



Contents lists available at ScienceDirect

Journal of Archaeological Science: Reports

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

From the inside out: Upscaling organic residue analyses of archaeological ceramics

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ARTICLE INFO

Article history:

Received 3 October 2015

Received in revised form 14 March 2016

Accepted 13 April 2016

Available online xxxx

Keywords:

Lipid residue analyses

Herding strategies

Plant processing

Trade

Food technology

Subsistence

Dating

ABSTRACT

Investigations of organic residues associated with archaeological pottery using modern analytical chemical methods began in the 1970s. There was early recognition that the analysis of lipids (i.e. fats, waxes and resins) preserved in surface residues or the fabric of single pottery sherds, representative of single vessels, was a powerful method for defining pottery use at higher specificity. Subsequent developments saw a significant change of scale with studies usually involving lipid analyses of tens to hundreds of sherds per archaeological assemblage, providing information which extends beyond pottery use. The identification of animal and plant foodstuffs processed in pots lends insights into herding and farming; while trade in exotic organic goods can also be detected. Information about environment and climate can be derived from the isotopic composition of compounds detected in sherds, providing potentially novel avenues of investigation. The direct dating of lipids in pottery sherds is opening up new opportunities for building archaeological chronologies. The integration of lipid residue analyses with other environmental and cultural proxies in interdisciplinary projects is already providing unprecedented insights into past lifestyles from site to regional scales.

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1. Introduction

The maxim 'absence of evidence is not evidence of absence' is often invoked in archaeology, and holds especially true for organic remains, which generally degrade over archaeological timescales. Developments in organic residue analysis have been largely driven by developments in analytical chemistry, particularly advances in chromatographic and mass spectrometric instrumentation. The key advantage of modern analytical methods is their capacity to resolve the many biomolecular components that typically comprise organic residues in archaeology. Furthermore, the high sensitivities of the instruments are especially compatible with the extremely low concentrations of organic components preserved. Determination of the structures of compounds, or suite(s) of compounds, originating from plant and animal sources, known as "biomarkers" (Philp and Oung, 1988) in sediments, ceramics and other matrices substantially widens the available evidence base for archaeology (Evershed, 2008b).

Visible organic residues are well-known to archaeologists, appearing as encrusted deposits adhering to the interior or exterior surface of a vessel, which may derive from burnt residues (Fig. 1a), soot, etc. deposited by heating of the vessel over fire, or from materials used as decoration, sealants or adhesives (Fig. 1b). However, these visible residues

occur rarely in pottery assemblages and are prone to post-burial and post-excavation loss during cleaning of sherds and/or contamination (Fig. 2). Since avoiding or removing soil/handling contamination from visible or surface residues is so fraught with difficulties, such residues are not the preferred choice for this type of analysis.

Hence, the majority of analyses now target absorbed organic residues preserved in unglazed ceramic vessels, which generally originate from the original contents either stored or processed in the vessels, representing either a single use or an accumulation of cooking events over a vessel's life history. Early attempts to extract and analyse absorbed residues revealed the presence of highly unstable compounds, such as unsaturated fatty acids and human-derived lipids such as cholesterol (Rottländer, 1990), likely arising from modern contamination. Thorough mechanical removal of sherd surfaces before powdering the sub-surface fabric allows contaminating compounds from the burial environment and/or handling to be removed (Condamin et al., 1976; Heron et al., 1991). Critically, migration of lipids from the soil to the buried pottery sherd has been shown to be negligible (Heron et al., 1991). A sampling protocol involving the removal of potentially contaminated surface layers is now an accepted approach, and has been applied to thousands of sherds to date. The approach appears applicable to both freshly excavated sherds and those from museum collections. Experience has shown that sub-sampling between 2 and 3 g of clean pottery sherd affords sufficient lipid to work with, while still providing a reasonable area of the vessel wall to overcome local heterogeneity factors.

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Traditionally lipids have been extracted from visible residues and powdered fabric using organic solvent mixtures (Evershed et al., 1990; Evershed, 2008a). However, a recently introduced protocol involving the direct hydrolysis and methylation of lipids allows both high-throughput and higher recoveries of lipids from archaeological sherds to be achieved (Correa-Ascencio and Evershed, 2014). The loss of compositional information when complex lipids such as triacylglycerols or wax esters are hydrolysed is the only disadvantage of this method – although sherds can often be re-examined using solvent extraction if needed (Correa-Ascencio and Evershed, 2014).

The high sensitivities of instrumental methods such as gas chromatography and mass spectrometry allow μg to ng amounts of compounds to be detected and identified (Fig. 3). Higher sensitivity (pg) can be achieved using selected ion monitoring (SIM) methods e. g. for the detection of specific marine biomarkers (Evershed et al., 2008a; Cramp and Evershed, 2013). The advent of gas chromatography-combustion-isotope ratio mass spectrometry in the 1990s introduced the possibility of accessing stable isotope information from individual biomarker structures, opening a range of new avenues for the application of organic residue analysis in archaeology (Evershed et al., 1994, 1997a). The advent of analytical methods, which allow amorphous and invisible organic materials from archaeological contexts to be detected and identified has thus contributed significantly to answering hitherto intractable archaeological questions, across both temporal and spatial scales. This article provides a review of the current status of the contributions organic residue analysis has made across the themes of *Vessel technology*, *Subsistence and foodways*, *Movement and connectivity*, *Palaeoenvironment and palaeoecology*, and *Chronology*.

2. Vessel technology

The analysis of organic residues from pottery has been highly effective in gaining insights into a range of different aspects relating to ceramics, including production, use, repair and technological change and specialisation. Technologies involved in the production of ceramic vessels, that can be identified using organic residue analysis, include manufacture (including sealing), decoration and repair. For example, unglazed fabrics offer the highest potential for the retention and survival of absorbed residues, yet their surfaces need to be sealed to decrease the permeability of the fabric and make effective containers for liquids. To date, a range of sealants such as waxes, resins and bituminous materials have been identified on archaeological ceramics, dating back to the Neolithic. A known sealant, beeswax, largely undetectable by the naked eye, was identified as a waterproofing agent on Early Neolithic collared flasks (Fig. 3d; Salque et al., 2013) and Bronze Age East Mediterranean Red Lustrous Wheel ware (Knappett et al., 2005). A further commodity used as a sealant was birch bark tar, used to line the interior walls of handled jugs from Makriyalos (Late Neolithic, Greece; Urem-Kotsou et al., 2002) and to seal vessels at the Iron Age site of Grand Aunay, France (Regert et al., 2003). Sometimes sealants are preserved as surface deposits, for example, black/brown sticky deposits found on four 6th century BC vessel sherds from Naukratis (Egypt)

were identified as pitch derived from conifer wood, possibly used to line the interior of the vessels (Stacey et al., 2010). Excavations at Anuradhapura, Sri Lanka, identified a number of buff ware vessels lined with bitumen, known as ‘torpedo jars’. Dated stylistically to between the 3rd and 9th centuries AD, it is thought that the coating was used to seal the permeable containers to allow them to transport liquid commodities, such as oils, perfumes or wine. The bitumen was sourced to Susa in Iran through biomarker distributions and isotopic signatures, suggesting the existence of long distance trade relationships between Sri Lanka and the Middle East (Stern et al. 2008).

An example of applied decoration is found on bitumen-painted ceramics from Late Neolithic Tell Sabi Abyad, northern Syria (Connan et al., 2004). Significantly, the bitumen used to paint the ceramics likely originated from two different source areas in northern Iraq, suggesting long distance trade and exchange networks existed at that time. The adhesive used to repair a Roman Ecton ware jar at West Cotton, Northampton was identified as birch bark tar (Charters et al., 1993) and bitumen, identified using distributions of sterane and terpane biomarkers, was also used to repair bowls at Tall-e Abu Chizan, a late prehistoric (Middle Susiana to Middle Uruk) settlement in southwestern Iran, dating from between 5000 and 3900 BCE (Connan et al., 2008). Bitumen had a long history of use at Kavuşan Höyük (Turkey), from the 14th to the 4th century BC, being mainly utilised as a waterproofing agent on pottery (on both the inner and outer vessel walls) but also as a glue to repair broken Kavuşan ceramics. Thick bituminous crusts found on some jars from the site may also represent the storage and processing of bitumen at the site (Connan et al., 2013). Significantly, it seems that the bitumen originates from the Eruh outcrops, situated 120 km east of the site, and was used at Kavuşan Höyük during the occupation span of the site, implying the existence of a long-established trade route (Connan et al., 2013).

Qualitative and quantitative considerations of lipid residues can demonstrate different methods of food technology associated with particular commodities or vessel types, e. g. boiling or roasting. Significantly, information about the use of vessels as cooking pots can be derived from the presence of ketones (Fig. 3a). These ketones, when present with odd carbon number distributions of $\text{C}_{29:0}$ to $\text{C}_{35:0}$ (with $\text{C}_{31:0}$, $\text{C}_{33:0}$ and $\text{C}_{35:0}$ particularly abundant), originate from the pyrolysis of acyl lipids and ketonic decarboxylation reactions which occur in unglazed ceramic vessels possibly during cooking, when the temperature exceeds $300\text{ }^\circ\text{C}$, and are thought to accumulate gradually with repeated use (Evershed et al., 1995; Raven et al., 1997). In some vessels, these ketones only appear to form in some parts of the vessel, providing clues as to how the pots were being heated. For example, jars from some Romano-British sites bear distinctive patterning in ketone distribution, likely arising from cooking practices (Cramp et al., 2012). The ketones were concentrated around the rims of cooking jars, indicating that the vessels were used for boiling. In fact, the lower part of vessels are cooled down by the evaporation of water through the ceramic porous walls during boiling. The upper part of vessels reach higher temperatures conducive to the formation of ketones.

Relationships between form and function can usefully be examined by organic residue analysis as the fabrication and style of vessels are

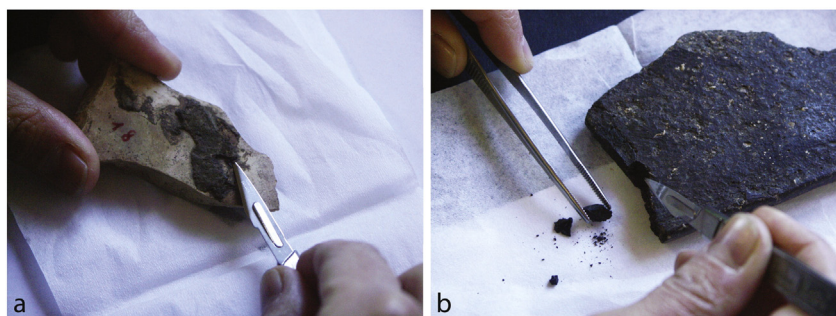


Fig. 1. Sampling of visible residues adhering on sherds and interpreted as a. burnt food crust and b. an adhesive used in pottery repairation.

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