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Gone to seed? Early pottery and plant processing in Holocene north Africa

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Abstract

Plant foods play an important role in the human diet and the ability to grow, store and extract nutritive potential from plants has had a transformative role in human history. During the Holocene, the invention of thermally resistant ceramic vessels, regarded as a crucial step in human technological progress, provided new opportunities to boil plants such as wild grasses, fully unlocking the potential of such plants as foodstuffs. This allowed a broadening of subsistence bases, increased dietary diversity, a greater variety of nutrients and more stable and palatable foods. Pottery was invented early in north Africa, at *c.* 12000 cal BP, where it was first made by semi-sedentary hunter-gatherers, raising questions as to what this early pottery was used for. Combined molecular and isotopic techniques revealed the presence of diagnostic plant lipids, including leaf waxes and seed oils, in pottery from Holocene sites in the Libyan Sahara and Mediterranean north Africa, suggesting the processing of grasses, seeds and aquatic plants. In combination with archaeobotanical evidence from sites across these regions, these data provide insights into the wide range of plants exploited in Holocene north Africa, providing information on dietary and subsistence practices of human groups across north Africa and confirming the importance of plant processing in the earliest pottery vessels in both regions.

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1. The invention of pottery

Discoveries made over the last decades suggest that pottery was invented independently in many different regions of the world. Although pottery is often associated with early farming communities, the first ceramics, invented by hunter-gatherers, appear much earlier, in two very geographically different regions. Of these, the oldest known pottery to date originates from the Xianrendong Cave, Jiangxi, China, at around 20,000 to 19,000 years cal BP (Wu *et al.* 2012), produced by hunter-gatherers. Here, many sherds bear signs of burning on their exterior surface, possibly indicating their use in cooking, and the large number of fragmented animal bones at the site suggest the pottery was used in the extraction of marrow and grease (Prendergast *et al.* 2009). Further evidence for early pottery in South China emerged at the Yuchanyan Cave, Hunan Province, dated to between 18,300 to 15,430 cal BP, and at the sites of Diaotonghuan in Jiangxi Province and Miaoyan in Guangxi Province, with dates ranging between *c.* 16,000 - 10,000 cal BP (Zhao 1998; Kuzmin 2006). However, inferred dates seem to vary widely, depending on the period of investigation, research focus, and techniques adopted for geochronological reconstruction, and are somewhat contested. Pottery also appeared in the Russian Far East and Japan between *c.* 17,300-15,000 cal BP where, in Japan, it is associated with the extensive exploitation of marine and freshwater resources, by hunter-gatherers, in the Incipient Jomon period (Kuzmin 2006; Craig *et al.* 2013; Shoda *et al.* 2020).

In the Levant, pottery emerged during the PPNB (around 9200 - 9000 cal BP), at sites such as Tell el Kherkh, Gritille, Assouad and Tepe Guran (Le Mièrre and Picon 1998; Mazurié de Keroualin 2003) and, by 8000 cal BP, pottery is found throughout the Fertile Crescent and is beginning to spread across Europe (Jordan *et al.* 2016). Lipid residue analyses shows, across these early agricultural societies in the Near East and Europe, the importance of early pottery in processing (or storing) dairy products, such as butter, cheese and yoghurt, and carcass products from both ruminant and non-ruminant domesticates, cattle, sheep, goat and pig (e.g. Evershed *et al.* 2008).

This pottery may have been influenced by the early pottery traditions of North Africa (Gronenborn 2009) although there has been little investigation of this. Certainly, in north Africa, pottery production began earlier, first being made by hunter-gatherers during the period 11,000 to 10,000 cal BP (Huysecom *et al.* 2009). The earliest ceramics were found within a 4000 km-long band encompassing the southern Sahara and northern Sahel, with early dates clustering in the Middle Nile valley and the Western Desert of Egypt, in the vicinity of the

mountainous regions of southern Algeria and northern Niger (Jordan *et al.* 2016), although recent research now pinpoints the earliest pottery in Africa originating from south of the Sahara, at the site of Ounjougou (Fig. 1), Mali (Close 1995; Nelson *et al.* 2002; Jesse 2003; Huysecom *et al.* 2009). Here, stratigraphic and chronological evidence provided a *terminus ante quem* of 9400 cal BC (c. 9785 BP) for an archaeological assemblage containing ceramics and lithics, although it should be noted that only a small number of sherds were found in the early levels. Furthermore, Jesse (2003) demonstrated, through a survey of radiocarbon dates of sites with pottery, that early pottery tends to cluster in the southern part of the Sahara. In summary, recent statistical modelling of radiocarbon dates confirms that ceramic vessel technology had independent origins in two different hunter-gatherer societies, in East Asia at c. 16,000 cal BP and in north Africa at c. 12,000 cal BP (Jordan *et al.* 2016).

Here, possible reasons for the early invention of pottery in North Africa are examined, using organic residue analysis in two different contexts, that of the early to mid-Holocene hunter-gatherer-fishers and mobile pastoralists of the Saharan desert and mid-Holocene foragers in Mediterranean north Africa. The importance of plant foods in the human diet is discussed and the value of the new technology of thermally resistant pottery in both facilitating innovative methods of cooking (e.g. boiling of wild grasses) and rendering, sometimes toxic, plants edible for consumption, is highlighted. Lipid and isotopic analysis, showing the exploitation of plants in pottery from both regions, is discussed in conjunction with the evidence for plant use, both at these sites and across the broader region, providing a holistic picture of plant processing in the earliest pottery vessels in North Africa.

2. Context of research

2.1 Why was pottery invented in North Africa?

In contrast to the Near East, where cereal agriculture preceded the use of ceramics by around 2000 years, in Saharan Africa, pottery was in use by semi-sedentary hunter-gatherers at least 2000 years before domesticated cattle appeared and several thousand years before the domestication of plants (Marshall and Hildebrand 2002; Garcea 2004, 2006; Neumann 2005; Marshall and Weissbrod 2011). This raises the question of why ceramics were invented in the region, with several authors suggesting theories to explain this. Close (1995) argues that pottery first appeared in the region between the Nile and the Hoggar Mountains, Algeria, in societies with a heavy reliance on aquatic resources and some exploitation of wild grasses. She suggests these earliest ceramics in north Africa were first prestige items which transformed into

everyday utilitarian vessels. Sutton (1974; 1977) posits that the invention of pottery occurred to facilitate the processing of the newly available aquatic resources, and suggested that pots were used to cook soup, porridge and fish stew. In contrast, Haaland (2007) proposes an origin in the Nile Valley, with pottery being developed to process grains to make porridge and beer. Finally, Huysecom *et al.*, (2009) suggest that ceramics were invented in the present-day Sahel-Sudanese belt as a technological advance emerging in response to the spread of new grasslands in the early Holocene, allowing pottery containers to be used to boil ground *Panicoidae* grasses. The common theme of these hypotheses regarding the early invention of pottery focuses on plant processing, specifically wild grasses, found widely across Holocene north Africa. Thus, it is useful to examine the importance of plants in the diet of Late Pleistocene and early Holocene groups in north Africa and what nutritional benefits might accrue from processing plants in the, newly invented, thermally resistant ceramic vessels.

2.2 The importance of plant foods in the human diet

The importance of plants in the evolution of human diet has long been known, and, today, plant foods often dominate the diets of many hunter-gatherer, agricultural and pastoral societies across the globe. These comprise a rich and diverse range of resources, including seeds, tubers, fruits, nuts, leaves, stems, and flowers. Plants are a valuable source of carbohydrates, polyunsaturated fatty acids, vitamin C, minerals and non-starch polysaccharides (dietary fibre). There are many biologically active components in plant foods (known as phytochemicals), thought to be both protective against a range of diseases and helpful in maintaining health and longevity, including plant sterols/stanols, phenolic compounds (e.g. lignans), glucosinolates and a range of minerals (Johnson 2013; Butterworth *et al.* 2016).

The human use of plants, for food, medicinal and veterinary purposes, animal fodder, clothing, fuel, shelter, containers, as dyes and for their intoxicating properties, requires an advanced understanding of both the botanical properties and local ecology of specific plants, described by Jones (2009) as ‘ecological intelligence’. Consequently, the ability to grow, harvest, store and extract nutritive potential from plants has had a transformative role in human history. Clearly, sophisticated culinary skills and knowledge which facilitate the successful transformation (processing) of raw foodstuffs, specifically plants, into edible meals, are also required. ‘Processing’ describes activities that change the physical and/or chemical properties of plant (or animal) foods in order to improve their suitability for eating and/or digestion (Johns and Kubo 1988; Wrangham *et al.* 1999; Wollstonecroft 2011). The enormous variation in

functional properties of plants and plant parts means that ‘multiple step processing’ (Jones 2009) is sometimes required to produce edible and safe plant foods. For example, the major means of detoxification are heat, solution (soaking or leaching), fermentation, adsorption (i.e. with clay), drying, comminution (grating, grinding, pounding etc), and chemical reaction due to pH change. In many cases, specific processes are used for particular plants, and sequences of these techniques are required, for example, grinding followed by washing and then cooking (Johns and Kubo 1988; Stahl 1989; Wollstonecroft 2011). Such technical choices increase the bioaccessibility and thus bioavailability of energy and nutrients in plant species (Wollstonecroft *et al.* 2008; Wollstonecroft 2011).

Across hominin history, the processing of plants using the techniques mentioned above undoubtedly provided humans with increased dietary diversity, a greater variety of nutrients and more stable and palatable foods, ultimately allowing them to move into new environments (Stahl 1989; Stahl *et al.* 2002; Carmody and Wrangham 2009; Wollstonecroft 2011; Butterworth *et al.* 2016). Technologies such as grinding stones, cooking pits, earth ovens, sieves and leaching devices were important in these processes, but it is the invention of thermally resistant ceramic vessels, long regarded as a crucial step in human technological progress, which I would argue fully unlocks the potential of some plants as foodstuffs.

The invention of pottery would have expanded the range of potential food sources within a habitat (including plants) available to human groups (Garcea 2004) and thus may well be related to the significant environmental and climatic change that accompanied the global transition from the Last Glacial Maximum (LGM) peak approximately 21 kya to the current Holocene interglacial period (Rehfeld *et al.* 2018). During this time, the Late Pleistocene was characterised by a broadening of the subsistence base of hunter–gatherers in the Near East and a similar ‘Broad Spectrum Revolution’ appearing among post-Pleistocene hunter-gatherers in many parts of the world, including Europe (Payne 1975; Morales *et al.* 1998). This was likely driven by the expansion in dietary plant diversity, clearly demonstrated by the increased complexity of plant palaeobotanical assemblages recovered from Pleistocene and Early Holocene hunter-gatherer sites across the world (Zeder 2012), which would have led, in turn, to a need for increased processing of plant foods. Plant assemblages recovered from early Near Eastern Epipalaeolithic sites suggest that, by this time, some groups had replaced generalised foraging strategies with intensive plant collecting practices, based on a wide range of resources (van Zeist and Bakker-Heeres 1984; Hillman *et al.* 1989b; Kislev *et al.* 1992; Hillman 2000; Colledge 2001). These included more than 250 wild plant taxa, identified from parenchymous

tissue, phytoliths, seeds, starch grains, charcoal and pollen. Following these changes in resource procurement, the relatively favourable climatic conditions of the Holocene, which commenced at 11.7 kya (Carlson 2013), are argued to have paved the way for the wholesale domestication of plants and animals and thus the beginnings of food production.

2.3 Why would plants be processed in pots?

Many plant species have either physical (e.g. thorns) and/or chemical (e.g. toxins) defences aimed at deterring possible predators. Furthermore, the tissues of many plants, such as pulses, nut kernels and underground storage organs (USOs) contain cell walls with physical and chemical properties that encourage nutrient encapsulation, hindering energy and nutrient release during chewing and digestion (Butterworth *et al.* 2016). This slows down or prevents interactions of critical carbohydrates, lipids and other micro- and macro-nutrients with digestive substances, meaning that plant foods would be excreted without human benefit of their nutrients. For example, the amount of lipid released from raw almonds (*Amygdalus communis* L.) during digestion is limited, due to cell wall encapsulation, even if chewed for a prolonged period (Ellis *et al.* 2004). Almonds are significantly more digestible in the finely ground form than at the raw or blanched stage (Mandalari *et al.* 2008). A similar study showed that cooking (thermal processing) increased the fraction of lipid released from peanuts (*Acachis hypogaea*), a lipid-rich oilseed, and increased the net energy gain, whereas blending (non-thermal processing) had no detectable energetic benefits (Groopman *et al.* 2015). Acorns, which have been a staple food for much of human history, have a characteristic bitter taste (because of their high tannin content) and can also be toxic for humans (Deshpande *et al.* 1984), suggesting they required processing, i.e. pounding, leaching, roasting or boiling, prior to consumption (Mason and Nesbitt 2009; Ayerdi *et al.* 2016; Morales 2018).

Plant processing methods show marked similarities worldwide and are classifiable according to the way in which they function to eliminate toxins (Johns and Kubo 1988). Plant foods can undergo short-term cooking over direct flames and on/in hot coal but the most common techniques used in plant detoxification worldwide are boiling and some form of roasting or baking. However, as Johns and Kubo (1988) note, roasting would have been the most common method throughout most of human history as boiling requires watertight and heat-resistant containers. Heated rocks could be added to watertight containers, such as baskets, to boil plants, but they could not be used for the prolonged boiling over direct heat often required for detoxification. Cooking stone technology (heated rocks), in earth ovens, dates back at least

30,000 years in Europe (Movius 1966; Straus 2006; Black and Thorns 2014) and stone boiling was identified at El Mirón Cave, northern Spain, at c. 15,500 B.P. (Nakazawa *et al.* 2009), confirming the antiquity of this technique. This suggests some early knowledge of the advantages of boiling foodstuffs, likely plants and meat products.

Thermally resistant ceramic vessels would also have facilitated the prolonged boiling of grains and seeds from legumes and monocot stems (Arnold 1985; Jones 2009) that previously needed long periods of soaking or cooking, although it should be noted that this could lead to a loss of nutrients (Stahl 1989). Furthermore, the cooking of high energy-yielding plant foods, such as starchy tubers (rich in carbohydrates) also leads to an increased speed of digestibility and consequent energy gain (Carmody and Wrangham 2009).

Thus, the investigation of prehistoric plant exploitation is of critical importance in unlocking diet and subsistence strategies of human groups, and, in broader terms, understanding their interaction and relationship with their environment. However, the ‘invisibility’ of plant processing, or what Hillman *et al.* (1989a; 1989b) call ‘missing foods’ has long been a feature of prehistoric archaeology, particularly in Africa. This is sometimes attributed to poor preservation of putative botanical remains and, until recently, the lack of systematic sampling of archaeobotanical remains at sites. This raises the question as to whether wild plant processing can be identified, using organic residue analysis, in the ‘new’ technology of thermally resistant ceramic containers, present from the early Holocene.

2.4 Organic residue analysis of archaeological pottery

Organic residue analysis has long contributed to a wide range of archaeological questions but is most notably known for its contribution to elucidating diet and animal management strategies worldwide (e.g. Copley *et al.* 2003; Craig *et al.* 2003; Copley *et al.* 2004b; Copley *et al.* 2005a; Copley *et al.* 2005b; Copley *et al.* 2005c; Evershed *et al.* 2008; Outram *et al.* 2009; Dunne *et al.* 2012; Cramp *et al.* 2014a; Cramp *et al.* 2014b; Heron *et al.* 2015; Smyth and Evershed 2015; Debono Spiteri *et al.* 2016; Dunne *et al.* 2017; Ethier *et al.* 2017; Mileto *et al.* 2017; Whelton *et al.* 2018; Dunne *et al.* 2019a). Employing these chemical analyses in the reconstruction of vessel use at sites worldwide has enabled the identification and separation of terrestrial animal fats (Evershed *et al.* 1997a; Mottram *et al.* 1999; Evershed *et al.* 2002), marine animal fats (Copley *et al.* 2004a; Craig *et al.* 2007; Cramp and Evershed 2014; Cramp *et al.* 2014b), plant waxes (Evershed *et al.* 1991; Dunne *et al.* 2016), beeswax (Evershed *et al.*

1997b; Roffet-Salque *et al.* 2015) and birch bark tar (Charters *et al.* 1993; Urem-Kotsou *et al.* 2002; Stacey *et al.* 2020).

However, similarly to the invisibility of archaeobotanical remains, and, despite the now commonplace identification of animal product processing in both prehistoric and historic period ceramics, the detection of lipid biomarkers denoting plant processing in ceramic vessels is rare. The obvious reason for this is that when meat and plants are processed in the same vessel, then fat-rich meats will ‘swamp’ the plant signal, as attendant animal fats contain several orders of magnitude higher lipid concentrations than the lipid abundance of plant-based foods (Charters *et al.* 1997; Evershed 2008).

2.5 The chemical and isotopic identification of plant biomarkers

There are a number of compounds that can be used to identify plant processing in archaeological ceramics. Likely the most common plant biomarkers are epicuticular (plant leaf) waxes, primarily found on the surface of plant leaves, sheaths, stems and fruits, although waxes are also found associated with other plant organs, i.e. seed oils and coats, flowers, bark and husks (Bianchi 1995). Plant waxes mainly comprise a mixture of long-chain aliphatic compounds (between 20 and 40 carbon atoms), derived from very long-chain fatty acids, and also include triterpenoids and sterols (Kunst and Samuels 2003). The major component classes are primary and secondary alcohols, ketones, fatty acids and aldehydes.

Fatty acid compositions of plant leaves are highly consistent among species. Even numbered long-chain *n*-alkanoic acids, with chain lengths ranging from C₁₆ to C₃₆ are common in the epicuticular waxes of plants (Tulloch 1976), however, the most common (normally leaf tissue) fatty acids are C_{16:0}, C_{18:2}^{Δ^{9,12}} and C_{18:3}^{Δ^{9,12,15}}, normally constituting >90% of total leaf fatty acids (Padley *et al.* 1986; Somerville *et al.* 2000), although these unsaturated components are unlikely to survive over archaeological timescales due to oxidation. Alkanes are also common components of waxes, usually occurring in low concentrations (Koch and Ensikat 2008), although occasionally they are the dominant lipid, e.g. the leaf wax of *Cotyledon orbicularis* is almost entirely comprised of alkanes (Eglinton and Hamilton 1967). Long-chain *n*-alkanes distributions occur in the range C₂₅ to C₃₅ (Chibnall *et al.* 1934), with an odd-over-even predominance (Eglinton and Hamilton 1967). The dominant chain lengths vary across plant taxonomic groups but the C₂₇, C₂₉, C₃₁ and C₃₃ homologues usually predominate (Diefendorf *et al.* 2011).

2.5.1 Diagnostic criteria and proxies

A range of diagnostic criteria and proxies, commonly used both to investigate modern plants and geological samples, such as oil deposits and ancient sediments, can be applied to investigate the origin of plant biomarkers in ancient vessels. These include the calculation of average chain lengths (ACL), based on the premise that different plants biosynthesis varying proportions of *n*-alkane homologues in their leaf waxes, allowing them to be used to provide chemotaxonomic information through linking the carbon chain-length distributions to plant species or genera (Maffei 1996) and photosynthetic pathways (Rommerskirchen *et al.* 2006). The carbon preference index (CPI) is used as a gauge of the dominance of odd-carbon numbered *n*-alkanes, where it is known that confirmed CPI values for all plant species have strong odd-chain preferences, with CPI values varying between 1.6 and 82.1 (Diefendorf *et al.* 2011). Mid-chain length *n*-alkanes (C₂₃ and C₂₅) characterise submerged and floating macrophytes (aquatic plants) and can thus be used in the P_{aq} (emergent and non-emergent aquatic macrophyte input) proxy ratio (Ficken *et al.* 2000) where $P_{aq} < 0.1$ corresponds to a terrestrial plant, P_{aq} 0.1-0.4 to emergent macrophytes and P_{aq} 0.4-1.0 to submerged or floating macrophytes.

2.5.2 Stable carbon isotope ($\delta^{13}C$) values of plants

The differences in the physiology and biochemistry, i.e., the modes of fixation of CO₂ in C₃ and C₄ plants, results in distinct stable carbon isotope signatures, with isotopic fractionation (kinetic isotope effect) between ¹²C and ¹³C making C₃ plants more depleted in ¹³C relative to C₄ plants (Smith and Epstein 1971; Cerling and Harris 1999; Tipple and Pagani 2007). The carbon isotope composition of plant tissues has been extensively investigated with current bulk $\delta^{13}C$ values for C₃ plants known to range from -32 to -20 ‰ and from -17 to -9 ‰ for C₄ plants (values taken from Boutton 1991 and references therein). However, these can vary considerably, with individual species displaying spatial and temporal variation, for example, seasonally. There will also be differences between species and forms in any given geographical location and between species in different regions (see Heaton 1999 for full review). Furthermore, different constituents within the same plant, such as the lignin, carbohydrates, proteins and lipids, all have different carbon isotope compositions, as do diverse parts of the plant such as the leaves, seeds and tubers (Collister *et al.* 1994; Dungait *et al.* 2008). It should also be noted that, during biosynthesis, leaf wax lipids become more depleted in ¹³C than the biomass, with $\delta^{13}C$ values ranging between -39 and -29 ‰ in C₃ plants and -26 and -14 ‰ in C₄ plants (Bi *et al.* 2005 and references therein). Measurement of the stable carbon isotope

values of plant-derived compounds, i.e. long-chain fatty acids or *n*-alkanes, can thus provide valuable information on the photosynthetic pathways (C₃ or C₄) of plants processed in archaeological vessels.

2.6 North African Holocene archaeological sites

2.6.1 Takarkori and Uan Afuda rockshelter, Libyan Sahara

Takarkori and Uan Afuda caves (Fig. 1) are located in the Libyan Sahara, in the region of the Acacus massif and Messak relict plateau, an area licensed to the Italian-Libyan Archaeological Mission, under the control of ‘*La Sapienza*’, the University of Rome. Takarkori is a large rock shelter, some 80 m long and around 10 to 15 m deep, set on a small, defined terrace, sheltered by a 30 m high cliff. It is located 100 m above the southern side of a large wadi joining the Tanezzuft valley to the eastern fringes of the Acacus Mountains (Biagetti *et al.* 2004; Biagetti and di Lernia 2007, 2013). The Uan Afuda cave site is located in a tributary of the Wadi Teshuinat, the Wadi Kessan, in the central Acacus Mountains. The cave is one of the largest in the Acacus, being 40 m wide and 20 m high (Cremaschi and di Lernia 1999).

Takarkori is noteworthy for the remarkable preservation of its archaeological deposit (about 2 metres depth, *c.* 200 m²), with a stratigraphic sequence extending from *c.* 10,200 to 4650 cal BP, with Late Acacus (hunter-gatherer) and Early, Middle and Late Pastoral occupation levels, demonstrating nearly 5000 years of human occupation (di Lernia 1999c; Biagetti *et al.* 2004; Biagetti and di Lernia 2007, 2009, 2013). Although faunal evidence is fragmented and poorly preserved at Takarkori, thus precluding herd reconstructions, the ¹⁴C dating of sheep/goat and cattle bones unambiguously indicates the presence of early herders in this and other sites of the Acacus Mountains (Biagetti and di Lernia, 2013). The material culture at Takarkori displayed a remarkable diversity and richness of archaeological finds, in particular, plant remains (Fig. 2) and other organic, usually perishable eco- and artefacts (di Lernia *et al.* 2012). The assemblage consists of lithic tools, grinding equipment, mortars and querns with ochre, potsherds, wooden and bone tools and ostrich eggshell beads. Also present were organic remains, including human and faunal remains, plant remnants, coprolites and dung, basketry and cordage and a leather artefact (di Lernia *et al.* 2012; Biagetti and di Lernia 2013).

Excavations at Uan Afuda yielded evidence of Early Acacus (Epipalaeolithic) hunters, utilising microlithic tool kits, with a subsistence economy based on hunting Barbary sheep. Late Acacus levels comprised hearths, stone structures and specialised areas. Assemblages, radiocarbon dated covering a period *ca.* 8900 - 8500 cal BP, include lithics, over 500 fragments

of grinding stones and rocker decorated pottery and basketry. The hunter gatherer groups occupying the site were living a mainly sedentary lifestyle, inhabiting the site for long periods during the year, but with a broader subsistence base, exploiting the mountainous areas more fully, hunting Barbary sheep but also other small and medium sized mammals, fish and birds (di Lernia 1999a; b; d; 2001). Considerable evidence of glumes, seeds and fragments of wild cereals suggests an exploitation of plants for human consumption in the Late Acacus, not seen in the earlier phase (di Lernia 1999a). Imprints of *Panicum* and other grasses were recorded in pottery (Magid 1999) and many grindstones (500 fragments) were found in the deposit (di Lernia 1999b), clearly demonstrating extensive knowledge of grass storage and processing and a systematic exploitation of wild grasses (Mercuri 1999).

2.6.2 Ifri Oudadane, Ifri n'Etsedda and Hassi Ouenzga, Morocco

The sites of Ifri Oudadane (OUD), Ifri n'Etsedda (ETS) and Hassi Ouenzga (HAS) lie in the Moroccan Rif (Fig. 1) region and are located on the northern coast of Morocco, west of the Melilla Peninsula and 5 km to the east of the Oued Kert River mouth (OUD), in north-eastern Morocco, at the eastern fringe of the Rif range, on the southern flank of the Kbdana mountains (ETS) and on the Plain of Gerrouaou, some 40 km south of the Mediterranean coast (HAS), respectively (Linstädter and Kehl, 2012; Linstädter *et al.* 2012; 2016).

At Ifri Oudadane, a rock shelter situated on the coast, archaeological deposits covered a period of around 5000 years, between 11,000 and 5700 cal BP, comprising two clear occupation phases, including the transition from the Epipalaeolithic to the Neolithic. Evidence from Epipalaeolithic deposits confirms the presence of hunter-gatherers who also exploited marine resources, including fishing and the consumption of mussels. The Early Neolithic saw the first appearance of decorated pottery with Cardium impressed designs and also marked the arrival of domesticated species such as sheep and goat, documented through bones and accumulation layers of calcite spherulites suggesting intensive penning of ovicaprids inside the shelter. Domesticated plants (Fig. 3), including cereals (*Triticum monococcum/dicoccum*, *Triticum aestivum/durum*, *Hordeum vulgare*) and pulses (*Lens culinaris*, *Pisum sativum*, *Vicia faba*) were also present, yielding the earliest date in Africa for a domesticated plant seed (lentil) at c. 7600 cal BP (Morales *et al.* 2013; Morales *et al.* 2016). In fact, Ifri Oudadane is one of the few sites on the Mediterranean coast of Morocco where the early arrival of full Neolithic 'traits', including domesticated plants and animals, is documented. However, the percentage of crops

in the seed assemblage was less than 1% (Morales *et al.* 2016), confirming the continuing importance of wild plants in the economy of those groups.

Occupation levels at Ifri n'Etsedda, an inland rock shelter, covered around 4000 years, between 10,000 and 6000 cal BP, including both Epipalaeolithic and Early Neolithic deposits. The Epipalaeolithic encompassed the period approximately 9900 to 8500 cal BP, with deposits comprising lithics and a large amount of shells, presumed to be food remains. The Early Neolithic was dated between 7200 - 6800 cal BP and, in these deposits, terrestrial snail shells remained a constant feature. Ceramics were also present, from typical impression decorated ware at the beginning of the Neolithic to later herringbone decoration. The latter also indicates an occupation during the Late Neolithic after 6000 cal BP (Linstädter *et al.* 2016; Stempfle *et al.* 2018). The faunal assemblage comprised domesticated ovicaprids, from the earliest Neolithic, although these were only present at a maximum of 10% in each stratigraphic unit. The assemblage was dominated by aurochs, wild boar, wild Barbary sheep and gazelle (Linstädter *et al.* 2016).

Hassi Ouenzga is a small rock shelter site with the earliest level dated to the early 9th millennium cal BP. Cardium impressed pottery, with a local influence, appeared from 7500 cal BP onwards (Linstädter 2003; Linstädter 2004) possibly indicating relationships with the Iberian coast and also that early levels at HAS are contemporaneous to the Early Neolithic phase at Ifri Oudadane, when Cardium pottery also appeared. Domesticated fauna were present, in low abundances, but botanical remains do not show any traces of domesticated species, although the lack of remains may be a methodological problem, since systematic sampling and flotation of the sediments was not carried out. In summary, a mixture of a late Epipalaeolithic lithic assemblage with Early Neolithic pottery and low abundances of domesticated species suggests this site belonged to a local hunter-gatherer community who had contacts to Neolithic groups at a time before food production had been adopted (Linstädter *et al.* 2012).

3. Materials and methods

3.1 Takarkori and Uan Afuda rockshelter – pottery

A total of 110 decorated potsherds from Early to Middle Holocene contexts at Takarkori ($n=81$) and Uan Afuda, Libya ($n=29$), were analysed (Fig. 4). Potsherds from Takarkori came from both Late Acacus hunter-gatherers (*c.* 10,200 - 8000 cal BP) and Pastoral herder contexts (*c.* 8300 - 4650 cal BP) whereas the potsherds from the Uan Afuda cave all originated from Late

Acacus stratigraphic contexts, dated by multiple radiocarbon measures (between approximately 8500 - 8900 years cal BP).

Late Acacus pottery mostly comprises semi-cylindrical or rounded bowls, decorated by the rocker technique, mostly of packed zigzags made with an evenly serrated edge instrument. The Dotted wavy line motif is often present in association with the packed rocker technique. Pottery form and decoration changes in the Pastoral period with vessels with a globular body and short neck or collar often being smoothed or burnished. Decoration shifts to the alternately pivoting stamp (APS) technique where a double-pronged implement is used in a rocker-like movement producing a variety of motifs, characterised by very regular patterns of rows of dots, incorporating paired lines, dashes or triangles (Caneva 1987; Eramo *et al.* 2020).

3.2 Ifri Oudadane, Ifri n'Etsedda and Hassi Ouenzga - pottery

A total of 306 potsherds from Ifri Oudadane ($n=130$), Ifri n'Etsedda ($n=81$) and Hassi Ouenzga ($n=95$) were analysed (Fig. 5). The majority of sherds analysed from Ifri Oudadane were from the ENB phase, representing the main occupation phase between 7100 and 6600 cal BP, where pottery was characterised by single Cardium impressions. (Linstädter and Kehl 2012). Pottery from the latest phase of the Early Neolithic C (ENC) were also sampled. Pottery sampled at Ifri n'Etsedda includes the earliest Cardium-decorated pottery which first appeared in layer INES-5 (*c.* 7200 cal BP) and also layer INES-6, dated to 6800 - 6500 cal BP. This is slightly later than the first appearance of pottery at both Ifri Oudadane and Hassi Ouenzga. Pottery sampled from the following phase, INES-7, the main occupation phase, dated between 6600 and 6100 cal BP, contained impressions which form horizontal, vertical and oblique lines, predominantly made using combs, although when the so-called herringbone motive appears, this is formed by marine shells. Pottery sampled from layer INES-8, denoting a Late Neolithic occupation, comprised comb impressions and undecorated, polished vessels. (Stempfle *et al.* 2018). The earliest pottery sampled at Hassi Ouenzga originated from levels dated to 7600 cal BP. The pottery displayed three distinct decorative motifs including, firstly, single impressions of Cardium shells, which have references to the Moroccan coast. Second were sherds decorated with cross-hatching, extremely similar to those found in the Oran area on the Algerian coast, together with a third group with decorations that only appeared at Hassi Ouenzga (Linstädter 2003; 2004).

3.3 Methods

Lipid analysis and interpretations were performed using well-established analytical procedures (Dudd and Evershed 1998; Correa-Ascencio and Evershed 2014). All solvents used were HPLC grade (Rathburn) and the reagents were analytical grade (typically > 98% of purity). Briefly, ~2 g of potsherd were sampled and surfaces cleaned with a modelling drill to remove any exogenous lipids.

3.3.1 Solvent extraction

The sherds were then ground to a powder, an internal standard added and solvent extracted by ultrasonication (chloroform/methanol 2:1 v/v, 2x10ml). The solvent was evaporated under a gentle stream of nitrogen to obtain the total lipid extract (TLE). Aliquots of the TLE were trimethylsilylated (*N,O*-bis(trimethylsilyl)trifluoroacetamide 80 µl, 70° C, 60 min), and submitted to analysis by GC and GC-MS. Further aliquots of the TLE were treated with NaOH/H₂O (9:1 w/v) in methanol (5% v/v, 70°C, 1 h). Following neutralization, lipids were extracted into chloroform and the excess solvent evaporated under a gentle stream of nitrogen. Fatty acid methyl esters (FAMES) were prepared by reaction with BF₃-methanol (14% w/v, 70°C, 1 h). The FAMES were extracted with chloroform and the solvent removed under nitrogen. The FAMES were re-dissolved into hexane for analysis by GC and GC-C-IRMS.

3.3.2 Acid extraction

The cleaned sherd powder was crushed in a solvent-washed mortar and pestle and weighed into a furnace culture tube (I). An internal standard was added (20 µg *n*-tetratriacontane; Sigma Aldrich Company Ltd) together with 5 ml of H₂SO₄/MeOH 2 - 4% ($\delta^{13}\text{C}$ measured) and the culture tubes were placed on a heating block for 1 h at 70 °C, mixing every 10 min. Once cooled, the methanolic acid was transferred to test tubes and centrifuged at 2500 rpm for 10 min. The supernatant was then decanted into another furnace culture tube (II) and 2 mL of dichloromethane extracted double distilled water was added. In order to recover any lipids not fully solubilised by the methanol solution, 2 x 3 mL of *n*-hexane was added to the extracted potsherds contained in the original culture tubes, mixed well and transferred to culture tube II. The extraction was transferred to a clean, furnace 3.5 mL vial and blown down to dryness. Following this, 2 x 2 mL *n*-hexane was added directly to the H₂SO₄/ MeOH solution in culture tube II and whirlmixed to extract the remaining residues. This was transferred to the 3.5 mL vials and blown down under a gentle stream of nitrogen until a full vial of *n*-hexane remained. Aliquots of the extracts (containing fatty acid methyl esters, FAME's) were derivatised using *N,O*-bis(trimethylsilyl)trifluoroacetamide (BSTFA) containing 1 % v/v trimethylchlorosilane

(TMCS; Sigma Aldrich Company Ltd.; 20 μ L; 70°C, 1 h). Excess BSTFA was removed under nitrogen and the extract was dissolved in *n*-hexane for analysis by gas chromatography (GC), GC-mass spectrometry (GC-MS) and GC-combustion-isotope ratio MS (GC-C-IRMS).

All FAMES initially underwent GC using a GC fitted with a HT non-polar column (DB1-HT; 100% dimethylpolysiloxane, 15 m x 0.32 mm i.d., 0.1 μ m film thickness). The carrier gas was helium and the temperature programme comprised a 50°C isothermal hold followed by an increase to 350°C at a rate of 10°C min⁻¹ followed by a 10 min isothermal hold. A procedural blank (no sample) was prepared and analysed alongside every batch of samples. Further compound identification was accomplished using GC-MS. FAMES were then introduced by autosampler onto a GC-MS fitted with a non-polar column (100% dimethyl polysiloxane stationary phase; 60 m x 0.25 mm i.d., 0.1 μ m film thickness). The instrument was a ThermoFinnigan single quadrupole TraceMS run in EI mode (electron energy 70 eV, scan time of 0.6 s). Samples were run in full scan mode (m/z 50–650) and the temperature programme comprised an isothermal hold at 50°C for 2 min, ramping to 300°C at 10° min⁻¹, followed by an isothermal hold at 300°C (15 min). Data acquisition and processing were carried out using the HP Chemstation software (Rev. C.01.07 (27), Agilent Technologies) and Xcalibur software (version 3.0). Peaks were identified on the basis of their mass spectra and GC retention times, by comparison with the NIST mass spectral library (version 2.0).

Carbon isotope analyses by GC-C-IRMS were also carried out using a GC Agilent Technologies 7890A coupled to an Isoprime 100 (EI, 70eV, three Faraday cup collectors m/z 44, 45 and 46) via an IsoprimeGC5 combustion interface with a CuO and silver wool reactor maintained at 850°C. Instrument accuracy was determined using an external FAME standard mixture (C₁₁, C₁₃, C₁₆, C₂₁ and C₂₃) of known isotopic composition. Samples were run in duplicate and an average taken. The $\delta^{13}\text{C}$ values are the ratios $^{13}\text{C}/^{12}\text{C}$ and expressed relative to the Vienna Pee Dee Belemnite, calibrated against a CO₂ reference gas of known isotopic composition. Instrument error was $\pm 0.3\%$. Data processing was carried out using Ion Vantage software (version 1.6.1.0, IsoPrime).

4. Molecular and isotopic identification of plant processing in Early to Middle Holocene ceramics from sites in the central Sahara and Mediterranean north Africa

4.1 Results – plant processing at Takarkori and Uan Afuda, Libya

Of the 81 sherds analysed from Takarkori, $n = 29$ displayed distributions typical of an animal fat origin (Evershed *et al.* 2002, Dunne *et al.* 2012), $n = 38$ displayed distributions strongly

indicative of a plant origin (Late Acacus, $n = 4$; Early Pastoral, $n = 2$ and Middle Pastoral, $n = 32$; Dunne *et al.* 2016) with the remainder likely reflecting either the processing of both plant and animal products in the vessels or the multi-use of vessels. Potsherd samples from the Uan Afuda cave totalled $n = 29$, of which $n = 22$ yielded appreciable lipid concentrations (76%). Of these, $n = 18$ of the TLEs yielded lipid profiles indicative of a plant origin (82%; Dunne *et al.* 2016).

The plant-derived lipid profiles (Fig. 6) are characterised by unusually complex mixtures of aliphatic compounds including short, medium and long-chain fatty acids, dicarboxylic acids, α,ω -hydroxyacids and n -alkanes (Dunne *et al.* 2016). The range of diagnostic criteria and proxies discussed above, not previously used in ceramic lipid residue analysis, aside from the palmitic/stearic acid ratios (P/S ratio), were used to explore the nature of the lipid distributions in the pottery, including average chain length (ACL; Eglinton and Hamilton, 1967), carbon preference index (CPI; Diefendorf *et al.*, 2011), the P_{aq} proxy ratio (Ficken *et al.* 2000) and compound-specific $\delta^{13}C$ values (Table 1). For example, relative abundance P/S ratio values, where greater than 1.3, indicate a plant origin, confirming the origin of Takarkori and Uan Afuda lipid residues, which range from 1.5 to 14.3 (Table 1) and not seen in animal fats, especially those of archaeological origin (Mills and White 1994). Where the CPI values could be calculated for the Takarkori and Uan Afuda residues, these ranged between 2.1 and 8.1 (Table 1), again confirming the plant origin of these n -alkanes.

Where possible (i.e. if all compounds were present such that the ratio could be calculated), the P_{aq} proxy ratio was applied to the values of the n -alkanes extracted from the Takarkori potsherd residues, to determine whether the lipid profiles, with maxima of C_{25} n -alkane, might originate from submerged or floating macrophytes. Significantly, seven lipid residues from Takarkori and two from Uan Afuda, where the C_{max} is the C_{25} n -alkane, have P_{aq} ratio values between 0.4-1.0 (Table 1) suggesting submerged or floating macrophytes may have been processed in the vessels (Ficken *et al.* 2000). Furthermore, the P_{aq} ratio was calculated for one other sample, TAK953 ($C_{max} = C_{31}$ n -alkane), revealing a P_{aq} value of 0.35 which falls within the range for emergent macrophytes (0.1-0.4). The ACL values were also calculated for Takarkori ($n=18$) and Uan Afuda ($n=7$). The values are extremely constant, at Takarkori, they range between 26.3 and 29.4, (mean 28.3) and for Uan Afuda, between 26.0 to 29.2, (mean 27.4). It is known that ACL values vary between plant functional types, for, example, the ACL range between the evergreen gymnosperms is quite large, between 26 to 34, and values for the deciduous angiosperms span from 27 to over 32 (Diefendorf *et al.* 2011). In Rommerskirchen's (2006)

chemotaxonomic study on the C₄ grasses, which are dominated by either C₃₁ or C₃₃ *n*-alkanes, he establishes that the mean ACL values for the Aristidoideae were 30.74, for the Chloridoideae 31.23, and the Panicoideae, 30.92. Interestingly, ACL values for both Takarkori (mean 28.3) and Uan Afuda (mean 27.4), suggest a mid-chain length origin, however, for the residues where the *n*-C₃₁ dominates, the ACL values are slightly longer than for those where the *n*-C₂₅ or *n*-C₂₇ predominates (Table 1).

4.1.1 The identification of leafy plants at Takarkori and Uan Afuda

In the lipid residue profiles two main signatures (Fig. 6a,b) dominated the extracts: i.e. either medium chain-length *n*-alkanes, C₂₅ or C₂₇, or longer chain *n*-alkanes, namely the C₃₁ *n*-alkane, providing further valuable information on the plants processed in the vessels. Those maximising at C₂₅ are highly unusual (Fig. 6b, *n*=9), and diagnostic as originating from aquatic plants, confirmed by the *P*_{aq} (emergent and non-emergent aquatic macrophyte input) proxy ratio (Ficken *et al.* 2000), as discussed previously. Those lipid profiles dominated by the C₃₁ *n*-alkane (Fig. 6a) likely originate from C₃ or C₄ wild grasses, for example, the Aristidoideae grasses (C₄ group) always maximise at the *n*-C₃₁ alkane while the Chloridoideae and Panicoideae (C₄ group) are dominated by either the C₃₁ or the C₃₃ *n*-alkane (Rommerskirchen *et al.* 2006) or lake-margin plants, such as sedges (Kolattukudy *et al.* 1976; Maffei 1996; Rommerskirchen *et al.* 2006).

Also present in a number of the lipid residues are a series of even-numbered long-chain fatty acids (LCFAs) up to C₃₀, often in high abundance (Fig. 6c). These are highly indicative either of an origin in leaf or stem epicuticular waxes (Kolattukudy *et al.* 1976; Tulloch 1976; Bianchi 1995; Kunst and Samuels 2003) or, possibly, suberin (Kolattukudy 1980; 1981; Walton 1990; Pollard *et al.* 2008), an aliphatic polyester found in all plants.

4.1.2 The identification of plant oils at Takarkori and Uan Afuda

The high concentrations of both short and long-chain fatty acids, and diacids, suggest the processing of a range of different plant types and organs, such as grains/seeds and leafy plants and stems, in the pottery. For example, well-known plant degradation products are evident in the gas chromatograms as short-chain fatty acids, such as *n*-nonanoic acid and diacids, e.g. azelaic acid (Fig. 6d). Also present in some of the Takarkori and Uan Afuda residues are lipid profiles displaying series of short-chain fatty acids, often starting at C₈, together with high abundances of C₁₂ and C₁₄ fatty acids. These distributions are unusual but were previously observed at the archaeological site of Qasr Ibrim, Egypt, where their presence was

demonstrated to originate from the processing of palm kernels in pots (Copley *et al.* 2001). However, although the vessels from the Libyan Sahara do include high abundances of C₁₂ and C₁₄, they are generally either present in similar abundances or the C₁₄ predominates, in contrast to the Qasr Ibrim residues where the C₁₂ is the most abundant compound.

In summary, the series of short and long-chain fatty acids seen in the potsherds are indicative of a plant origin and could derive from epicuticular waxes (in the case of the LCFAs) or from plant oils produced from seeds, or a mixture of both. Furthermore, long-chain fatty acids can also be found in plant oils, for example, groundnut oil comprises 4-7% of C₂₀, C₂₂ and C₂₄ saturated and monoene acids (Gunstone 2004). It seems likely that a number of the Takarkori and Uan Afuda residues, which include high abundances of the short chain fatty acids and diacids, together with high P/S ratios, denote processing of oils, perhaps by boiling of seeds to extract the oils (Dunne *et al.* 2016).

4.1.3 Stable carbon isotope ($\delta^{13}\text{C}$) values of plant-derived lipids

The extremely broad range of $\delta^{13}\text{C}$ values for both the C₁₆ fatty acid and the *n*-alkanes extracted from Takarkori and Uan Afuda vessels confirms mixtures of C₃ and C₄ plants were being processed in the vessels (Table 1). The individual $\delta^{13}\text{C}$ values for the leaf wax *n*-alkanes from both sites range from -30.0 to -17.7 ‰ for the C₂₅ *n*-alkane, from -32.6 to -23.1 ‰ for the C₃₁ *n*-alkane and from -27.4 to -13.8 ‰ for the C_{16:0} fatty acid. These extremely broad ranges reflect the known $\delta^{13}\text{C}$ values for both bulk plant lipids (from -32 to -20 ‰ for C₃ plants and from -17 to -9 ‰ for C₄ plants; Boutton, 1991) and for leaf wax lipids, which are more depleted in ¹³C than the biomass (between -39 and -29 ‰ in C₃ plants and -26 and -14 ‰ in C₄ plants; Bi *et al.*, 2005). These ranges also encompass the carbon isotope values of freshwater aquatic plants, which commonly display a C₄-like signature (Keeley and Sandquist 1992) but, as discussed above, are separable based on their respective *n*-alkane distributions. However, in summary, the leaf wax $\delta^{13}\text{C}$ values are significant, suggesting a more C₄ origin for the Takarkori leaf waxes in contrast to those from Uan Afuda which are mainly C₃.

4.2 Results - plant processing at Ifri Oudadane, Ifri n'Etsedda and Hassi Ouenzga, Morocco

Organic residue analysis of 306 potsherds from the sites of Ifri Oudadane, Ifri n'Etsedda and Hassi Ouenzga, in the Rif region, Morocco, yielded 47 interpretable lipid profiles (15% recovery overall) with approximately 25% of vessels from all sites yielding lipid profiles indicative of plant processing (Dunne *et al.* 2019b). These sherds all originated from Early

Neolithic levels (Ifri Oudadane, ENB phase, 7.1 to 6.7 cal BP, Ifri n'Etsedda ENB and ENC phases, 6.8 to 6.4 cal BP and Hassi Ouenzga EN layers 3 and 4, 7.6 cal BP). The lipid profiles comprised sequences of even-numbered long-chain fatty acids (LCFAs, Fig. 7a,b,c), containing C₂₀ to C₃₀ carbon atoms, generally dominated by the C₂₀ or C₂₄, which, as noted, are strongly indicative either of an origin in leaf or stem epicuticular waxes (Kolattukudy *et al.* 1976; Tulloch 1976; Bianchi 1995; Kunst and Samuels 2003). Also present in a number of these profiles is a series of odd-over-even long-chain *n*-alkanes, ranging from C₂₅ to C₃₃, dominated by the C₂₉ and C₃₁ *n*-alkanes (Fig. 7a,b), suggesting the possible processing of grasses in these vessels.

4.2.1 Stable carbon isotope ($\delta^{13}\text{C}$) values of plant-derived lipids

In the Moroccan Rif samples, the *n*-alkanes were present in low abundance and could not be measured isotopically but the $\delta^{13}\text{C}$ values of the long-chain fatty acids (C₂₀ to C₃₀) were measured to confirm whether the plant waxes processed in the pots had a C₃ or C₄ origin (see Table 2 for period and stratigraphic level). These fell within the known $\delta^{13}\text{C}$ values for C₃ bulk plant lipids, which range from -32 to -20 ‰ for C₃ plants (Boutton 1991), averaging as follows, -26.8, -28.4, -28.6, -30.3, -29.7 and -29.7 ‰ for the C₂₀, C₂₂, C₂₄, C₂₆, C₂₈ and C₃₀ fatty acids, respectively (Table 2). The combination of LCFAs and *n*-alkanes, and associated $\delta^{13}\text{C}$ values for the LCFAs, in *c.* 25% of the pottery lipid profiles at each site, strongly suggests the processing of C₃ plant material, likely leafy plants and/or wild grasses, within these vessels, possibly mixed with animal products (Dunne *et al.* 2019b).

5. Discussion - plant exploitation in the Libyan Sahara and Mediterranean north Africa.

Many archaeological sites in the Libyan Sahara have yielded deposits extraordinarily rich in pollen and plant macrofossil remains, for example, at Takarkori rockshelter, *c.* 30,000 desiccated seeds/fruits were identified in nine concentrations or 'spots' located close to fireplaces (Fig. 2; Olmi *et al.* 2011), demonstrating the systematic gathering of wild cereals (Mercuri 2001, 2008b). Furthermore, at Takarkori, remarkably well-preserved remains of 53 items of basketry (possibly used to gather and store wild cereals) and cordage, attests to the overall importance of wild grasses at the site. In fact, the main wild cereals collected for food and transported into the site (Olmi *et al.* 2011) were also the main plants used to make these items (di Lernia *et al.* 2012).

The significant abundances of specific plant biomarkers in the vessels from Takarkori and Uan Afuda is indicative of prolonged processing of large quantities of a range of plant material in

the vessels. This is confirmed by the wide range of exceptionally well-preserved archaeobotanical remains found at both sites (Castelletti *et al.* 1999; Olmi *et al.* 2011), including *Typha*, *Ficus*, *Cupressus*, *Tragus*, *Cassia* and *Balanites aegyptica* together with Panicoideae fruits (e.g. *Echinochloa*, *Panicum* and *Setaria*). Across the broader region, at archaeological sites across the Wadi Teshuinat region, such as Uan Tabu and Uan Muhuggiag (Fig. 1), grasses (Poaceae) comprised the most frequent fruit and plant fragments. The amount of florets and caryopses of dried and well-preserved Paniceae (mostly C₄ grasses) at these sites is striking, demonstrating the systematic exploitation of both C₃ and C₄ wild grasses, including *Urochloa*, *Brachiaria*, *Cenchrus*, *Echinochloa*, *Panicum* and *Setaria* (Wasylikowa 1992; 1993; Castelletti *et al.* 1999; Mercuri 1999; 2001; 2008a; 2008b; Olmi *et al.* 2011).

The archaeobotanical evidence from several sites in the Libyan Sahara and the Western Desert of Egypt also strongly suggests a persistent reliance on wild grasses in early pastoralist societies (Wasylikowa 1992; 1993; 2001; Barakat and Fahmy 1999; Castelletti *et al.* 1999; Mercuri 1999; 2001; 2008a, b; Mercuri and Trevison Grandi 2001; Olmi *et al.* 2011). At Nabta Playa (Fig. 1), in the Western Desert of Egypt (*c.* 8025 - 7400 BP), over 120 wild plant taxa were identified, including wild sorghum and several grasses from the subfamily Panicoideae, small seeded legumes, fruit and tuber plants (Wendorf *et al.* 1992; Wasylikowa 2001). Assemblages of charred wild grains, found within hearths, pits and potholes, were also found at Hidden Valley, near the Farafra Oasis, and Eastpans, near the Abu Ballas ridge (Fig. 1), dated between 6700 - 6200 BP (Barakat and Fahmy 1999). Of the eight types of wild grasses identified at these Western desert playa sites, five were found at all three sites. These likely grew in extensive stands in the vicinity of the playas. A similar record characterises sites in Upper Nubia and the Central Sahara, and the adoption of appropriate technology, such as pottery and large grinding stones, across the region, is well-documented (e.g. Wendorf *et al.* 1992; di Lernia 1996; Mercuri 2008b).

Grasses from dense stands close to these sites were likely gathered by prehistoric people, and used intensively, as staple food plants (Barakat and Fahmy 1999), but also for animal fodder and other purposes such as mats, bedding and basketry (e.g. di Lernia *et al.* 2012). The ability to boil these grasses for long periods of time in newly invented ceramic vessels would have significantly increased the range of plants prehistoric people could eat. This dovetails well with Huysecom's (2009) suggestion that ceramics were invented to boil ground Panicoideae grasses, following the spread of new grasslands in the early Holocene and Haaland (2007) who also argues pottery was developed to process grains to make porridge and beer.

Furthermore, many of the aquatic or emergent plants are edible, for example, *Typha* or cattail, found at Takarkori and Uan Afuda, have been used in many countries across the world, with many different parts of the plant, such as the rhizomes, lateral stems, the flower spike and pollen, being edible (Gott 1999). Tubers and rhizomes from the *Nymphaea* can also be eaten, and some varieties, such as *Nymphaea caerulea*, are known to have narcotic properties (Emboden 1978). Many of these plants would need to be boiled to soften and/or reduce any toxic properties. Significantly, Hather (1995) argues that tubers from *Cyperus rotundus* and rhizomes of *Typha* and *Nympheae* formed a significant part of the diet of the inhabitants at the Early Neolithic site E-75-6 at Nabta Playa, Western Desert. *Cyperus rotundus* was also identified at Uan Afuda (Castelletti *et al.* 1999) and the pollen record in the Tadrart Acacus demonstrating that flora such as *Typha*, *Potamogeton*, *Lemna* and *Scirpus*, plants that require permanent freshwater sources, were present c. 11,000 to 9200 cal BP (Mercuri, 1999). Leaves, stems and starchy rhizomes of *Potamogeton* are edible. Furthermore, Olmi *et al.* (2011) identified macro-remains from hygrophilous plants such as sedges, rushes and cattails at Takarkori.

Lipid and isotopic distributions from the Moroccan Rif pottery also confirm the importance of plant processing. This is in good agreement with plant macro-remains which were extremely well-preserved at Ifri Oudadane with archaeobotanical studies revealing the importance of exploiting a huge variety of both wild and domesticated plants, ranging from grasses to legumes and fruits of different tree species (Morales *et al.* 2013; Morales *et al.* 2016; Morales 2018). The Epipalaeolithic layers contained a significant number of wild plant remains (742) but the majority were found in Neolithic layers (7199) and only the Neolithic layers yielded domesticated plants. By far the most dominant species identified (5842) was seeds from the mastic tree (*Pistacia lentiscus*). Wild legumes (Fig. 3f,g; *Lathyrus* sp./*Vicia* sp.), large seeds with a high content of carbohydrates and proteins (Butler *et al.* 1998), were also commonplace throughout the stratigraphy, suggesting they were collected for consumption. The dwarf palm (Fig. 3e; *Chamaerops humilis*) was also common throughout the sequence (Morales *et al.* 2013) together with seeds from juniper (*Juniperus phoenicea*), wild olive (*Olea europaea*) and acorns (*Quercus* sp.). Although small (61 plant macroremains in total), the domesticated crop assemblage from Ifri Oudadane, only found in Neolithic levels, included einkorn (*Triticum monococcum*), emmer (Fig. 3b; *Triticum dicoccum*), hard wheat (*Triticum durum*), free-threshing wheat (*Triticum aestivum/durum*) and barley (Fig. 3d; *Hordeum vulgare*). Two domesticated pulses, pea (*Pisum sativum*) and lentil (Fig. 3c; *Lens culinaris*), were also present

(Morales *et al.* 2013). Radiocarbon dates on nine seeds, ranging from *c.* 7600 cal BP to 6400 cal BP, confirm their presence in the Early Neolithic phase. Flotation for plant remains was not carried out at Hassi Ouenzga but pollen samples were taken at Ifri n'Etsedda (Linstädter *et al.* 2016). These indicate that, during the ENB (*ca.* 6800 cal BP) wild olive (*Olea europaea*), carob tree (*Ceratonia siliqua*) and Atlantic terebinth (*Pistacia terebinthus*), present in the Late Epipalaeolithic, continue to be abundant, together with Phoenician juniper (*Juniperus phoenicea*), myrtle (*Myrtus communis*) and Mediterranean buckthorn (*Rhamnus alaternus*). The presence of cereal grains is also documented with percentages ($\geq 5\%$) sufficient to suggest local cultures in the vicinity of the shelter (López-Sáez and López-Merino 2005).

Excavations at other Mediterranean north Africa sites, for example, at Early Neolithic Khil and That el-Ghar, Morocco, (Fig. 1; Morales *et al.* 2016, 5500 to 5000 cal BC) also revealed domesticated cereals and at Grotte Capéletti, Algeria (*c.* 6400 to 5800 cal BP), and Hergla, Tunisia (Fig. 1, *c.* 7600 cal BP), abundant remains of wild plant species, such as acorns (*Quercus ilex*), wild legumes (*Vicia* sp.), and pine nuts (*Pinus pinaster*), although no signs of domesticated species were present (Roubet 1979; Mulazzani 2013), similar to the assemblage found in Neolithic layers at Haua Fteah, Libya (Fig. 1; Barker *et al.* 2010). Acorns, pine nuts and seeds of vetches, wild pulses, juniper and wild olive were also documented in the late Iberomaurusian levels of Taforalt (Fig. 1; Humphrey *et al.* 2014). Many of these plants provide the full range of nutrients required in the human diet, for example, wild legumes and grasses, junipers and acorns are high in carbohydrates, terebinth pistachio, wild olive and pine nut are rich in fats (lipids) and pine nuts, wild legumes and oats are relatively high in proteins (Morales *et al.* 2016).

6. Conclusion

Here, the combined lipid and isotopic data from Holocene sites in the Libyan Sahara and the Moroccan Rif confirms the potential for lipid residue analysis to provide insights into an aspect of dietary and subsistence practices of human groups across north Africa, namely plant exploitation. The findings of extensive plant wax and oil residues in prehistoric Libyan pottery provide a novel perspective on how early pottery was used in the Sahara, lending weight to arguments that its invention there may have been related to the processing of plants, likely wild grasses (Haaland 2007; Huysecom *et al.* 2009). However, plant lipid distributions also suggest the importance of a wider range of plants (both C_3 and C_4) for consumption, including seed oils and aquatic plants. Furthermore, combinations of diagnostic lipid distributions appear in

virtually all vessels with plant signals, suggesting that the pots were used to process several varieties of plants, either contemporaneously or over the life use of the container. These data all imply sophisticated strategies of plant gathering, processing and culinary use in the Libyan Sahara, beginning in the early Holocene.

Lipid and isotopic distributions from Rif pottery strongly suggests the processing of C₃ plant material, likely leafy plants and/or grasses. These could comprise wild legumes or wild and/or domesticated cereals, both found at Ifri Oudadane and Ifri n'Etsedda, although the domesticated cereals, present from at least 7600 cal BP, are generally found in much lower abundances (Morales *et al.* 2013; 2016).

Pottery found in both regions is often used to process plants in conjunction with other ingredients, either meat from both ruminant or non-ruminant animals, or dairy products, likely in the form of stews or porridges (Dunne *et al.* 2012, 2016), which together constitute a meal or dish (Hastorf 2016). However, whilst in the Libyan Sahara some pots are solely dedicated to plant processing, e.g. seed oil processing, this does not appear to be the case at Rif sites. Furthermore, a greater number of pots at Uan Afuda (82%) and Takarkori (47%) include evidence of plant processing, as opposed to Rif sites where plant lipids are only found in 25% of vessels, emphasising the differences in plant exploitation between the regions. This may be related to a number of factors, such as preservational conditions, although it should be noted that early plant processing in Mediterranean north African pottery may also have followed a different trajectory, likely garnering influences from close cultural contacts with groups from Holocene Iberia. The adoption of a Neolithic lifestyle in northern Morocco and south-western Spain is thought to be virtually contemporaneous (Cortés Sánchez *et al.* 2012; Linstädter *et al.* 2012). This is evidenced by close resemblances between *Cardium* impression ceramics found in the earliest Neolithic levels in the eastern Rif and Oran regions in the Maghreb and those from sites such as Nerja, Murciélagos and Carigüela in Andalusia, as opposed to typologies deriving from the Neolithic groups from the Sahara (Manen *et al.* 2007; Linstädter 2008; Ramos *et al.* 2008; Cortés Sánchez *et al.* 2012).

These results also show that plant processing maintained its importance in the subsistence strategies of these prehistoric groups across the Holocene, occurring both contemporaneously with, and following, the adoption of domesticates and the exploitation of secondary products (Dunne *et al.* 2012; 2019b). The plant lipid residues also correlate well with extensive archaeobotanical evidence for the gathering and processing of wild plants, including grasses,

seeds and aquatic plants, in both Holocene Saharan and Mediterranean Africa. The ‘new’ methods of cooking, in particular, boiling, in thermally resistant ceramics, likely expanded the range of plants available for consumption, leading to an increased importance for the role of wild plants in the subsistence strategies of these groups.

Finally, the advent of these ‘new’ methods of food processing, aided by the increased control of heat, facilitated access to increased amounts of digestible nutrients and energy, provided increased dietary breadth and allowed the production of safer foods. This would have led to improved nutrition for children (increasing juvenile survival rates), nursing mothers and the elderly, promoted longevity and health for adults, and, by increasing the range of available foodstuffs, would have expanded the carrying capacity of the environment (Ikawa-Smith 1976; Hoopes and Barnett 1995; Rice 1999; Blandino *et al.* 2003). Thus, in the wider scale, the development of, and improvement in, food processing techniques and consequent increased biological and ecological skills and knowledge had implications for changes in subsistence strategies, mobility and land use, ultimately contributing to shifts toward social complexity (Wollstonecroft 2011).

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Figures

Figure 1. Map of north Africa showing African sites mentioned in the text 1. Khil, 2. Kaf Taht el-Ghar, 3. Ifri Oudadane, 4. Hassi Ouenzga, 5. Ifri n'Etsedda, 6. Tatoralt, 7. Gueldaman, 8. Grotte Capéletti, 9. Hergla, 10. Haua Fteah, 11. Eastpans, 12. Hidden Valley, 13. Wadi Kubbaniya, 14. Nabta Playa, 15. Takarkori, 16. Uan Muhuggiag, 17. Uan Tabu, 18. Uan Afuda and 19. Ounjougou (from Google Maps).

Figure 2. Archaeobotanical remains from Takarkori rockshelter. 1. Spot accumulation of seeds from the Middle Pastoral layer. a. spot overview, b-e. dorsal and ventral view of the spikelets of b. *Panicum laetum*, c. *Echinochloa colona*, d. uncharred grain, e. *Sorghum bicolor verticilliflorum*, f. mix overview, g. split spikelets of *E. colona*, h. charred grains in dorsal and ventral view, i. outer glumes of *E. colona*. (Image courtesy of Anna Mercuri, University of Modena).

Figure 3. Archaeobotanical remains from the sites of Ifri Oudadane and Khil, Morocco. a. *Poaceae* (grass seed), b. *Triticum* cf. *dicoccum* (einkorn/emmer), c. *Lens culinaris* (lentil), d. *Hordeum vulgare* (barley), e. *Chamaerops humilis* (dwarf palm), f. *Vicia faba* (wild pulse) g. *Lathyrus* sp. (wild pulse) (all images courtesy of Jacob Morales).

Figure 4. Analysed potsherds from Takarkori rock shelter grouped by archaeological phases from Late Acacus to Late Pastoral periods (c. 10,200 to 4650 cal BP). (© The Archaeological Mission in the Sahara, Sapienza University of Rome)

Figure 5. Selection of pottery vessel units from a. Ifri Oudadane (Early Neolithic B) and b. Hassi Ouenzga archaeological sites (Images from a. Linstädter and Wagner, 2013 and b. Linstädter, 2004).

Figure 6. Partial gas chromatograms of trimethylsilylated total lipid extracts (TLEs) from potsherds excavated from Takarkori rock shelter. Chromatographic peak identities denoted by filled triangles comprise *n*-alkanes in the carbon change range C_{25:0} to C_{33:0} and filled circles indicate straight-chain fatty acids in the carbon chain range C_{9:0} to C_{30:0}, maximising at C_{16:0}, filled squares indicate dicarboxylic acid in the carbon chain range C_{5:0} to C_{11:0}. The distributions show leaf wax *n*-alkanes and plant fatty acids (a) *n*-alkanes maximising at C₃₁ originate from C₃ or C₄ wild grasses or lake-margin plants, such as sedges (b) *n*-alkanes maximising at C₂₅ characteristic of an aquatic plant origin, (c) plant fatty acid profile showing the predominance of the C_{16:0} over the C_{18:0} fatty acid and high abundance of C_{12:0} and C_{14:0} fatty acids, characteristic of plant seed oil lipids, together with LCFAs suggestive of plant leaf waxes and (d) series of dicarboxylic acids which are well-known plant degradation products IS, internal standard, C₃₄ *n*-tetratriacontane.

Figure 7. Partial gas chromatograms of trimethylsilylated total lipid extracts (TLEs) from potsherds excavated from a. Ifri n'Etsedda, b. Ifri Oudadane and c. Hassi Ouenzga. Chromatographic peak identities denoted by blue filled triangles comprise *n*-alkanes in the carbon change range C_{25:0} to C_{31:0} and red filled circles indicate straight-chain fatty acids in the carbon chain range C_{14:0} to C_{28:0}, maximising at C_{16:0}. The distributions show leaf wax *n*-alkanes and plant long-chain *n*-alkanoic acids. IS, internal standard, C₃₄ *n*-tetratriacontane.

Table 1. Sample number, archaeological phase, P/S ratios, stable carbon isotope values of *n*-alkanes, CPI, ACL, weighted mean and P_{aq} values from trimethylsilylated TLEs of potsherds from Takarkori rock shelter and Uan Afuda cave, Tadrart Acacus, Libyan Sahara. n/d, not determined, signal intensity too low.

Stable carbon isotope values ($\delta^{13}C$)												
Sample no.	Archaeological period	Palmitic/ Stearic ratio	C_{23} <i>n</i> -alkane	C_{25} <i>n</i> -alkane	C_{27} <i>n</i> -alkane	C_{29} <i>n</i> -alkane	C_{31} <i>n</i> -alkane	C_{33} <i>n</i> -alkane	CPI C_{23}/C_{33}	ACL C_{23}/C_{33}	Weighted mean $\delta^{13}C$	P_{aq}
TAK4	Late Acacus	3.08	n/d	-23.3	-23.2	n/d	n/d	n/d	2.1	27.7	-23.2	0.55
TAK14	Middle Pastoral	1.98	-24.8	-29.4	-29.7	-29.3	-28.8	n/d	n/d	28.9	-29.2	n/d
TAK23	Late Acacus	2.56	n/d	-18.2	-19.8	-23.0	-23.1	-23.2	5.7	28.7	-21.4	n/d
TAK24	Early Pastoral	5.38	-27.7	-20.1	-20.6	-24.1	-24.8	-23.4	n/d	28.5	-22.4	n/d
TAK82	Middle Pastoral	2.82	n/d	-19.0	-20.5	-21.5	-24.0	n/d	8.1	28.5	-21.2	n/d
TAK135	Middle Pastoral	4.96	-20.6	-25.0	-21.3	n/d	n/d	n/d	n/d	28.2	-23.3	0.55
TAK159	Middle Pastoral	4.20	-21.0	-22.9	-16.3	n/d	n/d	n/d	n/d	27.4	-20.7	0.61
TAK479	Middle Pastoral	4.55	-27.3	-27.2	n/d	n/d	n/d	n/d	5.2	27.7	-27.2	0.58
TAK709	Middle Pastoral	2.47	-17.7	-22.2	n/d	n/d	n/d	n/d	3.9	29.4	n/d	n/d
TAK730	Middle Pastoral	4.80	-21.8	-23.5	-24.0	-27.4	n/d	n/d	5.1	28.9	-25.0	n/d
TAK766	Middle Pastoral	2.93	n/d	-18.4	-19.0	-23.6	-24.1	n/d	8.1	26.3	-19.5	0.80
TAK860	Middle Pastoral	4.00	-18.0	-19.0	-19.7	-23.3	-23.7	n/d	n/d	28.6	-21.4	n/d
TAK873	Middle Pastoral	1.98	-19.9	-17.7	-20.2	-23.9	-24.1	n/d	4.6	29.5	-22.4	n/d
TAK953	Middle Pastoral	2.10	-21.0	-26.1	-21.4	n/d	n/d	n/d	7.6	28.9	-24.0	0.35
TAK1008	Middle Pastoral	2.46	n/d	-27.4	-24.0	n/d	n/d	n/d	n/d	27.9	-26.3	n/d
TAK1054	Middle Pastoral	3.13	-27.1	-25.5	-21.0	-24.1	n/d	n/d	n/d	28.4	-23.9	0.58
TAK1072	Middle Pastoral	2.86	-17.6	-18.3	-19.6	-23.8	-23.6	n/d	n/d	28.1	-20.7	0.57
TAK1531	Middle Pastoral	4.00	-21.6	-24.5	-23.4	-26.9	-25.0	n/d	n/d	28.4	-24.9	n/d
UAF A1	Late Acacus	3.67	-29.8	-29.3	-31.2	-32.0	-32.6	n/d	n/d	29.2	-24.9	n/d
UAF A3	Late Acacus	3.70	n/d	n/d	n/d	n/d	n/d	n/d	n/d	27.3	n/d	0.89
UAF20	Late Acacus	1.50	-26.2	-25.4	-22.2	-24.8	-26.2	n/d	n/d	28.7	-21.5	n/d
UAF45	Late Acacus	4.69	n/d	n/d	n/d	n/d	n/d	n/d	n/d	26.0	n/d	0.75
UAF46	Late Acacus	4.66	n/d	-27.7	-21.5	n/d	n/d	n/d	n/d	27.7	-26.4	n/d
UAF50	Late Acacus	4.23	-27.0	-30.0	n/d	n/d	n/d	n/d	n/d	26.3	n/d	n/d
UAF84	Late Acacus	14.33	n/d	n/d	n/d	n/d	n/d	n/d	n/d	26.9	n/d	n/d

Table 2. Sample number, period, stratigraphic level, stable carbon isotope values of measurable LCFAs, indicative of plant processing, of trimethylsilylated TLEs extracted from potsherds from three Moroccan sites, Hassi Ouenzga, Ifri n'Etsedda and Ifri Oudadane, Morocco.

Sample number	Period	Stratigraphic level	$C_{20}FA$	$C_{22}FA$	$C_{24}FA$	$C_{26}FA$	$C_{28}FA$	$C_{30}FA$
HAS303	EN	Layer 4	-27.4	-	-28.2	-	-	-
HAS306	EN	Layer 4	-26.8	-27.8	-30.7	-	-	-
HAS314	EN	Layer 4	-26.7	-27.9	-28.5	-29.9	-28.7	-28.3
HAS369	EN	Layer 3	-27.9	-	-	-	-	-
ETS229	ENB	INES-6	-29.4	-29.5	-30.7	-30.2	-30.7	-31.1
ETS252	ENC	INES-7	-26.9	-28.2	-28.8	-29.5	-	-
ETS273	ENC	INES-7	-27.2	-28.5	-	-	-	-
OUD114	ENB	-	-27.0	-28.9	-29.8	-31.6	-	-
OUD152	ENB	-	-27.0	-28.2	-	-	-	-
OUD215	ENB	-	-22.1	-	-23.3	-	-	-