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Compound-specific stable isotope analysis and radiocarbon dating of Irish bog butters reveal four millennia of dairy surplus and deposition

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Abstract

Bog butters are large white or yellow waxy deposits regularly discovered within the peat bogs of Ireland and Scotland. They represent an extraordinary survival of prehistoric and later agricultural products, comprising the largest deposits of fat found anywhere in nature. Often found in wooden containers or wrapped in animal bladders, they are considered to have been buried intentionally by past farming communities. While previous analysis has determined that Irish bog butters derive from animal fat, their precise characterisation could not be achieved due to diagenetic compositional alterations during burial. Via compound-specific stable isotope analysis, we provide the first conclusive evidence of a dairy fat origin for the Irish bog butter tradition, which differs from bog butter traditions observed elsewhere. Our research also reveals a remarkably long-lived tradition of deposition and possible curation spanning at least 3500 years, from the Early Bronze Age (c. 1700 BC) to the 17th century AD. This is conclusively established via an extensive suite of both bulk and compound-specific radiocarbon dates.

Introduction

Bog butters are large, white to yellow waxy deposits regularly recovered from the peat bogs of Ireland and Scotland, often found in wooden containers or wrapped in bark or animal membranes (Fig. 1). With recorded weights of up to 23 kg (and several examples that may be larger), bog butters were first documented in the 17th century; the total number recovered to date may approach 500 specimens (1, 2). Published radiocarbon determinations on Irish bog butters show activity spanning the Iron Age to the post-medieval period (3, 4) with folk accounts indicating survival into the 19th century (5, 6). While the reasons behind their

deposition continue to be debated (1, 2), the remarkable preservation properties of peat bogs are well known (7) and several post-medieval accounts mention the practice of storing butter in bogs to be consumed at a later date, whether by necessity or as a delicacy (8-10). Early medieval Irish law tracts list butter as one of the products payable as food rents (11), which may have needed to be stockpiled or stored. Parallels have also been drawn with the widespread deposition of metal and other objects in wetlands during the Bronze Age and Iron Age, often assumed to be votive or ritual acts (5, 12-14).

More than a century of chemical analyses has successfully determined that bog butters are derived from animal fat, although until recently the precise origins of bog butters could not be established due to diagenetic alterations during burial. The conclusion of early attempts was that they resembled adipocere rather than butter (15-19). Like bog butter, the chemical composition of adipocere is dominated by saturated fatty acids (mainly palmitic acid; C_{16:0} and stearic acid; C_{18:0}), with smaller amounts of unsaturated fatty acids (mainly oleic acid; Z-C_{18:1}), hydroxy fatty acids (mainly 10-hydroxystearic acid; C_{18:0}-OH) and intact triacylglycerols (19-22). In 2004, chemical analysis of nine Scottish bog butters using compound-specific stable carbon isotope measurements demonstrated six of the bog butters derived from a ruminant dairy source and three from ruminant carcass fat (tallow; 23). Here we report on analyses undertaken on 32 Irish bog butters (Table 1), with an accompanying programme of radiocarbon dating, to ascertain if similar practices took place in Ireland and if trends through time could be observed.

Classification of degraded animal fat remains using stable isotopes

In ruminant (e.g. cattle and sheep) and non-ruminant animals (e.g. pigs), adipose tissue is the main site for the storage of lipids, with triacylglycerols being by far the most abundant constituent, making up over 95% of the total lipids present (24). These triacylglycerols comprise of three fatty acids attached via ester linkages to a glycerol backbone, where the fatty acids mainly consist of an even number of acyl carbon atoms. In animal fats, acyl carbon chain lengths of C₁₆ and C₁₈ generally dominate (24, 25). In addition to adipose tissue, ruminant milk fats are also predominantly made up of triacylglycerols, but with a higher proportion of short chain fatty acids (26). The presence of these short chain fatty acids (C_{4:0} to C_{12:0}) in ruminant milk fats is in direct contrast to equivalent adipose fats, which contain very few fatty acids with chain lengths less than $C_{14:0}$ (26). Compared to other biochemical classes such as carbohydrates and proteins, the relative hydrophobic nature of lipids ensures their more frequent survival during archaeological timescales, with one of the most common finds being that of degraded animal fats (27). By using high-temperature gas chromatography (HTGC) and GC/mass spectrometry (GC/MS), such organic remains can easily be classified as deriving from animal fats based on the identification and distribution of free fatty acids (mainly C_{16:0} and C_{18:0}) and any remaining acylglycerols. Identification to species or fat type is much more difficult due to diagenetic transformations that occur during burial (28, 29). The heavier of the low molecular weight triacylglycerols, with acyl carbon numbers of C₄₀ and C₄₂, occasionally survive and can suggest a ruminant dairy fat origin rather than an adipose fat. However, dairy fat residues surviving archaeological timescales are often indistinguishable from adipose fat due to the increased solubility and hence preferential loss of the short chain fatty acids; for each additional methylene group of a fatty acid there is a fourfold decrease in its solubility (Fig. 2; 30).

With the aid of stable carbon isotope determinations (δ^{13} C values) on individual fatty acids, distinctions between the adipose fats of different animals can readily be achieved, first demonstrated by comparing the δ^{13} C values of lipid residues extracted from medieval dripping dishes and lamps with modern reference fats from pigs and ruminant animals (31, 32). Since then, traces of ruminant dairy fats (with δ^{13} C values ca. 2 to 4‰ less than adipose fat) have been successfully identified in a large number of pottery vessels from throughout Europe and the Near East (28, 33-35).

Results

a) Lipid residue analysis & stable carbon isotope measurements

Lipid compositions of the Irish bog butters were determined through GC and GC/MS analysis of each trimethylsilylated lipid extract (Supplementary Table S1). Similar to the findings of their Scottish equivalents (18, 23), free fatty acids with carbon numbers ranging from C_{12} to C_{20} (even over odd predominance) were the principal lipid components, with palmitic ($C_{16:0}$) and stearic ($C_{18:0}$) acids predominating (Fig. 2). Hydroxystearic acids (mainly 10-hydroxystearic) were also found to be present in seventeen of the bog butter samples with abundances ranging from 0.1% to 10.4% (mean 1.4%) of the total free fatty acids, respectively. These hydroxy fatty acids are known to be produced during adipocere formation (20) and were also identified in the Scottish bog butter samples (23). The other significant lipid components present in some of the Irish bog butters were acylglycerols, which demonstrates that hydrolysis to their component fatty acids had not gone to completion; fifteen of the bog butters contained triacylglycerols and eight contained diacylglycerols (Fig. 3). Nine (IB4-6, 13,

19, 20, 29, 30 and 32) of the bog butters were found to consist of triacylglycerol distributions with acyl carbon numbers ranging from C_{42} to C_{54} , while the remaining six (IB1, 10, 11, 22, 23 and 26) ranged from C_{44} to C_{54} acyl carbons. Those with acyl carbon distributions ranging from C_{42} to C_{54} are entirely consistent with a degraded dairy fat origin. However, those with acyl carbons ranging from C_{44} to C_{54} are more consistent with a ruminant adipose fat source, although a dairy origin cannot be discounted due to the presence and abundance of the C_{44} triacylglycerol. A more reliable approach in the identification of the Irish bog butter origins was through the measurement of the stable carbon isotope values ($\delta^{13}C$ values) of their $C_{16:0}$ and $C_{18:0}$ fatty acids. These values were compared against a global database of modern reference animal fats including animals from the UK raised on a pure C_3 diet (28; Supplementary Table S1).

The δ^{13} C values of the $C_{16:0}$ and $C_{18:0}$ fatty acids from the 32 Irish bog butters were plotted as a scatter graph with confidence ellipses (1 δ) representing ranges corresponding to reference non-ruminant adipose fats and ruminant adipose and dairy fats (Fig. 4a). Twenty-four samples plotted within the reference dairy fat ellipse and a further three (IB23, 30, 32) in very close proximity, suggesting a dairy fat origin. The precise origin for bog butter IB8 was unclear as a result of plotting between the reference ellipses of the ruminant adipose and dairy fats. IB1 and IB21 displayed δ^{13} C values more similar to a dairy fat origin; however, the δ^{13} C values for their $C_{16:0}$ fatty acids were approximately 1‰ more depleted in 13 C than reference dairy fats. Likewise, samples IB2 and IB28 also revealed values more similar to a ruminant dairy fat origin, but with both their $C_{16:0}$ and $C_{18:0}$ fatty acids more depleted in 13 C than reference dairy fat values. These bog butters may indeed have a dairy origin, with the observed values

occurring as a result of local isotopic differences in the diets of the ancient and modern reference animals. Such variations are negated by comparing the Δ^{13} C values (δ^{13} C_{18:0}- δ^{13} C_{16:0}) of the bog butters with the reference fat values (Fig. 4b), and here 26 of the 32 Irish bog butter samples were found to derive from a ruminant dairy origin with all values plotting within the range corresponding to a ruminant dairy fat. A further three bog butters (IB23, 30 and 32) were also likely to have derived from a dairy source as their Δ^{13} C values plotted just below the reference dairy fat range. A similar phenomenon has been noted for Δ^{13} C values from Irish Neolithic pot lipids and may be due to local environmental factors (35, 36). The remaining bog butters (IB1, 8 and 21) could not be precisely classified as their Δ^{13} C values plotted mid-way between the reference ranges for ruminant adipose and dairy fats. While no container was associated with IB1, both IB8 and IB21 were wrapped in animal bladders and their fatty acid δ^{13} C values may have been altered by the lipid content of these wrappings. As with the Scottish bog butters (23), where multiple samples were analysed from each bog butter mass, the homogeneity of bog butters and the robustness of the stable isotope methodology was confirmed. Analyses of sub-samples taken from the top (IB22tp) and bottom (IB22bm) of the same bog butter mass (IB22), revealed analogous results. Bog butter IB21, comprising two masses (IB21a and IB21b) wrapped separately, also provided very similar values.

b) Radiocarbon measurements on bulk bog butter samples

Radiocarbon dating undertaken for this study has provided 50 new measurements on 32 Irish bog butters, adding to the 19 previously published measurements on 19 examples (Supplementary Table S2). Together, they show bog butter manufacture and deposition spanning nearly four thousand years, from 1745-1635 BC (IB2; Knockdrin) to AD 1510-1800

(GrN-28728; Crovehy), generating new insights into both the butters and their associated containers (Fig 5a). Earwood's 1997 typology of kegs and churns (originally supported by six radiocarbon dates) holds up extremely well, with the 'Keg/Tub 2' type pushed back slightly from the Late to the Middle Iron Age, contemporary with the 'Keg/Tub 1' type. Most dramatic is the re-dating of the bowl from Killeenan More, Co. Galway (IB16), presumed to be medieval on the basis of its decoration (2, 3) but shown here to be Middle Iron Age. It joins the straightsided tubs from Glastonbury Lake Village as rare examples of prehistoric decorated wooden vessels (37, 38; Fig 5b). Two of the three bog butters returning Bronze Age dates (IB1, IB3) were measured using both bulk and compound-specific methods (Fig 5d), conclusively demonstrating that these unusually early dates are not due to contamination or other sample processing issues (39). However, there is further confirmation of the discrepancy between dates on butter and those on their containers, possibly due to contamination from the polyethylene glycol (PEG) used to consolidate wooden material (3). Bog butter samples from Rosberry (IB18) and Teernakill (IB19) are both several centuries younger than their containers, although a sample from Rosmoylan (IB12) is a similar age to its associated wooden keg (Supplementary Table S2). While the species (Alnus sp.) of the Rosberry keg is noted, none of the previously reported dates on wooden vessels include information on whether heartwood or sapwood was sampled (3, 40).

Discussion

Our analysis has confirmed that the substances known as 'bog butter' in Ireland are indeed butter, which is not as self-evident as one would suppose. While some 17th century sources mention the Irish burying butter in bogs, there are contemporary accounts of Faroe Islanders

burying sheep tallow prior to consumption (40), alongside clear evidence of adipose fat comprising many of the Scottish bog butters. Compound-specific stable isotope analysis provides the only method to conclusively establish Irish bog butter origins. Combining this analysis with radiocarbon measurements, we obtain unparalleled insight into an extremely long-lived activity. Clearly, it is unlikely there was a single reason for the deposition of bog butter over four millennia. Moreover, explanations which seek either a utilitarian or a ritual motivation perpetuate unhelpful categories that may not have applied in the past (41).

a) Bronze Age bog butter

Together with two recently dated samples (Supplementary Table S2), this study brings to five the number of Bronze Age bog butters recorded from Ireland. Their date is extremely significant and pushes back depositional activity by as much as 1500 years. Exact locational detail varies, but four of these five bog butters come from Co. Offaly: two are recorded approximately 8 km apart at Ballindown (IB3) and Drinagh townlands, while approximately 45 km to the northeast, two are recorded approximately 12 km apart at Esker More (IB1) and Knockdrin (IB2) townlands. A fifth example was recovered from Clonava townland, Co. Westmeath, approximately 35 km further to the northwest. These very early butter deposits may yet prove to be an isolated phenomenon, although the processing of milk is widespread in Ireland from the Early Neolithic onwards (35, 36) and gradual intensification of dairying over two millennia may have led to substantial surpluses being generated by the Early Bronze Age. The earliest dated sample, Knockdrin (IB2; 1745-1635 cal BC), and the Drinagh bog butter are both associated with bark, possibly a wrapping or container - a method of storage also evidenced in the Iron Age and Early Medieval period (Supplementary Table S2). In the early

2nd millennium BC, only small, round-based wooden bowls are known and pottery from non-funerary contexts is rare (3, 42), suggesting that deliberate choices were made about the materials used to store surplus food. While the acidic, anaerobic environment of bogs may have been utilised for temporary storage, there are wider patterns of depositional behaviour in the Early Bronze Age to be considered. Strict depositional rules have been observed for gold objects, axes and specialised bladed weapons (43); foods are an often-ignored category but may have also been infused with symbolism. In this regard, it may be no coincidence that both butter and gold are commonly deposited in bogs (cf 1).

b) Iron Age bog butter

Previous work has highlighted an apparent clustering of bog butter deposition in the Iron Age, as well as a possible focus on political and/or natural boundaries (14). Results from our study bring to 20 the number of recorded Iron Age bog butters (out of 46 dated samples), supporting this first observation although more research is needed to elucidate their relationship to boundaries. More than half (11/20) are associated with vessels, which in Iron Age Britain is recognised as a common category of votive object and argued to be linked to wider symbolic practices around food and agricultural fertility (13, 44-45). It is uncertain if such symbolism permeated the Irish examples, although we note fewer wooden vessels are associated with bog butter in the following Early Medieval period.

c) Early Medieval and Late Medieval bog butter

Just under half of bog butters examined are medieval in date (15 of the 32 samples analysed here; 22 out of 46 total dated samples). Both early and later medieval written sources contain

extensive references to dairy products, and butter is often portrayed as a luxury or upperclass food (11). It is generally included in food-rents, quantities ranging from the fist-size pats of butter listed in 7th/8th century law texts to the 'yearlie twenty fower methers of butter, and fiftie methers of barlie' exacted by Lisgole Abbey, Co. Fermanagh in 1609 (11, 46). Interestingly, texts do not mention the practice of depositing butter in bogs, although raiding of butter stores (*imenna*) is periodically recorded; food security must have been an issue for communities, with the storage of butter in bogs perhaps a wise precaution.

In terms of vessels, six of the thirteen Early Medieval (6th-12th centuries AD) bog butters were found in wooden containers, the remainder associated with leather, bladder or bark. Written sources indicate bark was commonly used for storing butter, with the Irish word *rúsc* meaning both bark and butter container (11). The *meadar* or mether, a distinctive quadrangular wooden vessel, appears in the Late Medieval period and is associated with two dated bog butters: Tawnagh Beg (IB10) and Goolamore, both from Co. Mayo (Supplementary Table S2). Recent research has distinguished between a 'drinking-type' and a 'container-type' mether (47), the former featuring spout-like or fluted corners with two to four handles positioned high on the vessel, and the latter with no fluted corners, two very large low-set handles, and larger. Tawnagh Beg (and possibly Goolamore) is one of the 'container-type' methers and its date of AD 1170-1280 provides the earliest known example of either vessel type.

d) Post-medieval/modern bog butter

The date ranges of up to three (out of 46) bog butters span the Irish post-medieval period (AD 1550-1850), with several 17th century accounts written by English observers (9, 10, 48-49)

providing the first mention of bog butter consumption. Experiments suggest that fresh butter deposited in bog conditions deteriorates relatively quickly and achieves a 'bog butter' condition in about two years (19), with aged or altered butters by no means inedible (50-51). Although our study has not identified any bog butters more recent than the 18th century, such a practice may have survived into the early 20th century in parts of rural Ireland (5), alongside the very widely documented folk superstitions and traditions associated with dairying and butter-making (6, 52-54).

Conclusions

Consistent with previous work, this investigation reveals that all sampled Irish bog butter were animal fats, which during burial had been diagenetically altered to resemble adipocere. GC-C-IRMS analyses of these substances revealed that twenty-six (81%) of the Irish specimens could confidently be assigned a ruminant dairy fat origin, with a further three samples (91% in total) probably deriving from a ruminant dairy fat. Only three samples (9%) could not be identified to origin, with their δ^{13} C values plotting between ranges expected for ruminant adipose and dairy fats. Deposition of butter in bogs in Ireland dates from at least the Early Bronze Age, a practice that may reflect intensification of a well-established dairying economy and thus increased likelihood of substantial surpluses of butter, a highly perishable but nutritionally valuable resource. Indeed, it may be that the burial of fats in the ground was much more widespread in antiquity than the archaeological record reflects. The survival of major hoards in bogs is consistent with their remarkable preservative properties; while burial of similar butter deposits in soil may well have achieved a similar goal of preservation in the

short term, these deposits would not survive to the present day. The Irish bog butters thus provide a unique encounter with a vitally important agricultural product.

Materials and methods

Samples from 32 bog butters (IB1-32) from various locations throughout Ireland were supplied by the National Museum of Ireland, Dublin (Fig. 2). Lipid analysis protocols and instrument conditions were described in detail previously (23). Briefly, bog butter samples (ca. 1 mg) were extracted in a mixture of chloroform and methanol (2:1 v/v; 10 ml) via ultrasonication and then centrifuged, filtered and evaporated under a gentle stream of N₂ to yield a total lipid extract (TLE). An aliquot of each TLE was trimethylsilylated using N,Obis(trimethylsilyl) trifluoroacetamide (20 µl; 70°C; 20 min) for analyses via HTGC and GCMS. Further aliquots were saponified using sodium hydroxide in methanol and double distilled water (9:1 v/v; 0.5M; 2ml; 70 °C, 1 h) and the fatty acids were converted into fatty acid methyl esters (FAME) using BF₃/methanol (14% w/v; 100 μl; 70 °C; 1 h) and analysed via GC-C-IRMS to determine their $\delta^{13}C$ values, which were corrected for added derivative carbon via a mass balance calculation (55). Bog butters were sampled from the middle of the mass, thus considered free of exogenous contaminant, and ca. 1.2 mg was directly weighed into tin capsules prior to graphitisation to obtain a bulk date. The compound-specific dates on single fatty acids were performed using preparative capillary gas chromatography for the isolation of single compounds (41). Bog butter samples (bulk and single compounds) were graphitised into an Automated Graphitisation Equipment (AGE3, Ion Plus) and the resulting graphite measured on the BRIS-MICADAS instrument (Ion Plus) at the BRAMS facility in Bristol.

Measurements were calibrated using OxCal v4.3.2 and the IntCal13 atmospheric curve (56, 57).

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Author contribution statement

Laboratory work was undertaken by RB (lipid analysis) and EC (radiocarbon dating); RPE, RB and JS designed the study, interpreted the data, and drafted the manuscript; JS compiled the

archaeological context to the study; FM, MS, IM and CS provided additional archaeological information and radiocarbon dates and commented on manuscript drafts. All authors gave final approval for publication.

Additional information

Datasets supporting this article have been uploaded as part of the supplementary material.

Each author declares the lack of conflicting interests/competing interests.



Figure 1: Examples of bog butter recovered from Irish contexts. Clockwise from top left: Rosberry, Co. Kildare (IB18), dated to 360-200 BC and deposited in a keg; Muckanagh, Co. Mayo (IB6), dated to AD 775-895 and associated with a wooden container; Tumgesh, Co. Mayo, deposited in a wooden mether; Shannagurraun, Co. Galway (IB8), dated to AD 960-1040 and wrapped in animal bladder. All dates this study. Images provided by kind permission of the National Museum of Ireland.

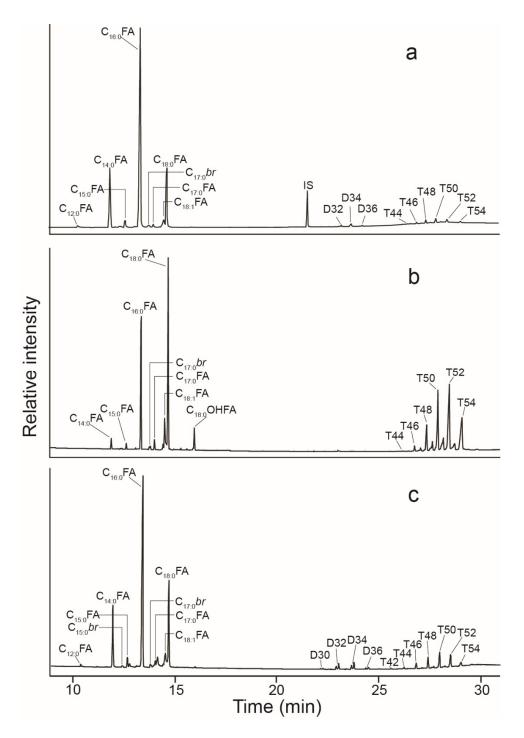


Figure 2: Partial high-temperature gas chromatograms of trimethylsilylated extracts from: (a) bog butter IB20, and adipoceres produced from b) mutton fat and c) New Zealand butter (19). Chromatographic peak identities are: $C_{12:0}$ FA to $C_{18:0}$ FA, saturated straight chain fatty acids with 12 to 18 carbons, respectively; $C_{18:1}$ FA, mono-unsaturated fatty acid with 18 carbon atoms; $C_{15:0br}$ and $C_{17:0br}$, branched chain fatty acids with 15 and 17 carbon atoms, respectively; $C_{18:0}$ OHFA, hydroxy fatty acid containing 18 carbon atoms; D_{32} to D_{36} , diacylglycerols with 32 to 36 acyl carbons, respectively; T_{44} to T_{54} , triacylglycerols with 44 to 54 acyl carbons, respectively; IS, internal standard, n-tetratricontane (n- C_{34}).

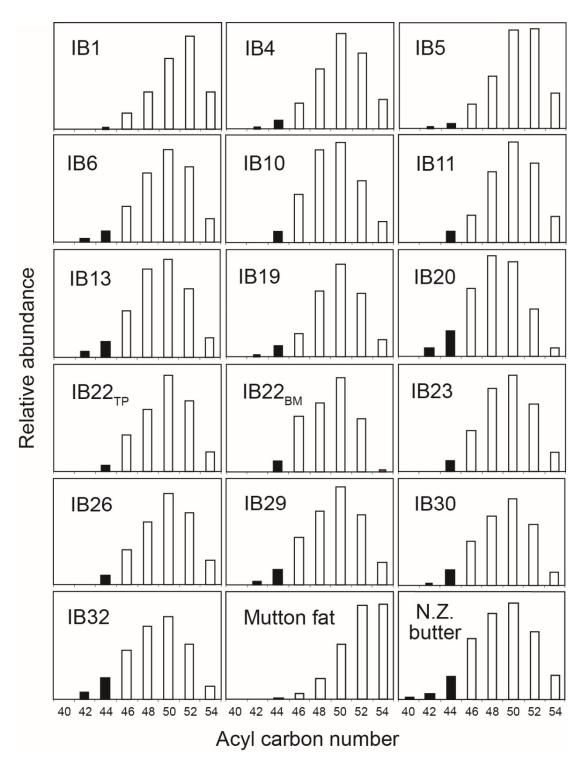


Figure 3: Histograms showing acyl carbon number distributions of triacylglycerols identified in the fifteen Irish bog butters that contained appreciable quantities and, for comparison, from previously reported in vitro adipocere formed from butter and mutton fat (19, 23). Shaded areas represent low molecular weight triacylglycerols, potentially deriving from a ruminant dairy fat. The abundance of each triacylglycerol component was calculated by integrating the peak areas in the HTGC profile.

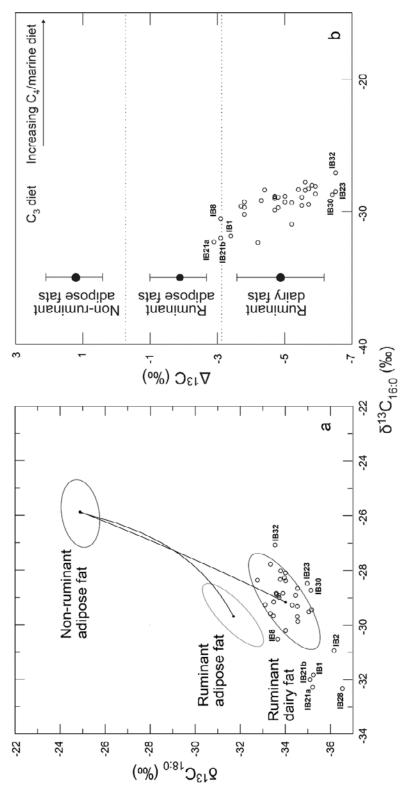


Figure 4: δ^{13} C values of methylated individual fatty acids ($C_{16:0}$ and $C_{18:0}$) from sampled Irish bog butters. 4a: δ^{13} C values plotted against reference ellipses (1 δ) derived from modern UK animal fats corrected for the contribution of post-industrial carbon by the addition of 1.2‰ (58). 4b: the same data with $\Delta\delta^{13}$ C values (= δ^{13} C_{18:0} - δ^{13} C_{16:0}) plotted against δ^{13} C_{16:0} values. Ranges of $\Delta\delta^{13}$ C values are based on a global database comprising modern reference animal fats from the UK, Africa, Kazakhstan, Switzerland and the Near East.

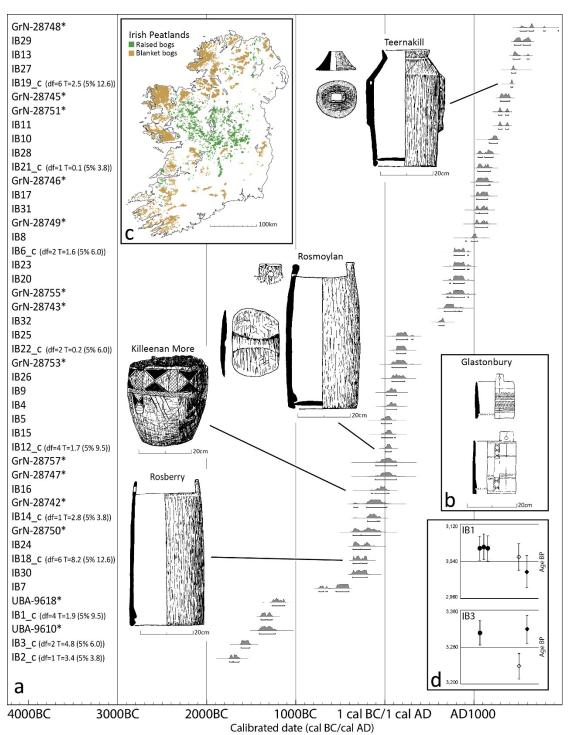


Figure 5: Dating of Irish bog butters. 5a: Radiocarbon date ranges for Irish bog butters, this study and additional specimens (marked with *; S2), with associated containers. Date ranges were calibrated using OxCal v4.3.2 and the IntCal13 atmospheric curve (56-57). 'C' denotes calculation of the weighted average of multiple measurements on the same bog butter, with the related chi-squared test results also displayed. T is the chi-squared value calculated and the value given in brackets is the level above which T should not rise in order to be acceptable. Degrees of freedom are given by df (number of dates minus one). All T results are below the respective values for rejection of the contemporaneity hypothesis at 2σ confidence level. 5b: Iron Age wooden vessels from Glastonbury Lake Village, UK; 5c: Irish

peatland distribution (59); 5d: Bulk and compound-specific radiocarbon measurements in years BP for Bronze Age bog butters IB1 and IB3. Dots correspond to bulk dates and diamonds to compound-specific dates. White diamonds = $C_{16:0}$ fatty acids, black diamonds = $C_{18:0}$ fatty acids. Error bars correspond to 1σ analytical uncertainty. Wooden vessels drawn by A. O'Sullivan, map drawn by C. McDermott.

Table 1: Irish bog butters sampled for this study

Code	County	Townland	Museum no.	Associated container
IB1	Offaly	Esker More	1998:62	
IB2	Offaly	Knockdrin	1998:63	Bark adhering to surface
IB3	Offaly	Ballindown	1986:125	
IB4	Laois	Baunaghra	1986:40	
IB5	Laois	Colt	1986:58	
IB6	Mayo	Muckinagh	2013:148.12	Wooden container?
IB7	Galway	Drinaun	1983:29.12	Bladder?
IB8	Galway	Shannagurraun	1983:28	Bladder
IB9	Kildare	Newtownbert	1967:102-103	Wicker basket
IB10	Mayo	Tawnagh Beg	1940:44	Mether
IB11	Mayo	Mullagh	1929:1343	Wooden
IB12	Roscommon	Rosmoylan	1962:101	Keg
IB13	Monaghan	Corlea	1965:275	Keg
IB14	Kerry	Tullamore	1954:16.12	Tub
IB15	Kildare	Hawkfield	1986:36	
IB16	Galway	Killeenan More	1939:994	Bowl

IB17	Mayo	Sheskin	1958:11	Bladder with bark
IB18	Kildare	Rosberry	1970:32	Keg with cord
IB19	Galway	Teernakill Bog	1925:14	Plunge churn
IB20	Donegal	Ards Beg	1987:112	Tub
IB21	Mayo	Ballyguin	1943:314-5	Two bladders in wooden container
IB22	Tipperary	Derrycoogh	1991:13	Bark wrappings
IB23	Mayo	Derryloughan	M1948:4	Wooden
IB24	Kildare	Killinagh	1929:1298	Wooden
IB25	Limerick	Glennacowan	1943:54	
IB26	Meath	Ardanew	1930:195	Keg
IB27	Mayo	Rosdoagh	1968:440A	Stave tub
IB28	Leitrim		2018:29-30	Bark
IB29	Mayo	Gowlaune	2018:27-28	Bark
IB30	Sligo	Cloncoose	2007:38	Bark
IB31	Mayo	Derragh	2007:39	
IB32	Mayo	Knockmoyle	1986:39	Keg

Compound-specific stable isotope analysis and radiocarbon dating of Irish bog butters reveal four millennia of dairy surplus and deposition

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Supplementary Table S1:

Lipid composition and $\delta^{13}C$ values of analysed Irish bog butters. Key to major lipid components: FFA C_x are straight chain free fatty acids of carbon length x, OHFA C_x are hydroxy fatty acids of carbon length x; DAG C_x are diacylglycerols of carbon length x; TAG C_x are triacylglycerols of carbon length x.

Code	Major lipid components	δ ¹³ C _{16:0} fatty acid (‰)	δ ¹³ C _{18:0} fatty acid (‰)	Assignment
IB1	FFA(C ₁₂ -C ₂₀); OHFA(C ₁₈); TAG(C ₄₄ -C ₅₄)	-31.84	-35.24	Possible dairy fat
IB2	FFA(C ₁₂ -C ₁₈); OHFA(C ₁₈); TAG(C ₄₄ -C ₅₄)	-30.95	-36.16	Dairy fat
IB3	FFA(C ₁₂ -C ₂₀); OHFA(C ₁₈); TAG(C ₄₄ -C ₅₄)	-29.70	-34.53	Dairy fat
IB4	FFA(C ₁₂ -C ₁₈); OHFA(C ₁₈); TAG(C ₄₂ -C ₅₄)	-28.89	-33.60	Dairy fat
IB5	FFA(C ₁₂ -C ₁₈); OHFA(C ₁₈); TAG(C ₄₂ -C ₅₄)	-29.17	-33.48	Dairy fat
IB6	FFA(C ₁₂ -C ₂₀); OHFA(C ₁₈); DAG(C ₃₂ -C ₃₆); TAG(C ₄₂ -C ₅₄)	-29.53	-35.04	Dairy fat
IB7	FFA(C ₁₂ -C ₁₈)	-28.92	-33.70	Dairy fat
IB8	FFA(C ₁₂ -C ₁₈)	-30.53	-33.66	Possible dairy fat
IB9	FFA(C ₁₂ -C ₁₈)	-28.03	-33.79	Dairy fat
IB10	FFA(C ₁₂ -C ₁₈); DAG(C ₃₂ - C ₃₆); TAG(C ₄₄ -C ₅₄)	-28.34	-33.77	Dairy fat
IB11	FFA(C ₁₂ -C ₁₈); DAG(C ₃₂ - C ₃₆); TAG(C ₄₄ -C ₅₄)	-29.59	-33.33	Dairy fat
IB12	FFA(C ₁₂ -C ₁₈)	-29.28	-34.30	Dairy fat
IB13	FFA(C ₁₄ -C ₂₀); DAG(C ₃₀ - C ₃₆); TAG(C ₄₂ -C ₅₄)	-29.01	-33.72	Dairy fat
IB14	FFA(C ₁₄ -C ₁₈)	-29.88	-34.55	Dairy fat
IB15	FFA(C ₁₂ -C ₁₈)	-28.39	-34.01	Dairy fat
IB16	FFA(C ₁₂ -C ₁₈)	-28.85	-33.89	Dairy fat
IB17	FFA(C ₁₂ -C ₁₈)	-29.27	-33.10	Dairy fat
IB18	FFA(C ₁₂ -C ₁₈)	-30.21	-34.01	Dairy fat
IB19	FFA(C ₁₄ -C ₁₈); DAG(C ₃₀ - C ₃₆); TAG(C ₄₂ -C ₅₄)	-28.37	-32.76	Dairy fat
IB20	FFA(C ₁₄ -C ₁₈); DAG(C ₃₂ - C ₃₆); TAG(C ₄₂ -C ₅₄)	-27.78	-33.34	Dairy fat

IB21a	FFA(C ₁₂ -C ₁₈); OHFA(C ₁₈)	-32.29	-35.21	Possible dairy fat
IB21b	FFA(C ₁₂ -C ₁₈); OHFA(C ₁₈)	-32.01	-35.09	Possible dairy fat
IB22tp	FFA(C ₁₂ -C ₁₈); OHFA(C ₁₈); TAG(C ₄₄ -C ₅₄)	-28.10	-34.00	Dairy fat
IB22bm	FFA(C ₁₂ -C ₁₈); OHFA(C ₁₈); TAG(C ₄₄ -C ₅₄)	-28.28	-33.96	Dairy fat
IB23	FFA(C ₁₂ -C ₁₈); OHFA(C ₁₈); TAG(C ₄₄ -C ₅₄)	-28.49	-34.97	Probable dairy fat
IB24	FFA(C ₁₂ -C ₁₈); OHFA(C ₁₈)	-28.92	-34.45	Dairy fat
IB25	FFA(C ₁₂ -C ₁₈); OHFA(C ₁₈)	-29.46	-35.16	Dairy fat
IB26	FFA(C ₁₀ -C ₁₈); OHFA(C ₁₈); DAG(C ₃₂ -C ₃₆); TAG(C ₄₄ -C ₅₄)	-29.33	-34.53	Dairy fat
IB27	FFA(C ₁₂ -C ₁₈); OHFA(C ₁₈)	-29.68	-33.47	Dairy fat
IB28	FFA(C ₁₂ -C ₁₈)	-32.34	-36.53	Dairy fat
IB29	FFA(C ₁₂ -C ₂₀); OHFA(C ₁₈); DAG(C ₃₀ -C ₃₆); TAG(C ₄₂ -C ₅₄)	-28.67	-34.54	Dairy fat
IB30	FFA(C ₁₂ -C ₂₀); OHFA(C ₁₈); TAG(C ₄₂ -C ₅₄)	-28.74	-35.13	Probable dairy fat
IB31	FFA(C ₁₂ -C ₁₈); OHFA(C ₁₈)	-28.85	-33.59	Dairy fat
IB32	FFA(C ₁₂ -C ₂₀); OHFA(C ₁₈); TAG(C ₄₂ -C ₅₄)	-27.08	-33.54	Probable dairy fat

Supplementary Table S2: Radiocarbon determinations for Irish and Scottish bog butters. All measurements calibrated using OxCal v4.3.2 and IntCal13 atmospheric curve (1, 2)

Code	Townland	County	¹⁴ C age BP and associated lab code (this study)	Calibrated date range (2σ)	Associated ¹⁴ C determination (years BP)	Calibrated date range (2σ)	NMI accession no.	Container * Earwood 1997 type	Refs	
IB1	B1 Esker More	Offaly	3069±16; BRAMS-1087.1 3050±30; BRAMS-1087.4.1 (C _{16:0} FA)	1405-1275 BC 1410-1220 BC		1998:	1998:62			
	201.01 111.01	J	3018±35; BRAMS-1087.4.2 (C _{18:0} FA)	1400-1120 BC						
IB2	Knockdrin	Offaly	3431±34; BRAMS-1088.1	1880-1840, 1830- 1640 BC	3368±22; UBA-9617; butter	1740-1715, 1695-1615 BC	1998:63	Bark adhering to surface		
IB3	Ballindown	Offaly	3308±34; BRAMS-1089.1 3239±28; BRAMS-1089.2.1 (C _{16:0} FA)	1670-1500 BC 1610-1440 BC			1986:125			
			3319±31; BRAMS-1089.2.2 (C _{18:0} FA)	1690-1510 BC						
IB4	Baunaghra	Laois	1924±34; BRAMS-1090.1	20 BC-AD 170, AD 200-210			1986:40		3	
IB5	Colt	Laois	1962±34; BRAMS-1091.1	50 BC-AD 80, AD 100-130	1880±40; GrN-28752; butter	AD 50-240	1986:58		3-5	
		Мауо	1153±25; BRAMS-1092.1	AD 770-970						
IB6	Muckanagh		1201±29; BRAMS-1092.2.1 (C _{16:0} FA) 1168±34; BRAMS-1092.2.2	AD 719-750, 760- 900, 930-940 AD 770-970			2013:148.12	Wooden container		
			(C _{18:0} FA)							
IB7	Drinaun	Galway	2422±26; BRAMS-1093.1	750-680, 670-640, 550-400 BC			1983:29.12	Bladder?	3	
IB8	Shannagurraun	Galway	1027±27; BRAMS-1153.1	AD 900-920, 960- 1040			1983:28	Bladder	3	
IB9	Newtownbert	Kildare	1923±27; BRAMS-1154.1	AD 20-140			1967:102-103	Wicker basket	3	
IB10	Tawnagh Beg	Mayo	814±25; BRAMS-1094.1	AD 1170-1270			1940:44	Mether	3	
IB11	Mullagh	Мауо	670±25; BRAMS-1095.1	AD 1270-1320, 1350-1390			1929:1343	Wooden		
		lan Roscommon	1971±15; BRAMS-1096.	1971±15; BRAMS-1096.1	25-10 BC, 5 BC-AD 75					
IB12	Rosmoylan		1919±37; BRAMS-1096.2.1 (C _{16:0} FA)	AD 0-180, 190-220		50 BC-AD 180, 190-220	1962:101	Keg *K2	3, 6-7	
			1967±40; BRAMS-1096.2.2 (C _{18:0} FA)	50 BC-AD 130						
IB13	Corlea	Monaghan	401±25; BRAMS-1097.1	AD 1430-1520, 1590-1620			1965:275	Keg *K5	3, 8	
IB14	Tullamore	Kerry	2087±26; BRAMS-1098.1	180-40 BC	2160±35; GrN-28744; butter	360-90 BC	1954:16.12	Tub *T2	3-5	

Code	Townland	County	¹⁴ C age BP and associated lab code (this study)	Calibrated date range (2σ)	Associated ¹⁴ C determination (years BP)	Calibrated date range (2σ)	NMI accession no.	Container * Earwood 1997 type	Refs
IB15	Hawkfield	Kildare	1959±26; BRAMS-1099.1	40 BC-AD 90, 100- 120			1986:36		3
IB16	Killeenan More	Galway	2035±26; BRAMS-1100.1	160-130 BC, 120 BC-AD 30, 40-50			1939:994	Bowl *FRB	9
IB17	Sheskin	Mayo	945±25; BRAMS-1101.1	AD 1020-1160			1958:11	Bladder with bark	3
			2192±15; BRAMS-1102.1	360-270, 260-190 BC					
			2165±29; BRAMS-1102.3.1 (C _{16:0} FA)	360-150, 140-110 BC	2270±30; GrN-23287;	400-350, 310- 210 BC			
IB18	Rosberry	Kildare	2124±29; BRAMS-1102.4.1 (C _{16:0} FA)	350-320, 210-50 BC	butter		1970:32	Alder keg *K1 with alder cord	3, 10-11
			2199±37; BRAMS-1102.3.2 (C _{18:0} FA)	380-170 BC	2740±50; GrA-5457; wood	1000-800 BC			
			2211±35; BRAMS-11-2.4.2 (C _{18:0} FA)	380-190 BC					
			509±15; BRAMS-1103.1	AD 1410-1440					
			536±37; BRAMS-1103.2.1	AD 1310-1360,					
			(C _{16:0} FA)	1380-1450		AD 1040-1100.			
IB19	Teernakill Bog (Teernakill North or South)	Galway	499±32; BRAMS-1103.3.1 (C _{16:0} FA)	AD 1320-1340, 1390-1450	840±50; GrA-6015; — wood	1110-1280 1925:	1025:14	Plunge churn *PC1	3, 9
1019			486±38; BRAMS-1103.2.2	AD 1320-1350,			1925.14		3, 9
			(C _{18:0} FA)	1390-1470					
			476±29; BRAMS-1103.3.2 (C _{18:0} FA)	AD 1410-1460					
IB20	Ards Beg	Donegal	1198±25; BRAMS-1104.1	AD 720-740, 760- 900			1987:112	Tub *T	7
IB21a		Mayo	899±27; BRAMS-1155.1	AD 1040-1210				Two bladders	
IB21b	Ballyguin		912±27; BRAMS-1155.2	AD 1030-1190			1943:314-5	in wooden container	3
IB22tp IB22bm	Derrycoogh	Tipperary	1827±27; BRAMS-1156.1 1829±27; BRAMS-1156.2	AD 90-100, 120-260 AD 90-100, 120-250	1850±40; GrN-28754; butter	AD 60-260	1991:13	Bark wrappings	3-5
IB23	Derryloughan	Mayo	1177±25; BRAMS-1105.1	AD 770-900, 920- 950			M1948:4	Wooden	
IB24	Killinagh	Kildare	2173±26; BRAMS-1106.1	360-160 BC			1929:1298	Wooden	
IB25	Glennacowan	Limerick	1813±26; BRAMS-1107.1	AD 120-260, 300- 320			1943:54		
IB26	Ardanew	Meath	1865±26; BRAMS-1108.1	AD 70-230			1930:195	Keg *K2	7
IB27	Rosdoagh	Мауо	448±25; BRAMS-1109.1	AD 1420-1470			1968:440A	Hazel stave tub	3, 7, 12
IB28		Leitrim	887±25; BRAMS-1110.1	AD 1040-1100, 1110-1220			2018:29-30	Bark	
IB29	Gowlaune	Mayo	357±25; BRAMS-1111.1	AD 1450-1530, 1550-1640			2018:27-28	Bark	
IB30	Cloncoose	Sligo	2211±26; BRAMS-1112.1	370-200 BC			2007:38	Bark	
IB31	Derragh	Mayo	959±25; BRAMS-1113.1	AD 1020-1160			2007:39		
IB32	Knockmoyle	Mayo	1399±26; BRAMS-1114.1	AD 600-670			1986:39	Keg *K4	3

Code	Townland	County	¹⁴ C age BP and associated lab code (this study)	Calibrated date range (2σ)	Associated ¹⁴ C determination (years BP)	Calibrated date range (2σ)	NMI accession no.	Container * Earwood 1997 type	Refs
	Inchimacteige	Kerry			2090±50; GrN-28742	360-300 BC, 210 BC-AD 30	1941:1118	'Firkin'	4, 5
	Lonart	Kerry			1265±35; GrN-28743	AD 660-780, 790-870	M1948:1	Wooden	4
	Goolamore	Mayo			615±30; GrN-28745	AD 1290-1410	1955:45.12	Mether	4, 5
	Enagh	Cavan			925±30; GrN-28746	AD 1020-1190	1959:747	Bladder	4, 5
	Rathbaun	Sligo			1980±50; GrN-28747	120 BC-AD 130	1967:144	Bladder	4, 5
	Crovehy	Donegal			265±30; GrN-28748	AD 1510-1600, 1610-1670, 1780-1800	1967:212.12	Animal skin?	4, 5
	Canburrin	Kerry			970±30; GrN-28749	AD 1010-1160	1971:1006	Cylindrical bark envelope	4
	Annaghbeg	Kerry			2150±50; GrN-28750	360-50 BC	1975:235	Tub	4
	Corragarrow	Longford			670±25; GrN-28751	AD 1270-1320, 1350-1390	1984:152	Wooden keg	4, 5
	Killoran	Tipperary			1840±35; GrN-28753	AD 70-250	1990:119	Wicker	4, 5
	Inchiboy	Kerry			1205±30; GrN-28755	AD 710-750, 760-900, 930- 940	1995:97	Bark	4
					1970±50; GrN-28757	110 BC-AD 140			4, 5
	Clonava	Westmeath			3061±33; UBA-9610	1420-1230 BC	2016:223		
	Drinagh	Offaly			2981±22; UBA-9618	1270-1120 BC	1977:2176	Bark and withies?	
	Morvern	Argyle			1802±35; UB-3185	AD 120-330	NMS ME166	Alder keg	3, 4, 13
	Kyleakin	Skye			1730±35; UB-3186	AD 230-400	NMS ME167	Alder keg	4, 13-14
	High Borve	Lewis			949±20; UB-3737	AD 1025-1155		Two balls wrapped in intestine	3-4, 15

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