Organic Geochemistry 117 (2018) 31–42

Contents lists available at ScienceDirect

Organic Geochemistry

journal homepage: www.elsevier.com/locate/orggeochem

Reconstruction of prehistoric pottery use from fatty acid carbon isotope signatures using Bayesian inference

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ARTICLE INFO

Article history:
Received 11 August 2017
Received in revised form 23 November 2017
Accepted 30 November 2017
Available online 7 December 2017

Keywords:
Fatty acids
Carbon isotopes
Pottery use
Bayesian mixing models
FRUITS

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.

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 ($\delta^{13}C$) 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

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https://doi.org/10.1016/j.orggeochem.2017.11.014

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 Eq. 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:

$$H_k \sim N\left(\mu_{H,k}, \sigma^2_{H,k}\right) \tag{1}$$

where

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

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: $0 \leqslant \alpha_i \leqslant 1$ for $i = 1, \ldots, n$ 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, $T_{ik} \sim N(\mu_{T,ik}, \sigma^2_{T,ik})$; 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 Eq. 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 (Eq. 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.

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

To link an inequality relationship, a parameter l (Eq. 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.

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

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 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 (Eq. 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 5000 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