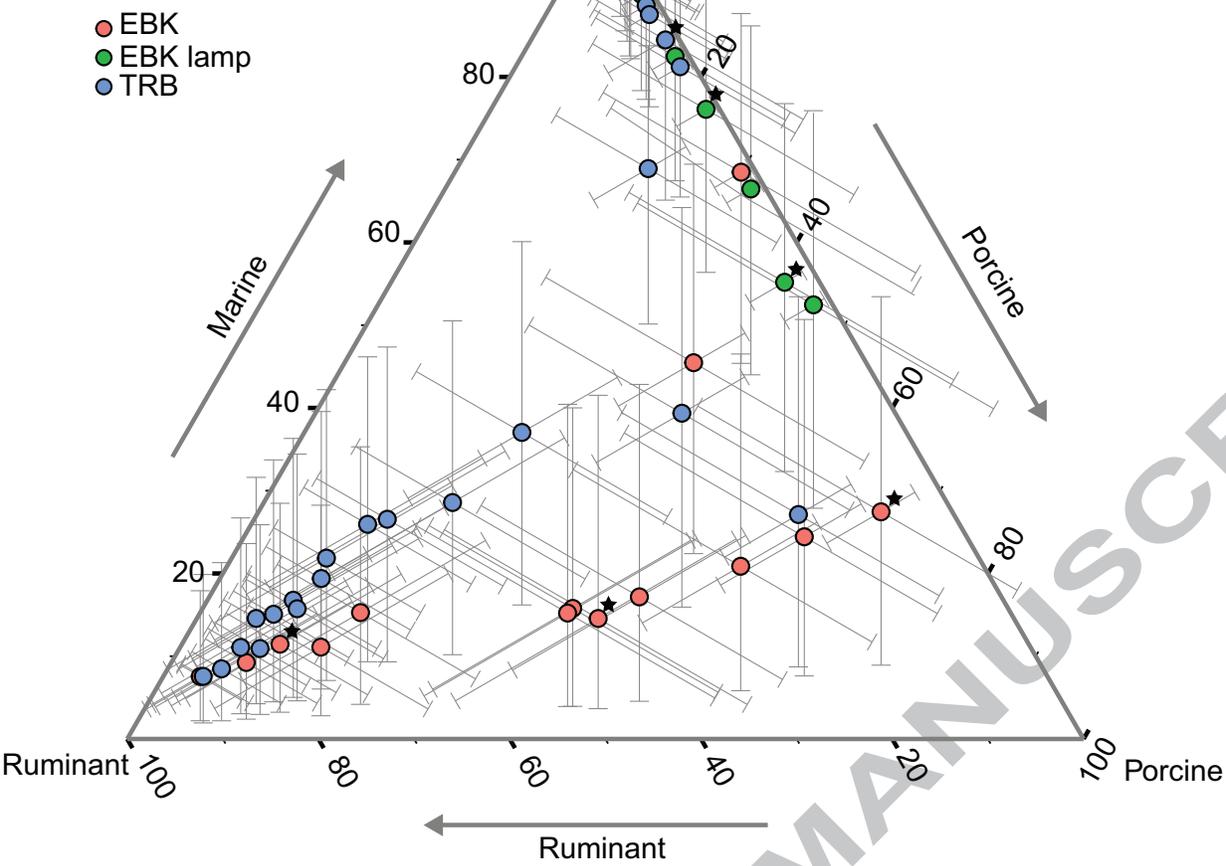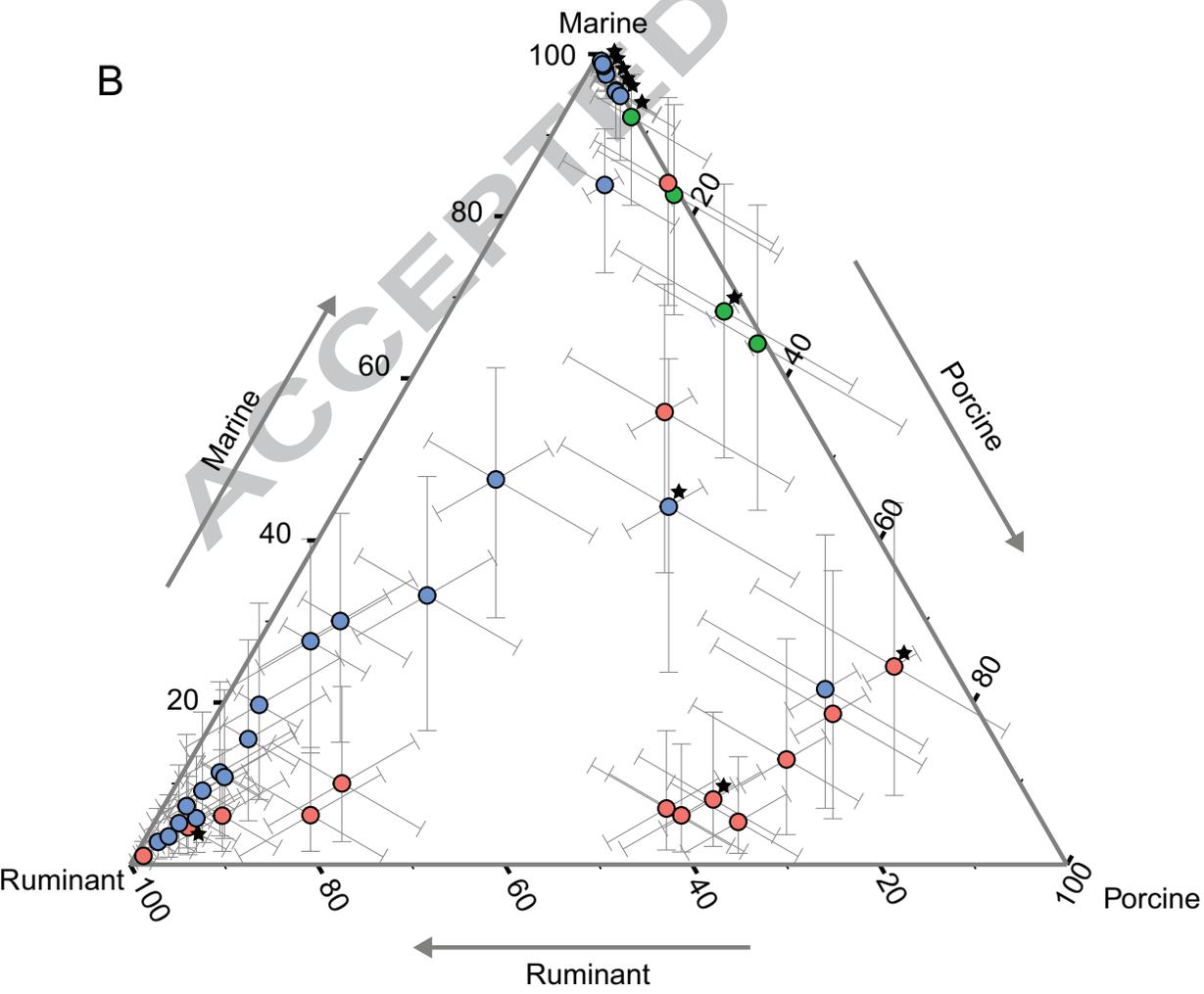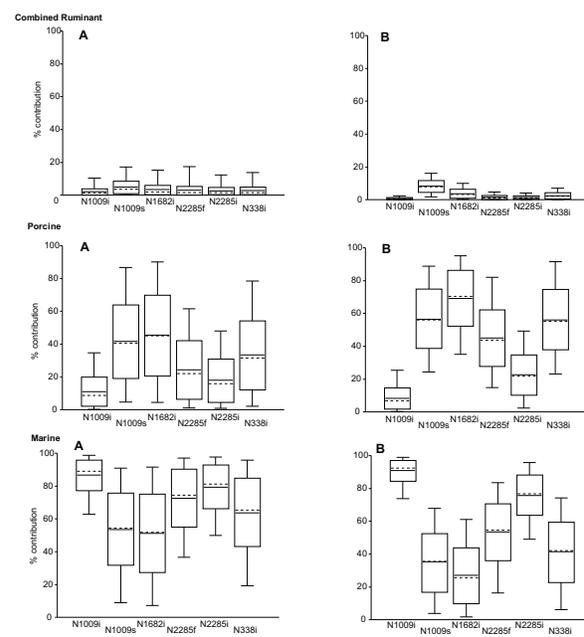
Tables

Table 1. Mean plus standard deviation fatty acid isotopic and concentration values for different lipid groups.

	Marine fats	Dairy fats	Domesticated ruminant adipose	Porcine adipose	Wild ruminant adipose	Combined ruminant
$\delta^{13}C_{16:0}$ (‰)	-21.6±2.7	-29.2±1.0	-29.6±0.7	-25.9±0.7	-29.4±0.9	-29.4±0.9
$\delta^{13}C_{18:0}$ (‰)	-22.1±2.2	-34±0.9	-31.7±0.8	-24.9±0.5	-33±0.8	-32.7±1.3
n (isotopes)	20	10	17	9	10	37
$C_{16:0}$ (% total FA)	13.6±3.9	34.6±7.2	24.2±5.0	24.1±2.1	17.2±3.6	26.3±8.6
$C_{18:0}$ (% total FA)	1.9±1.2	18.4±8.4	28.5±6.5	11.5±2.4	15.7±1.7	22.4±8.6
n (concentration)	15	10	13	9	6	29

Table 2. Known mixtures of foodstuffs added to the experimental pots as part of the YEAR cooking experiment.

Mass wet tissue added (g)	Experimental number										
	1	2	3	4	5	6	7	8	9	10	11*
Chestnut	22.5	0.0	0.0	14.2	13.8	0.0	14.2	13.0	3.9	6.5	0.0
Deer	0.0	21.3	0.0	12.7	7.5	18.6	0.0	9.2	17.0	9.1	0.0
Salmon	0.0	0.0	19.8	6.1	0.0	7.0	8.4	3.1	2.6	8.8	0.0
Total	22.5	21.3	19.8	33.0	21.3	25.6	22.6	25.2	23.4	24.4	0.0
% by mass in each pot											
Chestnut	100%	0%	0%	43%	65%	0%	63%	52%	16%	27%	0%
Deer	0%	100%	0%	38%	35%	72%	0%	36%	73%	37%	0%
Salmon	0%	0%	100%	18%	0%	28%	37%	12%	11%	36%	0%
Measured isotope value (‰)											
$\delta^{13}C_{16:0}$	-35.8	-33.5	-26.4	-30.6	-35.7	-27.2	-29.3	-31.1	-30.4	-27.9	n/a
$\delta^{13}C_{18:0}$	-37.9	-35.7	-26.3	-29.9	-36.5	-30.2	-27.6	-31.1	-34.6	-30.6	n/a

*Pot processed as a control containing only water

Table 3. Carbon isotope values and concentration of $C_{16:0}$ and $C_{18:0}$ fatty acids from the foodstuffs used in the cooking experiments.

Foodstuff	Carbon stable isotope value $\delta^{13}C$ (‰)		Fatty acid concentration (mg/g of wet mass)	
	$C_{16:0}$	$C_{18:0}$	$C_{16:0}$	$C_{18:0}$
Chestnut	-35.8	-38.1	0.631	0.095
Deer	-33.6	-35.3	0.839	0.872
Salmon	-26.6	-26.6	7.314	1.640

Highlights

Fatty acid (FA) stable carbon isotopes were used to investigate ancient pottery use

A Bayesian approach was applied to these data to estimate source contributions

This approach was tested using simulated, experimental, and archaeological data

Results obtained illustrate the advantages of a Bayesian approach

A probabilistic approach to the reconstruction of pottery use is recommended