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AMS dating of ancient plant residues from experimental stone tools: a pilot study

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ABSTRACT

Residue analyses on stone artefacts have contributed to resolving functional questions in stone tool research. Although identifying the function of tools through the analysis of their micro-residues is possible, the establishment of a sound numerical chronology for stone tools lacking a clear stratigraphic sequence, such as surface scatters, remains a challenge. While radiocarbon dating of blood residue on stone artefacts has been published previously (Loy 1987, 1990, 1993; Loy et al., 1990; Nelson et al.1986), this paper reports on an experiment designed to assess the possibility of directly dating residues on stone artefacts by accelerator mass spectrometry (AMS) based radiocarbon measurements. Innovative with this approach is (1) the use of mid and late Holocene pre-dated plant material (wood and peat), processed with contemporarily manufactured stone flakes under controlled laboratory conditions and (2) the use of very small carbon masses (less than 22 μg) for radiocarbon dating. The ^{14}C results of the wood residues are in excellent agreement with the original sample, whereas the ^{14}C results of the peat residues yield a wider age variation as expected due to the inhomogeneity of the material, but nevertheless, provided dates within an expected age range. Preliminary results demonstrate the feasibility of dating very small amounts of plant residue on lithics directly when contaminants are confined.

1. Introduction

This study examines the feasibility of dating plant residues on stone artefacts by AMS radiocarbon dating. The majority of stone tools, both in Australia and abroad, are found in open sites and are referred to as 'surface scatters'. For most of these artefacts, it is difficult to achieve a sound chronology in the absence of datable organic material or cultural markers (e.g. characteristic typology). Even in the best scenario, where additional parameters can be identified, only a relative dating with a broader time frame can be deciphered. Recent results of organic residues analyses on stone tools from open sites (Barton, 2009; Cooper and Nugent, 2009; Langejans, 2010) have identified their potential for direct dating of artefacts.

Previous research focussing on dating stone tools residues was limited to dating blood residue (Nelson et al., 1986; Loy, 1987, 1993). The first attempt was conducted on two stone artefacts containing sufficient amounts of blood residue. Results were consistent with their radiocarbon dated stratigraphic position (Nelson et al., 1986). Some analysts experimentally tested the practicability of detecting blood components, such as the proteins haemoglobin, immunoglobulin G and albumin on artefacts and found it difficult, but possible (e.g. Cattaneo et al., 1993; Gurfinkel and Franklin, 1988). Others doubt the plausibility of preservation of protein and haemoglobin after several hundred years' burial, and in particular, the possibility to differentiate between species based on blood residue (Smith and Wilson, 1992). The survival of proteinaceous residue, however, appears to be related to burial conditions. Clay rich soils, along with other conservative conditions such as an alkaline pH and cation exchange capacity have shown to be beneficial for their survival (Jones, 2009; Loy, 1987: 58; Gurfinkel and Franklin, 1988). Further dating of blood residue on stone tools has established the perturbing effect of contaminants on the AMS dates and possibilities for their chemical removal were discussed (Loy, 1987, 1993). An additional limitation for blood residue dating was the actual quantity required: between 50 μg and 1 mg carbon was required to achieve high enough precision (Loy, 1987: 62; Loy, 1993: 46; Vogel et al., 1989: 608).

We are aware of no further attempts to either date residues other than blood or to address the problem of contamination of non-use-related residues on the samples extracted for dating. In this experimental study, we investigate the potential of directly dating plant residues from stone tools.

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Potential sources of contamination resulting from artefact environmental exposure, retrieval, handling and storage were addressed by using experimentally produced stone tools and predated plant materials. In a real–artefact context factors such as fungal activity (e.g. Barton, 2009: 134), the precipitation of calcite on the artefact (or hard water/freshwater reservoir effect) (Fischer and Heinemeier, 2003; Long et al., 1992), insect remains (e.g. Cooper and Nugent, 2009: 217) post-depositional related soil components (Wadley and Lombard, 2007: 1003; Langejans, 2011) all must be addressed. Care must also be taken with sample preparation (e.g. Barton and Matthews, 2006; Fullagar, 2006b: 191). By excluding these factors we have focused on testing the possible influence of introduced modern carbon during sample preparation, the impact on dated results, and consequently, the significance of applying the method on dating residues.

1.1. Identification and preservation of residue on stone artefacts

Although use-related residues on stone artefacts have been reportedly preserved for up to 2 million years in Africa (Loy, 1998; Jones, 2009; Dominguez-Rodrigo et al., 2001; Lombard and Wadley, 2009) and tens of thousands of years abroad (Hardy et al., 1997, 2008; Hardy and Svoboda, 2009; Loy and Hardy, 1992; Pawlik, 2004) in this study we focus on the 40,000 year dating time frame of AMS radiocarbon dating.

In this context, in mainland Australia, residues have been found preserved on stone tools dated between 30,000 and 37,000 years BP (Dodson et al., 1993; Fullagar and David, 1997), on backed artefacts, dating from 1500 to 8500 years BP (e.g. Attenbrow et al., 2009; Robertson et al., 2009) and in Tasmania 6000 years BP (Fullagar and Jones, 2004). In the Pacific region, 28,000-year-old starch residues were found on stone artefacts (Loy et al., 1992).

While not all residues found on stone artefacts are use-related, residue and use–wear analyses aim to identify and interpret ancient residues. Pioneering research on, use–wear and residue analyses was conducted by Semenov (1964), Kamminga (1977, 1979, 1982), Fullagar (1986) and Loy (1987, 1993). Thomas Loy also started integrating interdisciplinary methods and initiated several new directions in residue analyses – with detection of ancient blood residue on stone artefact surfaces and attempted species and DNA identifications (Loy, 1983; Loy and Remington, 1994). However, the reliability of species identification through the analysis of blood residues has been debated (Fiedel, 1996; Smith and Wilson, 1992) and in the case of immunological techniques, such as protein radioimmunoassay on ancient samples, is still subject to misidentification caused by taphonomic and diagenetic alterations of proteins (Potter et al., 2010).

Haslam (2009) reviewed published microscopic residue analyses over the 30-year span from 1976 to 2006. He pointed out the differences in sample sizes examined within assemblages and the lack of transparency indicating the ratio of artefacts with no residue to artefacts with residue. The need to refine sampling protocols for residue analysis was identified. Relevant to this are the recent finds of residue analysed stone artefact assemblages from archaeological contexts which show that more than half of the examined inventories contain residues (Hardy et al., 2008: 652; Hardy and Svoboda, 2009: 165; Robertson, 2009: 300, 302).

Research over the last decade has focused on the occurrence of residues on particular implements, such as backed artefacts (Attenbrow et al., 2009; Fullagar et al., 2009; Hardy et al., 2008; Robertson et al., 2009; Robertson, 2011), bondi points (Robertson, 2011), pieces esquillée (Langejans, 2012) or the detection of hafting locations on lithic points (Lombard and Wadley, 2009; Parr, 2006). Further analyses concentrated on preservation (Barton, 2009; Cooper and Nugent, 2009; Field et al., 2009; Fullagar et al., 2009; Hardy and Svoboda, 2009; Jones, 2009; Langejans, 2010), contamination of residues (Barton, 2009: 134; Cooper and Nugent, 2009: 209, 217; Haslam, 2004, 2006: 1717; Kononenko, 2008: 33; Langejans, 2011; Wadley and Lombard, 2007: 1003) and starch residue research (Barton, 2009; Haslam, 2004; Lentfer, 2009; Torrence and Barton, 2006; Torrence, 2006).

While many earlier experimental studies researched blood residues (e.g. Cattaneo et al., 1993; Gurfinkel and Franklin, 1988; Hyland et al., 1990; Shanks et al., 2004), recent experimental residue analyses have increasingly focused on preservation and contaminant issues (Barnard et al., 2007; Barton, 2009; Haslam, 2004; Jones, 2009; Langejans, 2010, 2011; Loy and Barton, 2006; Wadley and Lombard, 2007).

In the context of residue preservation previous studies have illustrated that use-related residues, such as plant residues, can be very resistant to extraction and even withstand several washes (Barton et al., 1998: 1233; Fullagar, 1986, 1993; Fullagar et al., 1996; Shanks et al., 2004). The observations of some analysts indicate that residues can build a *shield* (Barton, 2009; Loy, 1987: 58; Loy, 1990: 650) once preserved. This protective barrier is considered to be hydrophobic and defiant to microbial attacks (Barton, 2007; Barton and Matthews, 2006; Loy, 1990).

The studies discussed above outlining the recent improvement in residue identification coupled with recent advances in AMS dating which have reduced the carbon size limit down to a 5 μg of carbon (e.g. Smith et al., 2007, 2010; Yang et al., in press) has inspired the objective of the current study to further examine residue dating.

The aim of this study is to test the feasibility of dating residues on stone tools, having excluded post-depositional related contaminants and – for the first time – to systematically investigate the influence of possible introduced contaminants during sample preparation for AMS radiocarbon dating. It was decided to use experimentally produced stone flakes, so avoiding the physical alteration of ancient artefacts by residue extraction and the possibility of extracting any ancient remaining residues on the tools. This also permitted duplication of the experiments (Loy, 1993: 46; Fullagar et al., 1996: 741).

2. Materials and methods

2.1. Materials and procedure

The experimental design had two parts. The aim of the first part of the study was to gain experience with residue extraction procedures and to quantify the amount of residue collected. In the initial trial, stone flakes were produced from flint cores using an antler as a percussion stick and a leather leg/lap protection. Organic residue was applied to stone flakes in the form of fresh plant materials of wood, and fern (Fig. 1). The flakes with the applied residue were dried in a closed clean room for 2 weeks before being stored in re-sealable plastic bags. Residues were then extracted to estimate the amount of organic material that can be retrieved from a single flake. Extraction was conducted in two ways: by physically removing residue with a scalpel whilst monitoring it under microscope, and by using a sonic bath (Fullagar, 2006a: 213) (Fig. 1).

We elected to use a jewellery sonicator (LEO Ultrasonic Cleaner, LEO-50), rather than an industrial sonicator, because the sonication would be gentler and less likely to dislodge parts of the stone itself. Following this preliminary procedure we were prepared for the second part of the experiment, involving the application and extraction of ancient residue under controlled laboratory conditions on stone flakes produced in the laboratory.

Organic material, comprising peat and wood, was chosen from previous environmental studies of the North East Coast of New

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