Archaeological science and object biography: a Roman bronze lamp from Kavastu bog (Estonia)

Ester Oras[1,2,*], Thomas F.G. Higham[3], Lucy J.E. Cramp[4] & Ian D. Bull[5]

[1] *Institute of Chemistry, Ravila 14A, 50411, University of Tartu, Tartu, Estonia*

[2] *Department of Archaeology, Jakobi 2, 51014, University of Tartu, Tartu, Estonia*

[3] *Oxford Radiocarbon Accelerator Unit, Research Laboratory for Archaeology and the History of Art, University of Oxford, South Parks Road, Oxford OX1 3QY, UK*

[4] *Department of Archaeology and Anthropology, University of Bristol, 43 Woodland Road, Bristol BS8 1UU, UK*

[5] *School of Chemistry, University of Bristol, Cantocks's Close, Bristol BS8 1TS, UK*

[*] *Author for correspondence (Email: ester.oras@ut.ee)*

Analytical protocol for fuel residue analysis

Subsamples of the lamp fuel (1–10mg) were crushed using a pestle and mortar. The powdered deposit was ultrasonically extracted with organic solvent (chloroform/methanol, 2:1 *v/v*, 3 × 5 ml). Solvent was then evaporated from the combined extract under a gentle stream of nitrogen to yield a total lipid extract (TLE). An aliquot of the resulting extract was trimethylsilylated prior to analysis (30 µl *N,O*-bis(trimethylsilyl)trifluoroacetamide + 1% trimethylchlorosilane, 60°C, 1 h). The TLE obtained from the lamp deposit was analysed by gas chromatography/mass spectrometry (GC/MS) and GC/combustion/isotope ratio MS (GC/C/IRMS) to identify the compounds present and determine their δ^{13}C values, respectively. GC/MS analyses were conducted using a ThermoQuest TraceMS instrument (ThermoQuest, Hemel Hempstead). The derivatized sample was injected (1.0 µl) onto a column (CPSil-5CB, 50 m × 0.32 mm × 0.12 µm, Agilent J&W), as solutions in ethyl acetate, *via* a programmable temperature vaporising (PTV) injector; ramping from 70°C to 300°C at a rate of 14°C s^{-1} and maintaining this temperature for the duration of the oven programme (see below). Helium was used as the carrier gas. The GC oven temperature was held at 70°C for 2 min, following injection, then programmed to 200°C at a rate of 10°C min^{-1} then to 300°C at a rate of 3°C min^{-1} with a final hold time of 20 min. The ion source was maintained at 200°C and the transfer line at 300°C. The emission current was set to 150 A and the electron energy to 70 eV. The analyser was set to scan *m/z* 50–650 with a duty cycle time of 0.6 s. GC/MS peak assignments were made by comparison with known mass spectra. A ThermoFinnigan

Delta^{Plus} XP instrument (Thermo Electron Corporation) was used to determine the $\delta^{13}C$ values of the compounds identified in the lamp deposit TLE. The MS (EI, 100 eV, three Faraday cup collectors *m*/*z* 28, 29 and 30) was interfaced to a Thermo Electron Trace 2000 GC *via* a ThermoElectron GC combustion III interface (CuO/NiO/Pt oxidation reactor maintained at 940 °C and Cu reduction reactor maintained at 600 °C). Derivatised TLE was introduced using a PTV injector as above. Helium was used as the carrier gas whilst column and temperature programmes were the same as those used for the GC/MS analyses.

Discussion of dating discrepancies

All the Kavastu lamp fuel samples were dated at the Oxford Radiocarbon Accelerator Unit (ORAU). Samples were taken at the museum using a clean scalpel and stored in aluminium foil to avoid contamination from plasticisers from packaging. The bulk samples went through the standard sample preparation protocol (Brock *et al*. 2010). However, the total lipid extract (TLE) sample was obtained after solvent extraction at the University of Bristol School of Chemistry laboratory as part of the lipid residue analysis and was essentially combusted and dated at ORAU with no additional preparation.

The AMS date of the TLE gave a result of 2319±31 BP, cal. 430–356 BC (88.4% probability). This predates the earliest occurrence of this type of lamp according to artefact chronology by several centuries. It is also almost a millennium earlier than the two bulk fuel dates and it seems unrealistic to conclude that a passage of ~800–1000 years had passed between the two purported events being dated.

Contamination is sometimes a serious issue in the field of radiocarbon dating and it may be significant in explaining our TLE dating result. Contaminants affect samples in quite different ways, and much depends on the ‘true’ age of the sample and the age of the contaminant. They can be both old and young, and come from the site or from excavation, conservation, storage and from the analysis of the sample in the laboratory (Gillespie & Hedges 1984; Lanting & van der Plicht 1998; Stott *et al*. 2001; Fischer & Heinemeier 2003; van der Plicht *et al.* 2014; Yates *et al*. 2014, 2015a & b). It is noteworthy that in the case of microsamples (our TLE weight was 1.31mg) the issues of contamination can be very critical, affecting the measurements significantly, and lipids in general tend to be more susceptible to contamination (Yates *et al*. 2015a).

For relatively recent material such as the Kavastu lamp fuel, old carbon contaminants are much more significant in causing aberrant determinations compared with modern carbon contaminants. A 10% contribution of old carbon will make an AMS sample ~800 years too old, regardless of its age. For near modern samples, on the other hand, a much greater proportion of modern carbon is required to shift the age significantly. Thus a more likely explanation for the discrepancy in our bulk and TLE dating results is the introduction of old carbon to the TLE sample.

Exactly how this has happened is difficult to pin down. In the lipid residue analysis we only see the compounds that are amenable to GC fitting into the analytical window previously defined. However, the TLE also contains compounds that have a higher molecular weight and/or are much more polar, and indeed, may be older. The AMS date for a TLE is therefore not truly comparable to a few fatty acids, but probably also includes substances unseen and unknown to us. The latter can easily contain old carbon resources affecting the dating results of the TLE. For a true comparison one would need to date the fatty acid fraction or specific fatty acids which we cannot, as yet, separate for the AMS dating. In addition, unremoved solvent from the TLE might also play a role in producing ages that are older than they should be.

Although the AMS results of the two bulk samples do not overlap entirely, they are very close, falling into the first half of the first millennium AD and as such do not contradict the typo-chronological context of the lamp. On the contrary, the dating of the TLE precedes the occurrence of this lamp type by several centuries. Therefore we consider the TLE age erroneous for dating the usage of lamp for illumination purposes and favour the two bulk determinations—1699±33 BP and 1561±25 BP—as more likely accurate providing a *terminus post quem* for the deposition of the object.

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