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Remains of the day-preservation of organic micro-residues on stone tools

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ABSTRACT

Here I report on the decay processes of microscopic organic residues left on stone tool surfaces after their use. Residue analysis on ancient stone tools facilitates reconstruction of past activities. This study enables predictions about the circumstances under which ancient residues preserve. Experimental tool sets with modern residues were buried for a year in separate deposits at Sterkfontein, Sibudu (South Africa) and Zelhem (the Netherlands) whose pH and geomorphology varied, they were then analysed using light microscopy. Biological weathering mainly causes residue decay. In unstable environments rich in microbes and micro-organisms, residues decay quickly. From an archaeological perspective this means that sites that are stable, desiccated, waterlogged, extremely acidic or alkaline and extremely cold or hot sites. Different residue types have different preservation optima and this may lead to a preservation and perhaps interpretation bias. The preliminary predictive models presented in this paper could aid in the considered selection of sites and samples.

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1. Introduction

It has been argued that researchers are unable to describe the mechanism of residue decay (Grace, 1996; Odell, 2001). This paper aims to investigate some of these taphonomic issues. Grace observes that residue analysts "... simply record a phenomenon (...) and then make functional interpretations by analogy without having explained the process that gave rise to the phenomenon." (Grace, 1996, 214). Odell argues that residue analysts "are inclined to consider what they see, not what is missing" (Odell, 2001, 63).

Although these issues are acknowledged and in some cases examined (e.g. Barton, 2009; Barton et al., 1998; Fullagar, 1988, 1998; Haslam, 2004; Jones, 2009; Lombard and Wadley, 2007; Lu, 2006; Rots and Williamson, 2004; Wadley and Lombard, 2007), they remain essentially unresolved. Potentially the results from residue studies may be oversimplified, or worse inaccurate.

Stone tool micro-residue analysis aims to identify microscopic remains or traces that are left on a tool's surface after use. Analysts have identified organic plant and animal remains and inorganic deposits. Residue analysis is generally conducted with a direct or indirect light microscope, using a range of magnifications (between 50 and 800 times), an experimental comparative collection and sometimes an ethno-archaeological comparative collection

(Anderson, 1980; Bruier, 1976; Fullagar et al., 1996, 1999; Loy, 1997; Miller, 1979; Rots and Williamson, 2004; Williamson, 2000a,b).

The identification of processed materials is of relevance to a wide range of research questions (Bruier, 1976; Dominguez-Rodrigo et al., 2001; Fullagar, 2006; Fullagar and Jones, 2004; Hardy et al., 2008; Horrocks et al., 2007; Hurst et al., 2002; Lombard, 2005; Piperno et al., 2004; Staller and Thompson, 2002). Residues themselves may also serve as a source for further investigation, for example DNA analysis (e.g. Hardy et al., 1997; Nelson et al., 1986; Williamson, 2000b) and they can be used to study rare artefacts for additional information (e.g. Barton, 2007; Loy, 1998; Loy and Dixon, 1998).

As yet, it is unclear to which extent residues on tools are the result of use or of taphonomic processes. As residues are expected to be influenced by taphonomy, direct analogies between use and remains are problematic (Grace, 1996; Haslam, 2006; Odell, 2001), especially when quantitative spatial analyses are not conducted on representative samples (e.g. Lombard, 2005, 2007; Wadley and Lombard, 2007). When analysts are able to describe the mechanism of residue preservation and predict under which circumstances (what) residues preserve, residue analysis has great potential.

There have been many investigations into the molecular decay of blood and fat (e.g. Cattaneo et al., 1993, 1991; Eisele et al., 1995; Evershed, 2008; Evershed et al., 2001; Evershed and Tuross, 1996; Gurfinkel and Franklin, 1988; Kooyman et al., 1992; Tuross et al., 1996). The decay and taphonomy of blood and starch has been

studied to some extend with microscopy (Barton, 2009; Barton and Matthews, 2006; Barton et al., 1998; Haslam, 2004; Hortolà, 2002; Lu, 2003, 2006; Therin et al., 1999), but thus far little microscopy study has been conducted on the preservation of other microremains, such as muscle tissue and woody plant tissue.

It has been suggested that biochemistry and biomolecular analysis will furnish insight into the circumstances under which residues preserve (Barton et al., 1998; Fullagar, 1988). Considering the strong visual aspect of residue analysis, the issue of preservation may be best resolved using conventional microscopy. This paper presents the results of such a study. The first part of this study consists of a literature review of decay processes and the circumstances in which decay takes place. I consider several material groups that are of interest to residue specialists. In the second part two experiments that test several variables of residue decay are discussed. Towards the end of the paper I review predictions made earlier and provide a preliminary model that may aid site and sample selection.

2. Setting the stage: preservation of macro- and micro-remains

In many circumstances ancient organic materials do not preserve, but occasionally they do. In these instances the decay process is incomplete; this may be caused by one missing key factor or a combination of circumstances.

Biological weathering is caused by micro-organisms, such as bacteria, fungi and insects that feed on organic remains. Enzymes secreted by these microbes break down (complex) molecules into absorbable molecules (Blanchette, 2000; Deacon, 1997; Warren, 1996). To do so they need oxygen and water (e.g. Tibbett and Carter, 2003; Wilson et al., 2007). In general, fungal decay takes place in moderate environments (pH 6.5–7.5 and 0–25 °C) and is rapid; bacterial decomposition also occurs in extreme situations (for example pH 3) and it is slow. Most organic materials are preserved in environments hostile to fungi. To stop biological decay, oxygen or water must be absent or the environment is extremely acidic, alkaline, hot or cold. Additionally, the presence of heavy metals also interferes with microbial activity and may ensure preservation (Janaway, 1985, 1987).

During deterioration, chemical reactions take place that may change and dissolve components of a material or object. Chemical weathering is hindered when there is no water to transport or dilute by-products such as carbon dioxide, oxygen, ions and humic acids (Chesworth, 1992; Kars, 2003). Without water, hydrolysis is not possible. Chemical weathering is also problematic when there are few free cations in the environment, for example when sediment is depleted of nutrients. If there is no oxygen available, there will be no oxidation and thus less weathering. A neutral pH may impede chemical weathering as many chemical reactions take place in highly alkaline or acidic environments.

Mechanical weathering does not change the chemical composition of material, but objects tear and break (Kars, 2003). In a stable environment with little erosion objects may be protected from mechanical weathering. In general the following environments are advantageous to preservation:

- 1. Oxygen free.
- 2. Water free.
- 3. Extreme pH values, particularly low values hinder biological decay (for example in bogs).
- 4. Moderate pH values impede chemical weathering.
- 5. Low to extremely low temperatures.
- 6. Nutrient or cation depleted.

- 7. Anti-bacterial; can be extremely salty (extreme pH) or in the presence of metals.
- 8. Stable, non-erosive.

Below follows a review of the preservation of a limited material group in freshwater and terrestrial environments. Where possible I refer to studies on residues, but generally the data are drawn from conservation studies on macro-remains.

2.1. Preservation of woody plant tissue

Wood consists of systematically arranged cells with cell walls. The cell walls are composed of varying amount of cellulose, hemicellulose and lignin (Blanchette, 2000). Lignin in plant tissue can be damaged by long exposure to UV light (Smit, 2003a,c). Mechanical weathering leads to tears and breaks in the wood and eventually wood can crumble and turn into dust (Smit, 2003a). Wood is also damaged in an extreme salty and acidic environment (Smit, 2003a). Moist plant tissue combined with iron or copper leads to crevice corrosion (Smit, 2003a).

Fungi and bacteria decay woody plant tissue (Blanchette, 2000; Blanchette et al., 1994; Clausen, 1996; Cronyn, 2001; Greaves, 1969; Smit, 2003a). The different types of decay lead to different physical, chemical and morphological changes in plant tissue. Fungi, causing white rot, brown rot and soft rot, are the most common microbes to degrade wood. These fungi have a pH tolerance between 3 and 7. Bacteria can degrade wood in aquatic environments, in oxygendeprived environments and in arid environments. The bacteria that cause wood decay have pH tolerance between 3 and 10.4 (Blanchette, 2000; Deacon, 1997; Smit, 2003a,c). Different types of deterioration can occur simultaneously. Woody tissue preserves best in extremely dry, oxygen depleted and alkaline environments.

Macro-botanical remains are preserved in poorly developed soils and in soils that underwent leaching of nutrients and minerals, such as ranker soils and podzols. Botanical remains can also preserve in the nutrient rich brown earth soils and luvisols, but to a lesser extent (Goldberg and Macphail, 2006).

2.2. Preservation of starch

Starch grains are the nutrition storage cells in plants and they are energy-dense (Gott et al., 2006). They only have a primary cell wall and are not difficult to break down, as the plant needs easy access to its energy. Studies demonstrate that unprotected starch quickly deteriorates (Adu and Oades, 1978a,b; Barton, 2009; Cheshire et al., 1969; Lu, 2003, 2006; Martin, 1971). However, starch has been found in diverse environments, ranging from extremely dry to wet and in soils and sediments with moderate pH values and extremes; starch is found in (sub- and tropical) sediments and on artefacts; in temperate and warm climates; and even in coprolites (e.g. Lentfer et al., 2002; Tuross and Dillehay, 1995; Ungent et al., 1981, 1986). The following resistant groups are recognised (Lehmann and Robin, 2007; Sajilata et al., 2006; Samuel, 2006): first, physically inaccessible starches, for example in indigested partly milled grains and seeds, in soils with small aggregates, when protected by artefacts (e.g. under potsherds and grindstones) and when protected by resilient plant matter (e.g. resins, desiccated and charred tubers) (Babot and Apella, 2003; Barton and Matthews, 2006; Haslam, 2004; Horrocks, 2006; Parr, 2003; Piperno and Holst, 1998; Piperno et al., 2000, 2004; Ungent et al., 1981, 1986). Secondly, starch in which the grains are packed so tightly together that enzymes cannot access them (e.g. Fullagar et al., 2006). This is similar to the first point, but here starch itself is the barrier. Thirdly, thermally stable retrograded starch formed during cooling of gelatinised starch. The hydrated grains dry into a stronger

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