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Analysis of adhering organic residues of two ''coupes-à-socles'' from the Neolithic funerary site ''La Hougue Bie'' in Jersey: evidences of birch bark tar utilisation

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Abstract

The ''coupes-à-socle'' (footed cups) are ceramics which can be found in domestic Neolithic contexts in Brittany (Castellic, Auzay-Sandun), but more generally in non-domestic areas. Their ceremonial function has often been evoked, in particular as a perfume burner. The analysis by GC and GC/MS of two samples from the ''Hougue Bie'' (Jersey) passage graves showed the presence of vegetable fatty acids, degraded *n*-alkanes and terpenoïds, biomarkers of birch bark tar. The presence of such substances could be explained either by the use of the ''coupes-à-socle'' for burning birch bark tar as an odoriferous product or their use as an everyday vessel for heating and increasing tar plasticity. However, the limited occurrence of this type of ceramic in habitat sites supports the former utilisation. This first report of the presence of birch bark tar in ''coupes-à-socle'' thus suggests use of this tar as an odoriferous product, in addition to its numerous, previously recognised, more technical uses.

Keywords: Neolithic; coupe-à-socle; Organic residues; Birch bark tar; GC/MS

1. Introduction

The objects designed as ''footed cups'' (coupe-à-socle) are, in France, an emblematic form of the ceramic productions at the beginning of the 4th millenary BC, in particular within the Chassean culture, in the broad meaning of the term, which covers a large area of the national territory. Their function was inadequately known when their existence was first acknowledged in the 1870s (''vase support''). A more precise and ceremonial designation (''perfume burner'') (Baudon, 1911; de Mortillet, 1901) was proposed in the 1900s. However, the first term was still in use until the 1980s, even though the more neutral term ''coupe-à-socle'' had been recommended at the end of the 1960s (Thevenot, 1969).

The successive excavations in the Morbihan department of the monuments of the ''Table des Marchands'' at Locmariaquer (1986–1994) (Cassen and L'Helgouac'h, 1992) and of ''Lannec er Gadouer'' at Erdeven (1993–1997) (Boujot and Cassen, 2000) displayed this ceramic form and its decorative system in the chronological table. The ''coupes-à-socle'' were abundant in the paleosol of the first site, but absent in the paleosol of the second one. These excavations thus validated a theoretical periodisation stipulating two chronological stages in the Castellic entity, the most recent of which contains the ceramic form in question (Boujot and Cassen, 1992).

These two excavations also suggest a similar purpose for these potteries: both in funerary contexts (passage grave) and in extraordinary sites (stele alignments and enclosures Le Rouzic, 1930), they were found in great number and

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excessively fragmented, especially in non-domestic places unlike the remainder of the ceramic evidence.

The illustrations engraved on steles and the reinterpretation of the main signs engraved on these Armorican standing stones may suggest more acceptable functions for these potteries (Cassen, 2000). In particular, the identification of a sperm–whale on some steles suggested that the ambergris of this animal could be used in procedures of scented fumigation carried out in the hollow of the cups of these ceramic forms. Indeed, their openwork barrels (feet) suggest they could be used as uncovered stoves.

Ambergris, when relatively fresh, was described as ''bitumen'' when it was still running aground on the Vendean coasts in the 18th century (Baudouin, 1931). This same term was used by diggers at the time of the discovery of some organic materials in the Neolithic tombs of the Carnac area. In the 19th century, in tumulus of La Vigie excavations (Davy de Cussé et al., 1867), the collected product could be match-ignited to release a pleasant odour, but was not identified chemically. All these observations led us to examine the possible use of ambergris in the ''coupes-à-socle''. Ambergris contains up to 46% of cholestanol type steroids and 25–45% of ambreine triterpene (Sell, 1990). So the presence of these molecules and their by-products are biomarkers of ambergris utilisation. In consequence, the analysis of chemical components of organic residues in two ''coupes-à-socle'' was performed for examining the presence of ambergris, or other components, in this type of potteries.

The first study undertaken in 1999/2000 on tens of cups preserved in the museums of the west of France (Bretagne, Pays de Loire and Poitou-Charentes) proved to be unfruitful in terms of residues visible to the naked eye (cleanings and restorations strongly deteriorate these residues). Finally, only the specimens previously described as presenting carbonized residues on the bottom of the saucers (in the megalithic passage grave of la Hougue Bie in Jersey) (Toulmin Nicolle, 1926) were the most reliable for successful extraction and analysis.

Several specimens were collected during the early excavation, five were published with drawings (Hawkes, 1938) but black traces were only observed on two individuals (LHB 57 and LHB 7305) (Fig. 1).

2. Materials and methods

2.1. Extraction and separation

Surface residues from two coupes-à-socle (LHB 57 and LHB 7305) out of the four specimens of the tomb were collected by scrapping with a scalpel blade. Three distinct area (about 1 cm^2) that appeared darker (as carbonized) were thus scrapped under a binocular magnifying glass. The dust was stored into plastic vials. Laboratory (and plastic vial) blanks were regularly performed in order to test for the absence of recent contamination of the analysis extracts.

The samples (51.2 mg for LHB 57 and 25.6 mg for LHB 7305) were extracted twice with 30 mL of chloroform/methanol solution (2:1) by ultrasonication for 90 min at 40 °C. The obtained solution (38% and 43%, dry weight of extract, of total sample weight) was then separated according to the method of McCarthy and Duthie (1962) by column chromatography on potassium hydroxide impregnated silica gel (10 g). The neutral compounds were eluted first with ethyl ether (100 mL). The acidic compounds, eluted with 2% formic acid in ether (100 mL) were derivatized into their methyl ester by treatment with acetyl chloride in methanol solution for 20 min at 80 °C. The neutral fraction was further separated according to functional group by column chromatography (5 g of silica) in four subfractions by elution with 50 mL of heptane/ether mixtures of increasing polarity (1:0, 3:1, 1:1, 0:1). Each subfraction contains, respectively, hydrocarbons, ketones, alcohols and no significant amounts of compounds were observed in the last subfraction. The separation of the neutral fraction allowed better identification of minor components.

The four obtained subfractions and the acidic fraction were then analyzed by GC (gas chromatography) and gas

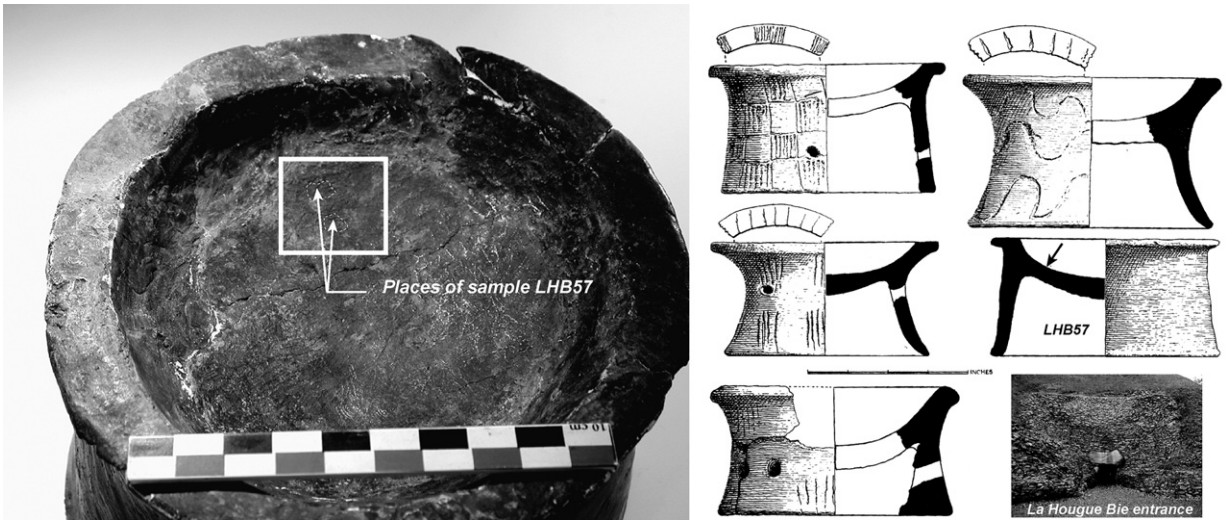

Fig. 1. ''Coupes-à-socles'' and samples of la Hougue Bie.