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Chasan, R.

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Rivka Chasan

A THESIS SUBMITTED

FOR THE DEGREE

"DOCTOR OF PHILOSOPHY"

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Department of Archaeology

July, 2022

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By: Rivka Chasan

Supervised by: Prof. Danny Rosenberg and Prof. Cynthianne Spiteri

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(Supervisor)

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(Supervisor)

Approved by: Dr. Yitzchak Jaffe Date: 18/07/22

(Chairperson of PhD studies Committee)

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Food choices and dietary practices during the 7th to 5th Millennium BC in the southern Levant: Evidence from organic residue analysis

Rivka Chasan

Abstract

The archaeological record of the protohistoric southern Levant (6,400–3,800 cal. BC) reflects small-scale communities, which strongly relied on domesticated farm animals, cereals and legumes. The time span also witnessed numerous technological innovations including most notably the introduction of pottery and later copper metallurgy. Prior studies suggest a gradual increase in socio-economic complexity, culminating with craft specialization and the introduction of specialized horticultural and pastoral economies, with an emphasis on olives and milk.

This dissertation tested these suggestions and discerned specific dietary and economic choices by applying organic residue analysis and synthesizing the results with the related archaeozoological and archaeobotanical assemblages and material culture remains. Lipid biomarker and isotope analysis were applied to a corpus of vessels from archaeological sites in Israel dating to the Pottery Neolithic and Chalcolithic periods. This was done using Gas Chromatography-Mass Spectrometry and Gas Chromatography-combustion-Isotope Ratio Mass Spectrometry.

The results indicate dietary continuity, regardless of chronology, culture, settlement scale and, typically, environmental conditions. Throughout the entire sequence, meat from domestic farm animals and the core cultivator crops were the main food sources. These were cooked in various combinations in ceramic vessels, with no clear task specific vessel forms identified. Some variation is noted in the use of dairy products. Lipid residue analysis and animal kill-off patterns suggest that milk was first used starting in the Middle Chalcolithic in limited frequency, and its use continued into the Late Chalcolithic period. Dairy residues were identified only in semi-arid regions, reflecting an environmentally-bound economic strategy, linked perhaps to climatic constraints. There is at present no lipid evidence for olive exploitation, contrary to prior suggestions for the time span. However, there is some suggestion for the use of wild resources, including beeswax and certain plants, indicating that even after the Neolithization process, specific wild resources were still exploited for their unique properties.

The general uniformity indicates that for thousands of years after the initial domestication processes, the various cultures shared a core economic and dietary tradition. The Pottery Neolithic

and Chalcolithic periods represent one long economic continuum. The chronological attributions then relate mainly to cultural preferences rather than differences in lifestyle in relation to economy and food ways.

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List of Abbreviations

4THTph10: 4-(5-Decyltetrahydrothiophene-2-yl) butanoic acid
5THTph9: 5-(5-Nonyltetrahydrothiophene-2-yl) pentanoic acid
6THTph8: 6-(5-Octyltetrahydrothiophene-2-yl) hexanoic acid
7THTph7: 7-(5-Heptyltetrahydrothiophene-2-yl) heptanoic acid
8THTph6: 8-(5-Hexyltetrahydrothiophene-2-yl) octanoic acid
APAA: ω-(*o*-alkylphenyl)alkanoic acid
COS: Cyclic octaatomic sulfur
Cr: Crust
$C_{x:y}$: Fatty acid with X carbon atoms and Y unsaturations
D_x: Diacylglycerol with X carbon atoms
DCM: Dichloromethane
E: Exterior
FID: Flame ionization detector
GC: Gas Chromatography
GC-c-IRMS: Gas Chromatography-Combustion-Isotope Ratio Mass Spectrometry
GC-MS: Gas Chromatography-Mass Spectrometry
In: Interior
K_x: Ketone with X carbon atoms
L2: Layer 2
$M_{x:y}$: Monoacylglycerol with chain length X and Y number of double bonds
MeOH: Methanol
MS: Mass Spectrometry
MSD: Mass spectra detector
NISP: Number of identified specimens
NIST: National Institute of Standards and Technology
P: Plastic
S: Soil
SS: Sunscreen
SHFA: Sulfur heterocyclic fatty acid
T_x: Triacylglycerol with X carbon atoms
TLE: Total lipid extract
WE_x: Wax ester with X carbon atoms

Chapter 1. Dissertation organization

This dissertation discusses food ways throughout the various stages of the protohistoric southern Levant using organic residue analysis (ORA) as a lens to test for the presence of diachronic and regional variation in dietary patterns. To that end, the following articles were included:

1. Chasan, R., Rosenberg, D., Klimscha, F., Be'eri, R., Golan, D., Dayan, A., Galili, E. and Spiteri, C. 2021. Bee products in the prehistoric southern Levant: evidence from the lipid organic record. *Royal Society Open Science* 8: 210950. Doi: 10.1098/rsos.210950.
2. Chasan, R., Klimscha, F., Spiteri, C. and Rosenberg, D. 2022. Foodways of an agro-pastoral community: Organic residue analysis of pottery and stone vessels at Middle Chalcolithic Tel Tsaf. *Journal of Archaeological Science: Reports* 43: 103491. Doi: 10.1016/j.jasrep.2022.103491.
3. Chasan, R., Spiteri, C. and Rosenberg, D. 2022. Dietary continuation in the southern Levant: A Neolithic-Chalcolithic perspective through organic residue analysis. *Archaeological and Anthropological Sciences* 14: 49. Doi: 10.1007/s12520-022-01519-0.

The first article explores the earliest archaeological evidence for the use of bee products in the southern Levant in relation to pottery (Chasan *et al.* 2021). While hints for beeswax exploitation were already identified in the Late Chalcolithic period, *vis-à- vis* a prior residue analysis study (Namdar *et al.* 2009a) and identification of the lost-wax technique in copper metallurgy (*e.g.* Golden 2009), this study indicates that bee products were used in a low frequency throughout the Chalcolithic and already by the Early Chalcolithic phase. Beehive products likely originated from wild bees, and these were used probably as a vessel surface treatment and/or as part of the diet. Alternatives were preferred as wild resources contrasted with the socio-economic system, centered on domesticated resources, controlled production and standardization. Bee products only became a more important part of the economic canon several millennia later (Chasan *et al.* 2021).

The second article focuses on the Middle Chalcolithic site of Tel Tsaf (Chasan *et al.* 2022a). Tel Tsaf is located in the Jordan Valley, and it plays a seminal role in understanding the transition between the Neolithic and Late Chalcolithic lifestyle. Organic residue analysis of 100 pottery vessels and three stone vessels and comparison to the botanical and faunal remains of earlier excavations shows that vessel use was unspecialized; they were used for various meat and plant products. There is however the earliest direct evidence found so far for dairy in the region. Additionally, there is a suggestion for strategic herding based on the fatty acid isotope signatures.

Combined, the data supports the ideas that Tel Tsaf was a liminal site, with a standard 'Neolithic' diet paired with the initial steps toward a reliance on renewable and storable food sources.

The third article marks the synthesis of this study, detailing the results of a diachronic and interregional analysis of ceramic utilization in the late prehistory of the southern Levant (Chasan *et al.* 2022b). Organic residue analysis was applied to 226 pottery vessels and three stone vessels from sites dating from the Pottery Neolithic to the Late Chalcolithic. The results when integrated with the related botanical and faunal remains and material culture indicate that there was a uniform diet throughout the entire sequence, despite notable socio-economic developments (including the introduction of pottery and later copper metallurgy). Regardless of chronological, cultural and environmental variation, the diet included primarily meat, originating mainly from domestic ruminants, and cultivated vegetal resources. Minor variation is seen in the use of dairy products, which were introduced during the Middle Chalcolithic period and used only in semi-arid regions. Wild plant resources were used minimally throughout the sequence, indicating that even after the introduction of the Neolithic, wild resources were still valued, likely for task-specific roles. The overarching dietary uniformity suggests that the time span represents one economic continuum, punctuated by distinct cultural entities with a shared dietary tradition that was not altered by the socio-economic processes characteristic of this time span.

These articles are linked methodologically, and they revolve around different aspects of economic behavior during the Pottery Neolithic and Chalcolithic periods. They detail the use of varying plant and animal products, while showing an almost direct continuation of the Neolithic diet. There are rare signs of increasing economic complexity, primarily within the botanical remains. At the same time, these studies show the use of previously unidentified wild plants and beeswax and reinforce the use of dairy starting in the Chalcolithic period.

To supplement these articles, this dissertation is presented. The dissertation comprises the following chapters, in addition to this Chapter 1. Chapter 2 details the theoretical and archaeological background as well as the research questions, and Chapter 3 details the methodological background. Following this, Chapter 4 describes the methodology applied in extracting and analyzing the lipids from the ceramic and stone vessel samples. Chapter 5 lists the articles published as part of this PhD. The information specified includes the article title, journal information, publication date and all related documents. Following this, Chapter 6 presents a

discussion and synthesis of the results. The dissertation concludes with several appendices that provide additional data on the analyzed archaeological sites, pottery and lipids.

Chapter 2. Food ways in the southern Levant

2.1. Food ways

Food is a fundamental aspect of self-identification and human interaction, part of daily practice. What people eat and how they eat results in part from individual decisions made within a complex of social contexts. Therefore, food choices may relate to social factors such as status, ethnicity, gender and religion, and these choices can unite and divide groups of people (Camp 1982; Smith 2006; Twiss 2012). The archaeology of food ways studies how foods were selected, produced, stored, processed, served and consumed as well as the greater picture framing these practices – culinary traditions, food production, economy and environmental regulations (*e.g.* Bray 2003; Costin and Earle 1989; D'Altroy and Earle 2002; Gumerman 1997; Pfalzner 2002; Smith 2006; Twiss 2012; Welch and Scarry 1995; Yasur-Landau 2005).

The mechanisms behind foodways change as social complexity rises. In a village level society, dietary choices reflect economic and social concerns, but in more complex stratified economies and societies, the role of the 'political economy' increases, with food resource management used to control and legitimize power (Costin and Earle 1989). Further, there is increased emphasis on the production of surplus and storable resources using intensification processes; the surplus can be used to support various institutions and specialists (D'Altroy and Earle 2002; Zeder 1988), and they can be converted into social currency (Twiss 2012; Welch and Scarry 1995). These behaviors are commonly associated with the introduction of urbanization in the Levant (Fall *et al.* 2002; Zeder 1988), but the foundations for this are present well beforehand.

2.2. The 'Neolithic Revolution'

In the southern Levant, the Neolithic Revolution marks the starting line in the transition toward complex societies. During this time span, there was a gradual transition from hunting and gathering to a mixed subsistence economy based on domesticated plants and animals (*e.g.* Asouti and Fuller 2012; Bar-Yosef 1998; Bar-Yosef and Belfer-Cohen 1989; Kuijt and Goring-Morris 2002; Moore 1982; Wright 1994). The pathway toward domestication was multi-trajectory, varying regionally (Moore 1982). Why these changes occurred is debated; however, the transition was likely multi-causal (Zeder 2015).

The earliest domesticates included staple plant foods – wheat (*Triticum*) and barley (*Hordeum volgare*). While some (Bar-Yosef and Meadow 1995:66–67) argued that cereals were domesticated as early as the Pre-Pottery Neolithic A (*ca.* 11,700–10,500 cal. BC; Kuijt and Goring-Morris 2002:

table 1), the domestic status was only identified with certainty in the Pre-Pottery Neolithic B (*ca.* 10,500–8,700 cal. BC; Kuijt and Goring-Morris 2002: table 1) based on the high frequency of spikelet forks with rough disarticulation scars and evidence of broad kernels (Zeder 2011; Zohary *et al.* 2012:1). Cereals were quickly popularized because they thrive on open ground, complete their life cycle in less than one year and have high nutritional value (Zohary *et al.* 2012:20). Emmer wheat (*Triticum turgidum*) is well adapted for irrigation, so it can survive in hotter irrigated lowlands, while einkorn wheat (*Triticum monococcum*) is better suited for dry cultivation in cool settings (Fall *et al.* 2002). Barley is also multi-functional, used in beer production and as animal feed (Zohary *et al.* 2012:51–52). These cereals became the backbone of the subsistence economy because they can be stored (*e.g.* Garfinkel *et al.* 2009; Genz 2003; Golani and Yannai 2016; Mazar 2001), allowing for surplus, the accumulation of wealth and the formation of a safety net that could be exchanged as a commodity (*e.g.* Kuijt 2009; Winterhalder and Goland 1997).

Pulses were also an essential domesticate. These were attractive because they add nitrogen to the soil, so by practicing crop rotation or by mixing pulses with cereals, the soil maintains a higher level of fertility. The pulses themselves are nutritionally rich, including high amounts of protein. The earliest domesticated pulses included lentils (*Lens culinaris*), peas (*Pisum sativum*) and chickpeas (*Cicer arietinum*) (Zohary *et al.* 2012:75–99). Identification of domestication is impaired by the morphometric similarities of wild and domesticated forms and their general morphometric variability (*e.g.* Weiss and Zohary 2011; Zeder 2011).

In tandem, a set of animals was domesticated by the Pre-Pottery Neolithic B. Goats (*Capra hircus*) were domesticated first, followed shortly thereafter by sheep (*Ovis aries*) (Kolska Horwitz *et al.* 1999; Zeder 2011). Cattle (*Bos taurus*) were likely domesticated in the late Pre-Pottery Neolithic B or the Pre-Pottery Neolithic C (*ca.* 8,600–8,250 cal. BC; Kuijt and Goring-Morris 2002: table 1). This was demarcated by a sudden increase in their frequency (Kolska Horwitz and Ducos 2005; Kolska Horwitz *et al.* 1999). Pig (*Sus scrofa*) domestication occurred at earliest during the Pre-Pottery Neolithic C; this was supported by the increased frequency and young kill-off pattern of pigs (*e.g.* Galili *et al.* 1993). Domestication however is not definitive until later periods (Haber and Dayan 2004). The domesticated animals were likely exploited initially only for their meat, bone and hide (Greenfield 2010; Sherratt 1983). This so-called Neolithic revolution culminated with the Pottery Neolithic (6,400–5,800 cal. BC; Garfinkel 2009: table 14.1) and the introduction of pottery. Pottery was considered a significant innovation because ceramic vessels could be easily produced and fulfill various household necessities such as storage, food preparation, cooking and serving

(e.g. Brown 1989; Garfinkel 2019:2, 4; Gibbs 2105; Rice 1999). An entire set of ceramic vessels was introduced during this time span, with various open and closed forms represented and specific decoration styles (Garfinkel 1999:16–103).

2.3. The secondary product revolution and the introduction of the Mediterranean diet

The secondary product revolution theory (Sherratt 1981, 1983) states that only several thousand years after initial animal domestication people started exploiting animals for their secondary products – milk, wool, dung and labor. The secondary product revolution was an economic and social insurgency that allowed people to expand into less desirable areas and form plough- or pastoral-based economies (Sherratt 1981). Some question its status as a revolution, noting earlier indicators, which suggest that secondary products were introduced gradually (*e.g.* Chapman 1982; Greenfield 2017), and this has become one of the dominating research themes in the study of early agricultural communities.

These secondary products are beneficial because they are renewable resources, each with their own benefits (*e.g.* Redding 1981:107; Sherratt 1981). A milk-based economy is more efficient in terms of protein and energy output in comparison to a meat-based economy. Milk also provides calcium, sugar and vitamin D (Davis 1987:155; Greenfield 2010; Redding 1981: table IX-2). Milk is further beneficial because it can be converted into storable and transportable products such as cheese and yogurt. These products are easier to digest than raw milk because the lactose is broken down (Davis 1987:155; Simoons 1970). Wool is also a significant resource because it offers a new raw material for fabric production with unique properties. Dung, while commonly considered waste, can also be viewed as a resource. It can be used for fuel (*e.g.* Lancelotti and Madella 2012) and as manure, improving agricultural yields (*e.g.* Jones 2012). Animal labor also plays a key role in boosting the economy. Plowing increases production and allows poorer soils to be cultivated by preventing the loss of moisture, and pack and drought animals increase long distance trade (Sherratt 1980, 1981).

Identifying milk, wool and animal labor in the archaeological record is difficult because in contrast to animal bones and even animal dung, evidence for these secondary products preserves poorly. However, the economic strategy can be identified in part based on the animal kill-off pattern. Theoretical models show that in a meat-based economy, most animals are butchered after reaching full size near the end of immaturity (Payne 1973). In a milk or wool-based economy, a large portion of animals is killed off in adulthood right before milk and wool production decline at four to seven years of age (Davis 1987:158). Further, in a milk-based economy, surplus males are killed off at a

young age even before maturity, while the females are kept as breeding stock, and in a wool-based economy, there is less of a gender bias (Payne 1973; Sherratt 1981). Wool production can also be supported by light spindle whorls that are complementary to the spinning of fine delicate wool fibers (Barber 1991:52). Animal labor is commonly identified through bone pathologies that result from the pressure of labor (*e.g.* Armour-Chelu and Clutton-Brock 1982; Baker and Brothwell 1980; de Cupere *et al.* 2000; Davis 1987).

In tandem, the Mediterranean diet also played a key role in changing food ways. The Mediterranean diet is characterized by a high consumption of vegetables, a moderate intake of wine, fish, poultry and dairy and a low consumption of meat. Olive oil is the main fat source (Hu 2003; Ortega 2006; Trichopoulou and Vasilopoulou 2000; Willet 2006). Each element in the diet has health benefits that work in tandem to create greater effects (Ortega 2006). The diet is renowned for its ability to lower the overall mortality rate and lessen the risk of heart disease (Hu 2003; Ortega 2006; Willet 2006).

Olives are at the heart of this diet, and they have a long history in the Mediterranean and southern Levant. In the Old Testament, Israel is described as a land of olive oil (Deuteronomy 8:8), and this emphasis is reinforced by other ancient texts (Bennett 1958; Ventris and Chadwick 1973:133, 272–273). Olives are economically important because they can be processed, stored and traded (Genz 2003; Salavert 2008), allowing for surplus and the accumulation of wealth. Olives can be transformed into oil or pickled. In addition, olive wood and *jift*, a mixture of the olive pulp and crushed olive stones formed after olive pressing, can be used for fuel (Genz 2003; Liphschitz *et al.* 1991; Rowan 2015; Salavert 2008; Vossen 2007), and *jift* can be further used as fertilizer and livestock fodder (Rowan 2015). This increases the monetary intake and decreases the waste from olive cultivation and olive oil production.

The introduction of these food ways is important because both require a greater investment of time and effort and a higher degree of organized production in comparison to a primary product-based economy. After milking the animals and picking the olives, additional labor is required to process these into storable resources. Further, it requires several generations of selection to breed milk livestock (Davis 1987:156), and olive trees take five to six years to bear fruit after planting (Zohary *et al.* 2012:114). Therefore, the introduction of these food ways required and reinforced a sedentary lifestyle in which people were strongly tied to their land and invested for the future generations.

2.4. Archaeological evidence for food ways in the Chalcolithic southern Levant

The Chalcolithic period in the southern Levant is divided into three phases. The Early Chalcolithic period incorporates the Wadi Rabah culture, which dates to approximately 5,800–5,200 cal. BC (Banning 2007). The culture had a northern orientation, extending south only to the Shephelah (Gopher and Gophna 1993: fig. 15). The Middle Chalcolithic period ranges from approximately 5,200–4,700 cal. BC (Garfinkel 2009) and is characterized by numerous regional entities including the Qatifian, Besorian, Tsafian, Natzurian and Beth Shean XVIII. These are distinguished geographically and by certain aspects of their material culture (Garfinkel 2009; Gilead 2007, 2009). The Late Chalcolithic period dates to *ca.* 4,500–3,800 cal. BC (Gilead 1994; Joffe and Dessel 1995). The dominant culture is the Ghassulian, but there are other smaller regional entities differentiated by distinct features within their material culture (Levy 1986; Lovell 2001:51). This time span was thoroughly studied because of the significant changes observed in the social organization, settlement patterns, cult and mortuary practices, subsistence economy and craft production (*e.g.* Gilead 1988; Levy 1986; Rowan and Golden 2009), including the introduction of metallurgy (*e.g.* Golden 2009). The economy and dietary patterns of the Chalcolithic can in part be elucidated by the faunal, botanical and material culture remains.

2.4.1. Faunal remains

During the Early Chalcolithic period, there was a decreased reliance on hunting and gathering and an increased reliance on domesticated animals (Gopher 2012a:1553–1554; Gopher and Gophna 1993). While there was an emphasis on caprines (*e.g.* Davis 2012; Khalaily *et al*. 2016: table 6; Kolska Horwitz 2002; Kolska Horwitz *et al.* 2006), there was also a minor utilization of cattle and pigs (*e.g.* Davis 2012; Kolska Horwitz 2002; Kolska Horwitz *et al.* 2006), the latter domesticated by this time span. This pattern continued in the Middle Chalcolithic and Late Chalcolithic periods (*e.g.* van den Brink *et al.* 2016; Grigson 1987:221–235, 1995; Hill 2011:108; Kolska Horwitz 2007; Milevski *et al.* 2015: table 7; Nativ *et al.* 2014: table 3). These animals were exploited mainly for meat (Namdar *et al.* 2021), but during the Middle Chalcolithic period, bone pathologies at one site suggest the use of cattle for labor (Hill 2011:148–156). During the Late Chalcolithic period, bone pathologies show that people continued using animal labor (Grigson 1995:402, 2006:231; Price *et al.* 2013), and the prevalence of adult sheep and goat livestock at some sites suggests dairy or wool exploitation (*e.g.* Grigson 1987:225, fig. 7.6, 1995:389, fig. 10.4; Kolska Horwitz 2007; al-Zawahra 2008:437).

Some (Levy 1983) use this data to argue for the application of a specialized pastoral economy in the northern Negev during the Late Chalcolithic period. Analysis indicates two settlement types

with different subsistence strategies. Permanent village settlements were in the arid foothill zones; these communities cultivated the soils near the wadis and used irrigation systems. Such sites relied on a mixture of caprine, cattle and pigs (Grigson 1989; Levy 1983). Seasonal sites were located around springs on the coastal plain. Mobile herders or pastoralists who were specialized segments of the permanent villages used these and relied exclusively on sheep and goats (Grigson 1989; Levy 1983).

2.4.2. Botanical remains

Botanical remains from various Wadi Rabah sites indicate that communities relied primarily on cereals and pulses; a preference for emmer wheat and barley was observed (Kislev and Hartmann 2012: table 32:1). This preference is repeated at the Middle Chalcolithic site of Tel Tsaf (Graham 2014) and the Middle Chalcolithic layers at Teleilat Ghassul (Meadows 2015: table 5.6), although further botanical studies from additional sites are needed to characterize the plant products used during the Middle Chalcolithic as whole. During the Late Chalcolithic period, the botanical remains indicate that the primary agricultural products were emmer wheat and two-row barley, with lentils exploited at a lower frequency (Kislev 1987:252–254; Lovell *et al.* 2006; Zaitschek 1959, 1961; Zohary *et al.* 2012:23–59, 77–89). There are some signs of intensification in crop cultivation, using irrigation and plowing (*e.g.* Hill 2011:148–156; Marom 2011:92–93; Rosen and Weiner 1994), which can generate surplus and allow settlement in areas that could not previously be occupied.

Olive stones were uncovered at several Early Chalcolithic (*e.g.* Galili *et al.* 1989, 1997; Kislev 1994–1995; Kislev and Hartmann 2012: table 32:1; Namdar *et al.* 2014), Middle Chalcolithic (Graham 2014; Meadows 2005: table 5.6; Rosenberg *et al.* 2021) and Late Chalcolithic sites (*e.g.* van den Brink *et al.* 2001; Epstein 1998:24–48, 110, 123; Liphschitz 2004: table 10.2; Liphschitz *et al.* 1996; Lovell *et al.* 2006; Meadows 2005: table 5.6; Zaitschek 1961). These were likely exploited for oil. Evidence includes crushed olive stones with smoothed edges, broken in antiquity and worn down by post depositional processes (Epstein 1993; Galili *et al.* 1997; Lovell *et al.* 2006: table 2; Neef 1990:298). Pickling of olives is suggested at a few sites (Galili *et al.* 2021). During the Early Chalcolithic period, wild olives were likely exploited. This is supported by the high degree of olive stone morphometric variation, which is characteristic of wild forms (Dighton *et al.* 2017; Galili *et al.* 2021; Kislev 1994–1995; Terral *et al.* 2004). However, by the Middle and Late Chalcolithic, olive domestication and olive horticulture are supported by the identification of olive stones and olive wood at sites in regions where wild olives could not grow without management (*e.g.* Langgut and Garfinkel 2022; Lovell *et al.* 2006; Meadows 2005; Zaitschek 1961).

Domestication and horticulture are reinforced in the Late Chalcolithic period by the decreased metric variation of olive stones at some sites (Dighton *et al.* 2017; Meadows 2005:160, appendix f). This may suggest multiple independent and non-contemporaneous domestication events.

In addition, during the Chalcolithic period, there is some evidence for other fruits. Figs were identified at each stage of the Chalcolithic (Kislev and Hartmann 2012: table 32:1; Kislev 1987: table 9.1; Meadows 2015: table 5.6). In the Late Chalcolithic period, a wider array of fruits was identified alongside some vegetables – dates, walnuts, pomegranates, almonds, onions and garlic (Lovell *et al.* 2006: table 2; Melamed 2002; Zaitchek 1961; Zohary and Spiegel-Roy 1975). These were found at limited sites, and they may represent later intrusions.

2.4.3. Material culture

The pottery forms displayed during the Chalcolithic period are typified by a variety of bowls and storage jars (Garfinkel 1999). However, several specialized forms were noted in low frequencies during the Early and Middle Chalcolithic periods. These include platters, chalices, spouted vessels, strainers and proto-churns (van den Brink *et al.* 2021a; Epstein 1984: fig. 3:5; Garfinkel 1999:111, 123, 166, 167, fig. 68; Gopher and Eyal 2012: figs. 10.73:6, 10.75:14, 10.79; 10.83:8, 10.86; Gophna and Sadeh 1988–1989: fig. 10:10; Gustavson-Gaube 1986: fig. 15:55; Leonard 1992: pl. 5:3).

These forms became more common during the Late Chalcolithic period. Churns in particular were found at many sites and were potentially used in dairy processing (*e.g.* Burton 2004:126–127, 214–215; Garfinkel 1999:254–257; Gilead and Goren 1995:165–171; Kaplan 1954; Scheftelowitz 2004:45). This is suggested based on their similarities to ethnographic churns produced from animal hide (Kaplan 1954). Alternatively, the shape would also be ideal for transport (Commenge 2006:444). Cornets, horned shaped goblets and spouted vessels were also uncovered (*e.g.* Burton 2004:118–120, 193–194, 203, 222; Epstein 1998:164; Garfinkel 1999:219, 226, 237; Gilead and Goren 1995:158–163; Scheftelowitz 2004:43). The spouted vessels may have been used in olive oil storage (Epstein 1993). Strainer vessels were also found, the strainer incorporated most commonly in churns and jars (van den Brink *et al.* 2021a).

The ground stone tools industry is characterized in part by a predominance of small-scale lower and upper grinding stones. These were made from primarily local raw materials (*e.g.* Bekker 2017: tables 5, 8, 31; van den Brink *et al.* 2016; Gilead 1995:326–330; Gopher 2012b:1056–1059, 1073–1076; Ilan *et al.* 2015:86–87; Milevski *et al.* 2015; Nativ *et al.* 2014; Rosenberg 2011:202–264;

Rosenberg *et al.* 2017; Rowan *et al.* 2006:578–580). During the Late Chalcolithic period, there was a high frequency of flat-based and fenestrated pedestal V-shaped basalt vessels (Rowan 1998). While their function is unclear, internal polish on some vessels suggests that the vessels were used in processing activities (Hruby *et al.* 2021). In addition, throughout the Chalcolithic period, there are few spindle whorls used for textile production (*e.g.* Bekker 2017:60–61, table 19; Rosenberg 2011: table 8.1; Rowan *et al.* 2006:592–594).

2.5. Research objectives

This dissertation investigates these changes in dietary practices in the late prehistoric southern Levant using organic residue analysis to identify residues preserved in pottery and stone vessels. While the food ways are well documented by faunal and botanical remains, organic residue analysis will provide a new layer to our understanding of the archaeological record by aiming to identify food products that do not commonly preserve in the macro-analytical record and how various vessel types and tool forms were used in food preparation.

Moreover, dietary preferences may be linked to cultural and environmental factors (*e.g.* Bray 2003; Costin and Earle 1989; Gumerman 1997; Mintz and du Bois 2002; Smith 2006; Twiss 2012). Considering this, the research documents diachronic and regional variation in the way food was selected, produced, stored, cooked and consumed and delves into why certain changes in food ways occurred, while others remained consistent, incorporating additional external data into the analysis. This includes faunal and botanical remains, material culture and climatic markers. The research also elucidates on the economic systems and strategies of this time span by discussing why settlements employed specific economic strategies.

Chapter 3. Organic residue analysis

Organic residues are amorphous organic remains that rely on chemical analysis for identification (Heron and Evershed 1993). Organic residue analysis aims to detect and identify lipids using molecular and compound specific carbon isotope analysis (*e.g.* Evershed 2008a; Heron and Evershed 1993). Unlike other organic residues, lipids often preserve due to their strong chemical structure and hydrophobic nature (Debono Spiteri 2012:70; Heron and Evershed 1993).

The most commonly studied archaeological material carrying organic residues is pottery. The original contents can preserve as visible crusts or, more commonly, as residues absorbed within the porous ceramic matrix (Evershed 2008a; Heron and Evershed 1993). Non-porous lithics are less likely to retain a chemical signature due to their impermeability, although some studies on stone tools were successful (*e.g.* Buonasera 2007, 2013, 2016; Namdar *et al.* 2009b; Quigg *et al.* 2001).

The versatility of organic residue analysis is wide ranging. It can identify a variety of substances, including ruminant, porcine and fowl adipose, dairy fats, fish oils, plant oils, wine, resins and waxes (*e.g.* Colonese *et al.* 2017a, b; Craig *et al.* 2007; Debono Spiteri *et al.* 2016; Evershed *et al.* 2008; Heron *et al.* 1994; Pecci *et al.* 2013) through comparison to lipid biomarkers and carbon isotope signatures of modern references. This includes food products that preserve poorly in the macro-archaeological record, such as leafy plants and liquids. The results can elucidate significant economic trends, such as the shift from hunter-gather-fisher economies to agrarian economies (Craig *et al.* 2007, 2011) and the introduction of dairying (*e.g.* Copley *et al.* 2003; Cramp *et al.* 2014; Debono Spiteri *et al.* 2016; Evershed *et al.* 2008). Organic residue analysis also provides direct information for what the studied vessels were used for (*e.g.* Evershed *et al.* 2003; Salque *et al.* 2013) and can identify mixtures of products (Fernandes *et al.* 2018; Isaksson and Hallgren 2012; Miller *et al.* 2020). This methodology was so far minimally applied to protohistoric Levantine pottery, and it was used to suggest the identification of meat, olive oil, beeswax and, in the northern Levant, dairy residues (*e.g.* Burton 2004:566–621; Evershed *et al.* 2008; Gregg *et al.* 2009; Namdar *et al.* 2009a, 2014). These results were achieved using Gas Chromatography-Mass Spectrometry (GC-MS) and Gas Chromatography-combustion-Isotope Ratio Mass Spectrometry (GC-c-IRMS)

3.1. Gas Chromatography-Flame Ionization Detector and Gas Chromatography-Mass Spectrometry

Gas Chromatography (GC) separates the molecules in a sample based on their weight, functional groups and molecular shape. The molecules pass through the GC capillary column carried by a gas in what is defined as the mobile phase. The stationary phase is a thin layer of a liquid lining the interior of the capillary columns. The rate at which each molecule moves through the mobile phase is determined by how much the molecules interact with the stationary phase (Malainey 2011:436–438; Pollard *et al.* 2007:137–138, 142–146).

When the molecules elute, they pass through a flame ionization detector (FID), which quantifies the relative abundance. These results are then plotted on a chromatogram, which compares molecule relative abundance to column retention time. Molecules have a consistent retention time because they interact in set ways with the stationary phase. This allows for molecular identification of unknown molecules through comparison to the retention times of known molecules. The GC also identifies the relative quantity of a given molecule based on the peak area.

Mass Spectrometry (MS) is coupled with GC to enhance identification. In a mass spectrometer, the separated molecules are bombarded with high-energy electrons; this removes an electron and generates a positive molecular ion fragment. The fragments' mass to charge ratios (*m*/*z*) are plotted against their intensity, forming a characteristic molecule fingerprint. The molecule is identified based on specific fragments that represent the molecule's functional groups and chain length or more simply by comparing the mass spectrum to the fragmentation pattern of known molecules (Malainey 2011: table 14.1; Pollard *et al.* 2007:174–177). GC-MS can therefore separate and identify the specific components preserved within an unidentified residue.

3.1.1. Application of GC-MS to identify residues and cooking processes

Based on the components identified through GC-MS, the preserved residue may be discerned. The sections below detail how specific products and cooking techniques relevant to the archaeological record are identified.

3.1.1.1. Terrestrial animal fat

Terrestrial animal fat residues are comprised mainly of saturated fatty acids, with a predominance of palmitic ($C_{16:0}$) and stearic acid ($C_{18:0}$). Generally, stearic acid is more abundant than palmitic acid (Copley *et al.* 2005), although the ratio varies based on species, and this is complicated by degradation. In addition, animal fat is indicated by cholesterol and cholesterol degradation markers (deMan 1999:54; Evershed 1993; Hammann *et al.* 2018). Using cholesterol as a biomarker is

problematic because it can also result from post-excavation handling. In such a case, cholesterol is usually paired with squalene (Archer *et al.* 2005), allowing the contamination to be differentiated.

3.1.1.1.1. Ruminant fat

Ruminant fat contains odd-numbered long-chain fatty acids – pentadecanoic acid ($C_{15:0}$), heptadecanoic acid ($C_{17:0}$) and nonadecanoic acid ($C_{19:0}$), sometimes with *iso* and *anteiso* branches (Doreau and Chilliard 1997; Regert 2011). These are synthesized through bacterial activity in the rumen (Doreau and Chilliard 1997). Odd-numbered fatty acids are formed by reducing the chain length of fatty acids through alpha-oxidation of propionyl-CoA (Doreau and Ferlay 1994). Branched-chain fatty acids similarly form from isobutyrate and isovalerate (Doreau and Chilliard 1997). In addition, $C_{18:1}$ appears in multiple isomers, with double bonds at the 9, 11, 13, 14, 15 or 16 positions (Evershed *et al.* 1997).

Ruminant dairy fat has the same markers but is further identified by the presence of short- and mid-chain fatty acids ($C_{4:0}$–$C_{14:0}$) (Dudd and Evershed 1998; Gunstone and Harwood 2007: table 2.61). These are produced through *de novo* synthesis in the mammary gland (Gunstone and Harwood 2007:94). The short-chain fatty acids are more susceptible to hydrolysis because of the reduced steric effects at the ester linkages in triacyclglycerols, and they are more water soluble (Evershed *et al.* 1999, 2002). Once degraded, this may result in a signature similar to ruminant adipose.

3.1.1.1.2 Porcine fat

Degraded porcine adipose has no key biomarkers. However, in contrast to ruminant adipose, palmitic acid is more abundant than stearic acid (Regert 2011). Porcine adipose may be further distinguished from ruminant fat by the presence of exclusively $C_{18:1}$ *cis*-9 (Evershed *et al.* 1997). Identification based on $C_{18:1}$ is problematic because it is also highly characteristic of plant products (Pollard and Heron 2008:385, table 11.2).

3.1.1.2. Aquatic resources

Unlike terrestrial animal fats, fish oil has minimal saturated fatty acids, consisting primarily of $C_{16:0}$ (deMan 1999:41; Regert 2011). The key fraction of the fatty acids is mono- and polyunsaturated C_{20} and C_{22} with up to six double bonds and multiple isomers of each unsaturated fatty acid (Cramp and Evershed 2014:321; deMan 1999:37; Gunstone and Harwood 2007:103l; Heron and Craig 2015). Eicosapentaeonoic acid ($C_{20:5}$ *n*-3) and docosahexaeonoic acid ($C_{22:6}$ *n*-3) are most common (Cramp and Evershed 2014:321). These however are highly susceptible to degradation. To identify fish residue with certainty, specific biomarkers must be present. These include vicinial dihydroxy

acids, isoprenoid acids and ω-(*o*-alkylphenyl)alkanoic acids (APAA; Cramp and Evershed 2014:322–325). Vicinal dihydroxy acids are the oxidation products of unsaturated fatty acids; for aquatic resources, dihydroxy acids with C_{16}–C_{22} are expected (Cramp and Evershed 2014:322–323). Isoprenoid structures include phytanic acid (synthesized from phytol, an alcohol moiety of chlorophyll in algae), pristanic acid and 4,8,12-trimethyltridecanoic acid. Of these, only 4,8,12-trimethyltridecanoic acid is found exclusively in aquatic environments, so it is particularly indicative (Ackman and Hansen 1967; Cramp and Evershed 2014:323; Heron and Craig 2015). Further, APAAs with C_{16}–C_{22} are formed upon heating unsaturated fatty acids common in fish oil (Cramp and Evershed 2014:323–325; Hansel *et al.* 2004; Heron and Craig 2015).

3.1.1.3. Plant oils

Identifying plant lipids is problematic because plants have lower lipid yields than animal fat, so they are easily masked and are less likely to preserve (Debono Spiteri 2012:74, 128–129; Evershed 2008b). Plant oils are comprised primarily of unsaturated fatty acids, with C_{18} fatty acids ($C_{18:1}$, $C_{18:2}$ and $C_{18:3}$) usually the most common (Pollard and Heron 2008:385, table 11.2). Seed oils also commonly contain gondoic acid ($C_{20:1}$ *n*-11) and erucic acid ($C_{22:1}$ *n*-13) (Gunstone and Harwood 2007: table 2.26). When heated, these unsaturated fatty acids can produce APAAs with C_{18} and more rarely C_{20} (Bondetti *et al.* 2021). A smaller percentage of the total lipid content is formed of even-numbered long-chain saturated fatty acids ($C_{14:0}$–$C_{36:0}$). Among these, $C_{16:0}$ followed by $C_{18:0}$ is the most abundant (Copley *et al.* 2005; Scrimgeour and Harwood 2007: table 1.14; Tulloch 1976:245). Plant products are also identified by phytosterols: β-sitosterol, campesterol and stigmasterol (deMan 1999:51; Evershed 1993). In addition, plants are characterized by odd-numbered alkanes, ranging from C_{21}–C_{35}, with C_{29} and C_{31} the most common, and even-numbered alcohols ranging from C_{22}–C_{34} (Scrimgeour and Harwood 2007: table 1.14). Using the *n*-alkane and alcohol profiles, more details can be gleamed about what type of plant the lipids originated from. C_4 plants are unique, and their *n*-alkanes and alcohols maximize at C_{33} and C_{32} respectively (Rommerskirchen *et al.* 2006). Aquatic plants and halophytes also have a unique signature; unlike most plants, these contain a high amount of the C_{23} and C_{25} *n*-alkanes (Eley *et al.* 2014: SI table 1; Ficken *et al.* 2000). Additional indicators of plant products are terpenoids – cyclic compounds found in plant resins. Specific suits of terpenoids are linked to different wood sources (*e.g.* Mills and White 1977; Otto and Wilde 2001)

3.1.1.3.1. Olive oil

Degraded olive oil is composed primarily of unsaturated fatty acids, with oleic acid ($C_{18:1}$ *cis*-9) the most abundant. There are also high amounts of palmitic, stearic and linoleic acid ($C_{18:2}$ *cis, cis*-9,12) (Paiva-Martins and Kiritsakis 2017:83–84). These may oxidize into characteristic dicarboxylic acids, hydroxy acids and dihydroxy acids (Passi *et al.* 1993; Regert *et al.* 1998).

3.1.1.3.2. Cereals

Cereals are difficult to identify because they contain trace quantities of lipids (Colonese *et al.* 2017a). Cereal fatty acids are composed primarily of $C_{16:0}$, $C_{18:0}$, $C_{18:1}$ and $C_{18:2}$ (Hammann and Cramp 2018; Hammann *et al.* 2019). The main phytosterols are β-sitosterol and campesterol (Hammann *et al.* 2019). Key markers are alyklresorcinols – 1,3-dihydroxy-5-alkylbenzene molecules with an odd number of carbons (C_{17}–C_{25}) (Ross *et al.* 2003). While generally each cereal contains the entire suite of alyklresorcinols, the cereals contain them in differential frequencies, allowing for identification. Barley for instance has a high proportion of C_{25}, while wheat has a high proportion of C_{21} (Hammann and Cramp 2018; Hammann *et al.* 2019). In addition, cereals have tocopherols and tocotrienols. Barley is unique, with predominantly the γ-isomer; in all other cereals, the β-isomer is dominant (Hammann *et al.* 2019).

3.1.1.3.3. Legumes

Legumes are difficult to identify due to their low lipid yields and high amount of unsaturated fatty acids, which easily degrade. Up to 70% of the fatty acids are polyunsaturated, with a predominance of $C_{18:2}$. Monounsaturated fatty acids form up to 40% of the fatty acids. This is predominated by $C_{18:1}$. The main saturated fatty acid is $C_{16:0}$ (Caprioli *et al.* 2016).

Chickpeas have notably high quantities of linoleic acid and lower quantities of linolenic acid ($C_{18:3}$ *cis*-9,12,15) (Caprioli *et al.* 2016). Chickpea processing by soaking or germination further increases this difference by increasing $C_{18:2}$ and decreasing $C_{18:3}$ (Vasishtha and Srivastava 2012). The primary sterol in chickpeas is β-sitosterol. Chickpeas also have a high amount of α-tocopherol (Jukantil *et al.* 2012).

Lentils contrast, with a less distinct difference in the quantity of linoleic and linolenic acid (Caprioli *et al.* 2016; Paucean *et al.* 2018). Further, lentils have phytic acid and are high in polyphenols, with a predominance of tannin. The main sterol is β-sitosterol (Faris *et al.* 2012), and the main tocopherol is γ-tocopherol, with only trace amounts of δ- and α-tocopherol. Lentils also have carotenoids. The major carotenoids are trans-lutein and trans-zeaxanthin (Zhang *et al.* 2014).

3.1.1.3.4. Garlic, mustard seed oil and sulfur-rich plants

Garlic, mustard seed oil and likely other sulfur-rich plants are characterized by a unique biomarker – sulfur-heterocyclic fatty acids (SHFAs) (Dembitsky *et al.* 2007; Eibler *et al.* 2017; Wijesundera and Ackman 1988). These are alkanoic acids with a tetrahydrothiophene ring in their aliphatic chain. Different SHFA 'species' are identified based on the chain length and the placement of the tetrahydrothiophene ring (Eibler *et al.* 2017). Because of the structural similarities, in a chromatogram one peak represents all positional isomers with a shared chain length. The following SHFA families were identified: SHFA1 (C_{18}), SHFA2 (C_{20}), SHFA3 (C_{22}) and SHFA4 (C_{24}) (Eibler *et al.* 2017). Mustard seed oil can be further differentiated based on the presence of $C_{20:1}$ *n*-11 and $C_{22:1}$ *n*-13 (Colombini *et al.* 2005), while *allium* plants, like garlic, may be further characterized by the presence of the ketone 16-hentriacontanone (Rabah *et al.* 2020; Raven *et al.* 1997).

3.1.1.4. Beeswax

Beeswax is composed of long-chain even palmitate wax esters (WE), with C_{40}–C_{52} and C_{46} the most abundant (Heron *et al.* 1994; Jackson and Blomquist 1976:218–219; Regert *et al.* 2001), and odd-numbered alkanes, with C_{21}–C_{33} and C_{25}, C_{27} and C_{29} the most common (Jackson and Blomquist 1976:216; Regert *et al.* 2001). Within the *n*-alkanes, long-chain *n*-alkanes are preferentially preserved because they have decreased volatility and are more abundant initially (Regert *et al.* 2001). Wax ester degradation products are also frequent – alcohols and palmitic acid. Alcohols are even-numbered and range from C_{20}–C_{36} (Jackson and Blomquist 1976:220; Regert *et al.* 2001). Fatty acids found naturally in beeswax, prior to wax ester hydrolysis, include $C_{14:0}$–$C_{36:0}$, with $C_{24:0}$ and $C_{26:0}$ the most common (Jackson and Blomquist 1976:219–220; Regert *et al.* 2001).

3.1.1.5. Heating

Heating of vessels to high temperatures (over 300–400°C) or prolonged heating to lower temperatures is recognized by the presence of ketones (K) with C_{29}–C_{35}. In particular, when palmitic and stearic acid are heated, 16-hentriacontanone (K_{31}), 16-tritriacontanone (K_{33}) and 18-pentatriacontanone (K_{35}) are formed. While other ketones are found in leafy plant waxes, this combination is exclusively formed from the dehydration and decarboxylation of carboxylic acid salts at high temperatures in the presence of palmitic and stearic acid (Evershed *et al.* 1995; Raven *et al.* 1997). In addition, these ketones are distinguishable from plant ketones as K_{31}, K_{33} and K_{35} are commonly found in a 1:2:1 ratio, and they are asymmetrical unlike most plant ketones (Raven *et al.* 1997). Unsaturated ketones can also form from the pyrolysis of unsaturated fatty acids. So

far, $K_{33:1}$ and $K_{35:1}$ have been identified. These form from the pyrolysis of $C_{18:1}$ with $C_{16:0}$ and $C_{18:0}$ (Evershed *et al.* 1995). Ketones have so far been used to link vessels to cooking (e.g. Nieuwenhuyse *et al.* 2015; Raven *et al.* 1997) and post-firing vessel treatments (Drieu *et al.* 2019).

3.2. Gas-Chromatography-Combustion-Isotope Ratio Mass Spectrometry (GC-c-IRMS)

GC-c-IRMS of lipids preserved within pottery determines the $\delta^{13}C$ value of individual components, providing additional information on the source. The $\delta^{13}C$ values of $C_{16:0}$ and $C_{18:0}$ are typically analyzed in archaeological samples because these are the most commonly encountered fatty acids, they are easily extracted from archaeological samples and their isotopic signals remain unaltered over time (Evershed *et al.* 2002).

Variation of the $\delta^{13}C$ values occurs in animal fats because of differences in animal metabolic pathways. The isotopic signature of non-ruminants directly reflects the diet. The signature of ruminant fat is altered by the biohydrogenation of unsaturated fats in the rumen, forming a more negative value. The signature of ruminant dairy fat is further altered by the synthesis of more enriched saturated fatty acids with up to 16 carbon atoms in the mammary gland (Copley *et al.* 2003). Considering this, different animal fats may be identified by comparing the isotopic signatures of archaeological samples to modern reference fats (Fig. 1). In this process, the modern $\delta^{13}C$ values are corrected for the post-industrial revolution effect on the atmospheric carbon (Friedli *et al.* 1986; Regert 2011).

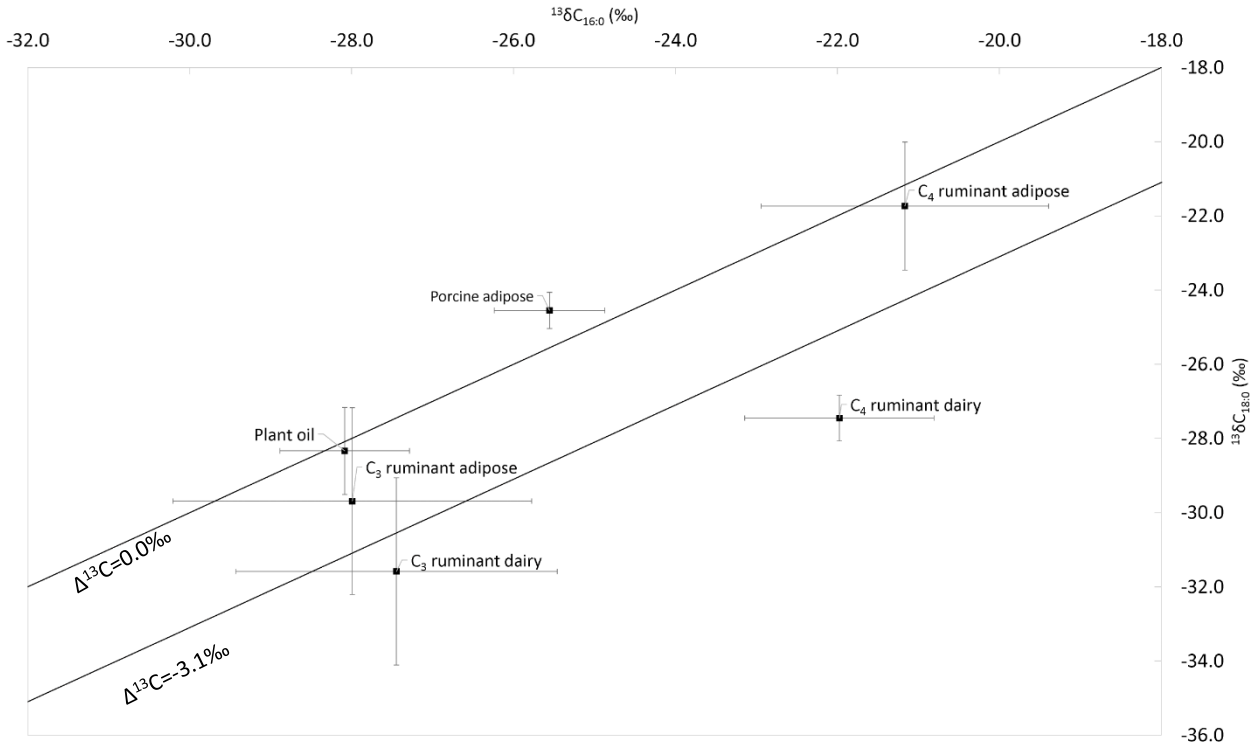

Figure 1. Mean ± 1 s.d. of the $\delta^{13}C_{16:0}$ and $\delta^{13}C_{18:0}$ values for modern references of terrestrial animal fat (Debono Spiteri 2012; Dudd 1999; Gregg *et al.* 2009; Outram *et al.* 2009) and plant oils (almond oil, argan oil, olive oil, moringa oil, sesame oil and walnut oil; Steele *et al.* 2010). The animals originate from the UK (animals raised on a pure C_3 diet), Germany (animals raised on a pure C_3 diet), Italy (animals raised on a pure C_4 diet), Malta (animals raised on a pure C_3 diet and animals raised on a pure C_4 diet), Israel, Jordan and Kazakhstan (animals raised on an unspecified diet). All references had a post-industrial carbon (PIC) correction of 1.14‰ (Friedli *et al.* 1986).

Animal diet (C_3 vs. C_4 plants) also affects the isotope signature. The $\delta^{13}C$ values of C_4 plants are more positive than in C_3 plants (Meier-Augenstein 2002; Tieszen 1991), causing isotope enrichment in the animals who consume these. However, in arid and saline environments, C_3 plants have more positive values as well because as an adaptive strategy to conserve water, they use a modified photosynthetic pathway that results in less alteration to the atmospheric carbon (Meier-Augenstein 2002; Tieszen 1991: table 1). Further, marine resources have enriched values from the consumption of ocean bicarbonates as phytoplanktons (Cramp and Evershed 2014). To control for this, $\delta^{13}C_{16:0}$ is plotted against $\Delta^{13}C$ ($\delta^{13}C_{18:0}$–$\delta^{13}C_{16:0}$) (Fig. 2; Copley *et al.* 2003). Non-ruminant adipose has a $\Delta^{13}C$ ratio greater than 0.0‰, ruminant adipose has a $\Delta^{13}C$ ratio of -3.1 to 0.0‰ and ruminant dairy has a $\Delta^{13}C$ ratio under -3.1‰ (Fig. 2) (Regert 2011; Roffet-Salque *et al.* 2016).

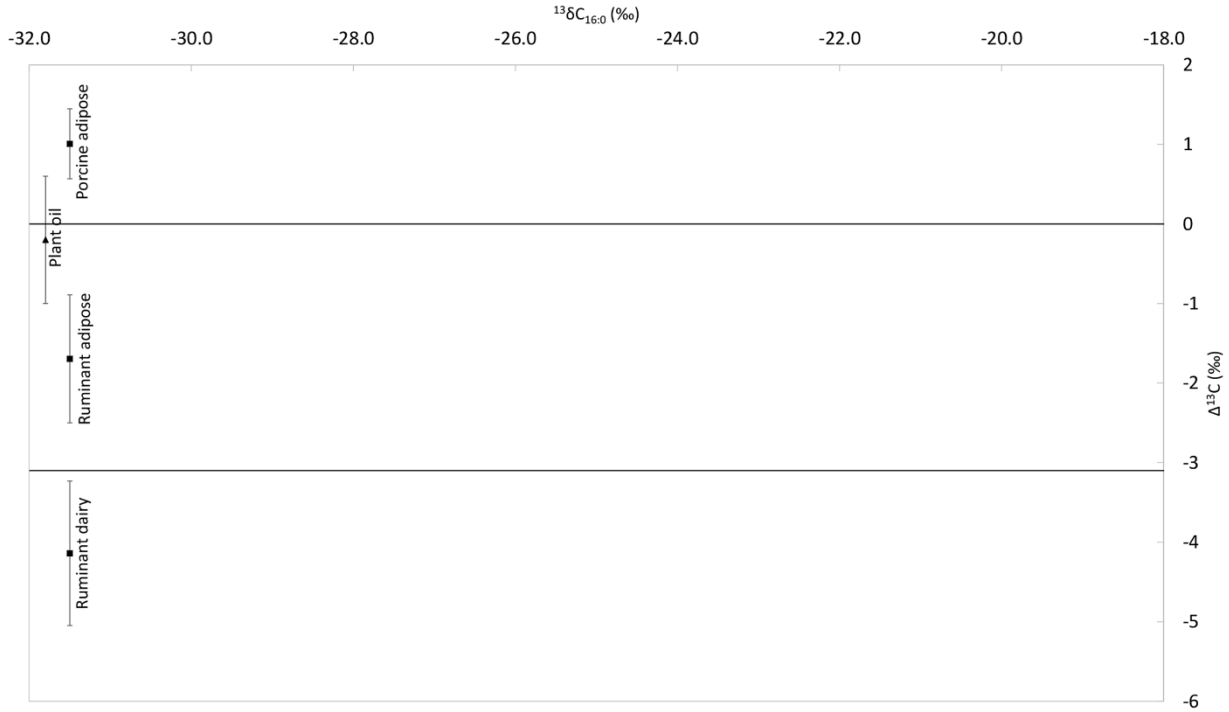

Figure 2. Mean of the $\delta^{13}C_{16:0}$ values and mean ± 1 s.d. of the $\Delta^{13}C$ ($\delta^{13}C_{18:0} - \delta^{13}C_{16:0}$) values for modern references of terrestrial animal fat (Debono Spiteri 2012; Dudd 1999; Gregg *et al*. 2009; Outram *et al.* 2009) and plant oils (almond oil, argan oil, olive oil, moringa oil, sesame oil and walnut oil; Steele *et al.* 2010). The animals originate from the UK (animals raised on a pure C_3 diet), Germany (animals raised on a pure C_3 diet), Italy (animals raised on a pure C_4 diet), Malta (animals raised on a pure C_3 diet and animals raised on a pure C_4 diet), Israel, Jordan and Kazakhstan (animals raised on an unspecified diet). All references had a post-industrial carbon (PIC) correction of 1.14‰ (Friedli *et al.* 1986).

Plants are difficult to differentiate using isotopic analysis as they plot between the ruminant and non-ruminant adipose categories (Figs. 1–2; Steele *et al.* 2010). Further, theoretical models, which are yet to be validated by cooking experiments, predict that when C_3 plants are mixed with milk or ruminant adipose fat, the $\Delta^{13}C$ can shift into the range of ruminant adipose fat and non-ruminant adipose fat respectively (Hendy *et al.* 2018: fig. 5b–c). Because of this, one must employ caution in interpreting isotope results when plant lipid contribution is suspected.

Chapter 4. Method and sample preparation

This section details the selection and analysis procedure undertaken in this study.

4.1. Pottery and ground stone tools selection

In total, 248 pottery fragments were sampled from several archaeological sites (Fig. 3; Appendix A and B). When possible, pottery sherds with evidence of heating (*e.g.* soot marks) were selected for analysis. Three stone vessels were also selected for analysis. Most of the sherds were chosen from ongoing excavations at the Middle Chalcolithic site of Tel Tsaf. A smaller sample of fragments from sites in the northern Negev, Mediterranean coastal plain, Jordan Valley and Galilee was also selected to compare environmental and diachronic variation (Fig. 3). In addition, one Pottery Neolithic site was sampled to gain a pre-Chalcolithic perspective. This includes material from ongoing excavations (Ashqelon Agamim, Tel Bene Beraq (South), Ein Gedi, Fazael 7, Nissim Aloni Street and Tsomet Shoket) and old excavations, for which post-excavation handling must be considered (Sha'ar Hagolan, Neve Yam, Tel Hreiz, Abu Matar, Azor, Bir es-Safadi, Gilat, Peqi'in and Yehud).

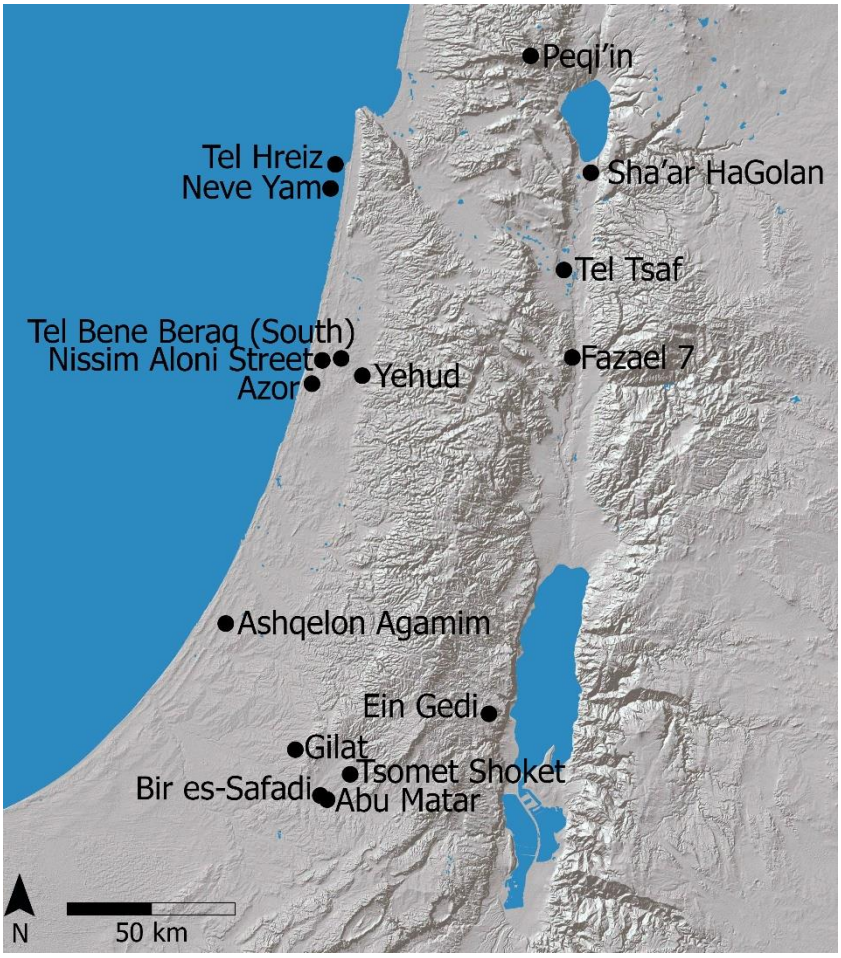

Figure 3. Map of sites analyzed.

4.2. Sampling

Artifacts were sampled at the Zinman Institute of Archaeology, University of Haifa. The following protocol was used:

1. To control for exogenous contamination, the area to be sampled on the vessel interior was lightly cleaned by removing the initial surface layer to a depth of *ca.* 1–2 mm with a Dremel drill using a tungsten bit.

2. Following, *ca.* 2.0 g of powder was sampled from the interior. The powder was collected on sterilized aluminum foil, weighed and transferred to a sterile scintillation vial for analysis.

3. Any adhering crusts were removed using a sterilized scalpel and homogenized. The powder was collected on sterilized aluminum foil, weighed and transferred to a sterile scintillation vial for analysis.

4. The exterior surface and soil from the surrounding context when possible were sampled in the same fashion and tested to control for exogenous and post-depositional contamination. The powder and soil were collected on sterilized aluminum foil, weighed and transferred to a sterile scintillation vial for analysis.

To avoid contamination during sampling and laboratory procedures, the following precautions were followed:

1. Nitrile gloves were always worn.
2. No plastic apparatuses were used.
3. Prior to use, the drill bits were placed in dichloromethane (DCM) and sterilized via sonication in an ultrasonic bath for 20 minutes, repeated three times.
4. Glassware was sterilized at 500°C for 8 hours in a muffle furnace prior to use.
5. HPLC (high-performance liquid chromatography) grade reagents and high purity standards were used.

4.3. Laboratory sample preparation: extraction and derivatization

The samples were extracted and analyzed in the organic residue analysis facilities at the University of Tübingen. Nitrile gloves were worn at all times, glassware was sterilized at 500°C for 8 hours in a muffle furnace before use and HPLC grade reagents and high purity standards were used. Two different extraction techniques were applied: solvent extraction and saponification. Solvent extracts were silylated, and the acid fractions of the saponified samples were methylated. Direct acidified methanol extraction using sulfuric acid (Correa-Ascencio and Evershed 2014) was not applied because, while successfully applied to other pottery, procedural blanks and isotopic standards, the methodology proved incompatible with Chalcolithic pottery and limestone samples from Israel. The pottery is characterized by calcite inclusions (*e.g.* Burton *et al.* 2019; Cohen-Weinberger 2003, 2019; Roux *et al.* 2011) and clay likely heavily influenced by the alkaline Israeli bedrock (Singer 2007), which prevented the mandatory reaction with sulfuric acid from completing. Limestone is similarly composed of calcite, and the samples displayed the same issues.

4.3.1. Solvent extraction

The total lipid extract (TLE) of the samples was obtained through solvent extraction (Mottram *et al.* 1999). 5 mL of dichloromethane:methanol (DCM:MeOH, 2:1, *v*:*v*) was added to each sample.

The mixture was sonicated for 15 minutes and centrifuged at 3000 rpm for 10 minutes at room temperature. The solvent was then decanted into scintillation vials. This process was repeated twice more after which the TLE was dried under a gentle stream of nitrogen and mild heating (40°C). The TLE was partitioned for future analysis (50%).

4.3.2. Saponification

Following solvent extraction, the remaining ceramic and stone powder underwent saponification to release lipids bound to the matrix (*e.g.* Craig *et al.* 2004; Stern *et al.* 2000). 4 mL of 0.5 M of sodium hydroxide solution (MeOH:$H_2$0, 9:1, *v:v*) was added. This was heated for 90 minutes at 70°C and allowed to cool. The sodium hydroxide solution was decanted into a sterilized scintillation vial. From this, the neutral fraction was removed with a liquid-liquid extraction using 2 mL of cyclohexane. This process was repeated three times. The solvent was dried using mild heating (30°C) and a gentle stream of nitrogen, and the neutral fraction was not analyzed further. To the remaining aqueous solution, 2 mL of 1 M hydrochloric acid was added until reaching pH 3, forming the acid fraction. This was liquid-liquid extracted using 2 mL of cyclohexane three times. The solvent was dried under a gentle stream of nitrogen using mild heating (30°C), forming the acid fraction of polar compounds.

4.3.3. Silylation

Silylation was applied to 50% of the TLE of each solvent extract. 50 µL of *N,O*-bis(trimethylsilyl)trifluoroacetamide with 1% trimethylchlorosilane and 4 µL of pyridine were added. After heating for 30 minutes at 40°C, the vials were cooled, and the solvent was dried using nitrogen and mild heating. At this point, the samples were resolubilized in 50 µL of cyclohexane. These were analyzed alongside 1 µg of hexatriacontane (C_{36} alkane, 98%, Sigma-Aldrich) for quantification.

4.3.4. Methylation

The saponified acid fractions were methylated. Methylation was selected because saponified samples with high palmitic and stearic acid content would undergo GC-c-IRMS, and methylation adds fewer carbons than silylation. This produces more accurate GC-c-IRMS results (Docherty *et al.* 2001). These were methylated alongside 100 µg of palmitic (99%, Fluka) and stearic acid (99.5%, Sigma-Aldrich) standards of known isotopic composition. 250 µL of boron trifluoride methanolic solution (14%) was added to the archaeological samples, and 1 mL of boron trifluoride methanolic solution was added to fatty acid standards. These were heated for 60 minutes at 70°C.

Following this, the reaction was quenched with ultrapure water. The lipids were then liquid-liquid extracted three times using 2 mL of cyclohexane. The solvent was dried using mild heating (30°C) and a gentle stream of nitrogen until 1 mL remained. This was transferred into an autosampler vial and dried under nitrogen. The samples were resolubilized in 50 μL of cyclohexane, and the standards were resolubilized in 1 mL of cyclohexane. C_{36} alkane was measured as a standard for quantification.

4.4. GC-MS

GC-MS was performed using an Agilent 7890B GC coupled to an Agilent 5977A Mass Spectrometer (MSD) and Flame Ionization Detector (FID). Splitless injections were conducted using a GERSTEL multi-purpose sampler and cold-injection system. The samples were run on an Agilent J&W DB-5HT column (15.0 m x 0.32 mm i.d.; 0.1 μm film thickness) and the eluent was divided into two equal parts using 0.18 mm non-coated deactivated silica capillary columns (0.66 m splitter-column to the FID and 1.52 m splitter-column to the MSD) with the Three-Way Splitter Kit. The inlet temperature was ramped from 30°C to 240°C at 12°C s^{-1} (held isothermally for 5 minutes) and then increased to 350°C at 12°C s^{-1} (held isothermally for 10 minutes). The oven temperature was ramped from 40°C (held isothermally for 1 minute) to 100°C at 15°C min^{-1} and then to 240°C at 6°C min^{-1} and then increased to 350°C at 10°C min^{-1} (held isothermally for 20 minutes). The analysis used helium as the carrier gas. Mass spectra were acquired using election ionization at 70 eV. The mass range was from *m*/*z* 50–950 in 1.562 s. The temperature of the ion source, transfer line and FID was set to 300°C. The injection system was operated in splitless mode with a purge flow of 3.0 mL min^{-1} and a constant pressure at the head of the column of 8.4435 psi.

Mass spectra were identified using the National Institute of Standards and Technology (NIST) Library, 2014 edition. *n*-Alkanes were identified through comparison to an *n*-alkane standard mix (C_7–C_{40}, Sigma-Aldrich). The TLE (Appendix C and D) was calculated based on the FID signal using the following equation:

$$\mathrm{TLE} = \frac{\mathrm{Area(sample)}}{\mathrm{Area(C_{36})}} \times \frac{\mathrm{Weight(C_{36})}}{\mathrm{Weight(sample)}}$$

The full range of identified compounds is detailed in Appendix E and F.

4.5. GC-c-IRMS

Acid fractions with over 5 μg of palmitic and stearic acid in the acid fraction underwent GC-c-IRMS. 1 μL of each sample was injected by a CTC CombiPAL autosampler in PTV splitless mode

at 80°C and held for 0.5 minutes. For sample transfer the injector was ramped to 300°C at 14.5°C s^{-1}, and it was held for 2 minutes. Afterwards the injector was heated until the end of the run to 350°C for cleaning, and a split flow of 20 mL min^{-1} was applied.

In the GC, helium with a constant flow of 1.5 mL min^{-1} was used as the carrier gas. The compounds were separated on a DB-5ms column (30 m x 0.25 mm; 0.25 μm film thickness). The oven temperature was ramped from 50°C (held isothermally for 2 minutes) to 230°C at 5°C min^{-1} and to 310°C (held isothermally for 10 minutes) at 30°C min^{-1}. Compounds eluting from the GC between 31 and 38 minutes were combusted at 1000°C in a CuO/NiO/Pt combustion reactor. During the rest of the run, the backflush mode was activated to divert unwanted matrix components from the reaction and limit oxidation. The isotopic ratio mass spectrometer had a filament current set to 1.5 mA, the electron energy to 124 eV and the high voltage to 3 kV. During the measurements, a vacuum of *ca.* $6x10^{-7}$ mBar was maintained. Carbon dioxide was used as the reference gas.

Compound specific standards were measured at the beginning of each sequence and after every three runs. Samples were measured in duplicates. A precision of 0.5‰ was assumed for all samples. These results (Appendix D) were compared to a global reference database after correction by 1.14‰ (following Friedli *et al.* 1986). To account for environmental effects, the plot of $\delta^{13}C_{16:0}$ and $\Delta^{13}C$ was considered in tandem with the plot of $\delta^{13}C_{16:0}$ and $\delta^{13}C_{18:0}$. The samples were also plotted against mixing curves (following Woodbury *et al.* 1995). These were formed based on the isotopic values of $\delta^{13}C_{16:0}$ and $\delta^{13}C_{18:0}$ and the concentrations of $C_{16:0}$ and $C_{18:0}$ in each food source.

Chapter 5. Details of published articles

Article 1. Bee products in the prehistoric southern Levant: evidence from the lipid organic record

Journal: Royal Society Open Science

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Letter of acceptance:

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We hope you are keeping well at this difficult and unusual time. We continue to value your support of the journal in these challenging circumstances. If Royal Society Open Science can assist you at all, please don't hesitate to let us know at the email address below.

Dear Dr Chasan,

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Thank you for your fine contribution. On behalf of the Editors of Royal Society Open Science, we look forward to your continued contributions to the Journal.

Kind regards,
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openscience@royalsociety.org

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Name of Principal Author (Candidate)	Rivka Chasan		
Contribution to the Paper	R. Chasan conceptualised the study, carried out the laboratory work and data analysis and drafted the manuscript.		
Certification:	This paper reports on original research I conducted during the period of my Higher Degree by Research candidature and is not subject to any obligations or contractual agreements with a third party that would constrain its inclusion in this thesis. I am the primary author of this paper.		
Name and Signature		Date	27/06/2022

Co-Author Contributions

By signing the Statement of Authorship, each author certifies that:

i. the candidate's stated contribution to the publication is accurate (as detailed above);

ii. permission is granted for the candidate in include the publication in the dissertation

Name of Co-Author	Danny Rosenberg		
Contribution to the Paper	D. Rosenberg helped conceptualise the study, draft the manuscript and acquisition funds. He also provided ceramic vessels for analysis.		
Name and Signature	Danny Rosenberg 	Date	03.07.2022

Name of Co-Author	Florian Klimscha
Contribution to the Paper	F. Klimscha provided ceramic vessels for analysis.

Name and Signature	 Florian Klimscha	Date	28 06 2022

Name of Co-Author	Ron Be'eri		
Contribution to the Paper	R. Be'eri provided ceramic vessels for analysis.		
Name and Signature	 Ron Beeri	Date	10/7/2022

Name of Co-Author	Dor Golan		
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Name and Signature	 Dor Golan	Date	10/7/2022

Name of Co-Author	Ayelet Dayan		
Contribution to the Paper	A. Dayan provided ceramic vessels for analysis.		
Name and Signature	 Ayelet Dayan	Date	10/7/2022

Name of Co-Author	Ehud Galili
Contribution to the Paper	E. Galili provided ceramic vessels for analysis.

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Name of Co-Author	Cynthianne Spiteri		
Contribution to the Paper	C. Spiteri supervised the GC-MS analysis and helped conceptualise the study and draft the manuscript.		
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Author for correspondence:
Rivka Chasan
e-mail: rchasan@campus.haifa.ac.il

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Bee products in the prehistoric southern levant: evidence from the lipid organic record

Rivka Chasan[1], Danny Rosenberg[1], Florian Klimscha[2], Ron Beeri[3], Dor Golan[3], Ayelet Dayan[3], Ehud Galili[4,5] and Cynthianne Spiteri[6]

[1]Laboratory for Ground Stone Tools Research, Zinman Institute of Archaeology, University of Haifa, Haifa, Israel
[2]Archaeology Division, Research/Collections, Lower Saxony State Museum, Hanover, Germany
[3]Israel Antiquities Authority, Jerusalem, Israel
[4]Zinman Institute of Archaeology, and [5]Leon Recanati Institute for Maritime Studies, University of Haifa, Haifa, Israel
[6]Institute of Pre- and Protohistoric Archaeology, Eberhard Karls Universität Tübingen, Tübingen, Germany

RC, 0000-0002-0150-9250; CS, 0000-0003-1773-3895

Beehive products have a rich global history. In the wider Levantine region, bees had a significant role in Egypt and Mesopotamia, and intensive beekeeping was noted in Israel during the Biblical period when apiaries were first identified. This study investigates the origins of this extensive beekeeping through organic residue analysis of pottery from prehistoric sites in the southern Levant. The results suggest that beehive products from likely wild bees were used during the Chalcolithic period as a vessel surface treatment and/or as part of the diet. These functions are reinforced by comparison to the wider archaeological record. While the true frequency of beeswax use may be debated, alternatives to beehive products were seemingly preferred as wild resources contrasted with the socio-economic system centred on domesticated resources, controlled production and standardization. Bee products only became an important part of the economic canon in the southern Levant several millennia later.

1. Introduction

Bees produce a variety of products that can be readily exploited, including beeswax, honey and propolis, each having an extensive history of use [1]. Beeswax was used for several

purposes, including lighting [2], gluing [3], medicine [4–6], art [7,8], pottery sealing [9], embalming [10] and metallurgy [11]. Honey was frequently used as a sweetener and preservative, but it also played a role in medicine and embalming [1]. Propolis was similarly used as medicine [12] and as an adhesive [1].

The use of bee products may be identified in the archaeological record using varying lines of data including textual sources and, more recently, organic residues. Organic residue analysis identifies bee products by comparing the archaeological lipid residues to the lipid profile of modern beeswax. The lipid signature of fresh beeswax is characterized by its wax ester, saturated fatty acid, *n*-alkane and *n*-alcohol profiles. The long-chain even-numbered palmitate wax esters (C_{40}–C_{52}) are the most characteristic marker, maximizing at C_{46} [13]. Saturated fatty acids are even-numbered and long-chain ($C_{14:0}$–$C_{36:0}$) with a predominance of lignoceric acid ($C_{24:0}$). Once degraded, there is an increasing amount of palmitic acid ($C_{16:0}$) formed from the hydrolysis of palmitate wax esters [13–15]. Beeswax is also characterized by odd-numbered *n*-alkanes ranging from C_{21}–C_{35}, maximizing at C_{27} [13,15], as well as even-numbered *n*-alcohols ranging from C_{24}–C_{36}, maximizing at C_{30} [15].

Using this biomolecular approach, beeswax was widely identified in the global archaeological record. Evidence for the exploitation of beeswax was found already in Middle Palaeolithic Europe and South Africa, where it was used for hafting flint tools by Neanderthals and anatomically modern humans [16–18]. A wider exploitation was documented during the 7th–3rd millennium cal. BC. Beeswax was used in dentistry [5] and hafting [19], and while no specific function was proposed, beeswax lipid residues were found in relation to pottery at over 50 Neolithic sites in Eurasia and North Africa (figure 1*a*; [20] and see references therein). The frequency of beeswax residue at these sites is highly variable, ranging from 1% to well over 30% of the tested vessels with significant lipid yield [20] (although the proportion may be influenced and biased by the number and types of vessels sampled as well as the preservation conditions offered by the climate and sediment characteristic of each site). The tested sites are from topographically and climatically variable areas, showing that bees thrived in diverse environmental and ecological niches and that their exploitation was a common and shared practice.

With a few potential south Levantine Late Chalcolithic (*ca* 4500–3700 cal. BC) exceptions [38], indications for the use of beeswax before the 3rd millennium BC in the wider Levantine region are rare. The first evidence for bees stems from later written sources. Textual evidence from the Egyptian First Dynasty described the Egyptian god Min as the 'master of wild bees' [1] and the king of Upper and Lower Egypt as 'he of the sedge and bee' [39]. Shortly thereafter (*ca* 2400 BC), beekeeping was depicted in bas-reliefs [1]. In Anatolia and Mesopotamia, bees were also highly valued. Evidence from Hittite legal documents described the fine for beehive theft as lofty and nearly equal to the fine for stealing a sheep [1,40], and other texts suggest that Sumerian honey was used as religious offerings [1]. Honey was also mentioned numerous times in the Bible, which described Israel as 'a land flowing with milk and honey' [Exodus 3.8]. Archaeological evidence for beekeeping in the southern Levant was first noted at the Iron Age II site of Tel Rehov (tenth–ninth century BC), located in the Jordan Valley, Israel. The site includes several beehives formed from hollow clay cylinders that would have been capable of producing hundreds of kilograms of honey annually. Their use as hives was reinforced by organic residue analysis (ORA) [41].

Other contemporary and slightly earlier examples of beeswax in the southern Levant were recently identified based on lipids preserved within ceramics and characterized by Gas Chromatography-Mass Spectrometry (GC-MS). Some of these identifications relied primarily or exclusively on the *n*-alkane profiles, and as such, beeswax identification is tenuous. Highly degraded beeswax was suggested as identified on a Late Bronze Age II clay coffin from Tel Shadud in the Jezreel Valley, Israel. Beeswax may have been applied to the coffin as a coating or a sealant [42]. Similar markers were observed from a storage jar found at the Late Bronze Age III site of Tel Azekah in the Shephelah, and it was suggested that these markers were related to bee products and that the jar was used to store honey [43]. Potential evidence for beeswax was also recovered from the Iron Age I burial site of Horvat Tevet in the Jezreel Valley, and it was suggested that heated beeswax was used in burial ceremonies [44]. Finally, a sub-set of beeswax markers was recovered from a large storage jar found at Jneneh, an Iron Age II site in north-central Jordan. The ceramic form is commonly associated with liquid storage, so it is possible that beeswax was used as a sealant; alternatively, the jar could have been used to store honey [45].

Beeswax is rare in the organic record of the prehistoric southern Levant although prior studies analysed ceramic vessels from many sites (figure 1*b*); beeswax residue has only potentially been identified in relation to cornets—a cone-shaped vessel characteristic of the Late Chalcolithic period. Cornets with lipid profiles similar to beeswax residue were recovered from three sites: Grar in the northern Negev and Moringa Cave and Ein Gedi in the Judean Desert, west of the Dead Sea. Based on the *n*-alkane profile, it was suggested that the cornets represent lamps used with beeswax candles

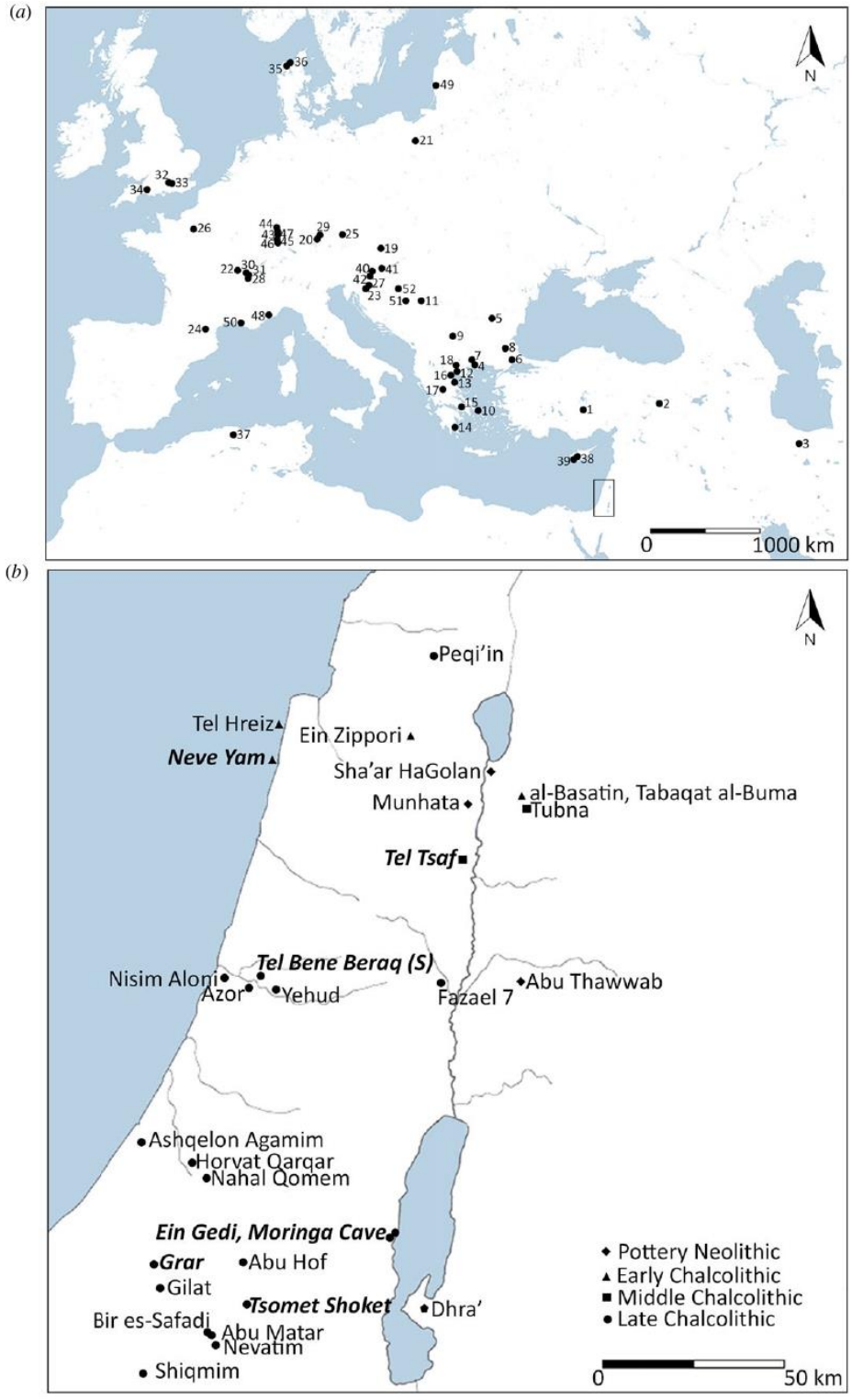

Figure 1. (*Caption overleaf.*.)

Figure 1. (*Overleaf*.) (*a*) Map of sites from the 7th–3rd millennium BC with lipid biomarker evidence for beeswax: 1. Çatalhöyük [20]; 2. Çayönü Tepesi [20]; 3. Tepe Sofalin [21]; 4. Liménaria [22]; 5. Măgura [20]; 6. Toptepe [20]; 7. Dikili Tash [20,23]; 8. Aşağı Pınar [20]; 9. Drenovac Turska Cesma [20]; 10. Ftélia [20]; 11. Vinča Belo Brdo [20]; 12. Paliambela [20]; 13. Rachmani [20]; 14. Kouvéléikès A [20]; 15. Profitis Ilias Rizoupolis [20]; 16. Vassilara Rachi [20]; 17. Théopetra [20]; 18. Balkan Export [20]; 19. Brunn am Gebirge [20]; 20. Niederhummel [20]; 21. Kuyavia region [24]; 22. Chassey-le-Camp [20]; 23. Moverna vas [20]; 24. Font-Juvénal [20]; 25. Künzing-Unternberg [20]; 26. Bercy [14]; 27. Ajdovska jama [20,25]; 28. Clairvaux XIV [20]; 29. Ergolding Fischergasse [26]; 30. Chalain 3 [20,27,28]; 31. Chalain 4 [20,27,28]; 32. Eton rowing lake [29]; 33. Runnymede bridge [20]; 34. Bulford Torstone [20]; 35. Åle [20]; 36. Bjørnsholm [20]; 37. Gueldaman [20]; 38. Sotira Teppes [30]; 39. Erimi Pamboula [30]; 40. Zgornje Radvanje [31]; 41. Turnišče [31]; 42. Spodnje Hoče [31]; 43. Bischoffsheim [32]; 44. Rosheim [32]; 45. Ensisheim [32]; 46. Sierentz [32]; 47. Colmar [32]; 48. Pendimoun rock-shelter [33]; 49. Šventoji 4 [34]; 50. Vernègues-Cazan-l'Heritière [35]; 51. Starčevo-Grad [36]; 52. Magareći Mlin [36]; (*b*) Map of sites in the southern Levant dating to ca. 6400–3700 BC [37] whose pottery assemblages were studied using ORA. Sites with evidence for beeswax are in bold and italics: Neve Yam (current study), Tel Tsaf (current study) Tel Bene Beraq (South) (current study), Moringa Cave [38], Ein Gedi [38], Grar [38] and Tsomet Shoket (current study).

[38], although the *n*-alkane profile alone is insufficient to support this with certainty. These few examples do not sufficiently explain the emergence of intensive bee exploitation in the southern Levant observed in the Late Bronze Age and Iron Age. Further archaeological and palynological evidence (e.g. [46]) to suggest the presence of beehives or bees is also absent, but additional archaeological investigation and palynological analysis is required.

The aim of this study is to use a lipid biomarker approach to investigate a possible earlier presence and use of beeswax in the Chalcolithic period of the southern Levant. This research is part of a wider study, which tested 247 ceramic vessels from 15 prehistoric sites in Israel using ORA. Emphasis was placed on pottery from the Chalcolithic period (including the Early Chalcolithic (*ca* 5800–5300 cal. BC), Middle Chalcolithic (*ca* 5300–4700 cal. BC) and Late Chalcolithic (*ca* 4500–3700 cal. BC)), with a few vessels from the Pottery Neolithic (*ca* 6400–5800 cal. BC) also tested. At the end of this timespan, important socio-economic transformations and technological advances occurred, such as developments in olive horticulture, the intensified use of secondary products, craft specialization and the introduction of copper metallurgy (e.g. [47–51]). The results obtained here together with previously published research are discussed to explain why during this timespan beehive products were used and, more importantly, why this versatile resource appears to have been underexploited.

2. Material and methods

In the course of the wider study, ceramic vessels were sampled from a geographically and chronologically diverse range of sites (table 1). Vessels were selected from ongoing excavations and assemblages stored in the Israel Antiquities Authority's collection. There was no targeted selection of specific vessels as prior studies on south Levantine sites identified beeswax inconsistently in both task-specific and unspecialized vessel forms, such as bowls, jars, cornets, chalices, flasks and lamps [38,43,44]. Correspondingly, we sampled a wide selection of vessel forms (e.g. bowls, holemouth jars, necked jars, bow-rim jars, pithos spouted vessels, platters, churns, cornets and strainers), which are commonly found at prehistoric sites [52], as well as typologically unidentified fragments.

Following cleaning the initial surface layer (*ca* 1–2 mm) to remove potential exogenous contamination, around 1–2 g of ceramic powder were collected from the internal surface of each vessel using a Dremel modelling drill fitted with a tungsten bit. An additional crust from an internal vessel surface was collected and analysed after being homogenized. Ceramic powder from the external surfaces or soil samples from the layer the ceramics were retrieved from were collected (*ca* 1–2 g) and analysed as controls to test for exogenous and post-depositional contamination.

Glassware was sterilized before use (500°C in a muffle oven overnight), and HPLC grade solvents were used. Lipids were extracted alongside a process blank to monitor for laboratory contamination. The 'unbound' lipid fraction was recovered by solvent extraction (following [53]), and this was followed for internal samples by saponification to release the 'bound' lipid fraction (following a modified Method D in [54]). This two-step approach was selected in favour of a direct acidified methanol extraction [55] to ensure that wax esters and acylglycerols could be analysed before hydrolysis.

Lipids were extracted three times using 5 ml of dichloromethane : methanol (DCM:MeOH, 2:1, *v:v*), assisted by sonication (15 min) and centrifugation (10 min at 3000 r.p.m.). The solvent was dried under a gentle stream of nitrogen and mild heating (40°C) to obtain the total lipid extract (TLE). An aliquot of

Table 1. Summary of the sites analysed by ORA.

site	period	N pottery vessels sampled	types of vessels sampled
Sha'ar HaGolan	Pottery Neolithic	16	bowls, jars and typologically unidentified vessel fragments
Neve Yam	Early Chalcolithic	17	jars, bow-rim jars, spouted vessels and typologically unidentified vessel fragments
Tel Hreiz	Early Chalcolithic	3	jars and typologically unidentified vessel fragments
Tel Tsaf	Middle Chalcolithic	100	bowl, v-shaped bowls, deep bowls, small bowls, hemispherical bowls, jars, holemouth jars, necked jars, platters and typologically unidentified vessel fragments
Abu Matar	Late Chalcolithic	3	holemouth jars and churns
Ashqelon Agamim	Late Chalcolithic	28	bowls, jars, cornets and typologically unidentified vessel fragments
Azor	Late Chalcolithic	1	strainer vessel
Tel Bene Beraq (South)	Late Chalcolithic	10	jars, holemouth jars and typologically unidentified vessel fragments
Bir es-Safadi	Late Chalcolithic	11	jars, holemouth jars, spouted vessels and churns
Fazael 7	Late Chalcolithic	8	jars, holemouth jars and typologically unidentified vessel fragments
Gilat	Late Chalcolithic	13	v-shaped bowls, jars, holemouth jars, churns and cornets
Nisim Aloni	Late Chalcolithic	9	bowls, v-shaped bowls, jars, churns and typologically unidentified vessel fragments
Peqi'in	Late Chalcolithic	3	v-shaped bowls
Tsomet Shoket	Late Chalcolithic	18	bowls, v-shaped bowls, jars, holemouth jars, pithoi and churns
Yehud	Late Chalcolithic	7	churns, strainer vessels and typologically unidentified vessel fragments

50% of the TLE was silylated using 50 µl of *N,O*-bis (trimethylsilyl) trifluoroacetamide with 1% trimethylchlorosilane and 4 µl of pyridine (40°C; 30 min). The samples were run as trimethylsilylated (TMS) derivatives. After derivitization, initially a known amount of hexatriacontane (C_{36} *n*-alkane; 98%, Sigma-Aldrich) was added to the samples to allow for TLE quantification; upon noticing that hexatriacontane occasionally co-eluted with other molecules, the methodology was changed, and it was run systematically in the GC-MS sequence after every four samples to allow for a more accurate quantification.

Following solvent extraction, the lipids bound to the ceramic matrix were saponified (70°C; 90 min). 4 ml of 0.5 M sodium hydroxide solution (MeOH:H_2O, 9:1, *v:v*) was used. The neutral fraction was extracted three times with cyclohexane and was not analysed further. To the remaining sodium hydroxide solution, 2 ml of 1 M hydrochloric acid was added until pH 3 was reached. The acid fraction was extracted three times with cyclohexane. It was then methylated using 250 µl of boron trifluoride methanolic solution (14%) (70°C; 60 min). The samples were run as methyl esters. As with the solvent extracts, initially a known amount of C_{36} *n*-alkane was added to the samples to allow for TLE quantification, and in later batches, the C_{36} *n*-alkane was run only externally after every four archaeological samples.

GC-MS was performed using an Agilent 7890B GC coupled to an Agilent 5977A Mass Spectrometer (MSD) and Flame Ionization Detector (FID). Injections were carried out using a GERSTEL multi-purpose sampler and a GERSTEL Cold-Injection System (CIS) 4. The samples were run on an Agilent J&W DB-5HT column (15 m × 0.32 mm i.d.; 0.1 µm film thickness), and the eluent was divided into two equal parts using 0.18 mm non-coated deactivated silica capillary columns (0.66 m splitter-column to the FID and 1.52 m splitter-column to the MSD) with the Three-Way Splitter Kit. The inlet temperature was ramped from 30°C to 240°C at 12°C s^{-1} (held isothermally for 5 min) and then increased to 350°C at 12°C s^{-1} (held isothermally for 10 min). The oven temperature was ramped from 40°C (held isothermally for 1 min) to 100°C at 15°C min^{-1} and then to 240°C at 6°C min^{-1} and then to 350°C at 10°C min^{-1} (held isothermally for 20 min). The analysis employed helium as the carrier gas. Splitless injection was applied, with a purge flow of 3.0 ml min^{-1} and a constant pressure at the head of the column of 8.4435 psi. Mass spectra were acquired using electron ionization at 70 eV. The mass range acquired was from *m/z* 50–950 in 1.562 s. The temperatures of the ion source, transfer line and FID were each 300°C. Mass spectra were identified using the National Institute of Standards and Technology Library, 2014 edition, and *n*-alkanes were identified through comparison to a saturated *n*-alkane standard mix (C_7–C_{40}) (Sigma-Aldrich).

3. Results

Lipid preservation was low as expected given the Mediterranean climate and alkaline soil [56] characteristic of the southern Levant. Both factors support microbial activity and correspondingly lipid degradation [57,58]. From the 247 vessels tested, only 22 vessel interiors and one calcified crust yielded more than 5 µg g^{-1} of lipids after solvent extraction, considered an interpretable residue [59]. While originally sampled as controls, an additional seven vessels had more than 5 µg g^{-1} of lipids exclusively externally. This may relate to spillage or more likely the application of a vessel treatment to alter the vessel surface or porosity [60] (post-depositional contamination, spillage and lipid migration from the vessel interior may likely be ruled-out because of the lack of lipids internally). The bound acid fraction had a much higher lipid yield. Within this fraction, 47% of the internal layers of the vessels tested ($n = 116$) and the crust contained over 5 µg g^{-1} of lipid.

Among these internally and externally solvent extracted vessels, evidence for beeswax was identified in only four vessels from four of the studied sites (figure 2): Early Chalcolithic Neve Yam, Middle Chalcolithic Tel Tsaf and Late Chalcolithic Tel Bene Beraq (South) and Tsomet Shoket (see site details in electronic supplementary material, S1). This includes the interior surface of a jar from Neve Yam (NY-12 interior layer 2), the calcified crust on a wall fragment from Tel Tsaf (TSF18-57 crust), the interior surface of a jar from Tel Bene Beraq (South) (BB-21 interior layer 2) and the exterior surface of a jar from Tsomet Shoket (TS-4 exterior) (table 2). The related biomarker evidence, which will be detailed below, includes even-numbered long-chain saturated fatty acids, odd-numbered long-chain *n*-alkanes, even-numbered long-chain *n*-alcohols and palmitate wax esters [13,15].

Beeswax markers were not present in a variety of other vessel forms tested, including bowls, holemouth jars, bow-rim jars, spouted vessels, platters, churns, cornets and strainers. The absence of beeswax markers in the six cornets analysed is notable considering the proposed positive

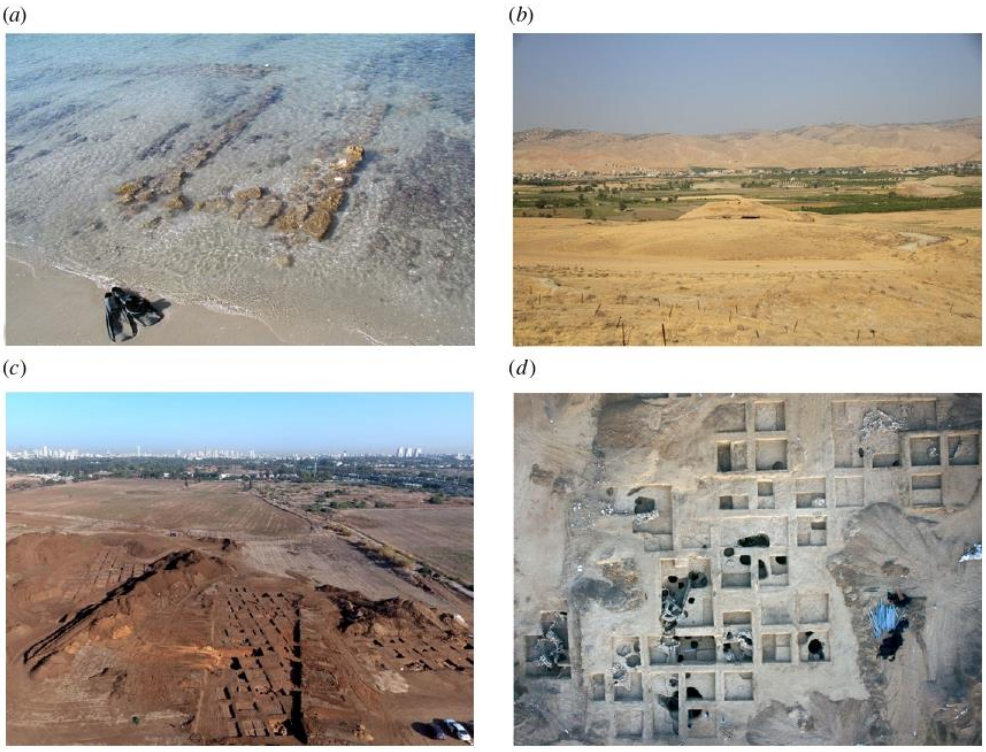

Figure 2. A view of the sites with evidence for beeswax: (*a*) Neve Yam (photo by E. Galili); (*b*) Tel Tsaf (photo by D. Rosenberg); (*c*) Tel Bene Beraq (South) (photo by I. Marmelstein (Israel Antiquities Authority)); (*d*) Tsomet Shoket (photo by A. Peretz (Israel Antiquities Authority)).

identification in an earlier study based on the *n*-alkane profiles [38]. This suggests that if cornets were used as candles, other fuel sources were used in addition to beeswax. Alternatively, the beeswax biomarkers in the cornets could relate to an entirely different use of beeswax, for instance as a sealant for drinking vessels (e.g. [61]).

A more in-depth biomarker analysis of the four vessels with evidence for beeswax was conducted to confirm this attribution and identify additional lipid inputs:

3.1. Jar wall fragment from Neve Yam (NY-12)

The TLE of the interior surface of the jar found at Neve Yam (NY-12 interior layer 2; figure 3; table 2) contained in part saturated fatty acids, including pelargonic ($C_{9:0}$), pentadecylic ($C_{15:0}$), palmitic ($C_{16:0}$), margaric ($C_{17:0}$) and stearic ($C_{18:0}$) acid, maximizing at $C_{16:0}$. The bound acid fraction revealed a wider range of saturated fatty acids ($C_{12:0}$–$C_{30:0}$), with high amounts of long-chain even-numbered fatty acids (electronic supplementary material S2). The *n*-alkanes in the TLE ranged from C_{20}–C_{29}, maximizing at *n*-nonacosane (C_{29}), with a clear odd over even preference as identified by the carbon preference index (CPI) (CPI 28.1) (following [62]). The *n*-alcohols ranged from C_{12}–C_{30}; all are even, and they maximize at 1-octacosanol (C_{28}). Even-numbered palmitate wax esters with 42–46 carbon atoms were present, with C_{44} the most abundant.

Some of the lipid compounds indicate contributions from animal fat and plant oil. The presence of animal fat may be indicated by the recovery of cholesterol [63]. No cholesterol degradation markers were identified, so this may suggest contamination, although no other contaminants that originate from human contact (e.g. [64]) were identified. Ruminant fat may be specifically suggested based on the odd-numbered and branched-chain fatty acids in the acid fraction as well as the high amount of $C_{18:0}$ (electronic supplementary material, S2; table 2) [65,66]. Odd-numbered and branched-chain fatty acids are formed by bacteria in the rumen [66]. The presence of a plant oil is suggested by the unsaturated fatty acids in the solvent extract ($C_{18:1}$, two isomers) and acid fraction (one isomer of $C_{16:1}$

Table 2. Lipid profile summary of the samples with evidence for beeswax and their controls (SE, solvent extract; AF, acid fraction; K, ketone). The maximum molecule in each category is bolded and the lipid marker sets that are characteristic of fresh beeswax are underlined.

vessel	sample	TLE ($\mu g\ g^{-1}$)		saturated fatty acids		unsaturated fatty acids		branched-chain fatty acids in AF	*N*-alkanes in SE	*N*-alcohols in SE	wax esters in SE	other molecules in SE and AF
		SE	AF	SE	AF	SE	AF					
NY-12	**NY-12 interior layer 2**	5.0	58.9	$C_{9:0}$, $C_{15:0}$, **$C_{16:0}$**, $C_{17:0}$, $C_{18:0}$	$C_{12:0}$, $C_{13:0}$, $C_{14:0}$, $C_{15:0}$, $C_{16:0}$, $C_{17:0}$, **$C_{18:0}$**, $C_{19:0}$, $C_{20:0}$, $C_{21:0}$, $C_{22:0}$, $C_{23:0}$, $C_{24:0}$, $C_{25:0}$, $C_{26:0}$, $C_{27:0}$, $C_{28:0}$, $C_{30:0}$	$C_{18:1}$	$C_{16:1}$, $C_{18:1}$, $C_{20:1}$	$C_{14:0}$, $C_{16:0}$	C_{20}, C_{21}, C_{23}. C_{24}, C_{25}, C_{27}, **C_{29}**	C_{12}, C_{14}, C_{16}, C_{18}, C_{24}, C_{26}, **C_{28}**, C_{30}	C_{42}, **C_{44}**, C_{46}	K_{31}, K_{33}, 1-monopalmitin, Cholesterol
	NY-12 exterior	1.4	—	**$C_{16:0}$**, $C_{18:0}$	—	—	—	—	C_{20}, C_{21}, C_{22}, C_{23}, C_{24}, C_{25}, C_{26}, C_{27}, C_{28}, **C_{29}**, C_{31}	C_{14}, C_{16}, C_{18}, C_{20}, C_{22}, C_{24}, **C_{26}**, C_{28}	C_{42}, **C_{44}**	1-monopalmitin
TSF18-57	**TSF18-57 internal crust**	6.8	12.8	$C_{16:0}$, $C_{18:0}$	$C_{14:0}$, $C_{15:0}$, $C_{16:0}$, $C_{17:0}$, **$C_{18:0}$**, $C_{19:0}$, $C_{20:0}$, $C_{21:0}$, $C_{22:0}$, $C_{23:0}$, $C_{24:0}$, $C_{25:0}$, $C_{26:0}$, $C_{27:0}$, $C_{28:0}$, $C_{30:0}$	—	$C_{18:1}$, $C_{18:2}$	$C_{14:0}$–$C_{16:0}$	C_{23}, C_{25}, C_{26}, C_{27}, C_{28}, **C_{29}**, C_{30}, C_{31}, C_{32}, **C_{33}**	C_{20}, C_{21}, C_{22}, C_{23}, C_{24}, C_{25}, C_{26}, C_{27}, **C_{28}**, C_{30}, C_{32}, C_{34}	C_{40}, C_{42}, **C_{44}**, C_{46}, C_{48}	K_{31}
	TSF18-57 interior layer 2	3.1	5.6	$C_{9:0}$, $C_{10:0}$, $C_{15:0}$, **$C_{16:0}$**, $C_{18:0}$, $C_{20:0}$, $C_{22:0}$, $C_{24:0}$, $C_{26:0}$, $C_{28:0}$	$C_{14:0}$, $C_{15:0}$, $C_{16:0}$, $C_{17:0}$, **$C_{18:0}$**, $C_{19:0}$, $C_{20:0}$, $C_{21:0}$, $C_{22:0}$, $C_{23:0}$, $C_{24:0}$, $C_{25:0}$, $C_{26:0}$, $C_{27:0}$, $C_{28:0}$	$C_{18:1}$	$C_{18:1}$, $C_{18:2}$	-	C_{23}, C_{25}, **C_{27}**, C_{29}, C_{31}	C_{18}, C_{20}, C_{22}, C_{23}, C_{24}, C_{26}, C_{27}, **C_{28}**, C_{30}, C_{32}	C_{40}, C_{42}, **C_{44}**, C_{46}	K_{31}, 1-monopalmitin, 1-monostearin
	TSF18-57 exterior	1.5	—	$C_{16:0}$, **$C_{18:0}$**, $C_{28:0}$	—	—	—	—	C_{14}, C_{23}, C_{25}, C_{26}, C_{27}, **C_{29}**, C_{30}, C_{31}, C_{33}	C_{22}, C_{24}, C_{26}, C_{27}, **C_{28}**, C_{30}, C_{32}	—	—

(*Continued*.)

Table 2. (*Continued.*)

vessel	sample	TLE ($\mu g\ g^{-1}$)		saturated fatty acids		unsaturated fatty acids		branched-chain fatty acids in AF	*N*-alkanes in SE	*N*-alcohols in SE	wax esters in SE	other molecules in SE and AF
		SE	AF	SE	AF	SE	AF					
BB-21	**BB-21 interior layer 2**	55.3	86.4	**$C_{16:0}$**, $C_{18:0}$, $C_{24:0}$	$C_{9:0}$–$C_{13:0}$, $C_{14:0}$, $C_{15:0}$, $C_{16:0}$, $C_{18:0}$, $C_{20:0}$, $C_{21:0}$, $C_{22:0}$, $C_{23:0}$, **$C_{24:0}$**, $C_{25:0}$, $C_{26:0}$, $C_{27:0}$, $C_{28:0}$, $C_{30:0}$	—	$C_{18:1}$	$C_{14:0}$, $C_{17:0}$	C_{23}, C_{24}, C_{25}, C_{26}, **C_{27}**, C_{28}, C_{29}, C_{33}, C_{35}, C_{37}	**C_{24}**, **C_{26}**, C_{28}, **C_{30}**, C_{32}	C_{40}, C_{42}, C_{44}, **C_{46}**, C_{48}, C_{50}	K_{33}, C_{14}–C_{30} dicarboxylic acids
	BB-21 soil	0.7	—	**$C_{16:0}$**, $C_{18:0}$, $C_{22:0}$, $C_{24:0}$, $C_{26:0}$	—	—	—	—	C_{27}, **C_{29}**	C_{16}, C_{18}, C_{22}, C_{24}, **C_{26}**, C_{28}, C_{30}	—	1-monopalmitin, β-Sitosterol, Campesterol, Stigmasterol, Ferruginol, 2,3-Dehydroferruginol, Sempervirol, Totarol
TS-4	**TS-4 interior layer 2**	0.6	2.8	$C_{16:0}$	$C_{14:0}$, $C_{15:0}$, **$C_{16:0}$**, $C_{17:0}$, $C_{18:0}$, $C_{20:0}$, $C_{22:0}$, $C_{23:0}$, $C_{24:0}$	—	$C_{16:1}$, $C_{18:1}$, $C_{18:2}$, $C_{20:1}$, $C_{22:1}$	$C_{14:0}$, $C_{15:0}$	C_{27}, **C_{29}**	C_{16}, C_{18}	—	1-monopalmitin, β-Sitosterol, Cholesterol
	TS-4 exterior	6.6	—	—	—	—	—	—	C_{25}, C_{26}, C_{27}, C_{28}, C_{29}, C_{30}, **C_{31}**, C_{33}	C_{18}, C_{24}, C_{26}, C_{28}, **C_{30}**, C_{32}	C_{40}, C_{42}, **C_{44}**, C_{46}	—

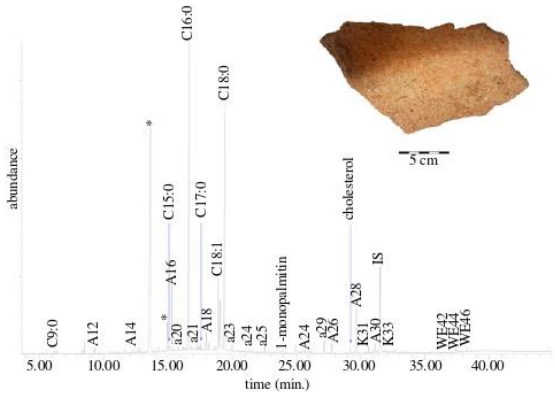

Figure 3. Total Ion Chromatogram (TIC) of the solvent extracted TLE released from the interior surface of a jar wall fragment from Neve Yam (NY-12 interior layer 2) analysed as TMS derivatives (*, plasticisers; ax, *n*-alkane with chain length X; Ax, *n*-alcohol with chain length X; Cx:y, fatty acid with chain length X and Y number of double bonds; Kx, ketone with chain length X; WEx, wax ester with chain length X).

and $C_{20:1}$ and two isomers of $C_{18:1}$), although small amounts of $C_{18:1}$ are also present in animal fat [67,68]. Ketones were also identified, including hentriacontan-16-one (K_{31}) and tritriacontan-16-one (K_{33}).

The external control sample (NY-12 exterior) had many of the same molecules in insignificant amounts, suggesting that lipids may have migrated through the ceramic matrix and that the lipid signature in the interior did not result from post-depositional contamination. This includes $C_{16:0}$, $C_{18:0}$, *n*-alkanes (C_{20}–C_{31}), even-numbered *n*-alcohols (C_{14}–C_{28}) and palmitate wax esters (C_{42} and C_{44}) (electronic supplementary material, S2; table 2).

3.2. Crust on wall fragment from Tel Tsaf (TSF18-57)

The TLE of a crust from Tel Tsaf (TSF18-57 crust; figure 4; table 2) contains trace amounts of saturated fatty acids, including $C_{16:0}$ and $C_{18:0}$. In the bound acid fraction, significant amounts of saturated fatty acids were identified, ranging from $C_{14:0}$–$C_{30:0}$, with high amounts of long-chain even saturated fatty acids (electronic supplementary material, S2). In the TLE, the *n*-alkanes ranged from C_{23}–C_{33}, with C_{29} the most abundant. There is a clear odd over even preference (CPI 9.4). The *n*-alcohols ranged from C_{20}–C_{34} and maximize at C_{28}. The few odd-numbered *n*-alcohols present were identified in trace amounts. Palmitate wax esters are even-numbered with 40–48 carbons atoms. C_{44} is the most abundant wax ester.

Additional markers in the solvent extraction and bound acid fraction are suggestive of animal fat and plant oil. Ruminant fat may be suggested based on the acid fraction, which contains high amounts of $C_{18:0}$, odd-numbered saturated fatty acids and branched-chain fatty acids (electronic supplementary material, S2; table 2) [65,66]. $C_{18:1}$ (two isomers) and $C_{18:2}$ (one isomer) were also identified and may be used to suggest a potential plant oil (e.g. [68]), although $C_{18:1}$ may also originate from animal fat [67,68]. Additionally, a single ketone was identified – K_{31}.

The TLE of the internal ceramic sample (TSF18-57 interior layer 2; electronic supplementary material, S2; table 2) and the external control sample (TSF18-57 exterior; electronic supplementary material 2; table 2) had an insignificant lipid yield. In the TLE of the internal ceramic sample, there is a wide range of saturated fatty acids with 9–28 carbon atoms; many of these same fatty acids were released by saponification, paralleling those in the crust. The identified odd-numbered *n*-alkanes (C_{23}–C_{31}), even-numbered *n*-alcohols (C_{18}–C_{32}), even-numbered palmitate wax esters (C_{40}–C_{46}) and K_{31} also parallel the molecules in the crust. This suggests that the lipids were absorbed into the ceramic matrix during the use episode that formed the crust. In the external sample, there is $C_{16:0}$, $C_{18:0}$, $C_{28:0}$, a wide range of *n*-alkanes, including C_{14}–C_{33}, and *n*-alcohols, with C_{22}–C_{32}. Wax esters were not identified on the vessel exterior, reinforcing that these originated from vessel use and not post-depositional contamination.

3.3. Jar wall fragment from Tel Bene Beraq (South) (BB-21)

The TLE from the interior surface of the jar found at Tel Bene Beraq (South) (BB-21 interior layer 2; figure 5; table 2) contains the following saturated fatty acids: $C_{16:0}$, $C_{18:0}$ and $C_{24:0}$. In the bound

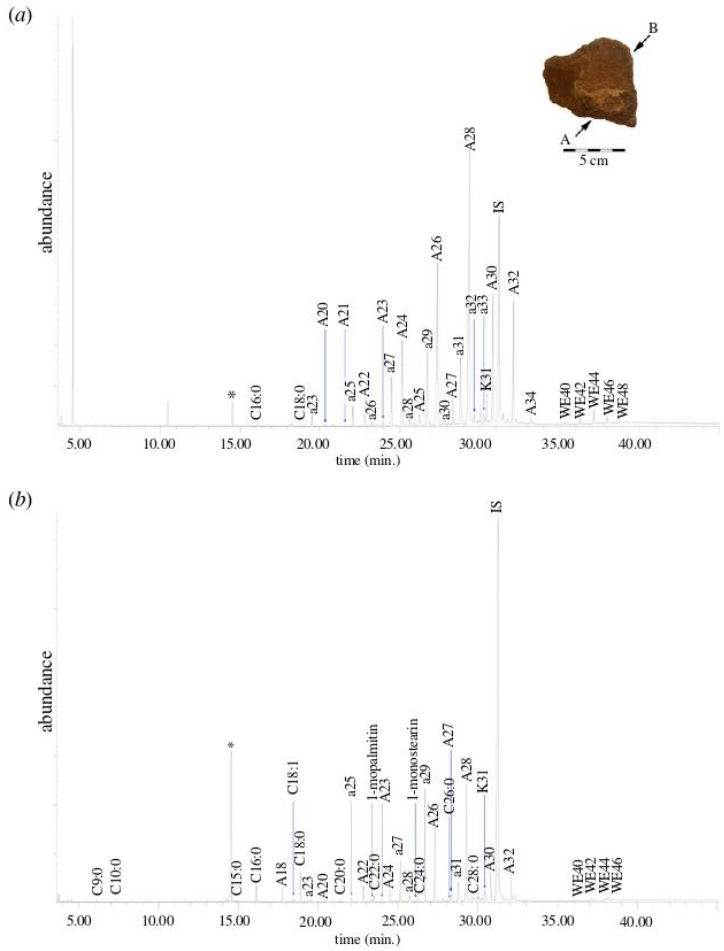

Figure 4. TIC of the solvent extracted TLE released from (*a*) the crust (TSF18-57 internal crust); (*b*) the interior wall (TSF18-57 interior layer 2) of a fragment from Tel Tsaf analysed as TMS derivatives (*, plasticisers; ax, *n*-alkane with chain length X; Ax, *n*-alcohol with chain length X; Cx:y, fatty acid with chain length X and Y number of double bonds; Kx, ketone with chain length X; WEx, wax ester with chain length X).

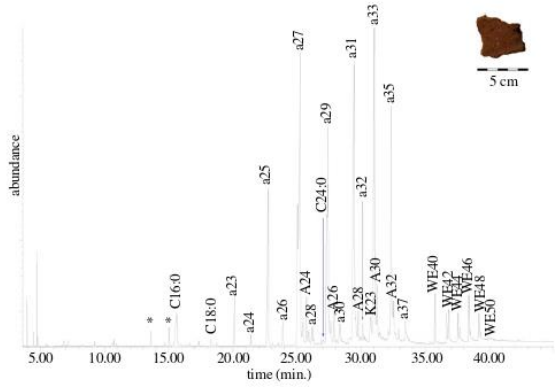

Figure 5. TIC of the solvent extracted TLE released from the interior surface of a jar wall fragment from Tel Bene Beraq (South) (BB-21 interior layer 2) analysed as TMS derivatives (*, plasticisers; ax, *n*-alkane with chain length X; Ax, *n*-alcohol with chain length X; Cx:y, fatty acid with chain length X and Y number of double bonds; Kx, ketone with chain length X; WEx, wax ester with chain length X).

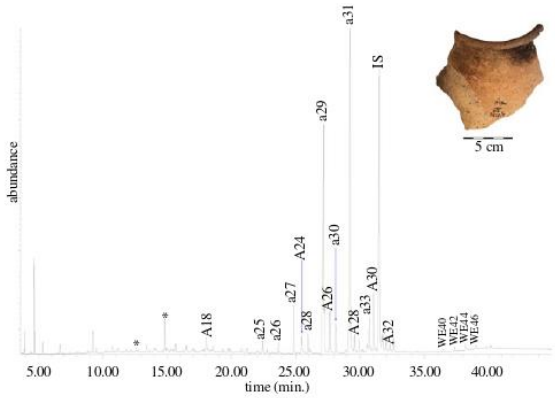

Figure 6. TIC of the solvent extracted TLE released from the exterior surface of a jar rim fragment from Tsomet Shoket (TS-4 exterior) analysed as TMS derivatives (*, plasticisers; ax, *n*-alkane with chain length X; Ax, *n*-alcohol with chain length X; IS, internal standard; WEx, wax ester with chain length X).

fraction, there was a wider range of saturated fatty acids with 9–28 carbon atoms (electronic supplementary material S2). Primarily, these are long-chain and even-numbered, and they maximize at $C_{24:0}$. The *n*-alkanes in the TLE ranged from C_{23}–C_{37}, maximizing at *n*-heptacosane (C_{27}) with a predominance of odd-numbered *n*-alkanes (CPI 23.3). The *n*-alcohols are exclusively even and range from C_{24}–C_{32}, maximizing at 1-triacontanol (C_{30}). Wax esters are even-numbered and palmitate, with 40–50 carbon atoms, maximizing at C_{46}. Most of the wax esters (C_{42}–C_{50}) have a variant eluting immediately after. Based on the retention time, these should be hydroxy wax esters (e.g. [26,69]), but the mass spectras lack a clear base peak of *m*/*z* 117, deterring identification.

Additional markers link the sample to animal and plant products. In the acid fraction, high amounts of $C_{18:0}$, odd-numbered saturated fatty acids and branched-chain fatty acids are suggestive of ruminant adipose fat (electronic supplementary material, S2; table 2) [65,66]. $C_{18:1}$ and long-chain dicarboxylic acids with 14–30 carbon atoms were also identified in the acid fraction. Long-chain dicarboxylic acids are found naturally in different plant products (e.g. hardwood, nuts and some wild plants; [70–72]) and are unlikely oxidation products because very long-chain unsaturated fatty acids, which are rare, would be required to produce these. K_{33} was also identified in the solvent extract.

By contrast, in a control soil sample from the same context (BB-21 soil; electronic supplementary material S2; table 2), there are trace amounts of lipids. These include fatty acids ($C_{16:0}$–$C_{26:0}$), *n*-alkanes (C_{27} and C_{29}), *n*-alcohols (C_{16}–C_{30}), phytosterols and terpenoids (ferruginol, 2,3-dehydroferruginol, sempervirol and totarol). These are consistent with degraded plant products in the soil and do not match the archaeological signature. Specifically, the terpenoids may originate from certain conifers including *Cupressaceae* and *Cedrus* [73,74].

3.4. Jar rim fragment from Tsomet Shoket (TS-4)

In the jar from Tsomet Shoket, significant amounts of lipids were only identified in the TLE of the external surface (TS-4 exterior; figure 6; table 2), with traces of $C_{16:0}$, *n*-alkanes (C_{27} and C_{29}) and *n*-alcohols (C_{16} and C_{18}) in the interior sample (TS-4 interior layer 2; electronic supplementary material, S2; table 2). In the exterior, there are no fatty acids. *n*-Alkanes range from C_{25}–C_{33}, maximizing at triacontane (C_{31}). All are long-chain, and most are odd-numbered (CPI 16.5). *n*-Alcohols are even-numbered and range from C_{18}–C_{32}, maximizing at C_{30}. The palmitate wax esters are even with 40–46 carbon atoms, maximizing at C_{44}. Because the exterior was originally sampled as a control, it was not saponified, so it is unknown if fatty acids were bound to the ceramic matrix.

3.5. Comparison to modern beeswax

The identification of beeswax was reinforced by comparing the archaeological samples to fresh beeswax published in previous studies (figure 7) (following [13,15]). Fresh beeswax is characterized by an abundance of long-chain even-numbered fatty acids, long-chain odd-numbered *n*-alkanes, long-chain even-numbered *n*-alcohols and long-chain even-numbered palmitate wax esters [13,15]. As such, the

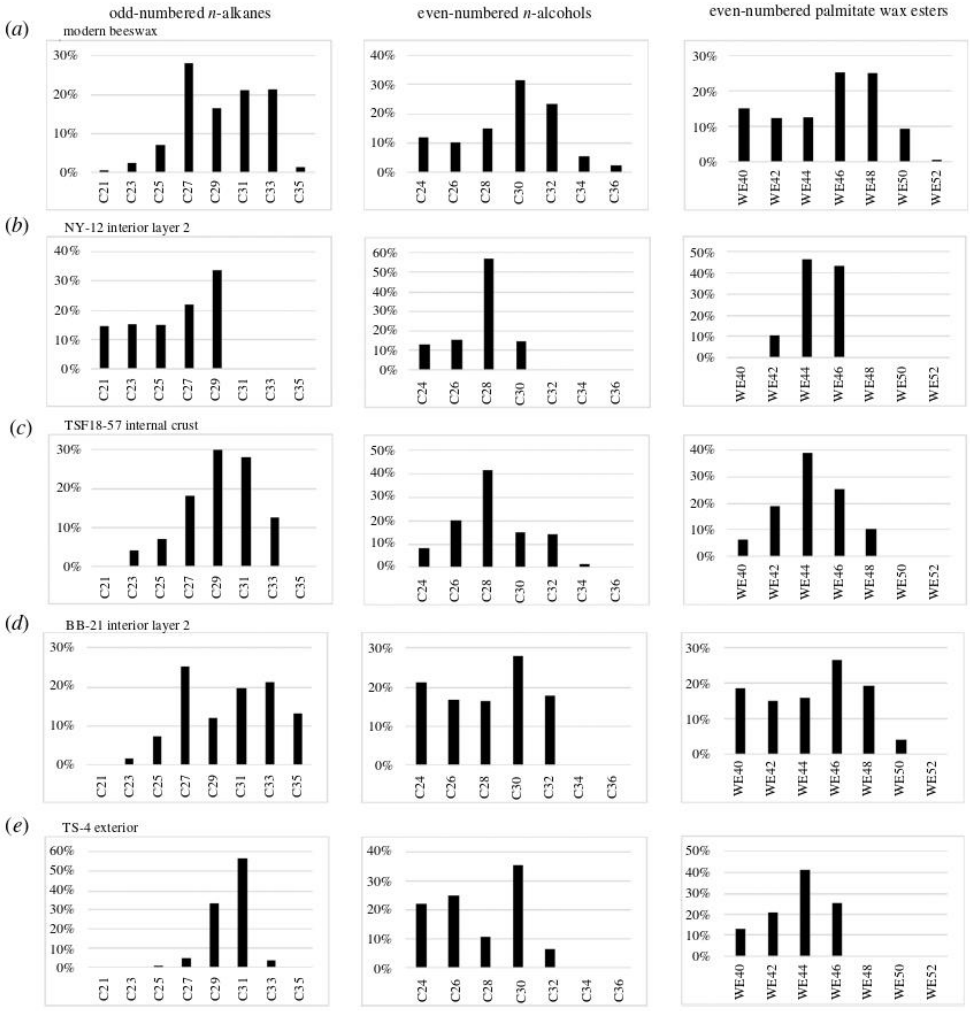

Figure 7. Histograms of the relative abundance of long-chain odd-numbered *n*-alkanes, even-numbered *n*-alcohols and palmitate wax esters of: (*a*) modern beeswax (following [13] tables 4, 6 and [15] table 2); and the archaeological samples with evidence for beeswax in this study: (*b*) Neve Yam; (*c*) Tel Tsaf; (*d*) Tel Bene Beraq (South) and (*e*) Tsomet Shoket.

fatty acid profile and the abundance of each odd-numbered *n*-alkane (C_{21}–C_{35}), even-numbered *n*-alcohol (C_{24}–C_{36}) and even-numbered palmitate wax ester (C_{40}–C_{52}) were calculated and compared. Parallels can be observed in all four samples, but the identification of beeswax for NY-12 and TS-4 is a bit more tenuous.

Comparison of the saturated fatty acid profiles to fresh beeswax [13–15] shows some clear parallels. In two of the solvent extracts and in all of the acid fractions, $C_{16:0}$ is abundant (although not always the maximum saturated fatty acid), reflecting hydrolysis of the palmitate wax esters. The long-chain even-numbered fatty acids characteristic of beeswax were primarily preserved bound to the ceramic matrix (electronic supplementary material S2; table 2), and in the unbound lipid fraction, they were unpreserved. In the TLE of the jar from Tsomet Shoket and the crust from Tel Tsaf, fatty acids are less common and may have been lost through sublimation [14].

Like fresh beeswax (figure 7*a*), in the archaeological samples, the *n*-alkanes are primarily odd-numbered and long-chain, maximizing at C_{27}, C_{29} or C_{31} (figure 7*b–e*). The shorter odd-numbered *n*-alkanes that are characteristic of beeswax were less common in the archaeological samples and were seemingly lost due to the decay processes. The longer-chain *n*-alkanes likely preferentially preserved due to their decreased volatility [14] and their greater abundance in fresh beeswax. In a few samples, there are short-chain *n*-alkanes that suggest bacterial input [75] and even-numbered *n*-alkanes with an unknown origin.

Comparison of the *n*-alcohol profiles to fresh beeswax (figure 7*a*) also shows distinct parallels. The *n*-alcohols in the ceramics are even-numbered and long-chain, consistent with the release of *n*-alcohols after hydrolysis of the palmitate wax esters (figure 7*b–e*). The source of the few short-chain and odd-numbered *n*-alcohols identified in the samples from Neve Yam, Tel Tsaf and Tsomet Shoket is unclear.

The long-chain even-numbered palmitate wax esters characteristic of fresh beeswax (figure 7*a*) [13] were also identified in the archaeological samples (figure 7*b–e*). Unusually, there seems to be a preferential loss of some of the longer wax esters, with the C_{52} wax ester never preserved and the C_{48} and C_{50} wax esters absent or in low frequencies; this is seen most clearly in the samples from Neve Yam and Tsomet Shoket (figure 7*b*,*e*). The lack of the longer-chain wax esters in these samples may relate to their low abundance in fresh beeswax (figure 7*a*).

These patterns differ from plant waxes. Plants have similar *n*-alkane and *n*-alcohol profiles to beeswax [62,76], but the wax esters differ, comprising 32–64 carbon atoms [77], and some plant waxes may be further distinguished by the presence of non-palmitate and unsaturated wax esters (e.g. [78,79]). Such wax esters were not identified in the archaeological samples characterized as beeswax. Additional palmitate wax esters were identified in 15 other vessels, but in these vessels, the lipid yield was too low or the suite of markers present was insufficient to confidently identify beeswax. While these may represent highly degraded beeswax, some of these vessels contain exclusively wax esters with 24–38 carbon atoms that are suggestive of a different wax origin. These wax esters were identified in specific vessel forms including strainers, cornets and churns from Late Chalcolithic sites, with none identified in jars where beeswax markers were found.

It is highly significant that only four of the 247 vessels tested here (13.3% of the internal, crust and external samples with significant lipid yields) contained evidence for beeswax. This result is consistent with previous work carried out in the southern Levant, which identified few to no instances of lipids characteristic of beeswax preserved in pottery (table 3). Despite the large number of pottery vessels analysed from past and ongoing studies, totalling 467 vessels, beeswax residues were suggested for only 3% of the vessels tested or 35% of the vessels with over 5 $\mu g\ g^{-1}$ of lipids in the solvent extracted interiors, crusts and exteriors (figure 1*b* and table 3). This frequency is even lower if the identifications by Namdar [38] based exclusively on the *n*-alkane profiles are ignored (10% of samples with over 5 $\mu g\ g^{-1}$ of lipid). While the frequency of beeswax residue in sherds with significant lipid yields is higher than at some contemporary sites in Eurasia and North Africa [20], it certainly cannot be considered commonplace as it was identified at very few sites. The true frequency is also complicated by samples for which the lipid concentration was not calculated [81–83], and it may be masked by preservation biases.

4. Discussion and conclusion

The lipids preserved in the three jars and the crust from Neve Yam, Tel Tsaf, Tel Bene Beraq (South) and Tsomet Shoket mark some of the earliest evidence for beeswax use in the southern Levant. Based on the current study and a previous study [38], beeswax was already used throughout Israel in small amounts during the Late Chalcolithic period in relation to jars and potentially cornets. However, as we demonstrated above, beeswax utilization goes back to the Early and Middle Chalcolithic. While a confident biomolecular identification of degraded beeswax is possible, discerning the exact function or functions of beeswax is more difficult to establish. This is particularly true as the vessels under discussion appear to have been used alongside beeswax as well as animal fat and plant products. The specific use of beeswax is only easily identifiable when the vessel form is task-specific (e.g. clay hives and lamps). In this study, several suggestions can be put forward to explain the use of the beeswax in the tested vessels.

The identification of beeswax in only the exterior surface of the jar from Tsomet Shoket suggests that it was used as part of a post-firing treatment to alter the external vessel surface, possibly to form lustre. By rubbing it on a surface, polish is created, enhancing the vessel's aesthetics [1]. This technique is applied today to a variety of materials, including wood, leather, stone and textiles [1], and written sources show that this technique was used already during the Classical period in the Mediterranean; melted beeswax was mixed with various plant oils to form coatings on marble and wood to create shine [1, Pliny, XXXIII.122]. Such a function for ceramics is difficult to support because the visible wax would not survive diagenesis.

Beeswax could also be applied to ceramic vessels internally and externally as a sealant or to fill in cracks. Beeswax can be applied post-firing to make the vessel water resistant because of its

Table 3. Summary of ORA results obtained from 7th–4th cal. BC sites in the southern Levant and the number of vessels with suggested evidence for beeswax residues (sites tested for the current study are marked in bold).

site	period	N vessels analysed	N vessels and crusts with > 5 µg g^{-1} of lipids in the solvent extract internally	N vessels with > 5 µg g^{-1} of lipids in the solvent extract exclusively externally	N vessels with evidence for beeswax	reference
Abu Thawaab	Pottery Neolithic	10	0	—	0	[80]
'Dhra	Pottery Neolithic	24	0	—	0	[80]
Munhata	Pottery Neolithic	13	0	—	0	[80]
Sha'ar HaGolan	**Pottery Neolithic**	16	0	—	**0**	[80]
		16	**1**	**1**		**current study**
al-Basatin	Early Chalcolithic	7	0	—	0	[80]
Ein Zippori[a]	Early Chalcolithic	3	—	—	0	[81]
Neve Yam	**Early Chalcolithic**	**17**	**1**	—	**1**	**current study**
		33	0	—		[80]
Tel Hreiz	**Early Chalcolithic**	**3**	**1**	—	**0**	**current study**
Tabaqat al-Buma	Early Chalcolithic	15	0	—	0	[80]
Tel Tsaf	**Middle Chalcolithic**	**100**	**14**	**1**	**1**	**current study**
Tubna	Middle Chalcolithic	22	0	—	0	[80]
Abu Hof[a]	Late Chalcolithic	10	—	—	0	[82]
Abu Matar	**Late Chalcolithic**	14	0	—	**0**	[80]
		3	**0**	**0**		**current study**
Ashqelon Agamim	**Late Chalcolithic**	**28**	**1**	**0**	**0**	**current study**
Azor	**Late Chalcolithic**	**1**	**0**	**0**	**0**	**current study**
Tel Bene Beraq (South)	**Late Chalcolithic**	**10**	**1**	**1**	**1**	**current study**
Bir es-Safadi	**Late Chalcolithic**	**11**	**0**	**0**	**0**	**current study**

(*Continued.*)

Table 3. (*Continued.*)

site	period	N vessels analysed	N vessels and crusts with > 5 µg g^{-1} of lipids in the solvent extract internally	N vessels with > 5 µg g^{-1} of lipids in the solvent extract exclusively externally	N vessels with evidence for beeswax	reference
Ein Gedi	Late Chalcolithic	10	6	—	6	[38]
Fazael 7	**Late Chalcolithic**	**8**	**0**	**0**	**0**	**current study**
Gilat	**Late Chalcolithic**	**13**	**3**	**2**	**0**	**current study**
		10[a]	—	—		[83]
Grar	Late Chalcolithic	4	2	—	2	[38]
Horvat Qarqar	Late Chalcolithic	4	0	—	0	[38]
Moringa Cave	Late Chalcolithic	4	2	—	2	[38]
Nahal Qomem	Late Chalcolithic	6	0	—	0	[38]
Nevatim	Late Chalcolithic	4	0	—	0	[80]
Nisim Aloni	**Late Chalcolithic**	**9**	**0**	**0**	**0**	**current study**
Peqi'in	**Late Chalcolithic**	**3**	**0**	**0**	**0**	**current study**
Tsomet Shoket	**Late Chalcolithic**	**18**	**0**	**1**	**1**	**current study**
Shiqmim[a]	Late Chalcolithic	11	—	—	0	[82]
Yehud	**Late Chalcolithic**	**7**	**1**	**1**	**0**	**current study**
Total		467	33[b]	7	14	

[a]The lipid concentration was not calculated, so preservation cannot be discussed.

[b]The number of samples with a TLE over 5 µg g^{-1} cannot be calculated accurately as the TLE was not reported in some published studies [81–83].

hydrophobic nature [1]. This has been frequently suggested in ORA studies that identified beeswax [9,21,30,44,84]. A beeswax sealant would benefit vessels meant to store liquids, and correspondingly, beeswax biomarkers were identified in vessel forms related to storage and liquids—jars in this study and amphorae, juglets, flasks and chalices in previous studies [24,44,84]. Cooking vessels would likely not receive a beeswax sealant because beeswax melts at 62–65°C [15], so the sealant would need to be frequently reapplied, and the flavour and texture of the sealant could be undesirably incorporated into the cooked food.

Beeswax present in the interior surface of the two jars from Neve Yam and Tel Bene Beraq (South) and the crust from Tel Tsaf may additionally relate to the storage and use of unfiltered honey. The signature of filtered honey would be unlikely to preserve because honey is formed primarily from carbohydrates [85], which easily degrade [57]. Honey is a desirable sweetener [1] with a long history of use. Honey consumption in the wider area was documented as far back as 2300–1300 BC, with examples noted from Egyptian texts and the Bible (e.g. [1,39,86,87], Deuteronomy 32.13, Judges 14.8, Psalm 81.16. Samuel 14.25-27). Honey may also be used as a preservative for fruits, vegetables and even meat. This practice is documented back to the Classical period in the Roman cookbook – *Apicus* [88]. While these examples post-date the Chalcolithic period by several millennia, they suggest that honey had a clear role in the wider Mediterranean diet, and an earlier root is expected. Further, if the residue relates to the use of honey, then when the entire lipid profile is viewed together, we may suggest that honey was used in complex recipes with various plant and animal products. A purer and more intensive utilization can be suggested for the jar from Tel Bene Beraq (South) because the saturated fatty acid, *n*-alkane, *n*-alcohol and wax ester signatures more directly matches those of fresh beeswax (figure 7*a*,*d*).

Another possible function for beeswax identified in internal samples from Neve Yam, Tel Tsaf and Tel Bene Beraq (South) is the use of unfiltered honey in medicine. Honey is renowned for its anti-inflammatory and antibacterial properties and is frequently used in ointments [1,12,89]. Using honey in medicine in the Levant is documented in several papyri from Egypt (dated to *ca* 1900–1550 BC). They show that honey was used as an ointment, a laxative and as part of contraception [90,91]. Because medicinal use is based on textual sources, it cannot be linked to vessel forms. However, studies on vessels from significantly later archaeological sites in Europe identified ointments in miniature vessel forms [4,92,93]. The small vessel size likely relates to the ointment's value, and it is therefore unlikely that medicine would be stored in the vessels discussed in this paper, which include three comparatively large jars.

Vessel surface alteration and a dietary role specifically may be further supported by the co-occurrence of beeswax residue with other lipids such as ketones, which are often used to link archaeological samples to cooking (e.g. [94]) and post-firing surface treatments [60]. 16-hentriacontanone (K_{31}), 16-tritriacontanone (K_{33}) and 18-pentatriacontanone (K_{35}) form by heating palmitic and stearic acid to temperatures over 300°C or at lower temperatures for extended time [60,94]. The two potential stimuli for ketone synthesis are difficult to distinguish and are not mutually exclusive. However, in the prehistoric southern Levant, certain jars are often linked to cooking based on the occasional presence of soot marks on the exterior [52], and it is therefore possible that the jars from these sites were similarly used for cooking, melting beeswax or heating honey. It is also possible that the single ketone homologue (K_{31}) identified in the sample from Tel Tsaf relates to a plant wax; plant waxes are generally dominated by one homologue that is exclusively symmetrical [94].

Outside of diet and foodways, beeswax can be used as fuel for lighting [2]. This was suggested in previous ORA studies on zoomorphic vessels from Neolithic Greece [23] and lamps and conical cups from Late Minoan Crete [95]. These vessels are shallow with an open form, ideal for holding small contents of fuel. Analogous vessel forms were not identified in the prehistoric southern Levant [52], but a similar function was attributed to cornets based on ORA [38] (although the identification of beeswax is debatable). While the vessels are not shallow, the conical form is suitable for holding small amounts of wax. The vessels under discussion in this study have a much greater volume and depth and were likely unsuitable for lighting.

The use of beeswax during the Late Chalcolithic period was also mentioned in relation to copper production using the lost-wax technique [11]. Fragments of the moulds required for the technique have not yet been recovered from Chalcolithic sites. Therefore, while beeswax clearly played a role in copper production, at present without the moulds, the direct connection between beeswax and metallurgy cannot be discussed using ORA. Beeswax biomarkers were also so far not identified in pottery vessels analysed by GC-MS from sites with evidence for copper production (e.g. Abu Matar, Fazael and Shiqmim; table 3) [80,82,96,97], but this cannot be used to discredit beeswax's role in the industry at these sites.

Regardless of how bees and bee products were used, evidence from the lipid record suggests that bee products were used in low frequencies. Evidence for beeswax was only found in four out of the 247 vessels tested as part of this study (13.3% of vessels with significant lipid yields internally or externally) or, if the results of prior studies based on *n*-alkane profiles are accepted [38], as much as 14 out of the 467 vessels analysed by GC-MS (35% of vessels with significant lipid yields internally or externally) (table 3). A low utilization must be suggested cautiously because lipids poorly preserve in the southern Levant (e.g. table 3; [80,98,99]) and bee products may have been used and stored in organic containers [1,100]. Therefore, the lack of evidence does not necessarily imply lack of use. Nonetheless, the number of sites with beeswax residues in the southern Levant (7/30 tested sites) is too little to consider a potential widespread utilization in the region. The absence is particularly clear at Tel Tsaf, from which 100 ceramic vessels were tested, but only one sherd yielded evidence for beeswax (table 3). This absence is paralleled in the northern Levant at Tel Sabi Abyad, from which 287 vessels were sampled and none of the 41 vessels with significant lipid yields contained evidence for beeswax [101].

The limited use of bee products is noteworthy because they have numerous benefits, and even without beekeeping, wild bees could have been exploited (e.g. [102,103]). *Apis mellifera* was certainly present in the region as it has a wide distribution, found in wet, dry, cold and warm environments in Africa, Europe and parts of Asia [104]. Why then were beeswax and honey rarely used in the prehistoric southern Levant? This question is particularly important as only a few millennia later there is textual evidence for bee exploitation, and apiaries were identified at Tel Rehov [41], but the organic and archaeological records offer no clear root for this extensive bee exploitation.

A potential hypothesis to explain this low use is that beeswax exploitation contrasted with the Chalcolithic socio-economic system. The Chalcolithic period is characterized by agro-pastoralism [105], with minimal evidence for hunting, gathering or fishing, even when wild resources were readily accessible [106,107]. Following the same trend, people likely exploited wild bee products in low amounts. Despite the numerous functions and benefits of bee products, the Chalcolithic communities may have used alternatives that were more complementary to their socio-economic system. Alternatives can be suggested in ceramic vessel surface treatment and foodways.

As shown previously, beeswax can be applied to ceramics as a sealant or to form a shiny surface. During the Chalcolithic period, ceramics were altered primarily with paint applied with a brush or a slip or by burnishing [52], and there is at present no evidence for techniques that employed organic materials (e.g. [60]). Shine, when present, was created primarily by burnishing using another object [52]. Furthermore, because the Chalcolithic period is characterized by increasing standardization in craft production (e.g. [108,109]) and within this ceramic production (e.g. [49]), techniques that did not conform to the technological norm, such as the application of beeswax, were likely used by limited individuals. The example from Tsomet Shoket may represent an exceptional case where beeswax was used as part of a surface treatment, especially considering that the interior and exterior bear no visible surface treatments (figure 6).

Evidence for alternatives in the Chalcolithic cuisine are difficult to identify because of biases toward charred seeds in botanic preservation, but food could be seasoned with wild herbs and plants and sweetened with wild fruits (e.g. figs and dates; [110]). One must also consider that sweet foods, while now commonplace, were originally considered a luxury in various cultures [1,111]. Honey's value in the wider Levant is evident because many civilizations used it as an offering to gods. This practice can be dated as far back as 2450 BC in Sumer, 1567–1085 BC in Egypt and 700 BC in Israel [1, II Chronicles 31.5]. Considering its supposed value, honey was likely not part of the daily diet, although this must be suggested with caution because of preservation biases against carbohydrates—the main component of honey [57,85].

To conclude, the current study brings direct evidence for early use of beeswax and, by association, the likelihood of the use of honey during the Chalcolithic period of the southern Levant. These seemingly played a minor role, with the potential exception of the copper metallurgy industry. The exact function of the vessels where beeswax was found is difficult to identify because the various uses of bee products leave identical lipid signatures. However, based on the vessel forms and comparison to the wider Eastern Mediterranean record, it can be suggested that the vessels were used to contain beeswax or honey or that the beeswax was applied to their surface to alter its appearance and properties. Our results, bearing in mind preservation issues, allow us to cautiously suggest that bee products were not frequently produced and used (at least not in an organized fashion) during the Chalcolithic period. This was probably the situation until the Iron Age, during which apiaries were clearly in use. Thus, while the ceramic vessels tested here mark an early evidence for bee product

residues in ceramics, they by no means mark the root of extensive bee exploitation that appeared millennia after the fading of the Chalcolithic period. Further analysis of additional vessels from Bronze Age sites is required to elucidate the trajectory of bee exploitation in the region and identify when bees became an important part of the economy.

Data accessibility. All data and research materials supporting the results are in the article and electronic supplementary material.

Authors' contributions. R.C. conceptualized the study, carried out the laboratory work and data analysis and drafted the manuscript. D.R. helped conceptualize the study, draft the manuscript and acquisition funds. D.R., F.K., R.B., D.G., A.D. and E..G. provided the ceramic vessels for analysis. C.S. supervised the GC-MS analysis and helped conceptualize the study and draft the manuscript. All authors gave final approval for publication and agree to be held accountable for the work performed therein.

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Foodways of an agro-pastoral community: organic residue analysis of pottery and stone vessels at Middle Chalcolithic Tel Tsaf

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Name of Co-Author	Florian Klimscha
Contribution to the Paper	F. Klimscha provided ceramic vessels for analysis and helped draft the manuscript.
Name and Signature	 Florian Klimscha Date: 28 06 2022

Name of Co-Author	Cynthianne Spiteri
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Name and Signature	Cynthianne Spiteri  Date: 04/07/2022

Name of Co-Author	Danny Rosenberg		
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Foodways of an agro-pastoral community: Organic residue analysis of pottery and stone vessels at Middle Chalcolithic Tel Tsaf

Rivka Chasan [a,*], Florian Klimscha [b], Cynthianne Spiteri [c], Danny Rosenberg [d]

[a] Laboratory for Ground Stone Tools Research, Zinman Institute of Archaeology, University of Haifa, Haifa, Israel
[b] Archaeology Division, Research/Collections, Lower Saxony State Museum, Hanover, Germany
[c] Institute of Pre- and Protohistoric Archaeology, Eberhard Karls Universität Tübingen, Tübingen, Germany
[d] Laboratory for Ground Stone Tools Research, Zinman Institute of Archaeology, University of Haifa, Haifa, Israel

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ABSTRACT

Tel Tsaf is a Middle Chalcolithic (ca. 5200–4700 cal BC) site located in the central Jordan Valley, Israel. The site reflects increasing long-distance trade, organized cereal crop cultivation and possibly olive horticulture. Organic residue analysis of lipids recovered from 100 pottery vessels and three stone vessels and comparison to the botanical and faunal remains suggest that the vessels were used to contain or process various combinations of domestic animal and plant products and that these food products were supplemented by local wild plants. This shows direct continuity of dietary traditions from the preceding Neolithic period; however, there were some developments within the suite of domesticated resources, including the earliest direct evidence for milk exploitation in the southern Levant. Culinary traditions incorporated recipes where these food products were cooked and consumed together in various combinations. These dietary patterns were a part of daily life at Tel Tsaf, adding another layer to our understanding of the village and the culinary traditions of the Middle Chalcolithic period in the southern Levant.

1. Introduction

The later prehistory of the southern Levant is discussed through the lens of increasing socio-economic complexity (e.g. Ben-Shlomo and Garfinkel, 2009; Gopher, 2012, pp. 1542–1543; Rowan and Golden, 2009). Research emphasizes the transition from the Pottery Neolithic to the Late Chalcolithic period, which is renowned for the development of metallurgy, increasing craft specialization and a widening symbolic repertoire (Rowan and Golden, 2009 and see references therein). Within this debate, the Middle Chalcolithic period bridges the gap between the Early Chalcolithic and the Late Chalcolithic periods, forming a timespan of approximately 500 years (ca. 5200–4700 cal BC). During the Middle Chalcolithic period, there are several regionally bound cultural entities defined by site clusters and unique material culture, forming islands of distinct cultural phenomena (Garfinkel et al., 2020, pp. 19–28; Gilead, 2011; Gopher, 2012).

The socio-economy and dietary patterns of the Middle Chalcolithic/Post-Wadi Rabah-Pre-Ghassul are minimally discussed in the literature, and most discussions are centered on a single site – Tel Tsaf (Ben-Shlomo et al., 2009; Garfinkel et al., 2009; Graham, 2014; Hill, 2011; Rosenberg and Klimscha, 2018). The faunal remains from Middle Chalcolithic sites reflect a reliance on caprines, cattle and pigs, with a predominance of sheep and goats (Baird et al., 1994, table 2; Hill, 2011, pp. 108; Milevski et al., 2015, table 7; Nativ et al., 2014, table 3). Botanical remains are seldom preserved or recovered, but there is some evidence for the founder crops – emmer wheat, barley and a variety of pulses (Graham, 2014; Meadows, 2005, pp. 121–123, table 5.6).

These faunal and floral remains represent a narrow window into the foodways of this timespan. What people eat and how they eat it results from individual decisions made within an array of social contexts and environmental constraints (Twiss, 2012). The analysis of foodways identifies how these foods were selected, produced, stored, processed, served and consumed as well as the greater picture framing these practices – culinary traditions, economy and environmental regulations (e.g. Bray, 2003; D'Altroy and Earle, 2002; Gumerman IV, 1997; Smith, 2006; Twiss, 2012). Consequentially, a more in-depth analysis of the Middle Chalcolithic foodways offers enhanced understanding of dietary habits, vessel and tool use and the factors that constrained these choices.

* Corresponding author.
E-mail addresses: rchasan@campus.haifa.ac.il (R. Chasan), florian.klimscha@landesmuseum-hannover.de (F. Klimscha), cynthianne.spiteri@uni-tuebingen.de (C. Spiteri), drosenberg@research.haifa.ac.il (D. Rosenberg).

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Tel Tsaf, the most widely researched Middle Chalcolithic site, can be used as a case study to elucidate the foodways of this timespan in the southern Levant. The site is dated to ca. 5200–4700 cal BC (Garfinkel et al., 2020, pp. 87) and is in the central Jordan Valley, Israel (Fig. 1). Tel Tsaf features intensive mudbrick constructions (Garfinkel et al., 2020, pp. 117–211; Rosenberg et al., 2020) and evidence for high-capacity grain storage (silos). Evidence for long-distance trade was also found (Garfinkel et al., 2007, 2009, 2014, 2020, pp. 219–225; Rosenberg and Klimscha, 2021; Rosenberg et al., 2014, 2021): Nilotic shells from Egypt and obsidian, Ubaid pottery and potentially specific pottery decoration styles from the northern Levant (Garfinkel et al., 2007, 2009). The silos and trade connections were interpreted as evidence for the reorganization of control over agricultural yields and increasing socio-economic complexity (Garfinkel et al., 2007, 2009, 2020, pp. 219–225). Tel Tsaf is associated with other sites in the Jordan Valley (Fig. 1) through parallels in the ceramic industry, including specifically the decoration style of the painted wear (Garfinkel et al., 2020, pp. 27, fig. 1.1).

Recent excavations at Tel Tsaf (Rosenberg and Klimscha, 2018, 2021; Rosenberg et al., 2014, 2020, 2021) focus heavily on identifying the site economy using multiple complementary lines of evidence. The occurrence of foodways similar to the 'Mediterranean diet' is hypothesized based on the emphasis on cereals, legumes and olives in the botanical remains (Graham, 2014; Rosenberg and Klimscha 2021; Rosenberg et al., 2021). The modern Mediterranean diet is characterized by a high consumption of vegetables and a lower intake of animal products. Olive oil is at the center of this diet as the main fat source (Ortega, 2006). The exploitation and domestication of olives in the protohistoric southern Levant is a focal area of research (e.g. Dighton et al., 2017; Liphschitz et al., 1991; Neef, 1990) because olives are economically significant; in addition to being consumed whole, they can be processed into oil, pickled, stored and traded (Genz, 2003; Salavert, 2008). Oil processing and the consumption of whole olives may be suggested at Tel Tsaf based on the co-occurrence of fragmented and complete olive stones; however, the scale of olive processing and consumption is unclear.

The current study aims to identify culinary practices at Tel Tsaf by applying organic residue analysis to a corpus of ceramic and stone vessels and linking the identified signatures to the faunal and floral remains. Organic residue analysis has been used in the past to identify a variety of animal fats and plant oils, including fish, dairy products and suggestions for olive oil (e.g. Copley et al., 2005; Cramp and Evershed, 2014; Debono Spiteri et al., 2016; Evershed et al., 2008; Namdar et al., 2014) – key components of the Mediterranean diet. By integrating our biomolecular results with the wider context of plant and animal exploitation systems, we discuss the foodways of the community at Tel Tsaf and their relation to the wider Middle Chalcolithic socio-economy.

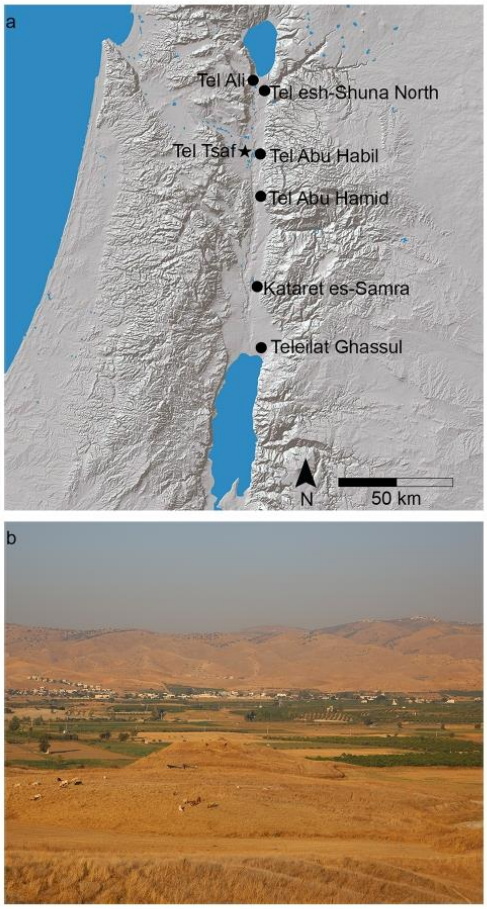

Fig. 1. Tel Tsaf: a) Map showing the location of Tel Tsaf and contemporary sites; b) A view of the site, looking east.

2. Materials and methods

One hundred pottery vessels and three stone vessels were sampled from various contexts at Tel Tsaf (Figs. 2 and 3; SI 1). The samples include typologically diverse bowls, jars and platters as well as non-diagnostic fragments; no emphasis was placed on particular forms as the ceramics from Tel Tsaf are often highly fragmented with little of the original vessel preserved, so it is difficult to identify typology. Most were collected from Sq. AR16 in Area C, a 20 m^2 probe, and from within this area, pottery fragments from accumulations, floors, hearths and a wall were selected. Samples were selected from this square, which was excavated to define the site stratigraphy, in order to sample from the entire archaeological sequence.

Lipids from the vessel interiors and a crust were extracted. Distinct crusts adhering to ceramic vessels are uncommon at Tel Tsaf, but this unique example was sampled because food crusts often contain a rich lipid signature related to the final vessel use (Miller et al., 2020). Samples from the external surfaces of the vessels were tested as controls, together with a soil sample taken from the area. The lipids were extracted using established protocols, namely solvent extraction (Mottram et al., 1999) and alkaline hydrolysis (Stern et al., 2000), which were applied to recover both the free and bound lipids. Samples were analyzed by Gas Chromatography-Mass Spectrometry (GC–MS), and all extracts with sufficient palmitic and stearic acid underwent Gas Chromatography-combustion-Isotope Ratio Mass Spectrometry (GC-c-IRMS). The GC-c-IRMS results were compared to a modern global reference database of plant oils, domestic ruminant adipose and milk and domestic pig adipose and their mixing curves (following Woodbury et al., 1995). A detailed description of the sampling criteria and the methodology followed are provided in the supplementary information (SI 2).

3. Results

3.1. Pottery

The average lipid yield after solvent extraction was 6.1 μg g^{-1} (ranging from 0.0 to 300.6 μg g^{-1}). Of the 100 vessels tested, 13% of the internal samples yielded a significant and interpretable residue, containing a total lipid extract (TLE) of over 5.0 μg g^{-1} of lipids (following

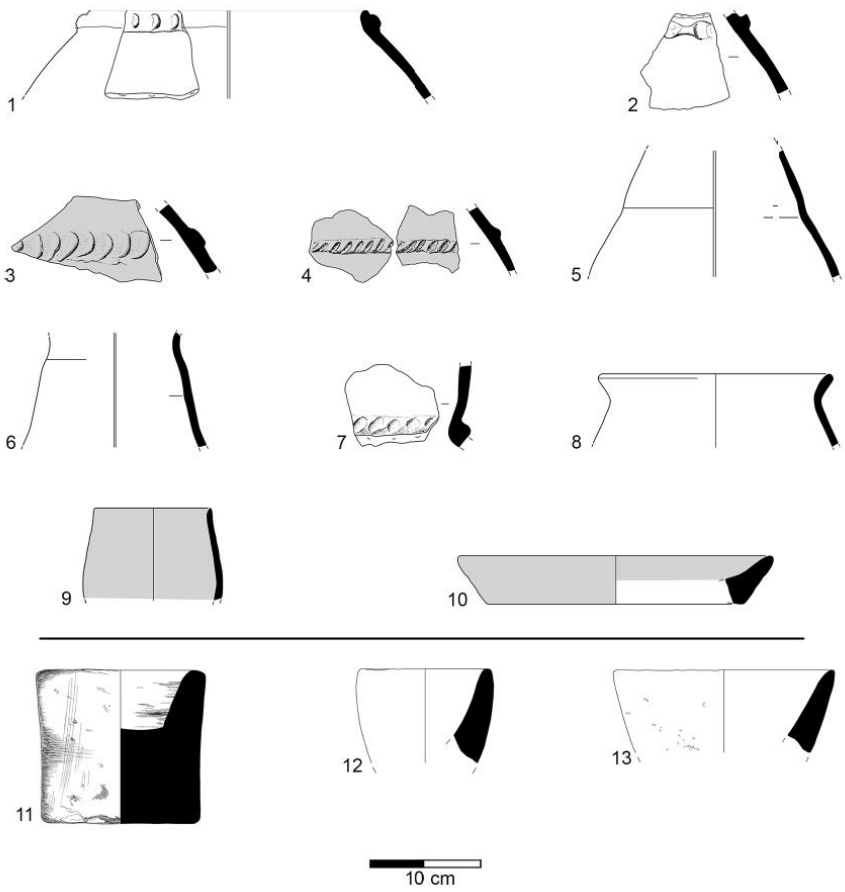

Fig. 2. Selection of sampled vessels from Tel Tsaf representative of the ceramic and stone vessel assemblage. This includes: Holemouth jars: 1) TSF17-4; 2) TSF17-1; 3) TSF18-45; 4) TSF18-59; 5) TSF17-30; 6) TSF17-16; Necked jars: 7) TSF18-73; 8) TSF19-3; Bowls: 9) TSF18-31; Platters: 10) TSF18-58; Stone bowls: 11) TSF19-14; 12) TSF17-9; 13) TSF19-13.

Evershed, 2008a). The sole crust sampled also contained a significant amount of lipids. The low preservation may be attributed to the environmental and sediment conditions. Tel Tsaf is in the Jordan Valley close to Beit Shean, with an average monthly temperature range of between 2°C and 42°C and an average annual rainfall of 270 mm (Israel Meteorological Service a, b). The pH of anthropogenic sediments at the site ranges from 7.71 to 8.06 (Hubbard, 2015, table 3.2). Both factors support microbial activity (Evershed, 2008b; Moucawi et al., 1981), which likely negatively affected lipid preservation. Quantification of the bound acid fraction obtained after alkaline hydrolysis displayed a higher lipid yield, averaging at 18.7 $\mu g\ g^{-1}$ (ranging from 0.5 to 195.0 $\mu g\ g^{-1}$). 54% of the internal samples and the crust have over 5.0 $\mu g\ g^{-1}$ of lipids. This shows that while the free lipids rarely preserved, the lipids polymerized to the ceramic matrix survived. The extraction method chosen was therefore essential to assess the entire lipid profile.

In the external control samples and the soil sample, negligible amounts of lipids were generally observed. Two vessels (TSF17-22 and TSF17-28) contained significant lipid yields in their external surface, with only one of these (TSF17-22) containing significant amounts of lipids internally as well. This can relate to the application of a treatment to the external vessel surface, spillage of the original vessel content or to lipids seeping through to the external surface due to the porosity of the ceramic. The soil sample contained a fingerprint characteristic of degraded plants and microbial activity (e.g. Heron et al., 1991). The process blanks extracted alongside the archaeological samples are devoid of lipids, showing that contamination did not occur during the extraction procedures.

A few obvious contaminants were identified in variable amounts. The primary example is plasticizers. The residue of sunscreen was also identified in one solvent extract (TSF19-9), and this sample was not analyzed further. High amounts of cyclic octaatomic sulfur were identified as well. While not a clear contaminant, this likely reflects the local saline groundwater (Salameh, 2001). Cyclic octaatomic sulfur was probably incorporated post deposition from the sediment or during ceramic production as part of clay processing (Reber et al., 2019). A variety of other lipids linked to the original vessel use was also identified.

3.1.1. Fatty acids

In the solvent extracts, saturated fatty acids with 6–30 carbon atoms were identified in nearly every sample. Most however contain only high amounts of palmitic ($C_{16:0}$) and stearic ($C_{18:0}$) acid. There is no clear

Fig. 3. Tel Tsaf: a) Roasting pit (Locus 2044) and surrounding mudbrick construction from which vessels TSF18-68 and TSF18-69 originated; b) *in situ* photo of a base fragment (TSF17-7); c) *in situ* photo of a holemouth jar rim fragment (TSF17-27).

preference for palmitic over stearic acid or *vice versa*, which could be used to suggest a plant or animal fat source (Copley et al., 2005). The very long-chain fatty acids are primarily even. Many samples contain unsaturated fatty acids. This is mainly restricted to $C_{18:1}$ in trace amounts; $C_{20:1}$ is noted in two samples (TSF17-22 and TSF18-51). The position of the double bond of the unsaturated fatty acids could not be identified because of the lack of the oxidation products.

In most acid fractions obtained after alkaline hydrolysis of the solvent extracted ceramic samples, there is a wide range of odd and even saturated fatty acids. These range from $C_{6:0}$–$C_{30:0}$, and palmitic and stearic acid are the most abundant, although other fatty acids are also profuse. Pentadecylic acid ($C_{15:0}$) and margaric acid ($C_{17:0}$) are particularly abundant. These are commonly related to ruminant fat, forming in the rumen through chain length reduction of long-chain even-numbered fatty acids (Doreau and Ferlay, 1994).

Unsaturated fatty acids were identified in nearly every acid fraction, but the specific isomer of these is unknown. This includes primarily $C_{16:1}$ and $C_{18:1}$. The ratio of $C_{18:1}$ to $C_{16:0}$ and $C_{18:0}$ can be used to identify olive oil; in fresh olive oil, oleic acid ($C_{18:1}$ *cis*-9) is the most abundant fatty acid (Dudd et al., 1998; Paiva-Martins and Kiritsakis, 2017). In the samples from Tel Tsaf, $C_{16:0}$ and $C_{18:0}$ are always more abundant than $C_{18:1}$, and the $C_{18:1}$ isomer could not be specified. Furthermore, $C_{18:1}$ may also be identified in other plant oils and animal fats (Mills and White, 1999, pp. 33; Pollard and Heron, 2008, table 11.2). Therefore, elucidation of the $C_{18:1}$ source was not possible. If the Tel Tsaf vessels were used in relation to olive oil, the related lipid signature was not preserved. The lack of direct evidence for olive oil is particularly interesting because hundreds of olive seeds were uncovered at Tel Tsaf (Rosenberg et al., 2021).

A smaller number of samples contain unidentified isomers of $C_{18:2}$ and $C_{18:3}$, which may be linked to plant products. Linoleic acid ($C_{18:2}$ *cis*, *cis*-9, 12) in particular is found in olive oil (Paiva-Martins and Kiritsakis, 2017, pp. 83–84), although it can be found in other plant oils and some animal fats (Mills and White, 1999, pp. 33). An additional set of samples contain $C_{20:1}$ and $C_{22:1}$. These could be related to seed oils from *Brassicaceae* plants, which contain high amounts of gondoic ($C_{20:1}$ *cis*-11) and erucic ($C_{22:1}$ *cis*-13) acid (Colombini et al., 2005). The primary example is mustard seed oil, with various types of mustard seed plants local to Israel (Danin, 2004, pp. 111). No oil producing seeds were so far found in the Tel Tsaf botanical assemblage (Graham, 2014), and the specific isomers of $C_{20:1}$ and $C_{22:1}$ were not identified, so the source of the unsaturated fatty acids is unclear. $C_{20:1}$ and $C_{22:1}$ can also originate from fish oil; however, fish contain multiple isomers of these fatty acids (Shimizu and Ando, 2012, table 3), and in each sample, only one isomer was identified. Therefore, a marine origin is unlikely, although potential degradation of the various isomers must be considered.

Iso and *anteiso* branched-chain fatty acids ($C_{14:0}$–$C_{18:0}$) were identified in over half of the vessels, with a predominance of $C_{15:0}$ and $C_{17:0}$, and they were commonly identified alongside odd-numbed saturated fatty acids. These branched-chain fatty acids may form from microbial activity in the rumen (Doreau and Ferlay, 1994), so these may be linked to ruminant fat. Soil bacteria should also be considered as a potential source (Amblès et al., 1994) that we cannot rule out, so these are not definitive markers in this context.

3.1.2. Dicarboxylic acids

Sebacic acid was identified in one solvent extract of a jar (TSF19-4). This may originate from the oxidation of ricinoleic acid (Naughton, 1974), although this hydroxy acid is not found in plants identified at Tel Tsaf (Graham, 2014). Additionally, four methylated acid fractions have azelaic acid (TSF17-7, TSF17-23, TSF18-13 and TSF18-24). This forms from the oxidation of unsaturated C_{18} fatty acids (Passi et al., 1993), which are also found in these samples. C_{18} unsaturated fatty acids are found in both plant and animal products (Pollard and Heron, 2008, table 11.2), so azelaic acid does not suggest a particular lipid origin.

Few samples (TSF17-23, TSF17-28, TSF17-31, TSF18-2 and TSF19-3) contain long-chain dicarboxylic acids in the acid fraction, ranging from C_{16}–C_{26}, with an even over odd preference (Fig. 4). These are unlikely to be oxidation products because very long-chain unsaturated fatty acids would be required to produce these; rather, they may originate naturally from plant products, including nuts (Dembitsky et al., 2002) and certain hardwood trees (Fine et al., 2002; Gandini et al., 2006). Nut oils are an unlikely source as their identified dicarboxylic acids range is from C_{14}–C_{22} (Dembitsky et al., 2002), and nuts have not yet been identified in the Tel Tsaf botanical assemblage (Graham, 2014). To assess whether these originated from hardwood trees, additional analysis is required to characterize dicarboxylic acids from hardwood species identified at Tel Tsaf – acacia, poplar and oak (Liphschitz, 1988).

So far, the most geographically relevant plant species that contains the suite of dicarboxylic acids found here is *Equisetum ramosissimum* – branched horsetail. It contains dicarboxylic acids with C_{10}–C_{34}, with a predominance of even-numbered very long-chain dicarboxylic acids (maximizing at C_{24} and C_{26}) (Rezanka, 1998, table 1). This fern is present in the Jordan Valley (Danin, 2004, pp. 17) and is used to this day in

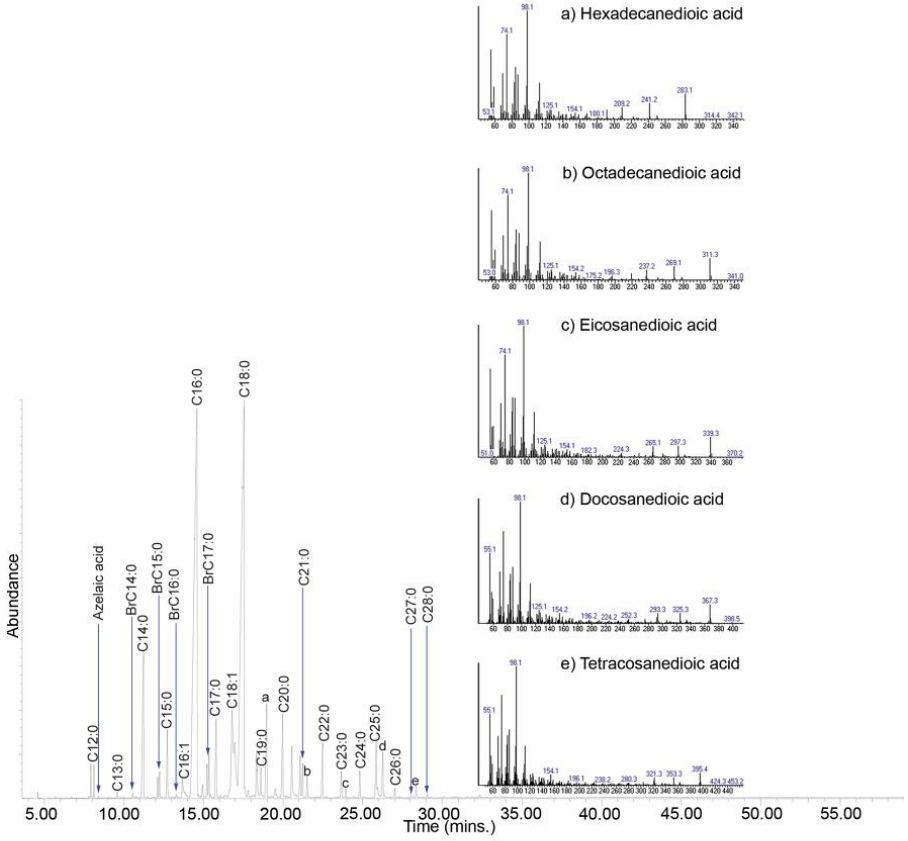

Fig. 4. Partial Total Ion Chromatogram of TSF17-23 In L2 run as methyl esters after saponification, showing long-chain dicarboxylic acids linked potentially to wild plant products (Br: branched; Cx:y: fatty acid with chain length X and Y number of double bonds); a–e are the long-chain dicarboxylic acids (C_{16}–C_{24}) and their corresponding mass-spectra.

Jordan for medicinal purposes (Lev and Amar, 2002). However, there is at present no evidence for this plant at Tel Tsaf in the macrobotanical or pollen record (Graham, 2014; Horwitz, 1988–1989). This is unsurprising as macrobotanical remains of leafy plants rarely preserve (e.g. Hillman et al., 1989, pp. 260).

3.1.3. *Acylglycerols*

Monoacylglycerols were identified in a few solvent extracts. These include primarily 1-Monopalmitin and 1-Monostearin. One sample (TSF18-69) contained a wider range: 1-Monomyristin, 1-Glycerol pentadecanoate, 1-Monopalmitin, 1-Glycerol heptadecanoate, 1-Monostearin and 1-Monooleoylglycerol. Dipalmitin was identified in one sample, although it has an insignificant lipid yield (TSF18-27). No triacylglycerols were identified. Triacylglycerols are important, representing the building blocks of fats and oils, with some animal and plant products having distinct triacylglycerol distributions (Dudd, 1999, pp. 131–152; Mirabaud et al., 2007). Their absence and the few acylglycerols identified suggest an advanced stage of triacylglycerol hydrolysis and degradation of the original lipid signature (Dudd et al., 1998).

3.1.4. *N-alkanes*

The *n*-alkanes present in the vessels tested range from C_{14}–C_{33}. To understand their origin, namely whether bacterial or botanical, the maximum *n*-alkane, carbon preference index (CPI), average chain length (ACL) and P_{aq} were identified (Diefendorf et al., 2011, equations 1–2; Ficken et al., 2000). The CPI is a weighted ratio of odd- to even-numbered *n*-alkanes (C_{23}–C_{33}), and values above 1.0 confirm a plant origin. Bacteria have low values, with mainly C_{15}–C_{20} *n*-alkanes (Diefendorf et al., 2011; Rao et al., 2009). The ACL is a weighted average of the odd-numbered *n*-alkanes (C_{23}–C_{37}); most terrestrial higher-plants have a high ACL (ranging from 27 to 34; Diefendorf et al., 2011), while macrophytes, halophytes and herbs have a lower ACL (Eley et al., 2014; Ficken et al., 2000; Luo et al. 2012). P_{aq} calculates the percent of the C_{23} and C_{25} *n*-alkanes among long-chain *n*-alkanes; this is used to differentiate aquatic plants. Values under 0.1 indicate terrestrial plants, values between 0.1 and 0.4 indicate emergent macrophytes and values over 0.4 indicate submerged or floating macrophytes (Ficken et al., 2000). Later studies suggest that some halophytes, with a maximum *n*-alkane of C_{23} and C_{25}, could also have high P_{aq} values (Eley et al., 2014, SI table 1). These general patterns, when paired with other specific lipids (fatty acids, alcohols and wax esters), can also be considered indicative of insect waxes (Blomquist and Jackson, 1979), so plants must be identified with some caution.

Few samples contained *n*-alkanes that could be definitively correlated with plant products of archaeological origin. This includes 10 vessels with significant lipid yields and a CPI value above 1.0 (ranging

from 1.2 to 25.2 and averaging at 9.4) or no even-numbered *n*-alkanes. Within these samples, most have a maximum *n*-alkane of C_{29} or C_{31} characteristic of higher-plants, although trace amounts of short-chain *n*-alkanes indicate bacterial input (Poynter et al., 1989). This is reinforced by their ACL, which ranges from 26.9 to 31.0 and averages at 28.9, like other higher-plants (Eglinton and Hamilton, 1967; Poynter et al., 1989, pp. 435). No samples maximized at C_{33} as characteristic of C_4 plants (Rommerskirchen et al., 2006). The P_{aq} could not be consistently calculated because most of these samples did not contain C_{23} and C_{25} *n*-alkanes. This absence reflects a terrestrial plant input. However, in samples with C_{23} and C_{25} *n*-alkanes, the P_{aq} ranges from 0.16 to 0.56, averaging at 0.34. These ratios were identified in two jars (TSF17-5 and TSF19-4), one wall fragment (TSF17-6) and one crust on a wall fragment (TSF18-57), and they correspond with macrophytes (emergent, submerged and floating) and halophytes. These plants were likely present in the local environment characterized by saline groundwater (Salameh, 2001) and freshwater sources, and halophytes were identified in the botanical record (e.g. *Atriplex* and *Chenopodium glaucum*; Graham, 2014). The typical cultivars (cereals and legumes) do not contain an indicative set of *n*-alkanes, so they cannot be identified using these methods (Maffei, 1996a, 1996b).

3.1.5. Alcohols

The alcohols present range from C_{12}–C_{34}. These are primarily even-numbered, with a predominance of C_{18} and C_{28}. This pattern is indicative of plant products (Eglinton and Hamilton, 1967; Poynter et al., 1989, pp. 435). No samples maximized at C_{32} as characteristic of C_4 plants (Rommerskirchen et al., 2006). This reinforces the *n*-alkane profile and the suggestion that C_4 plants were not consumed or that the signature is not preserved.

3.1.6. Sterols

Sterols were uncommon. The identified sterols include cholesterol, β-sitosterol and stigmasterol. Cholesterol may be linked to animal products, while β-sitosterol and stigmasterol are linked to plants. While no associated contaminants like squalene (e.g. Archer et al., 2005) were identified, sterol degradation markers (Hammann et al., 2018) were not identified. Therefore, contamination from post-depositional and post-excavation processes cannot be ruled out.

3.1.7. Terpenoids

Dehydroabietic acid and methyl dehydroabietate were identified in the solvent extracts of two vessels (TSF19-3 and TSF19-6) and the methylated acid fraction of two vessels (TSF18-1 and TSF18-6). These terpenoids were not identified in the soil sample, reinforcing their archaeological origin. Abietic-type acids are identified in resins from coniferous trees (Otto and Wilde, 2001, appendix 10). Pollen of several coniferous species was identified at Tel Tsaf – *Pinus halepensis*, *Cedrus libani* and *Cupressus sempervirens* (Horwitz, 1988–1989); however, without further terpenoids and dehydroabietic acid degradation products (e.g. Reber and Hart, 2008), the source of the resin cannot be identified.

3.1.8. Wax esters

Even-numbered palmitic wax esters with between 38 and 50 carbon atoms were identified in several vessels (Fig. 5). Such wax esters may be linked to beeswax (Tulloch, 1970). The identification of beeswax is reinforced in only one crust (TSF18-57) by the wax ester profile, found in combination with other lipids typical of beeswax (Chasan et al., 2021). However, the identification of beeswax at Tel Tsaf in other vessels is tenuous because of the low lipid yield of some of these samples (TSF18-7, TSF18-34, TSF18-35 and TSF19-3), although these signatures may represent highly degraded beeswax. A botanical origin was not considered because plants contain wax esters with 32–64 carbon atoms and non-palmitate and unsaturated wax esters (e.g. Reiter and Lorbeer 2001; Tulloch 1976), differing from the archaeological samples.

3.1.9. Ketones

Ketones were identified in several vessels, although not all had significant lipid yields. The majority contain hentriacontan-16-one (K_{31}), tritriacontan-16-one (K_{33}) and pentatriacontan-18-one (K_{35}). These form through the pyrolysis of fatty acids at temperatures over 300°C or prolonged exposure to low temperatures (Drieu et al., 2019a; Raven et al., 1997), and these are differentiated from plant-based ketones, which are generally symmetrical with only one homologue present (Raven et al., 1997). Some samples contained additional ketones with 29 (K_{29}), 33 ($K_{33:1}$), 34 (K_{34}) and 35 ($K_{35:1}$) carbon atoms (TSF18-36 and TSF18-69). $K_{33:1}$ and $K_{35:1}$ are formed from the pyrolysis of $C_{18:1}$ (Evershed et al., 1995).

These ketones are used to link vessels to cooking (e.g. Nieuwenhuyse et al., 2015; Raven et al., 1997) or post-firing vessel treatments (Drieu et al., 2019a). A combination of both situations is supported here because ketones were identified in vessel forms linked to diverse activities – bowls, jars and platters. A post-firing treatment is supported particularly in three vessels (TSF17-22, TSF17-28 and TSF18-69); the ketones are paired with darkened margins on the ceramic matrix

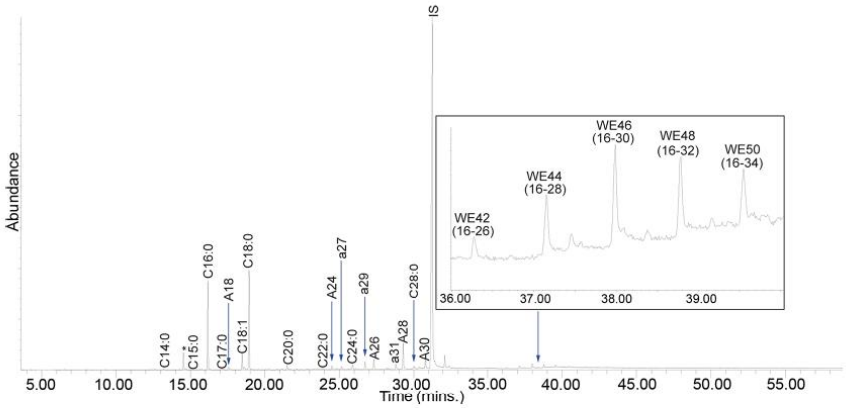

Fig. 5. Partial Total Ion Chromatogram of TSF18-35 In L2 run as a TMS derivative after solvent extraction, showing wax esters linked potentially to beeswax (*: phthalate plasticizers; ax: *n*-alkane with chain length X; Ax: alcohol with chain length X; Cx:y: fatty acid with chain length X and Y number of double bonds; IS: internal standard; WEx: wax ester with chain length X).

Fig. 6. Broken edge of pottery with darkened margins from potential exposure to a post-firing treatment: a) TSF17-22 (wall fragment); b) TSF17-28 (holemouth jar); c) TSF18-69 (wall fragment) (10x magnification, Zeiss Discovery V.8 stereoscope).

(Fig. 6). These form from the application of a liquid or melted fatty substance by soaking or rubbing post-firing. The margin darkens as the hot fat seeps into the ceramic (Drieu et al., 2019a). These techniques cut off the oxygen flow to the pot surface, and this can seal the vessel and change the vessel properties (e.g. porosity and hardness) (Drieu et al., 2019a; Skibo et al., 1997).

3.2. Stone vessels

Among the stone vessels tested are two limestone bowls (TSF17-9 and TSF19-14; Fig. 2:11–12) and one non-vesicular basalt bowl (TSF19-13; Fig. 2:13). The limestone vessels have a lipid yield of 0.5 and 0.1 μg g^{-1} in the solvent extract and 2.8 and 0.1 μg g^{-1} in the acid fraction obtained after hydrolysis of the solvent extracted stone powder. This low lipid yield is characteristic of stone, which rarely retains lipids because of its lower porosity (Evershed, 2008b). In the basalt vessel, the solvent extract and acid fraction lipid yield are 1.1 and 14.9 μg g^{-1} respectively, showing that lipids were bound to the basalt matrix.

This differential preservation may relate to the stone micromorphology. Basalt and limestone have different physical properties (Delgado-Raack et al., 2009; Manger, 1963; Prost and Prost, 1992, pp. 47, table 3.1), and the vessel porosity and mineral composition will affect preservation (Drieu et al., 2019b; Hammann et al., 2020). While further analysis is required, the differential lipid preservation may be explained by the pore size; studies suggest that lipids absorb and preserve best in matrixes with many small pores (Drieu et al., 2019b). Alternatively, the differential yields may relate to different functions of the vessels.

Few clear contaminants were identified in these samples. This includes primarily variable amounts of plasticizers. In addition, the solvent extract of one limestone vessel (TSF17-9) contained lipids from sunscreen, further deterring the interpretation of the lipid profile. As in the solvent extracts, cyclic octaatomic sulfur was also identified.

While none of the solvent extracts contained significant lipid yields, the saturated fatty acids include primarily traces of palmitic and stearic acid. In the acid fraction, a wider range of saturated fatty acids was identified ($C_{14:0}$–$C_{26:0}$) alongside unsaturated fatty acids – $C_{16:1}$, $C_{18:1}$ and $C_{18:2}$. 1-Monopalmitin and 1-Monostearin were identified in two solvent extracts. *n*-Alkanes range from C_{23}–C_{31}; however, their archaeological origin cannot be confirmed because of the low lipid yield. Alcohols are even-numbered and range from C_{18}–C_{30}. Cholesterol and ketones (K_{31}, K_{33} and K_{35}) were identified in one sample each (TSF17-9 and TSF19-13 respectively). Cholesterol may link the sample to animal products, but the low lipid yield and lack of degradation markers deters conclusive identification. The ketones may be linked to heating of a fatty substance.

3.3. GC-c-IRMS

GC-c-IRMS was applied to the saponified extract of 23 ceramic vessels and one stone vessel because only the saponified powder yielded consistently enough palmitic and stearic acid for analysis. Prior studies show that the GC-c-IRMS results of the saponified ceramic powder and the methylated solvent extracted TLE are similar (Craig et al., 2004), reinforcing the applicability of the methodology applied in this study. Animal fat identification was based on the plot of $\delta^{13}C_{16:0}$ and $\Delta^{13}C$ ($\delta^{13}C_{18:0}$-$\delta^{13}C_{16:0}$) (Fig. 7a). This was used because $\Delta^{13}C$ removes environmental biases and allows animal fats to be identified based solely on metabolic pathways (Copley et al., 2003). In the $\Delta^{13}C$ plot, samples with more positive $\delta^{13}C_{16:0}$ values reflect input of marine fat or C_4 plants and certain C_3 plants isotopically altered by the environment, usually as part of the animal diet. Marine resources have more positive $\delta^{13}C$ values from the incorporation of ocean bicarbonates (Cramp and Evershed, 2014). C_4 plants and C_3 plants from arid, water depleted and saline environments have more positive $\delta^{13}C$ values because of their unique photosynthetic pathways (Meier-Augenstein, 2002; Tieszen, 1991, table 2). A marine input is unlikely because of the site's inland location (ca. 70 km from the current Mediterranean Sea shore), the very few recovered fish bones (Hill, 2011, pp. 163) and the absence of fishing paraphernalia in the archaeological record (Bekker and Garfinkel, 2020, table 11.1). However, C_4 or enriched C_3 plant animal dietary input were considered because several plants that can undergo C_4 photosynthesis (e.g. species within the *Chenopodiaceae*, *Cyperaceae* and *Polygonaceae* families; Rudov et al., 2020) were identified in the Tel Tsaf botanical assemblage (Graham, 2014), and the Jordan Valley is characterized by some aridity and saline groundwater conducive to the isotope enrichment of C_3 plants.

$\delta^{13}C_{16:0}$ ranged from −18.9 to −30.1‰ and $\delta^{13}C_{18:0}$ ranged from –23.2 to −30.0‰ (Fig. 7b). In the $\Delta^{13}C$ plot (Fig. 7a), based on the average and ± 1 standard deviation of the modern reference fats, 25% of the samples plot within the range of ruminant adipose fat (n = 6), 16.7% plot within the range of ruminant dairy fat (n = 4) and 16.7% plot within the range of non-ruminant adipose fat (n = 4). An additional 33.3% of the samples plot between ruminant and non-ruminant adipose (n = 8), and 8.3% plot between ruminant adipose and dairy fat (n = 2). Among the samples that plot between ruminant and non-ruminant adipose, several clearly fall on the mixing curve of domestic ruminant and porcine adipose (Fig. 7b).

Further analysis shows that some samples that plot within the range of ruminant adipose and dairy fat ($\Delta^{13}C < 0‰$) have a more positive $\delta^{13}C_{16:0}$ value than samples that plot within the range of non-ruminant fat ($\Delta^{13}C > 0‰$) (Fig. 7a). As noted above, a marine lipid input is unlikely, and this instead may relate to the ruminants' diet containing C_4 grasses (Meier-Augenstein, 2002) or C_3 plants characteristic of arid, water depleted and saline environments (Tieszen, 1991, table 2). Lipid input of these enriched plants is supported by the mixing curves, which show that a few samples (TSF17-6, TSF18-3, TSF18-20 and TSF19-3) plot as mixtures of lipids from some ruminants consuming C_3 plants and some ruminants consuming C_4 plants or lipids from ruminants consuming both C_3 and C_4 plants (Fig. 7b).

A final point that must be considered is the role of plants in forming

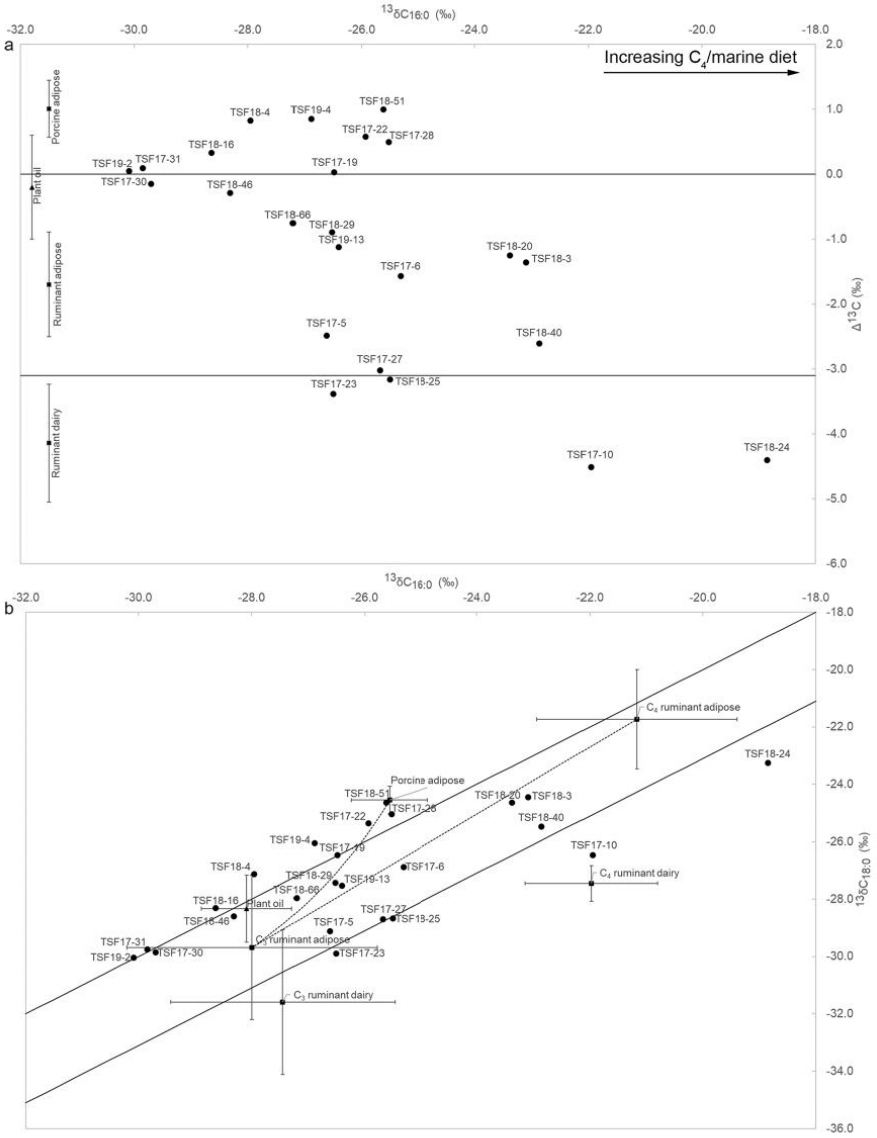

Fig. 7. Plot of GC-c-IRMS results from Tel Tsaf: a) $\delta^{13}C_{16:0}$ and $\Delta^{13}C$ ($\delta^{13}C_{18:0} - \delta^{13}C_{16:0}$) values; b) $\delta^{13}C_{16:0}$ and $\delta^{13}C_{18:0}$ values and theoretical mixing curves (following Woodbury et al., 1995); The animal reference fat ranges represent the mean ± 1 s.d. of the $\delta^{13}C_{16:0}$, $\delta^{13}C_{18:0}$ and $\Delta^{13}C$ values of modern references from the UK (animals raised on a pure C_3 diet), Germany (animals raised on a pure C_3 diet), Italy (animals raised on a pure C_4 diet), Malta (animals raised on a pure C_3 diet and animals raised on a pure C_4 diet), Israel, Jordan and Kazakhstan (animals raised on an uncontrolled diet) (Debono Spiteri, 2012; Dudd, 1999; Gregg et al., 2009; Outram et al., 2009). The plant oil reference range represents the mean ± 1 s.d. of the $\delta^{13}C_{16:0}$, $\delta^{13}C_{18:0}$ and $\Delta^{13}C$ values of modern almond oil, argan oil, olive oil, moringa oil, sesame oil and walnut oil (Steele et al., 2010). All references had a post-industrial carbon (PIC) correction of 1.14‰ (Friedli et al., 1986).

the isotope signature. Prior studies show that compound specific isotope signatures of select plant oils plot on the border between ruminant and non-ruminant adipose (Fig. 7; Steele et al., 2010). Although not yet verified by cooking experiments, theoretical models predict that when C_3 plants are mixed with ruminant adipose, the $\Delta^{13}C$ value shifts into the range of non-ruminant adipose (Hendy et al., 2018, Fig. 5c). Similarly mixtures of dairy fats and C_3 plants could plot within the range of ruminant adipose (Hendy et al., 2018, Fig. 5b). In this case, all of these situations are likely, given the wide fatty acid profile obtained for most samples and the botanical remains present at Tel Tsaf, so the GC-c-IRMS results obtained here need to be interpreted with caution.

4. Discussion and conclusion

Ongoing research on the Middle Chalcolithic site of Tel Tsaf attempts to synthesize the varying lines of data to discuss how the community's diet relates to the environmental constraints of the Jordan Valley. Archaeological evidence supports the use of various floral and faunal resources, intensified cereal production (Ben-Shlomo et al., 2009; Garfinkel et al., 2009; Graham, 2014; Hill, 2011) and olive exploitation (Rosenberg et al., 2021). New dietary patterns such as the Mediterranean diet were suggested as a working hypothesis (Rosenberg and Klimscha, 2018, 2021). The results of the current study add several significant layers to our understanding of what animal and plant products were consumed at Tel Tsaf and how pottery and stone vessels were used.

The organic residue analysis results indicate that the ceramic and stone vessels tested were used indiscriminately for both animal and plant products (Table 1). This is evidenced by the wide saturated fatty acid profile and the unsaturated fatty acids in most vessels. Most likely, the vessels were used for stewing or containing various mixtures of meats and plants. Cooking is supported by the identified ketones, the morphology of the vessels, dominated by various deep forms (medium-sized bowls and holemouth jars; Fig. 2; Shooval and Rosenberg, 2021), and rarely soot marks on the tested vessels (SI 1). Ketones were also identified in a basalt vessel, suggesting that this too may have been used for cooking. Basalt cooking pots are unknown in the local or even global archaeological record, but in regions rich in basalt, it may be used to form stone pit ovens (Carson, 2002; Nojima, 2008, pp. 17). Basalt is considered desirable in these ovens because it retains heat well and does not easily shatter (Carson, 2002; Gillies, 1979, pp. 50, 52). Based on these principles and the vessel's morphology and medium size (open form with a diameter of ca. 20 cm; Fig. 2:13), the example from Tel Tsaf is likely unsuitable for cooking, but the vessel could have been heated and used to keep its food contents warm for long time periods. Alternatively, the lipid profiles obtained for both the ceramic and stone vessels could represent subsequent use episodes rather than mixtures as even today pots are used for multiple purposes.

Differentiation between the two scenarios is complicated because both situations would generate similar lipid signatures, and most likely a combination of both is represented. This also holds true for interpreting the isotope signatures because, while mixtures can be resolved using Bayesian modeling (e.g. Courel et al., 2020; Cubas et al., 2020; Fernandes et al., 2018; Lucquin et al., 2018) and theoretical mixing curves (Woodbury et al., 1995), these methods cannot identify whether the isotope signature reflects complex recipes with multiple ingredients or subsequent vessel utilization. In addition, differentiating cooking episodes is complicated because recent studies show that the absorbed lipid signature recovered by acid extraction represents the entire vessel life history and not one specific event (Miller et al., 2020). This study applied solvent extraction followed by saponification, and experimental analysis is required to identify if there are different origins for the free and bound lipid fractions, especially because certain lipids are more susceptible to polymerization or are only preserved in the bound fraction (Hansel et al., 2011; Regert et al., 1998) (although other studies using acid extraction and saponification show parallels with the solvent extraction; Correa-Ascencio and Evershed, 2014; Craig et al., 2004), and the applied chemicals in the various extraction techniques do not recover all lipids equally (Reber, 2021). Therefore, the specific 'recipes' of these mixtures remain unknown.

The vessels' lipid signatures can also be used to discuss specific aspects of diet and site economy when reinforced by the faunal and floral remains. Among the tested vessels, the clearest lipid signature is animal fat. This may in part be attributed to the higher lipid yield of animal fat in comparison to plant products (Evershed, 2008a). The animal fat most likely relates to domesticated sheep, goats, cattle and pigs – the most common animals identified in the faunal remains. Wild animals are uncommon, with few ungulate and rare fish bones recovered; these include distinct concentrations of gazelle horn cores, which were correlated with specific events (Hill, 2011, pp. 214, 216, table 5.1).

Ruminants fulfilled a key dietary role. This is supported by the lipid profiles and GC-c-IRMS results (Table 1). Most vessels contain odd-numbered and branched-chain saturated fatty acids, which are characteristic of ruminant fat (Doreau and Ferlay, 1994), and several samples that underwent compound specific isotope analysis plot within the range of ruminant fat. This is supported by the faunal assemblage that shows that ruminants form over 50% of the number of identified specimens (NISP), with a prevalence of sheep and goat remains (Hill, 2011, table 5.1). The kill-off pattern of sheep and goats suggests that ruminants were used mainly for their meat as most were slaughtered immediately after reaching maturity (Hill, 2011, pp. 124–127, 164–165). This relates in part to risk management as this strategy reduces fluctuations in annual yield and ensures animal reproductive success by managing the ratio between males and females and killing off excess animals not required for breeding after reaching maturity when the meat yield is at its highest (Payne, 1973; Redding, 1981, pp. 175). Based on the wide range of isotope values attributed to ruminant fat, including the more positive values, we can also conclude that caprines were herded over a wide range, likely into the valley foothills, which are characterized by rocky shrubs (Grossman and Safrai, 1980). This style of pastoralism allowed for successful coeval herding and cultivation by preventing the ruminants from grazing on crops (Köhler-Rollefson, 1988).

So far, there is no clear indication for targeted milk exploitation based on the kill-off pattern of sheep and goats (Hill, 2011, pp. 129, 165). If animals were reared for targeted milk production, the kill-off pattern would show that surplus males were culled before reaching maturity, while the females were kept as breeding stock (Payne, 1973). However, there are indicators from the GC-c-IRMS results that dairy products were occasionally consumed at Tel Tsaf (Table 1). This so far marks the earliest direct evidence for the use of dairy products in the southern Levant, dating around 5200–4700 cal BC.

Prior studies on contemporary and earlier sites in the southern

Table 1
Summary of the products in the sampled vessels and supporting archaeological evidence (Br: Branched; SFA: saturated fatty acids; USFA: unsaturated fatty acids; +: even-numbered; –: odd-numbered).

Signature			Vessel types	Organic residue evidence	Additional evidence
Animal products	**Ruminant**	**Adipose**	Bowls, jars, unidentified vessel forms and a basalt bowl	Isotopes and SFAs (– and Br)	Fauna
		Dairy	Jars and unidentified vessel forms	Isotopes and SFAs (– and Br)	Fauna
	Porcine adipose		Jars and unidentified vessel forms	Isotopes	Fauna
Plant products	**Seed oils**		Bowls, jars and unidentified vessel forms	USFAs	–
	Wild plants		Jars and unidentified vessel forms	Dicarboxylic acids and *n*-alkanes	Flora
	Resins		Bowls, jars and unidentified vessel forms	Terpenoids	–
Beeswax			Crust on unidentified vessel form	Wax esters, SFAs (+), alcohols (+) and *n*-alkanes (–)	–
High heating			Jars, a platter, unidentified vessel forms and a basalt bowl	Ketones	Sooting and darkened ceramic margins

Levant did not find isotopic evidence for dairy, deterred in part by the low lipid yield from the tested vessels (Evershed et al., 2008, table 2; Gregg, 2009, pp. 98–121, figs. 5.3.1, 5.3.2, 5.5.1, 5.5.2). The few vessels identified here are significant because they show that animals were used for meat and milk by the Middle Chalcolithic period before the hypothesized intensified secondary product exploitation of the Late Chalcolithic period, based primarily on the rise of ceramic churns (Kaplan, 1954). Thus, our results suggest that milk exploitation had earlier roots, and it corresponds with the reinterpretation of the so-called 'Secondary Products Revolution' (Sherratt, 1983) as a gradual intensification (Greenfield, 2010). This is consistent with other residue analysis studies that identified dairying at earlier Neolithic sites in the Mediterranean and Anatolia (Debono Spiteri et al., 2016; Evershed et al., 2008).

At the same time, this differs from other Neolithic and Chalcolithic sites in the Mediterranean and Anatolia where there is isotopic evidence for dairy in higher frequencies (Debono Spiteri et al., 2016; Evershed et al., 2008). Milk may have been considered beneficial at these sites because it can be converted into storable and transportable products, and it is nutritionally beneficial, providing calcium, sugar and vitamin D (Davis, 1987, pp. 155). At Tel Tsaf, the low consumption may relate to the site's economy, which was centered on crop cultivation as evidenced by the numerous silos (Garfinkel et al., 2009). Cereal crops were considered desirable because they can be stored and produced in surplus (Genz, 2003), providing similar advantages to dairy products. In such an economy, dairy products would likely form a supplementary part of the diet. Additionally, the limited genetic studies on Late Chalcolithic individuals show that people did not yet have the genetic mutation for lactase persistence (Harney et al., 2018, supplementary material 7). This genetic mutation promotes increased dairy consumption, although populations without the lactase persistence gene may also consume dairy by processing it to reduce the amount of lactose (Simoons, 1970). While these factors would limit dairying, the rare examples of dairy residue at Tel Tsaf may also relate to the degradation of the lipid signature; the related triacylglycerols and short-chain fatty acid decay products easily degrade, leaving an unrecognizable signature (Dudd et al., 1998). In addition, the use of organic containers, which would not preserve, must also be considered. Therefore, the true frequency of dairy exploitation remains unclear.

Pork was also incorporated into the diet (Table 1). This is more difficult to identify because while porcine fat contains a distinct triacylglycerol distribution, without this preserved there are no usable biomarkers (Regert, 2011). However, several samples that underwent GC-c-IRMS plot within the range of porcine adipose fat or as mixtures of ruminant and porcine adipose fat, and pigs form ca. 30% of the faunal assemblage (Hill, 2011, table 5.1). The importance of pigs to the diet is supported mainly by finds from Building CI at Tel Tsaf (Ben-Shlomo et al., 2009). While no vessels uncovered in this building during earlier excavations were sampled, concentrations of pig bones were found deposited near large cooking facilities in the building, and this was used to suggest that feasting activities took place there (Ben-Shlomo et al., 2009).

The role of fish in the diet is so far unclear. Although targeted fish biomarker analysis is still underway, ω-(*o*-alkylphenyl)alkanoic acids and isoprenoids, key fish biomarkers, were not identified in the saponified acid fraction. ω-(*o*-alkylphenyl)alkanoic acids with C_{16}–C_{22} form from heating unsaturated fatty acids characteristic of fish, and phytanic, pristanic and 4,8,12 trimethyltridecanoic acid are isoprenoids found in fish (Cramp and Evershed, 2014). Among the isoprenoids, 4,8,12 trimethyltridecanoic acid is most indicative, with phytanic and pristanic acid also found in ruminant fat (Ackman and Hansen, 1967). Prior studies identified these in methylated samples even without targeted GC–MS programing (Heron et al., 2010), but the markers were not identified in these samples. Further, despite sifting all sediment with 2 mm sieves and floating ca. 7 tons of sediment in 250 μm nets, meager fish remains were recovered (Hill, 2011, pp. 163; Rosenberg et al., 2014, table 9). The lack of fish markers and the infrequency of fish bones suggest that fish did not fulfill a key dietary role despite the site's location near the perennial Jordan River and smaller freshwater streams.

Alongside meat and dairy products, plants were part of the diet (Table 1), but because of their low lipid yield, the lipid signature is often either masked by meat products or unpreserved (Evershed, 2008a). This is particularly true for cereals and legumes. Barley, wheat and diverse legumes are predominant in the botanical assemblage (Graham, 2014), and the silos at Tel Tsaf indicate intensive cereal cultivation (Garfinkel et al., 2009); however, their presence is invisible in the lipid record. Cereals contain low amounts of lipids and are composed mainly of starches (Colonese et al., 2017), and alkylresorcinols, which are unique cereal biomarkers, are highly susceptible to microbial degradation (Colonese et al., 2017; Hammann and Cramp, 2018). Similarly, legumes have a low lipid yield, with few distinct biomarkers (e.g. Caprioli et al., 2016; Faris et al., 2013; Zhang et al., 2014). This, coupled with the poor lipid preservation conditions at Tel Tsaf, prevents the identification of cereals and legumes with organic residue analysis.

Recent excavations also uncovered hundreds of olive stones (Rosenberg and Klimscha, 2021; Rosenberg et al., 2021), and olive oil processing may be suggested by the presence of crushed olive stones. However, no sherds contain indisputable evidence for olive oil based on the lipid signature. We hypothesize that the signature would only be identifiable if the vessels were used primarily and intensively for storing olive oil because this would create a more visible and unmasked signature in terms of the total lipid yield and preservation of the triacylglycerol profile and correlated fatty acids distinctive of olive oil. Therefore, organic residue analysis suggests that olive processing may have been low-scale, and this is reinforced by the lack of clear olive oil processing installations. Olives may have also been eaten whole as was proposed for other slightly earlier sites (Galili et al., 2021), but it is unclear what, if any, lipid signature this would leave in a vessel used to contain or store them.

Surprisingly, organic residue analysis did capture the use of wild plant products in low amounts. Seed oils, macrophytes, halophytes and other wild plants were identified based on the *n*-alkanes, unsaturated fatty acids and dicarboxylic acids. Many of these plant types are indigenous to the Jordan Valley. For example, in the Jordan Valley, oil producing *Brassicaceae* plants include *Erucaria hispanica*, *Sinapis alba* and *Sinapis arvenis*, macrophytes include *Juncus acutus* and *Juncus maritimus* and halophytes include numerous species within the *Chenopodiaceae*, *Compositae*, *Cruciferae* and *Cyperaceae* families, to name a few (Qasem, 2015, table 2). Additionally, two clear halophytes were found in the botanical assemblage from Tel Tsaf – *Artiplex* and *Chenopodium glaucum* (Graham, 2014). Branched horsetail, which is another potential lipid source, is also a species local to the Jordan Valley (Danin, 2004, pp. 17).

While some of these plant types are present but rare in the Tel Tsaf botanical assemblage (Graham, 2014), more were likely present. This may be suggested because the climatic and environmental conditions of the Chalcolithic are considered relatively equivalent to the present-day climate and environment (Bar-Matthews et al., 1997), with greater water flow in the Jordan River before the damming of the outlet of the Sea of Galilee and the Yarmuk River. Differentiating macrophytes and halophytes based on their *n*-alkane signatures is not possible, and most likely in the few vessels where their residues were identified with some certainty, a mixture of both plant types may be represented. The identification of these plants shows that the occupants of Tel Tsaf gathered and used wild plants from the open land around the site, given the groundwater characteristics, as well as from the Jordan River banks or from any of the streams or springs in the site vicinity.

Why these wild plants were used after the economic consolidation of domesticated resources is unclear. Wild plants could have formed a component of the daily diet, especially as spices to diversify the diet's flavor palette; however, wild plants unlikely fulfilled a significant nutritional role because as a dietary and calorie resource, they are eclipsed by the more stable cereals and legumes. An alternative

suggestion is that the wild plants were used for medicinal purposes, which is supported by ethnographic studies that show such a use for macrophytes, halophytes, horsetail and other plants (e.g. Ksouri et al., 2012; Lev and Amar, 2002; Qasem, 2015; Sanilkumar and Thomas, 2007). These plants represent a wide array of potential remedies. Wild plants in the botanical assemblage were also discussed in relation to cover crops, a fallow regime and animal fodder (Graham, 2014).

To conclude, the results of our organic residue analysis study reflect the multifaceted nature and complexity of diet and foodways of the inhabitants of Tel Tsaf during the Middle Chalcolithic period. This is especially true when viewed in tandem with the faunal and floral remains. While we see evidence for increasing storage capacity and the management of surplus (Garfinkel et al., 2009), by and large, the daily food-related economy was like the proceeding Neolithic period. However, as the current study well demonstrates, developments such as the earliest direct evidence for milk exploitation are noted. While this may have, as of yet, invisible roots in the Neolithic, the introduction of dairying is nonetheless significant because it shows a change in the way animals were viewed and exploited, although the scale was too low to require a specialized animal management strategy. The use of secondary products shows that people were now considering how to get the most out of their livestock in the long-term. The provided evidence for wild plant exploitation is also notable; it shows that even among agrarian communities, wild resources were still valued. Together with the relatively small component of wild game in the faunal assemblage, which was sometimes found in distinct caches (Hill, 2011, pp. 204), we can suggest that wild resources were now more appreciated for specific functions, rather than as a standard food source. On a more daily basis, pottery and stone vessels were used in relation to domesticated resources without discrimination of animal and plant products, and the foodways incorporated various culinary traditions where these were blended together.

CRediT authorship contribution statement

Rivka Chasan: Conceptualization, Methodology, Investigation, Writing – original draft, Writing – review & editing, Visualization. **Florian Klimscha:** Writing – review & editing. **Cynthianne Spiteri:** Conceptualization, Methodology, Writing – original draft, Writing – review & editing, Supervision. **Danny Rosenberg:** Conceptualization, Writing – original draft, Writing – review & editing, Supervision, Funding acquisition.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

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Article 3. Dietary continuation in the southern Levant: A Neolithic-Chalcolithic perspective through organic residue analysis

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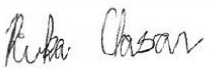
Name and Signature	Rivka Chasan 	Date	27/06/2022

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Name and Signature	Cynthianne Spiteri 	Date	04/07/2022

Name of Co-Author	Danny Rosenberg		
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ORIGINAL PAPER

Dietary continuation in the southern Levant: a Neolithic-Chalcolithic perspective through organic residue analysis

Rivka Chasan[1] · Cynthianne Spiteri[2] · Danny Rosenberg[1]

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Abstract
Foodways in the late prehistoric southern Levant evolved alongside changes in the social and economic organization of the communities occupying the region. In this paper, we present a comprehensive study of culinary practices from the Pottery Neolithic to the Late Chalcolithic period (ca. 6,400–3,800 cal. BC). The research examines changes and continuity in food-related organic residues preserved within ceramic and stone vessels along diachronic and interregional climatic lines in Israel. The results of lipid biomarker and compound-specific isotope analyses, together with the faunal and botanic records, indicate that while this timespan witnessed significant social and economic developments (including most notably the introduction of pottery and later copper metallurgy), there was a fairly uniform diet. Dietary preferences included meat, originating mainly from domestic ruminants, and cultivated vegetal resources, usually regardless of chronological, cultural, or environmental differences. Some chronological and potentially environmental variation was noted in the use of dairy products, which chemical residue analysis detected from samples dating to the Middle Chalcolithic period onwards in semi-arid regions. The overarching general uniformity in diet during the Pottery Neolithic and Chalcolithic periods suggests that the timespan was one long economic continuum separated by distinct cultural entities sharing a core dietary tradition. The socio-economic processes of the timespan had no significant effect on what people ate and how they used vessels to prepare and consume food.

Keywords Pottery Neolithic · Chalcolithic · Southern Levant · Culinary practices · Organic residue analysis · Lipids

Introduction

Foodways are reflections of individual and communal decisions formed within complex sets of internal and external factors such as culture, tradition, economy, and the environment (e.g., Graff 2018; Gumerman IV 1997; Twiss 2012; Zeder 1988). As these regulators change alongside increasing social complexity, foodways evolve to reflect economic and social concerns and eventually the "political economy" (Costin and Earle 1989). Food becomes a way of showcasing social differences (Twiss 2012), and the emphasis of foodways shifts from self-provisioning to the production of surplus and storable resources that can be used to support and maintain social institutions and individuals not involved in food production (D'Altroy and Earle 2002; Zeder 1988).

These changes are accompanied by new economic strategies. Intensification techniques such as manuring, irrigating fields, plowing, and using animals for secondary products (milk, wool, animal labor, and dung) increase crop and animal yield by investing more time and energy into the labor input (Morrison 1994; Sherratt 1983) and by expanding the types of soil that can be cultivated (Halstead 1995). Some of the most notable food products of intensified production are grains, olive oil, wine, and dairy products. In particular, additional labor is required to grow fruit and process them into liquid form (Genz 2003; Philip 2001: 185; Salavert 2008) and to milk animals and process the milk (Sherratt 1983). However, in exchange for these increased labor costs, there is an increased return of valuable storable and tradable resources (Morrison 1994). In the southern Levant, intensification is typically associated with later societies

✉ Rivka Chasan
rchasan@campus.haifa.ac.il

Cynthianne Spiteri
cynthianne.spiteri@uni-tuebingen.de

Danny Rosenberg
drosen@research.haifa.ac.il

[1] Laboratory for Ground Stone Tools Research, Zinman Institute of Archaeology, University of Haifa, Haifa, Israel

[2] Institute of Pre- and Protohistoric Archaeology, Eberhard Karls Universität Tübingen, Tübingen, Germany

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and urbanization processes (Fall et al. 2002; Philip 2001; Zeder 1988), but the foundations are present well before the Early Bronze Age during which the first urban societies are witnessed (Greenberg 2019: 24–135). The roots for intensified food production can be traced back to the Neolithic and Chalcolithic periods.

Evolving socio-economic systems of the southern Levant

Gradually during the Neolithic period, the primary subsistence strategy shifted from hunting and gathering to farming and herding domesticated plants and animals respectively. This economic transition was instigated by earlier climatic changes and an increasing population size in a narrow catchment area, which mandated intensive resource exploitation (Zeder 2015). The Neolithic period also heralds the introduction of pottery. Pottery marked a significant innovation that allowed diverse and improved storage, food preparation, cooking, and serving techniques to be developed (e.g., Brown 1989; Garfinkel 2019: 2, 4; Rice 1999). In the southern Levant, this is correlated with the Yarmukian culture (ca. 6,400–5,800 cal. BC; Garfinkel 2009: table 14.1, 2019).

The following Chalcolithic period (ca. 5,800–3,800 cal. BC; Garfinkel 2009: table 14.1) shows a continued reliance on domesticated animals and cultivated crops (e.g., Graham 2018; Levy 1983; Namdar et al. 2021), which arguably developed in some places into a distinct agro-pastoral economy (Levy 1983). The timespan is debatably divided into the Early, Middle, and Late Chalcolithic periods (Garfinkel 2009), and each of these phases contains unique geographically bound cultural entities defined by their material culture (Gilead 2007; Gopher 2012; Gopher and Gophna 1993; Rowan and Golden 2009). As a whole, the Chalcolithic period witnessed several notable changes, including regionalization (shown by geographically bound use of specific material culture items) and craft specialization (including copper metallurgy and certain aspects of the ceramic, lithic, and stone vessel industries) (e.g., Gilead 2007; Kerner 2010; Rowan and Golden 2009). Based primarily on the increased appearance of olives, churns, and spindle whorls, the economy is hypothesized to have expanded to include olive horticulture and secondary animal products (e.g., Dighton et al. 2017; Galili et al. 2017; Garfinkel 1999: 254–258; Gopher and Eyal 2012: 364; Heidkamp 2018; Levy and Gilead 2012; Meadows 2005; Rosenberg et al. 2021). The reason or reasons for these changes is unclear, but they may be related to suggested changes in social organization (Levy 1986), shifts in demographic and settlement patterns, including during the Late Chalcolithic period, population growth and consolidation and a southern settlement orientation (Holl 2019; Levy 1983; Winter-Livneh et al. 2010), or climatic amelioration (e.g., increased rainfall; Bar-Matthews and Ayalon 2011; Bar-Matthews et al. 1999; Frumkin et al. 1991).

In the current paper, we tested whether culinary preferences remained consistent throughout the Neolithic and Chalcolithic period or if they evolved alongside the noted socio-economic changes or if they followed lines of cultural or environmental variation. We achieved this by identifying lipids preserved within ceramic and stone vessels from Pottery Neolithic and Chalcolithic sites in Israel and by relating the results to published faunal and botanic remains. Synthesis of the results will enhance our knowledge on several previously unknown aspects of the diet by complimenting the already known macro-archeological remains with micro-residues.

Methodology

In total, 226 ceramic vessels and three stone vessels were sampled. These originate from one Pottery Neolithic site, two Early Chalcolithic sites, one Middle Chalcolithic site, and eight Late Chalcolithic sites that span diverse climatic and topographic zones in Israel (the northern Negev, Mediterranean coastal plain, Galilee, and Jordan Valley) (Fig. 1, Supplementary materials 1, 2). While most of the sampled vessel fragments were too small to identify typology, the sampled vessel forms include various bowls (V-shaped bowls, straight walled bowls, and inverted rim bowls,), jars

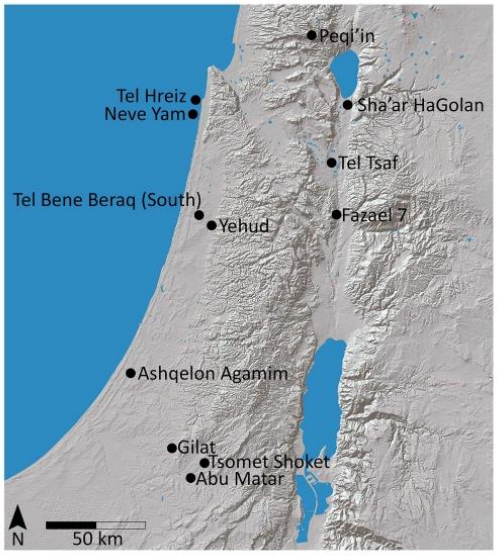


Fig. 1 Map of the sites included in the study

(holemouth and necked jars), pithoi, platters, churns, cornets, and strainers. For sampling and extraction, glassware was sterilized before use at 500 °C for 8 h in a muffle furnace, and HPLC grade solvents were used. Nitrile gloves were worn at all times. The interior was sampled using a Dremel modeling drill after removing and discarding the initial inner surface layer (ca. 1–2 mm deep) that has a higher potential for contamination. About 1–2 g of powder was collected. One crust was also collected and homogenized. The exterior surfaces and, when possible, soil from the surrounding archeological sediments were sampled as controls to test for post-depositional and post-excavation handling contamination.

Lipids were extracted alongside a process blank following established protocols (Mottram et al. 1999). The lipids were extracted three times using 5 mL of dichloromethane:methanol (DCM:MeOH, 2:1, *v:v*). The solvent was evaporated to dryness under a gentle stream of nitrogen and mild heating to obtain the total lipid extract (TLE). An aliquot of the TLE was silylated using 50 µL of *N,O*-bis(trimethylsilyl)trifluoroacetamide (BSTFA) with 1% trimethylchlorosilane (TMCS) and 4 µL of pyridine (40 °C; 30 min). The solvent was evaporated, and the samples were rehydrated in 50 µL of cyclohexane. One microgram of hexatriacontane (C_{36} *n*-alkane; 98% purity, Sigma-Aldirch) was used as a standard for quantification; it was initially added to the samples prior to analysis; but after observing that hexatriacontane occasionally co-eluted with peaks of interest, it was analyzed separately in the same sequence. Samples with over 5 µg g^{-1} of preserved lipids were considered significant interpretable residues (following Evershed 2008).

Saponification was applied to 225 of the 229 vessels tested to release lipids bound to the internal vessel matrix (following a modified method D; Stern et al. 2000) after it was solvent extracted (the remaining four were just solvent extracted). Four milliliters of 0.5 M sodium hydroxide solution (MeOH:H_2O, 9:1, *v:v*) was added to the samples and heated (70 °C; 90 min). The neutral fraction was extracted three times with 2 mL of cyclohexane, and it was not analyzed further. To the remaining fraction, 2 mL of 1 M hydrochloric acid (HCl:H_2O, 1:9, *v:v*) was added until pH 3 was reached. This was extracted three times with 2 mL of cyclohexane. The acid fraction was methylated with 250 µL of boron trifluoride methanolic solution (14%) (70 °C; 60 min). It was methylated alongside palmitic (99% purity, Fluka) and stearic fatty acid standards (99.5% purity, Sigma-Aldrich) of known isotope composition, which were used to correct for the carbon introduced during methylation after compound-specific isotope analysis. The reaction was quenched with ultrapure water, and the lipids were extracted three times with 2 mL of cyclohexane. The solvent was evaporated, and the samples were solubilized in 50 µL of cyclohexane. C_{36} *n*-alkane was measured as a standard. As in the solvent extracts, it was initially added to the samples prior to analysis and later analyzed separately in the same sequence.

Gas chromatography-mass spectrometry (GC–MS) was performed using an Agilent 7890B GC coupled to an Agilent 5977A Mass Spectrometer (MS) and flame ionization detector (FID). Injections were conducted using a GERSTEL multi-purpose sampler and a GERSTEL cold-injection system. The samples were run on an Agilent J&W DB-5HT column (15 m × 0.32 mm; 0.1 µm film thickness) and divided into two equal parts using 0.18 mm non-coated deactivated silica capillary columns (0.66 m splitter-column to the FID and 1.52 m splitter-column to the MSD) with the Three-Way Splitter Kit. The inlet temperature was raised from 30 to 240℃ at 12℃ s^{-1} (held isothermally for 5 min) and then raised to 350℃ at 12℃ s^{-1} (held isothermally for 10 min). The oven temperature was raised from 40℃ (held isothermally for 1 min) to 100℃ at 15℃ min^{-1}, to 240℃ at 6℃ min^{-1}, and then to 350℃ at 10℃ min^{-1} (held isothermally for 20 min). Helium was used as the carrier gas. The injection system was operated in splitless mode with a purge flow of 3.0 mL min^{-1} and a constant pressure at the head of the column of 8.4435 psi. Mass spectra were acquired using electron ionization at 70 eV. The mass range acquired was from *m/z* 50–950 in 1.562 s. The temperature of the ion source, transfer line, and FID was 300℃.

Mass spectra were identified using the National Institute of Standards and Technology Library, 2014 edition. *N*-alkanes were identified through comparison to an *n*-alkane standard mix (C_7–C_{40}, Sigma-Aldrich). They were further analyzed through comparison to various indexes: the maximum *n*-alkane, average chain length (ACL), carbon preference index (CPI), and P_{aq} (following Diefendorf et al. 2011: Eqs. 1 and 2; Ficken et al. 2000). The ACL and CPI are used to confirm a plant origin (Diefendorf et al. 2011; Eley et al. 2014; Ficken et al. 2000; Luo et al. 2012), and the P_{aq} is used to differentiate emergent and submerged macrophytes from terrestrial plants based on the percent of tricosane (C_{23}) and pentacosane (C_{25}) in relation to nonacosane (C_{29}) and hentriacontane (C_{31}) (Ficken et al. 2000). The maximum *n*-alkane of some halophytes is C_{23} and C_{25} (Eley et al. 2014: SI Table 1), so these can also be identified using the P_{aq} value.

Methylated extracts with over 5 µg of palmitic and stearic acid underwent gas chromatography-combustion-isotope ratio mass spectrometry (GC-c-IRMS). One microliter of each sample was injected by a CTC Combi-PAL autosampler in PTV splitless mode at 80℃ and held for 30 s. For sample transfer, the injector was raised to 300℃ at 14.5℃ s^{-1}, and it was held for 2 min. Afterwards, the injector was heated until the end of the run to 350℃ for cleaning, and a split flow of 20.0 mL min^{-1} was applied. In the GC, helium with a constant flow of 1.5 mL min^{-1} was used as the carrier

Table 1 Summary of lipid yield from interior samples from each analyzed site

Site	Period	*N* vessels sampled	Solvent extraction of internal samples				Acid fraction of saponified internal samples			
			Minimum TLE (μg g^{-1})	Maximum TLE (μg g^{-1})	Average TLE (μg g^{-1})	% with > 5 μg g^{-1}	Minimum TLE (μg g^{-1})	Maximum TLE (μg g^{-1})	Average TLE (μg g^{-1})	% with > 5 μg g^{-1}
Sha'ar Hagolan (SHH)	Pottery Neolithic	16	0.1	8.1	1.4	6.3	0.8	18.0	7.1	56.3
Neve Yam (NY)	Early Chalcolithic	17	0.0	5.0	1.2	5.9	0.4	58.9	5.7	11.8
Tel Hreiz (TH)	Early Chalcolithic	3	0.1	5.3	2.4	33.3	2.1	8.3	4.6	33.3
Tel Tsaf (TSF)	Middle Chalcolithic	103	0.0	300.6	5.9	12.6	0.1	195.0	18.4	53.4
Abu Matar (AM)	Late Chalcolithic	3	0.1	1.6	0.9	0.0	2.3	2.9	2.7	0.0
Ashqelon Aganim (AG)	Late Chalcolithic	28	0.0	14.8	1.1	3.6	1.1	49.3	7.2	46.4
Tel Bene Beraq (South) (BB)	Late Chalcolithic	10	0.0	55.3	6.5	10.0	4.0.0	86.4	17.6	60.0
Fazael 7 (FZ)	Late Chalcolithic	8	0.1	3.9	1.3	0.0	3.0	28.4	9.2	62.5
Gilat (GLT)	Late Chalcolithic	13	0.1	88.1	9.1	23.1	1.4	92.2	12.3	61.5
Peqi'in (PEQ)	Late Chalcolithic	3	0.0	0.5	0.3	0.0	6.7	10.7	8.0	100.0
Tsomet Shoket (TS)	Late Chalcolithic	18	0.1	2.1	0.7	0.0	0.9	30.3	6.1	38.9
Yehud (YEH and YV)	Late Chalcolithic	7	0.3	13.0	2.8	12.5	3.7	5.8	4.5	33.3
Total		**229**	**0.0**	**300.6**	**4.0**	**9.6**	**0.1**	**195.0**	**12.8**	**48.9**

gas. The compounds were separated on a DB-5 ms column (30 m × 0.25 mm; 0.25-μm film thickness). The oven temperature was raised from 50℃ (held isothermally for 2 min) to 230℃ at 5℃ min^{-1} and to 310℃ (held isothermally for 10 min) at 30℃ min^{-1}. Compounds eluting from the GC between 31 and 38 min were combusted at 1000℃ in a CuO/NiO/Pt combustion reactor. During the rest of the run, the backflush mode was used to divert unwanted matrix components and limit oxidation. In the MS, the filament current was set to 1.5 mA, the electron energy was set to 124 eV, and the high voltage was set to 3 kV. During the measurements, a vacuum of ca. 6×10^{-7} mBar was maintained. Carbon dioxide was used as the reference gas for compound-specific isotope analysis. Compound-specific palmitic and stearic acid standards were measured at the beginning of each sequence and after every three runs. Samples were measured in duplicate. A precision of 0.5‰ was assumed.

The compound-specific isotope results were plotted against a modern global reference database; all references had a post-industrial carbon (PIC) correction of 1.14‰ (following Friedli et al. 1986). The modern references included domestic ruminants and non-ruminants (Debono Spiteri 2012; Dudd 1999; Gregg 2009; Outram et al. 2009). Reference isotope signatures for ruminants raised on a C_3 and C_4 diet were included because in the Levant, there are some native C_4 plants (Rudov et al. 2020; Winter and Troughton 1978). Identifications were based on the plot of $\delta^{13}C_{16:0}$ and $\Delta^{13}C$ ($\delta^{13}C_{18:0}$-$\delta^{13}C_{16:0}$) because $\Delta^{13}C$ removes environmental biases and allows animal fats to be identified solely based on metabolic pathways (Copley et al. 2003). Mixing curves were plotted following Woodbury et al. (1995) based on the isotope values of $\delta^{13}C_{16:0}$ and $\delta^{13}C_{18:0}$ and the concentrations of $C_{16:0}$ and $C_{18:0}$ in each food source.

Results

GC–MS results

The lipid yield after solvent extraction (Table 1) ranged from 0.0 to 300.6 μg g^{-1}, and only 9.6% of the vessels ($n = 22$) had over 5 μg g^{-1}. The lipid yield of the bound acid fraction released by saponification (Table 1) ranged from 0.1 to 195.0 μg g^{-1}, and 48.9% of the vessels ($n = 110$) had over 5 μg g^{-1} of lipids. An additional crust on a vessel from Middle Chalcolithic Tel Tsaf had over 5 μg g^{-1} of lipids in both the solvent extract (6.8 μg g^{-1}) and acid fraction (12.8 μg g^{-1}).

In the external control samples, there are trace amounts of lipids. The lipids include primarily various fatty acids, *n*-alkanes, and alcohols (Supplementary material 2). These lipids may originate from the original vessel content, which could have spilled over during vessel use or seeped through

the vessel walls; alternatively, these may result from exposure of the sherd after discard to various sedimentary debris. Distinguishing these is complicated because of the often insignificant lipid yield in the vessel interior. However, eight vessels from Pottery Neolithic Sha'ar Hagolan, Middle Chalcolithic Tel Tsaf, and Late Chalcolithic Tel Bene Beraq (South), Gilat, Tsomet Shoket, and Yehud had significant lipid yields externally, with most of these containing insignificant amounts of lipids internally. This discrepancy suggests that the lipids relate to the application of an external surface treatment or to spillage of the original vessel content rather than contamination. Exogenous contamination is further excluded based on the lipid profiles obtained from the soil controls, which represent the lipid "background noise" from post-depositional contexts. The identified lipids include primarily saturated fatty acids, *n*-alkanes, and alcohols (Supplementary material 2), which relate to plant detritus.

Phthalates were found in varying amounts in the archeological samples; these may be related to vessel contact with plastic bags and crates. Octocrylene and oxybenzone, components of sunscreen, were noted in small amounts from vessels from Middle Chalcolithic Tel Tsaf and Late Chalcolithic Tel Bene Beraq (South), Tsomet Shoket, and Yehud. The lipid profiles of samples containing sunscreen were not analyzed further. Vessels from nearly every site contained cyclic octaatomic sulfur. This may be found in the sediment, which is often saline (Singer 2007: 231–248), and it could be incorporated during ceramic production as part of clay processing (Reber et al. 2019) or through post-depositional processes.

Other lipids include fatty acids, dicarboxylic acids, sulfur-heterocyclic fatty acids, acylglycerols, *n*-alkanes, alcohols, sterols, terpenoids, wax esters, ketones, and lactones. Most of these are likely related to the original vessel function.

Fatty acids

In the solvent extracts of the samples, saturated fatty acids range from $C_{6:0}$ to $C_{30:0}$, with usually only high amounts of palmitic ($C_{16:0}$) and stearic ($C_{18:0}$) acids. There is no clear preference for palmitic over stearic acid that would be indicative of plant products in arid conditions (Copley et al. 2005). Specifically, from Late Chalcolithic sites located in the northern Negev, there are a few samples with exclusively short-chain fatty acids, including enanthic acid ($C_{7:0}$), caprylic acid ($C_{8:0}$), pelargonic acid ($C_{9:0}$), and capric acid ($C_{10:0}$) (AM-2, GLT-1, GLT-3, GLT-10, GLT-12, GLT-13, and TS-20). This may attest to the limited utilization of a specific product or contamination. In contrast, in the acid fraction, most samples have a wide range of saturated fatty acids ($C_{8:0}$–$C_{30:0}$). Palmitic and stearic acid are the most abundant, and the majority of the vessels contained both odd- and even-numbered saturated fatty acids in variable amounts. The very long-chain saturated fatty acids are typically even, while the odd-chain saturated fatty acids include mainly pentadecylic acid ($C_{15:0}$) and margaric acid ($C_{17:0}$). These odd-chain saturated fatty acids are commonly related to ruminant fat (Doreau and Chilliard 1997).

Unsaturated fatty acids in the solvent extracts include primarily $C_{16:1}$ and $C_{18:1}$. $C_{20:1}$ was identified in only two vessels dating to the Middle Chalcolithic. Unsaturated fatty acids are more common in the acid fractions. These include primarily $C_{16:1}$ and $C_{18:1}$, but $C_{18:2}$, $C_{18:3}$, $C_{20:1}$, $C_{22:1}$, and $C_{24:1}$ were also identified. The position of the unsaturation could not be identified through oxidation products because, while there are some dicarboxylic acids, in most samples, oxidation products were absent. The amount of $C_{18:1}$ was always less than $C_{16:0}$ and $C_{18:0}$, and its origin is unclear as $C_{18:1}$ is known to be present in plant oils and animal fats (Pollard and Heron 2008: table 11.2). The presence of $C_{18:2}$ and $C_{18:3}$ in some vessels (including various bowls, jars, churns, strainers, and typologically unidentified vessel forms) is consistent with plant products (Pollard and Heron 2008: 385, table 11.2). $C_{20:1}$, $C_{22:1}$, and $C_{24:1}$ were identified in bowls, various jars, churns, cornets, and typologically unidentified fragments from only Chalcolithic sites. Specific isomers of these unsaturated fatty acids may stem from seed oils, such as from *Brassicaceae* plants (e.g., Colombini et al. 2005a). A marine origin could also be considered, although it is unlikely as the samples each have one isomer, and fish contain multiple isomers of each monounsaturated fatty acid (Shimizu and Ando 2012: table 3).

Furthermore, in the acid fractions, *iso*- and *anteiso*-branched-chain saturated fatty acids ($C_{14:0}$–$C_{18:0}$) were commonly identified. These may be synthesized by bacteria in the rumen (Doreau and Chilliard 1997) or originate from sediment bacteria (Amblès et al. 1994).

Dicarboxylic acids

Dicarboxylic acids were identified only in samples dating to the Middle and Late Chalcolithic period. From the Middle Chalcolithic site of Tel Tsaf, sebacic acid was identified in one jar (TSF19-4). This may form from the oxidation of ricinoleic acid (Naughton 1974). Furthermore, in the acid fraction of four vessels from Tel Tsaf (TSF17-7, TSF17-23, TSF18-13, and TSF18-24), azelaic acid was identified. This is an oxidation product of C_{18} unsaturated fatty acids (Passi et al. 1993).

Additionally, in the acid fraction of jars and typologically unidentified vessel fragments from the Middle Chalcolithic period (TSF17-23, TSF17-28, TSF17-31, TSF18-2, and TSF19-3; Fig. 2) and jars and a cornet from the Late Chalcolithic period (AG-32, BB-1, BB-4, and BB-21), there are long-chain dicarboxylic acids ranging from C_{14} to C_{30}. The long-chain dicarboxylic acids are primarily

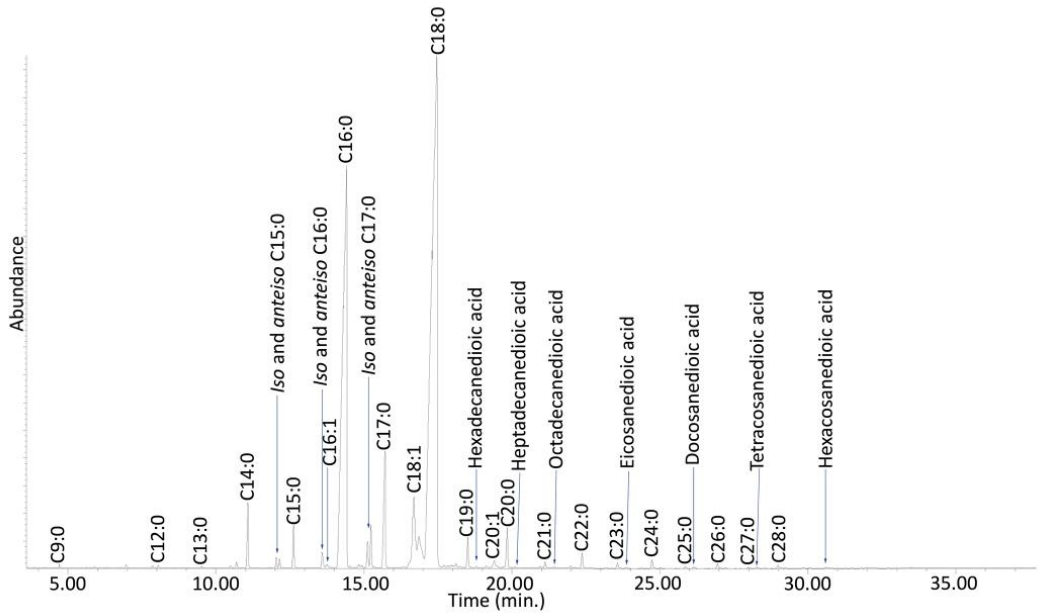


Fig. 2 Partial total ion chromatogram of the methylated acid fraction obtained after saponification of the ceramic powder from the internal surface of a necked jar rim fragment from Tel Tsaf (TSF19-3), showing the long-chain dicarboxylic acids (Cx:y = fatty acid with chain length *X* and *Y* number of double bonds)

even-numbered. They are unlikely to represent oxidation products because these could only have been formed from very long-chain unsaturated fatty acids, which are rare. Instead, these may originate naturally from nuts (e.g., almonds, pistachio, and walnuts; Dembitsky et al. 2002), hardwood trees (Fine et al. 2002; Gandini et al. 2006), and wild plants (Rezanka 1998, Table 1). Nut lipids were not considered as a potential source because their dicarboxylic acids range only from C_{14} to C_{22} (Dembitsky et al. 2002), unlike the archeological samples. Further analysis of modern reference samples is required to identify if this set of dicarboxylic acids could be indicative of local hardwood trees and/or wild plants.

Sulfur-heterocyclic fatty acids

In five vessels from Middle Chalcolithic Tel Tsaf and Late Chalcolithic Gilat, sulfur-heterocyclic fatty acids (SHFAs) were identified (Fig. 3). This includes one V-shaped bowl, a churn, and three typologically unidentified vessel fragments (TSF18-4, TSF18-6, TSF18-16, TSF18-55, and GLT-5). The SHFAs present are all SHFA1 (C_{18}) based on the mass to charge ratio (*m/z*) of the M-15 – *m/z* 313. This is an alkanoic acid with 18 carbon atoms and a tetrahydrothiophene ring (Eibler et al. 2017). The placement of the alkyl chain was identified based on the *m/z* of the α-ion and the alkyl chain + 44 u (following Eibler et al. 2017). In three vessels from Tel Tsaf (Fig. 3a), there is 6THTph8 (6-(5-Octyltetrahydrothiophene-2-yl) hexanoic acid; *m/z* 215 and 157) and 5THTph9 (5-(5-Nonyltetrahydrothiophene-2-yl) pentanoic acid; *m/z* 201 and 171). One vessel from Tel Tsaf (Fig. 3b) contains these in addition to other isomers – 7THTph7 (7-(5-Heptyltetrahydrothiophene-2-yl) heptanoic acid; *m/z* 229 and 143) and 4THTph10 (4-(5-Decyltetrahydrothiophene-2-yl) butanoic acid; *m/z* 187 and 185). Similarly, in the churn from Gilat, SHFAs include 8THTph6 (8-(5-Hexyltetrahydrothiophene-2-yl) octanoic acid; *m/z* 243 and 129), 7THTph7, 5THTph9, and 4THTph10 (Fig. 3c).

These SHFAs have been identified so far in modern rapeseed oil, mustard seed oil, and garlic (Dembitsky et al. 2007; Eibler et al. 2017; Wijesundera and Ackman 1988) and rarely in archeological samples (Mathur et al. forthcoming), and further analysis may identify these in other plants. As rapeseed is not native to the Levant, mustard seed, garlic, and other wild *Allium* and *Brassicaceae* plants were considered. All of these are present in Israel (Danin 2004: 91–115, 395–401) and have been identified in the

prehistoric archeological record. Garlic was identified at two Late Chalcolithic sites in the Judean Desert—Nahal Mishmar and Cave V/49, which also contained Roman remains (Melamed 2002; Zaitschek 1961). Mustard seeds were identified at Netiv Hagdud, a Pre-Pottery Neolithic site in the Jordan Valley (Kislev 1997: table 8.1), and Shiqmim, a Late Chalcolithic site in the northern Negev (Kislev 1987: table 9.2). Mustard seed oil is reinforced in some samples by the concurrence of SHFAs with $C_{20:1}$ and $C_{22:1}$, which are indicative of seed oils (Colombini et al. 2005a). However, in the samples without these monounsaturated fatty acids, a specific origin cannot be suggested because the SHFA pattern identified here is not specific to either mustard seed or garlic (Dembitsky et al. 2007; Eibler et al. 2017). Garlic is potentially identifiable in residues that contain just 8THTph6 and 5THTph9 (Dembitsky et al. 2007), and in all of the archeological samples that contain these, there are also other positional isomers, so the SHFA source could not be distinguished.

Acylglycerols

Acylglycerols were identified in samples from Pottery Neolithic and Early, Middle, and Late Chalcolithic contexts. Generally, these are restricted to monoacylglycerols (MAGs) and within this, 1-Monopalmitin and 1-Monostearin. A few unique samples contained additional MAGs. From the Pottery Neolithic site of Sha'ar Hagolan, SHH-1 and SHH-11, a jar and a typologically unidentified vessel fragment, have 1-Monomyristin and 1-Glycerol pentadecanoate, and one typologically unidentified vessel fragment, SHH-17, has 1-Monocaprylin. From the Early Chalcolithic sites of Neve Yam and Tel Hreiz, 1-Monomyrstin was identified in a few samples (NY-8, NY-11, NY-16, and TH-4), and 1-Monooleoylglycerol was identified in one vessel (TH-4). From the Middle Chalcolithic site of Tel Tsaf, one sample contained 1-Monomyristin, 1-Glycerol pentadecanoate, 1-Monopalmitin, 1-Glycerol heptadecanoate, 1-Monostearin, and 1-Monooleoylglycerol (TSF18-69). Finally, in the Late Chalcolithic period, 1-Monomystin was identified in one strainer from Yehud (YEH-1), and 1-Monooleoylglycerol was identified in two jars and one cornet from Ashqelon Agamim, Tel Bene Beraq (South), and Tsomet Shoket (AG-32, BB-28, and TS-7). The odd-numbered MAGs identified in the vessels from Sha'ar Hagolan and Tel Tsaf are particularly remarkable in that they reflect the degradation of odd-numbered triacylglycerols (TAGs). Such TAGs are rare, but TAGs with $C_{15:0}$ and $C_{17:0}$ have so far been recovered from ruminant adipose and dairy fat (e.g., Lísa et al. 2011; Ruiz-Sala et al. 1996). Diacylgylcerols (DAGs) are less common, but they were also identified throughout the entire sequence. They are restricted to isomers formed from $C_{14:0}$ (myristic acid), $C_{16:0}$, and $C_{18:0}$.

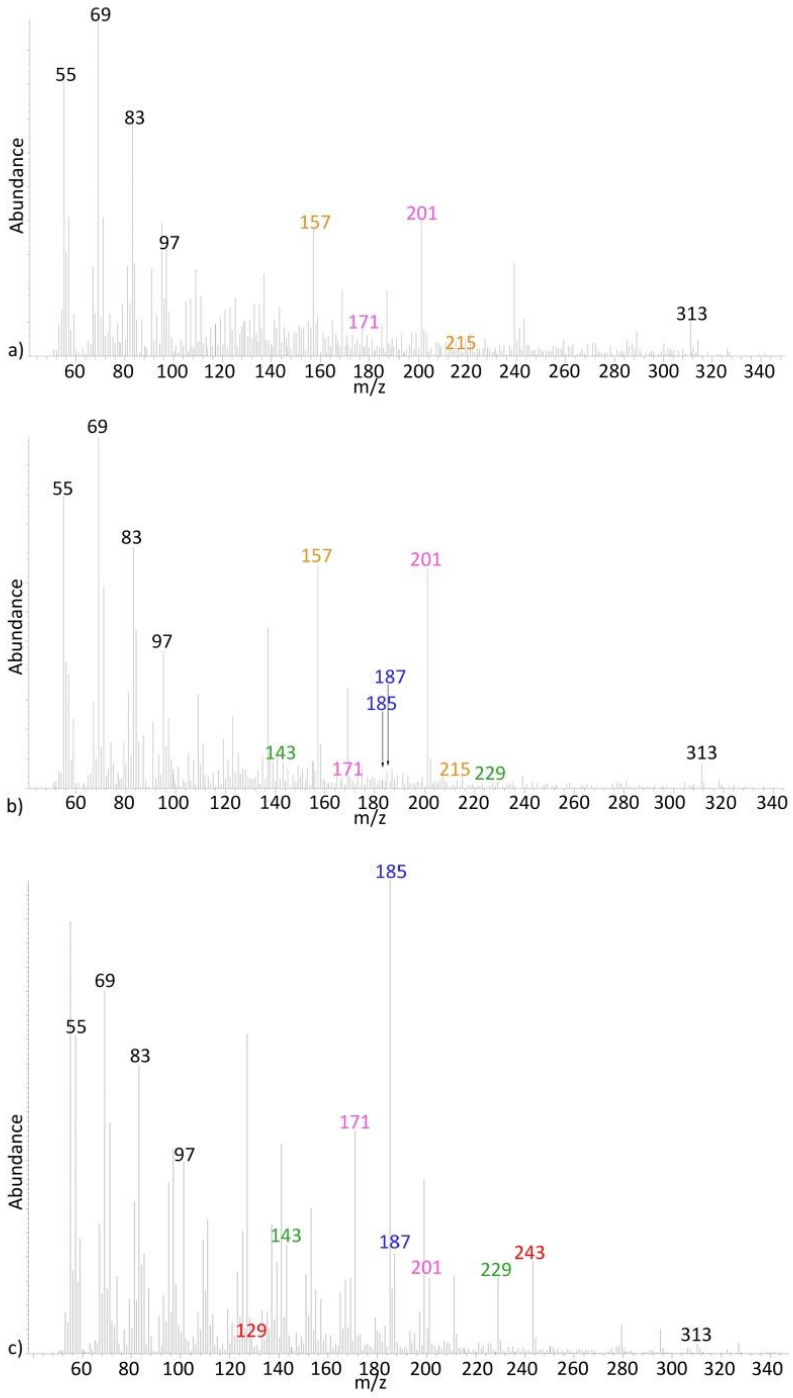
From Pottery Neolithic Sha'ar Hagolan, TAGs were identified in one sample taken from a vessel interior (SHH-17) and one sample taken from a vessel exterior (SHH-1). These include TAGs with 24 to 30 carbon atoms, formed from caprylic acid ($C_{8:0}$) and capric acid ($C_{10:0}$) (Fig. 4a). Additional samples from internal vessel surfaces with insignificant lipid yields (SHH-1, SHH-4, SHH-7, SHH-19, and SHH-20) contain similar TAG profiles. The correlated degradation products (saturated fatty acids, MAGs, and DAGs) are absent, suggesting that the TAGs may result from contamination or that the degradation products were leached out from the ceramic matrix after TAG hydrolysis.

From the Late Chalcolithic period, TAGs were identified on the interior surface of one vessel from Ashqelon Agamim (AG-26). This typologically unidentified vessel fragment has even-numbered TAGs with 46–54 carbon atoms, formed from various combinations of $C_{14:0}$, $C_{16:0}$, and $C_{18:0}$ (Fig. 4b), as well as some of the correlated degradation products. The TAG profile however is non-indicative. TAGs were also identified in two other vessels with insignificant lipid yields (GLT-5 and YEH-1), deterring a secure archeological attribution.

n-Alkanes

N-alkanes range from C_{14} to C_{37}. Short-chain *n*-alkanes (C_{15}–C_{29}, maximizing at C_{15} and C_{25}) characteristic of bacterial input (Poynter et al. 1989: 439, 458) were identified occasionally. Most *n*-alkane profiles maximize at C_{29} as characteristic of higher plants (Eglinton and Hamilton 1967; Poynter et al. 1989). No samples maximized at tritriacontane (C_{33}), distinctive of C_4 plants (Rommerskirchen et al. 2006). The ACL ranges from 23.4 to 31.4, averaging at 28.2. This is characteristic of higher plants (Eglinton and Hamilton 1967; Poynter et al. 1989: 435). The CPI could not be calculated in many samples because there were only odd-numbered *n*-alkanes. In the samples with even-numbered *n*-alkanes, the CPI ranges from 0 to 25.2, averaging at 4.1. This shows that while some samples with CPI values above 1.0 can be securely linked to plant products (Diefendorf et al. 2011), not all the *n*-alkanes originated from plants.

The P_{aq} could not be calculated in most samples because C_{23} and C_{25} are absent. In the samples with C_{23} and C_{25}, the P_{aq} ranges from 0.04 to 1.00 and averages at 0.41. This shows that in addition to terrestrial plants, there are also lipids from macrophytes and halophytes (Eley et al. 2014: SI Table 1; Ficken et al. 2000). Within these samples, some P_{aq} values are between 0.10 and 0.40, relating to emergent macrophytes (Ficken et al. 2000), and others are over 0.40, relating to submerged macrophytes (Ficken et al. 2000). These P_{aq} values were identified in vessels from nearly every site, but they are less common from Pottery Neolithic vessels. Such values are expected from vessels recovered from Early


a)
Abundance
m/z
69
55
83
97
157
201
171
215
313
b)
Abundance
m/z
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83
97
157
201
187
185
143
171
215
229
313
c)
Abundance
m/z
185
69
55
83
171
97
143
187
201
229
243
129
313

◂**Fig. 3** Mass spectra of SHFAs in the methylated samples of the internal surface of (**a**) a V-shaped bowl rim fragment from Tel Tsaf (TSF18-4): 6THTph8 and 5THTph9; (**b**) a wall fragment from Tel Tsaf (TSF18-16): 7THTph7, 6THTph8, 5THTph9, and 4THTph10; and (**c**) a churn from Gilat (GLT-5): 8THTph6, 7THTph7, 5THTph9, and 4THTph10; 8THTph6 was identified by α-ion = 243 and alkyl chain + 44 u = 129 (colored red). 7THTph7 was identified by α-ion = 229 and alkyl chain + 44 u = 143 (colored green). 6THTph8 was identified by α-ion = 215 and alkyl chain + 44 u = 157 (colored orange). 5THTph9 was identified by α-ion = 201 and alkyl chain + 44 u = 171 (colored pink). 4THTph10 was identified by α-ion = 187 and alkyl chain + 44 u = 185 (colored blue)

Chalcolithic Neve Yam and Tel Hreiz because both sites are located under the present-day Mediterranean shoreline, and when the sites were in use, they were located along the coast (Galili et al. 2017), which is an ideal environment for both macrophytes and halophytes. However, their presence at other sites shows that these types of plants were available and used throughout Israel. The related plants were used in connection to bowls, jars, pithoi, churns, cornets, and strainers.

Alcohols

Alcohols range from C_{12} to C_{34}. Most are long-chain and even. This general pattern is indicative of plant products (Eglinton and Hamilton 1967; Poynter et al. 1989: 435). No vessels maximized at C_{32} as characteristic of C_4 plants (Rommerskirchen et al. 2006). It is unclear where the odd-numbered and short-chain alcohols originate from.

Sterols

Sterols were identified in limited vessels from the Pottery Neolithic though the Late Chalcolithic. Most of these contain exclusively cholesterol, but phytosterols (β-sitosterol, campesterol, and stigmasterol) were also identified in few samples. These relate to animal and plant products respectively (Evershed 1993). The low lipid yield of most of these vessels deters forming a concrete interpretation. Sterol oxidation products (e.g., Hammann et al. 2018) were only identified in one vessel from the Pottery Neolithic site of Sha'ar Hagolan (SHH-1); cholesterol was identified with 7-keto-cholesterol.

Terpenoids

Terpenoids were identified in samples from a few Pottery Neolithic, Middle Chalcolithic, and Late Chalcolithic sites. These are mainly restricted to abietic acids, which are characteristic of coniferous trees (Mills and White 1977), but these terpenoids alone cannot confirm the presence of a resin and rule out contamination. From the Pottery Neolithic site of Sha'ar Hagolan, dehydroabietic acid was identified in the solvent extract of the exterior of one vessel (SHH-5). In the Middle Chalcolithic site of Tel Tsaf, dehydroabietic acid was identified in the solvent extract or acid fraction of three vessels (TSF18-6, TSF19-3, and TSF19-6), and methyl dehydroabietate was identified in the acid fraction of one vessel (TSF18-1). This includes both jars and bowls. In the Late Chalcolithic sites of Ashqelon Agamim and Tel Bene Beraq (South), dehydroabietic acid was identified in two solvent extracted vessels (BB-3 and BB-28) and the acid fraction of two saponified vessels (AG-25 and AG-30). Within the Late Chalcolithic samples, two vessels are further unique (AG-30 and BB-19; Fig. 5). Both have methyl abietate, methyl dehydroabietate, and 7-oxodehydroabietic acid, and AG-30 (Fig. 5a) contains in addition pimaric acid and β-pimaric acid. The wider set of terpenoids is more characteristic of conifers (Otto and Wilde 2001), and 7-oxodehydroabiatic acid, an oxidation marker (Mills and White 1977; Ribechini et al. 2008), reinforces that the residues are archeological. Moreover, the presence of methyl dehydroabietate in the vessels from Tel Tsaf, Ashqelon Agamim, and Tel Bene Beraq (South) is potentially suggestive of a tar residue because it is a thermal degradation byproduct (Pollard and Heron 2008: 243).

Further in one jar from Late Chalcolithic Tel Bene Beraq (South), unique terpenoids were noted—sempervirol, ferruginol, totarol, and 2,3-dehydroferruginol (BB-1). These may originate from specific conifers including *Cupressaceae* and *Cedrus* (Mangeni and Caputo 1967; Otto and Wilde 2001: table 8). These terpenoids were also identified in the vessel exterior and surrounding sediment (BB-12 S; Supplementary material 2), and the general lipid yield is low, so post-depositional contamination must be considered.

Wax esters

Even-numbered palmitate wax esters were identified at sites throughout the entire sequence. These range from C_{24} to C_{50}. The interior ceramic surface (NY-12 and BB-21), exterior ceramic surface (TS-4), and crust (TSF18-57) of a few vessels contain long-chain palmitate wax esters paired with saturated fatty acid, *n*-alkane, and alcohol profiles, previously published, that are suggestive of beeswax (Fig. 6; Tulloch 1970). Others contain similar lipid profiles, but the identification of beeswax is tenuous because of the low lipid yield in these vessels.

Additional vessels from Pottery Neolithic Sha'ar Hagolan and Late Chalcolithic Gilat and Yehud contain exclusively shorter wax esters (C_{24}–C_{38}). These were identified in churns, cornets, strainers, and typologically unidentified vessel forms. While their source is unclear, they may originate from plant products, which are characterized by a wide

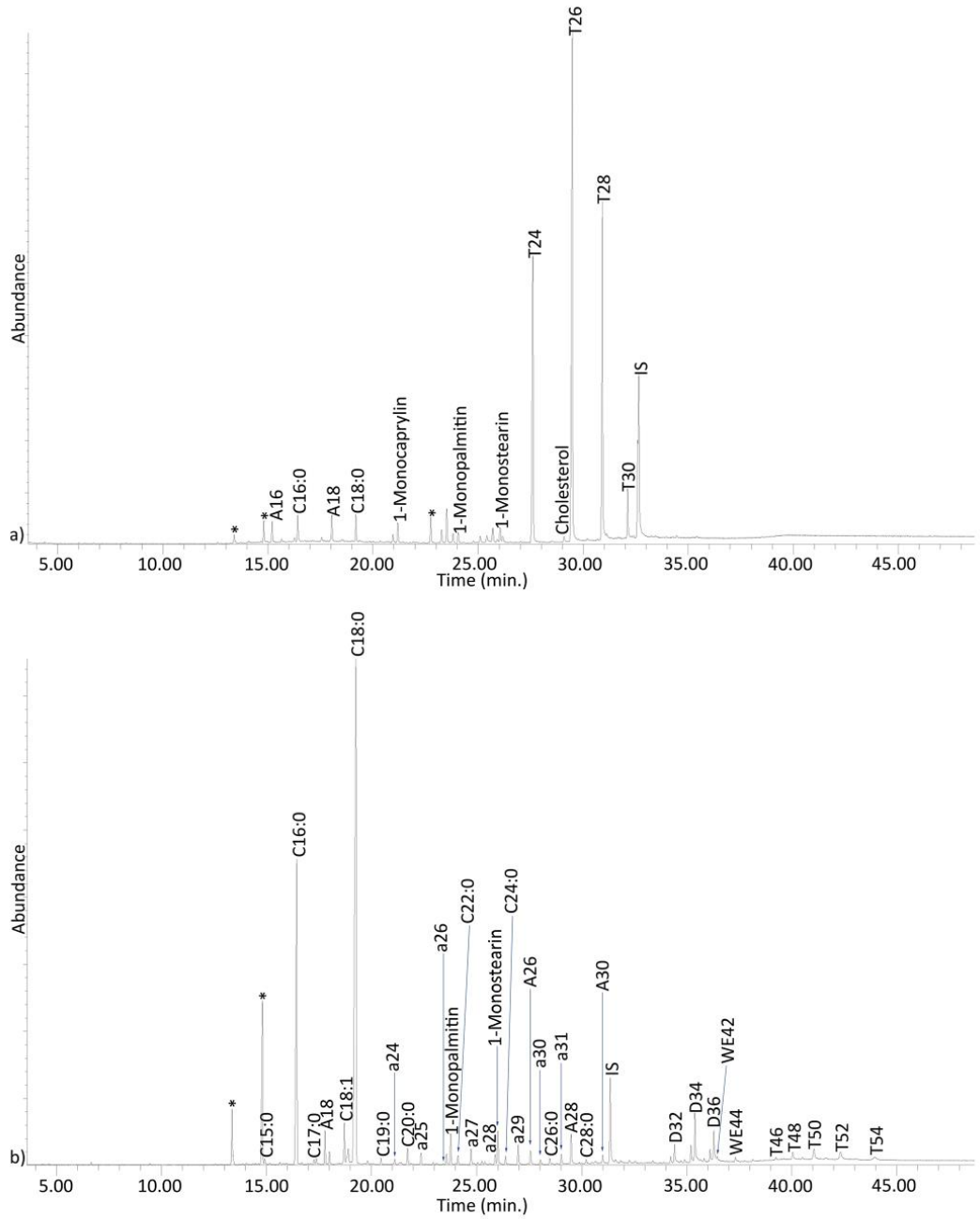


Fig. 4 Partial total ion chromatogram of wall fragments from (**a**) Sha'ar Hagolan (SHH-17); and (**b**) Ashqelon Agamim (AG-26) run as TMS derivatives, showing the triacylglycerol signatures (*=plasticizer; ax=*n*-alkane with chain length *X*; Ax=alcohol with chain length *X*; Cx:y=fatty acid with chain length *X* and *Y* number of double bonds; Dx=diacylglycerol with *X* carbon atoms; IS=internal standard; Tx=triacylglycerol with *X* carbon atoms; WEx=wax ester with chain length *X*)

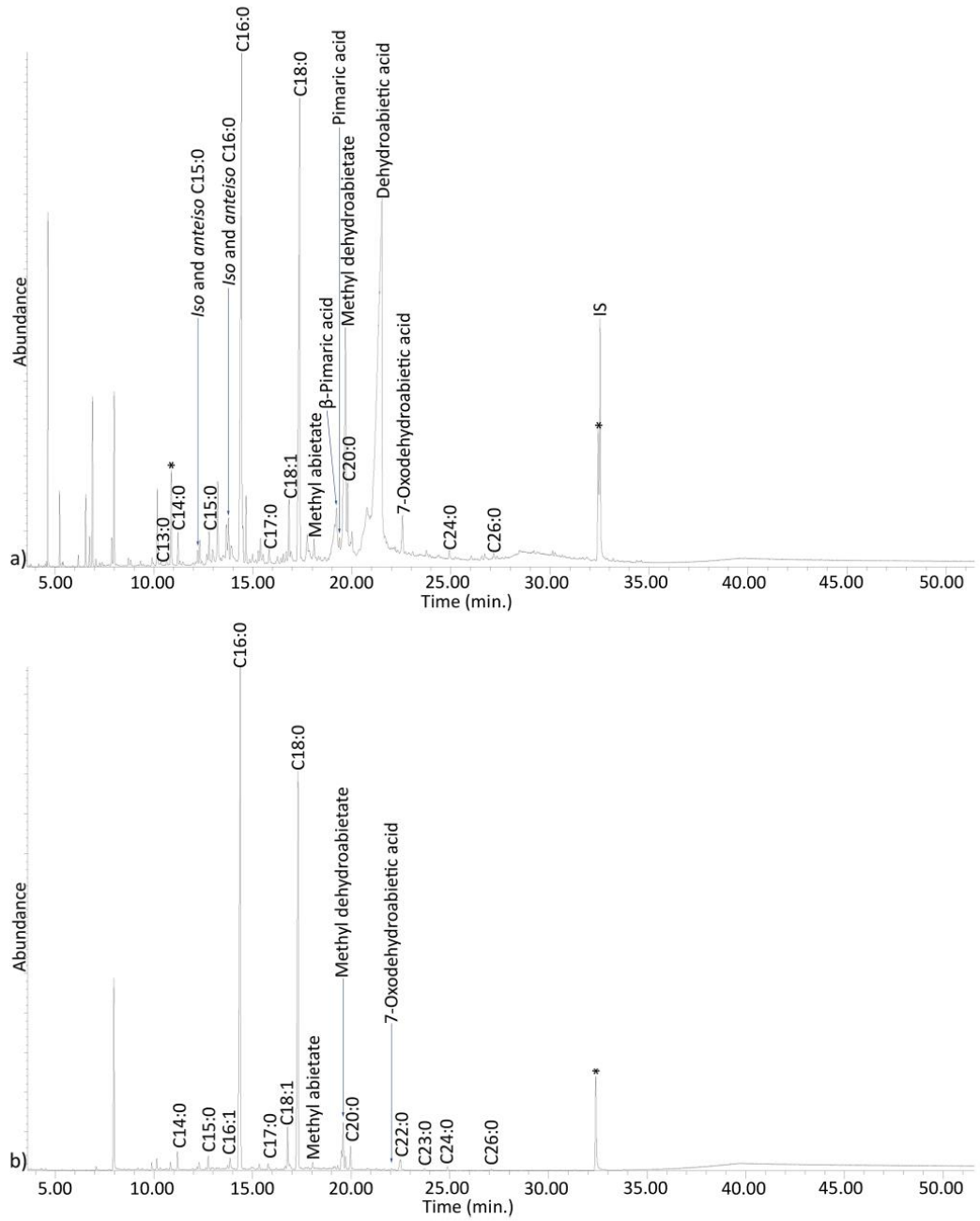


Fig. 5 Partial total ion chromatogram of (**a**) a cornet from Ashqelon Agamim (AG-30); and (**b**) a jar base fragment from Tel Bene Beraq (South) (BB-19) run as methyl esters after saponification showing terpenoids indicative of a plant resin (*=plasticizer; Cx:y=fatty acid with chain length *X* and *Y* number of double bonds; IS=internal standard)

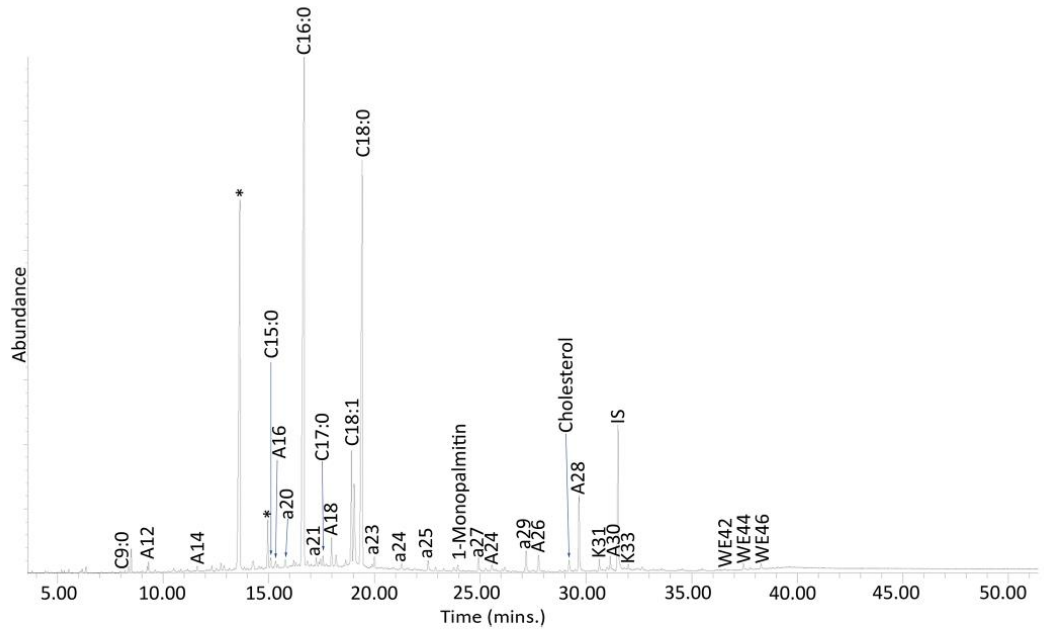


Fig. 6 Partial total ion chromatogram of a jar neck fragment from Neve Yam (NY-12) run as a TMS derivative. The lipid profile obtained is consistent with a beeswax residue (*=plasticizer; ax=*n*-alkane with chain length *X*; Ax=*n*-alcohol with chain length *X*; Cx:y=fatty acid with chain length *X* and *Y* number of double bonds; IS=internal standard; Kx=ketone with chain length *X*; WEx=wax ester with chain length *X*)

range of wax esters with typically 32–64 carbon atoms (Tulloch 1976).

Ketones

Ketones were identified at several sites in various jars, a platter, a churn, a basalt bowl, and typologically unidentified forms. The majority contain a suite of hentriacontan-16-one (K_{31}), tritriacontan-16-one (K_{33}), and pentatriacontan-18-one (K_{35}). These are pyrolysis markers formed from heating palmitic and stearic acid at temperatures over 300°C or prolonged exposure to lower temperatures (Drieu et al. 2019; Raven et al. 1997). When heated, K_{31}, K_{33}, and K_{35} form in a 1:2:1 ratio (Raven et al. 1997), although this precise ratio was not identified in most vessels. Few samples contain singular ketones (SHH-6, TSF18-57, BB-21, and GLT-1), which is considered indicative of higher-plant input (Evershed et al. 1995; Raven et al. 1997). In the sample from Middle Chalcolithic Tel Tsaf, just K_{31} was identified. This is found in *Allium* plants like garlic (Rabah et al. 2020; Raven et al. 1997); however, the origin cannot be confirmed using SHFAs (Dembitsky et al. 2007), which are absent in this sample. The sample from Pottery Neolithic Sha'ar Hagolan contains just K_{35}, and the samples from Late Chalcolithic Tel Bene Beraq (South) and Gilat contain just K_{33}; both of these are rare in plant products (Raven et al. 1997). Additionally, some samples from Tel Tsaf contained other ketones with 29 (K_{29}), 33 ($K_{33:1}$), 34 (K_{34}), and 35 ($K_{35:1}$) carbon atoms (TSF18-36 and TSF18-69). $K_{33:1}$ and $K_{35:1}$ are formed from the pyrolysis of $C_{18:1}$ (Evershed et al. 1995).

Lactones

C_7–C_{12} γ-lactones were identified in a few samples from Early and Late Chalcolithic sites. This includes one jar (TH-2), a holemouth jar (GLT-2), and a churn (GLT-3). There is a predominance of the C_9 γ-lactone. While lactones can be found naturally in several products, these specific lactones relate to the oxidation of unsaturated fatty acids (e.g., Wantabe and Sato 1969).

GC-c-IRMS results

GC-c-IRMS was applied to 46 vessels from Pottery Neolithic Sha'ar Hagolan, Early Chalcolithic Neve Yam, Middle Chalcolithic Tel Tsaf, and Late Chalcolithic Ashqelon

Agamim, Tel Bene Beraq (South), Fazael 7, Gilat, Peqi'in, and Tsomet Shoket (Fig. 7). $\delta^{13}C_{16:0}$ ranged from − 30.1 to − 18.9‰, and $\delta^{13}C_{18:0}$ ranged from − 30.5 to − 23.2‰. The animal fat source was characterized based on the $\Delta^{13}C$ values and comparison to the average value and the first standard deviation of the modern reference fats.

In the Pottery Neolithic and Early Chalcolithic sites, the samples plot within the range of adipose (Fig. 7a–b). Within these, based on the $\Delta^{13}C$ values (Fig. 7a), one sample plots within the first standard deviation of porcine adipose (SHH-7), the most likely source of non-ruminant adipose fat. The remainder reflect mixtures of ruminant and non-ruminant adipose. In particular, at Neve Yam, the tested vessel (NY-11) plots on the mixing curve of ruminant and porcine adipose (ca. 85% porcine fat and 15% ruminant fat) (Fig. 7b).

During the Middle Chalcolithic period, based on the $\Delta^{13}C$ values (Fig. 7c), 25% of samples ($n=6$) plot within the first standard deviation of ruminant adipose, 16.7% of samples ($n=4$) plot within the first standard deviation of ruminant dairy fat, and 16.7% of samples ($n=4$) plot within the first standard deviation of porcine or non-ruminant adipose. An additional 33.3% of samples ($n=8$) plot between ruminant and non-ruminant adipose, and 8.3% ($n=2$) of samples plot between ruminant adipose and dairy fat (Fig. 7c). According to the mixing curves, a large portion of the samples that plot between ruminant and non-ruminant adipose relate to lipid contribution from the domestic pig (Fig. 7d).

In the Late Chalcolithic period, based on the $\Delta^{13}C$ values (Fig. 7e), 27.8% of samples ($n=5$) plot within the first standard deviation of ruminant adipose and 16.7% of samples ($n=3$) plot within the first standard deviation of ruminant dairy fat. However, most samples, namely 55.6% ($n=10$), plot between ruminant and non-ruminant adipose (Fig. 7e). Few of these samples plot along the mixing curve of ruminant and porcine adipose (Fig. 7f).

Among these samples, several vessels from Middle Chalcolithic Tel Tsaf and, to a lesser extent, Late Chalcolithic Gilat and Tsomet Shoket have a distinct isotope signature. Some with a $\Delta^{13}C$ value less than 0‰ (characterized as ruminant adipose and dairy) have a more positive $\delta^{13}C_{16:0}$ value (Fig. 7c, e). From these sites, the $\delta^{13}C_{16:0}$ value is on average − 25.4‰, ranging from − 18.9 to − 28.2‰. This wide range of values likely relates to the inclusion of C_4 plants (Meier-Augensterin 2002) or C_3 plants characteristic of arid, water-depleted, and saline environments (Tieszen 1991: Table 1) into the animal diet. As part of an environmental adaption, the latter have a unique photosynthetic pathway to conserve water, resulting in values that are more positive (Tieszen 1991). C_4 plants are found throughout Israel (Rudov et al. 2020), and environmentally enriched C_3 plants were also probably present around these sites because they are located in regions with slight aridity (the Jordan Valley and the northern Negev) and saline soils (e.g., Singer 2007: 233–248).

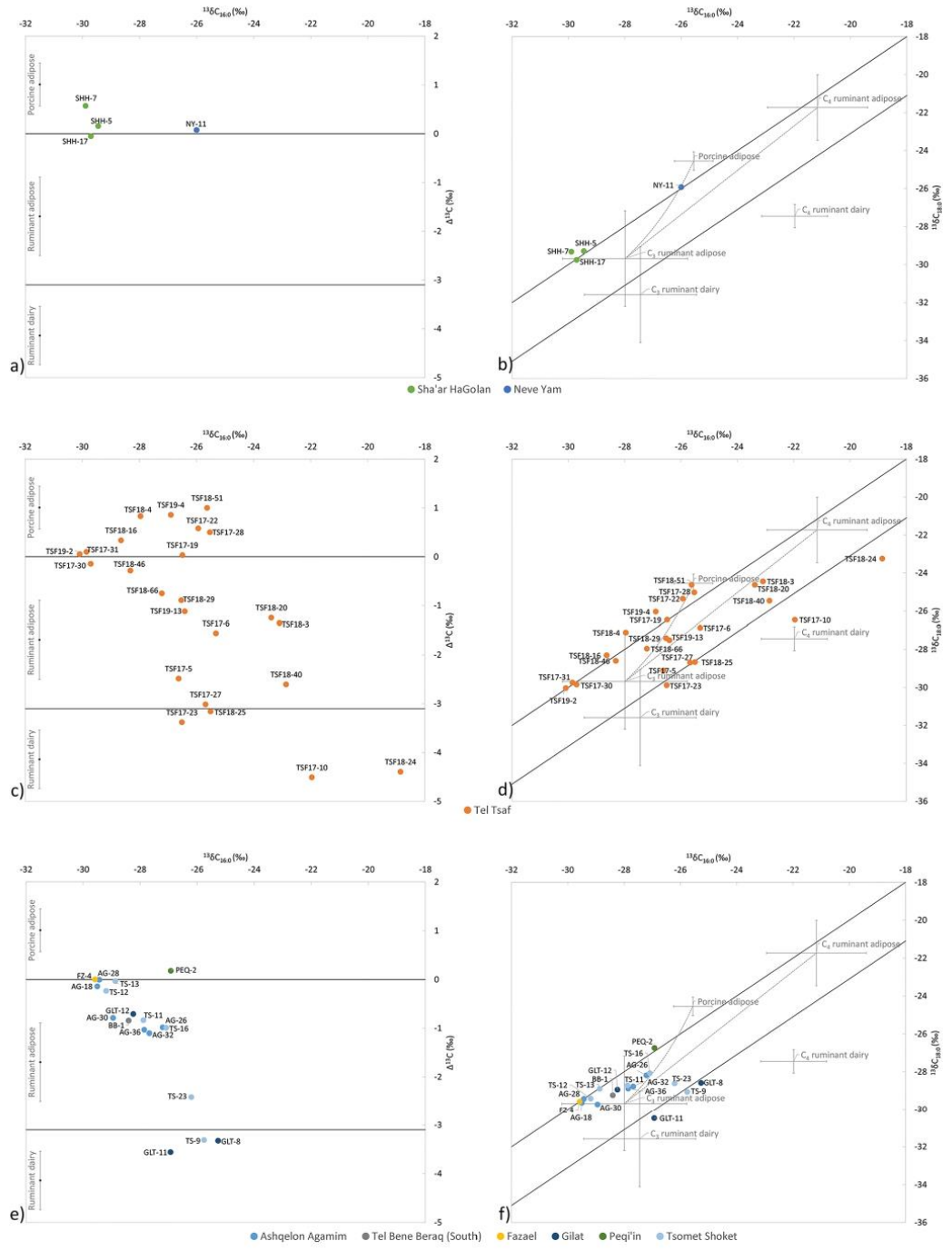
Furthermore, many of the samples that plot within the range of non-ruminant adipose fat ($\Delta^{13}C>0$‰) have relatively depleted $\delta^{13}C_{16:0}$ and $\delta^{13}C_{18:0}$ values. While the most probable source of non-ruminant adipose is pork (see discussion), this must be proposed cautiously because porcine adipose typically has somewhat more positive $\delta^{13}C_{16:0}$ and $\delta^{13}C_{18:0}$ values (Regert 2011) in comparison to the archeological samples. Freshwater fish is a potential alternative, with a more similar isotope signature (Regert 2011), but there is little corroborating lipid biomarker or archeological evidence for fish and fishing (see discussion). Mixtures of porcine and ruminant adipose as well as plant oils could also create similar signatures (e.g., Hendy et al. 2018) and should be considered as an alternative.

Discussion

Food economies and culinary practices are interrelated and regulated by a variety of internal and external factors including most notably environmental regulations and cultural preferences (e.g., Gumerman IV 1997; Twiss 2012; Zeder 2015). In the southern Levant, some of the most significant changes in foodways occurred at the onset of the Holocene, with the advance of agriculture (e.g., Bar-Yosef 1998; Bar-Yosef and Belfer-Cohen 1989). While this formed the basis of the economic strategy in the region for millennia, this study tested for micro-culinary variation in the Pottery Neolithic and Chalcolithic using organic residue analysis to identify whether culinary practices were affected by cultural variation (e.g., Garfinkel 1993; Gilead 2007; Gopher and Gophna 1993; Rowan and Golden 2009), changes in scale and structure of social organization (e.g., Kerner 2010; Levy 1986), or environmental variation between different regions in Israel. The current study indicates dietary continuation throughout the sequence in both animal and plant exploitation, although this must be suggested with some caution because the sample size for each period is variable (with further research needed on more sites dating to the Pottery Neolithic, Early Chalcolithic, and Middle Chalcolithic periods) and future residue and use-wear analyses may still confirm or rebut our findings.

Animal exploitation

The organic residue analysis results show that the vessels tested, regardless of typology, were commonly used in relation to the meat of domestic ruminants. The disassociation between vessel form and function must be suggested with caution because only few of the studied samples were from typologically distinct vessels and other studies, particularly


a)
b)
c)
d)
e)
f)
Porcine adipose
Ruminant adipose
Ruminant dairy
$^{13}\delta C_{16:0}$ (‰)
$\Delta^{13}C$ (‰)
$^{13}\delta C_{18:0}$ (‰)
C_4 ruminant adipose
C_3 ruminant adipose
C_3 ruminant dairy
Porcine adipose
Sha'ar HaGolan
Neve Yam
Tel Tsaf
Ashqelon Agamim
Tel Bene Beraq (South)
Fazael
Gilat
Peqi'in
Tsomet Shoket

◂**Fig. 7** GC-c-IRMS results: (**a**) $\delta^{13}C_{16:0}$ and $\Delta^{13}C$ values of Pottery Neolithic and Early Chalcolithic samples; (**b**) $\delta^{13}C_{16:0}$ and $\delta^{13}C_{18:0}$ values of Pottery Neolithic and Early Chalcolithic samples; (**c**) $\delta^{13}C_{16:0}$ and $\Delta^{13}C$ values of Middle Chalcolithic samples; (**d**) $\delta^{13}C_{16:0}$ and $\delta^{13}C_{18:0}$ values of Middle Chalcolithic samples; (**e**) $\delta^{13}C_{16:0}$ and $\Delta^{13}C$ values of Late Chalcolithic samples; and (**f**) $\delta^{13}C_{16:0}$ and $\delta^{13}C_{18:0}$ values of Late Chalcolithic samples. The animal reference fat ranges represent the mean ± 1 s.d. of the $\delta^{13}C_{16:0}$, $\delta^{13}C_{18:0}$, and $\Delta^{13}C$ values of modern references from the UK (animals raised on a pure C_3 diet), Germany (animals raised on a pure C_3 diet), Italy (animals raised on a pure C_4 diet), Malta (animals raised on a pure C_3 diet and on a pure C_4 diet), Israel, Jordan, and Kazakhstan (animal diet unclear) (Debono Spiteri 2012; Dudd 1999; Gregg 2009; Outram et al. 2009). All modern references had a PIC correction of 1.14‰ (following Friedli et al. 1986). Mixing curves were plotted following Woodbury et al. (1995)

on the Pottery Neolithic (Vieugué et al. 2016), apply different methodologies and suggest a correlation between vessel form and function. The current study shows this preference based on both lipid profiles and compound-specific isotope results (although only few samples, particularly from the Pottery Neolithic and Early Chalcolithic, had sufficient lipids for this analysis), and it is supported by the faunal remains from eight of the studied sites (Table 2). Parallel isotope signatures are observed from a contemporaneous site in Jordan—al-Basatîn (Gregg et al. 2009). According to the published faunal remains from eight of the sites analyzed in this study, domesticated sheep and goats form approximately 25–85% of the number of identified specimens (NISP), and domesticated cattle form ca. 4–35% of the NISP. Wild animals are less common, represented primarily by medium-sized ungulates and small vertebrates (Table 2). Based on the kill-off pattern of these domesticated ruminants, it was suggested that at Sha'ar Hagolan (Marom 2011: 79), Neve Yam (Horwitz et al. 2006), Tel Tsaf (Hill 2011: 124–127, 164–165), Fazael 7 (Bar et al. 2017), and Yehud (Itach et al. 2019) these animals were used specifically for meat because the ruminants were butchered after reaching maturity when the body weight plateaus and there is maximum carcass fat (Payne 1973).

In addition to meat, ruminants were exploited for milk, and in this regard alone chronological variation is noted. While there is at present no evidence for dairy exploitation in the south Levantine Pottery Neolithic and Early Chalcolithic based on prior residue analysis studies (Evershed et al. 2008; Gregg 2009: 115–120, 127–131; Gregg et al. 2009) and this study, the GC-c-IRMS results from Tel Tsaf indicate that dairy was used in the Middle Chalcolithic period. From the 24 samples, pure dairy residues were identified in 16.7% ($n=4$) of the samples, and mixtures with dairy residues were identified in an additional 8.3% of the samples ($n=2$). There is evidence for continued use in the Late Chalcolithic period, with dairy residues identified in 16.7% ($n=3$) of the 18 samples analyzed. These results confirm the earlier suggestion for dairying during the Late Chalcolithic period based on specific material culture items (e.g., churns; Kaplan 1954) and animal survivorship curves at few sites (e.g., Grigson 1987: 225, Fig. 7.6, 1995: 389, Fig. 10.4), and they indicate that dairy was used even before the Late Chalcolithic period.

Among the sites tested however, the faunal remains show only at Gilat that a high number of sheep and goats survived past maturity (Grigson 2006: 225–232) as characteristic of dairy and wool exploitation (Davis 1987: 158; Payne 1973; Vigne and Helmer 2007). Comparison of the faunal remains to these theoretical animal management models suggests that milk was used in such low quantities that a specialized animal management strategy aimed at maximizing milk yields was not required. This contrasts with other indicators for agriculture intensification or the "secondary product revolution" as debatably suggested for the Late Chalcolithic period (e.g., Commenge 2006: 444; Grigson 1987: 225, Fig. 7.6, 1995: 389, 402, Fig. 10.4, 2006: 225–228, 231; Sherratt 1983). Nonetheless, preservation biases and the probable use of organic containers must be considered as potentially masking the true frequency and early utilization of dairy, especially in the Pre-Pottery Neolithic B and C periods.

If we accept the limited use of dairy products during the Middle and Late Chalcolithic periods, one must question why dairy products were not used more frequently, especially as the related required technologies were already established. This question is notable because the results of our study contrast with many global studies, which identified dairy residues at the very beginning of their respective Neolithic periods in differential frequencies (e.g., Carrer et al. 2016; Craig et al. 2004; Cramp et al. 2014; Debono Spiteri et al. 2016; Evershed et al. 2008).

Dairy was probably valued by early farming communities because it is a storable food source with a high fat content paired with other nutritional benefits (Davis 1987: 155; Redding 1981: table IX-2). Depending on the processing technique, dairy products can be stored for extended time, and this allows dairy to supplement the diet when other resources are scarce in colder months without sacrificing animals for their meat (e.g., Bogucki 1984). This benefit of dairy products was likely less critical for Levantine communities because other products, such as cereals and legumes, were already being produced in surplus and stored for future utilization (e.g., Garfinkel et al. 2009; Kuijt 2008). So far, dairy residues were only identified in semi-arid regions of the southern Levant in this study. Although this may relate to preservation biases, the geographically restricted utilization may also relate to the presence of a specialized pastoral economy in some areas (Levy 1983) or to the drier environmental conditions that required the exploitation of all available resources.

Dairy processing during the Late Chalcolithic period was typically discussed in association with churns, a hallmark

Table 2 Frequency of the animals identified in the faunal remains of the studied sites based on the NISP

Site	Caprine (%)	Cattle (%)	Pig (%)	Marine fish (%)	Freshwater fish (%)	Wild animals, small vertebrates, and unidentified remains (%)	Reference
Sha'ar Hagolan	51.0	12.0	17.0[1]	0.0	0.0	19.9	Marom 2011: Fig. 3.4
Neve Yam	31.7	34.4	25.0	4.4	0.0	4.2	Horwitz et al. 2006: Table 1
Tel Tsaf	39.2	13.0	30.7	0.0	< 0.1	17.1	Hill 2011: table 5.1
Abu Matar	84.5	10.5	0.0	0.0	0.0	5.0	Josien 1955
Fazael 7	59.0	4.0	1.0	< 0.1		36.0	Bar et al. 2017: table 10
Gilat	63.0	13.8	12.3	1.3		9.6	Grigson 2006: table 6.1
Peqi'in	24.1	25.9	14.8	0.0	0.0	35.2	Bar-Oz 2013:408–409, table 16.1
Yehud Wienhaus	29.8	35.1	6.1	0.0	0.0	28.9	Itach et al. 2019: table 16

[1]Some of the pig remains here may represent wild boars

of the period with stylistic parallels to ethnographic dairy churns (e.g., Burton 2004: 594–595; Commenge 2006: 424–426, 443–445; Kaplan 1954). Organic residue analysis was unable to verify this association. While preservation biases and the sample size of churns analyzed in this study (10 with solvent extraction and saponification and 2 with only solvent extraction) must be considered, churns rarely preserved significant amounts of lipids. Only one churn contained a significant lipid yield in the solvent extract, and only three churns contained a significant lipid yield after releasing the lipids bound to the ceramic matrix. This low lipid yield is paralleled in a prior study by Gregg (2009: 227–228), which solvent extracted 18 churns. Among the vessels with significant lipid yields, lipid biomarker and GC-c-IRMS analysis complicate a functional interpretation, identifying residues potentially characteristic of *Allium* and *Brassicaceae* plants (based on SHFAs) and mixtures of ruminant and non-ruminant adipose (Supplementary material 2).

While preservation biases must be considered, based on the residue analysis results and the interpretations of the fauna demographics, the preferential use of ruminants for meat and less so for milk remains true when environmental variation is considered. Regardless of rainfall and terrain, ruminants were the preferred food source at all the tested sites. A mixed economy with a high percentage of sheep and goats is considered an effective form of risk management in the Near East because their maintenance level is lower than that of cattle and pigs (Marston 2011). Sheep and goats may forage and graze farther from home as well as on relatively steep slopes (Marston 2011; Saleh 2003: 34), allowing for the full exploitation of various topographies. They also require less water (4–9 L a day, depending on age, body size, and lactation; Saleh 2003) than cattle (16–71 L a day, depending on age, body size, and lactation; Saleh 2003) and pigs (ca. 8–12 L a day per 100-kg body weight; Ranjhan 1997). This is particularly important in the Beersheva basin in the northern Negev, which currently receives (Israel Meteorological Service) and during the Chalcolithic period received (Goodfriend 1990) only approximately 200 mm of rainfall annually (200 mm is generally considered the lower isohyet for pig exploitation; Grigson 2007). These environmental regulators worked to similarly structure animal exploitation and diet throughout the entire sequence.

Pigs were reared and consumed less frequently during the Pottery Neolithic and Chalcolithic (Grigson 2007: table 5.1). Although porcine adipose is difficult to identify because it does not contain a distinct lipid biomarker signature (Regert 2011), 10.8% of analyzed vessels could be connected with certainty to non-ruminant adipose using GC-c-IRMS, and an additional 45.6% of vessels show mixing of ruminant and non-ruminant adipose, with pork the most likely source as it is the sole non-ruminant consistently represented in the faunal record. In the fauna assemblages of the studied sites, pig remains form between 1 and 30% of the NISP (Table 2). Similar isotope signatures were identified in pottery from al-Basatîn in Jordan (Gregg et al. 2009), reinforcing the consumption of pork as well as its mixture with other animal fats. While porcine adipose and pig faunal remains were identified less frequently than ruminant adipose and caprine faunal remains, respectively, it is possible that pigs were cooked in earth ovens/roasting pits rather than in pottery, which would not create a lasting lipid signature. This practice was suggested at Tel Tsaf (Ben-Shlomo et al. 2009), and it is commonly observed in ethnography (e.g., Studer and Pillonel 2007: 325; Wiessner 2001: 131–137).

Furthermore, throughout the entire timespan, based on the lipid profiles, there are no indicators for fish consumption. Isoprenoids and ω-(*o*-alkylphenyl)alkanoic acids, key fish biomarkers (Heron et al. 2010), were not identified in the acid fractions, and, while sieving practices and the potential that fish were butchered off-site must be considered (with only extensive dry sieving in a 2 × 2 mm mesh

and floatation conducted at Tel Tsaf and Sha'ar HaGolan (Garfinkel et al. 2020: 43; Marom 2011: 37; Rosenberg et al. 2014)), fish always form less than 5% of the NISP (Table 2). Fishing paraphernalia is also uncommon, identified at very few of the tested sites and other Neolithic and Chalcolithic sites (Rosenberg and Chasan 2020; Rosenberg et al 2016). This is of note because many of the sites are located near perennial water sources, such as the Mediterranean Sea and the Jordan River and springs, and streams, which contained fish. This suggests that although water was a crucial factor in settlement establishment, it was viewed primarily as a resource necessary for drinking and, possibly, farming (e.g., Bourke 2001: 113; Levy 1983; Winter-Livneh et al. 2010).

Plant exploitation

The botanic remains show chronological continuity in plant use. Throughout the entire sequence, there is a reliance on domesticated cereals, such as emmer wheat and barley, and legumes, such as lentils, peas, fava bean, bitter vetch, and horse bean (e.g., Allen 2002: 239–242, table 17.2; Be'eri et al. 2017, 2019; Galili et al. 2017: 115–122; Graham 2014). However, drawing conclusions about the consumed plants based on the lipid profiles is difficult. Cereals have low lipids yields, and the signature is unlikely to preserve or remain unmasked (Colonese et al. 2017), and alkylresorcinols, a key cereal biomarker, are susceptible to microbial degradation (Hammann and Cramp 2018). Furthermore, cereals and legumes do not contain an indicative *n*-alkane signature. In these plants, the *n*-alkanes maximize at C_{29} and C_{31} (Maffei 1996a, b) like other higher plants (Diefendorf et al. 2011). Still, the high amount of very long-chain even-numbered saturated fatty acids, unsaturated fatty acids, odd-numbered long-chain *n*-alkanes, and even-numbered alcohols suggest that the vessels were used for plant processing and vegetable consumption alongside animal products. The overlapping signatures may result from different use episodes, but they could also result from various recipes that combined meat and vegetables.

No olive oil residue was detected despite the notable amounts of olives found at some of the Chalcolithic period sites (Be'eri et al. 2017, 2019; Galili et al. 2017: 116; Graham 2014; Rosenberg et al. 2021). This is not surprising because olive oil is difficult to identify. The key olive oil constituent—oleic acid ($C_{18:1}$ *cis*-9)—easily degrades, leaving behind an unrecognizable signature (Dudd et al. 1998). In the analyzed archeological samples, $C_{18:1}$ is never more abundant than $C_{18:0}$ as characteristic of olive oil (Dudd et al. 1998), and the oxidation products (Passi et al. 1993) are also rarely preserved, prohibiting isomer identification. Olive oil residue was so far only suggested as characteristic of three vessels from the Early Chalcolithic site of Ein Ziporri based on the abundance of $C_{16:0}$ and $C_{18:1}$ (Namdar et al. 2014). This identification is tenuous because the total lipid yield was not quantified, the specific isomer of $C_{18:1}$ was not identified, and the signature may relate to whole olives and not olive oil or to other plants (e.g., Pollard and Heron 2008: table 11.2).

Other fruits were also identified in the botanic assemblages of the Pottery Neolithic and Chalcolithic periods. This includes fig and *Pyrus* seeds at Sha'ar Hagolan (Allen 2002: table 17.2) and grape pips at Tel Bene Beraq (South) (Be'eri et al. 2019). Most likely these all originated from wild species, and these therefore suggest the opportunistic exploitation of fruit trees. Their signature however is not apparent in the lipid profiles, and lipid analysis, in general, is problematic for identifying fruits, which have limited biomarkers (see however Drieu et al. 2021: supplementary table S4).

Allium and *Brassicaceae* plants as well as macrophytes and halophytes appear to have potentially been exploited based on the unsaturated fatty acids, SHFAs, *n*-alkane patterns, and ketones. *Allium* and *Brassicaceae* plant residues were tentatively identified via monounsaturated fatty acids, SHFAs, and ketones in the Middle and Late Chalcolithic periods, although the related monounsaturated fatty acids were not identified to their specific isomers. Macrophytes and/or halophyte residues were identified based on the *n*-alkane profiles of samples dating primarily to the Chalcolithic period. This suggests that there was targeted exploitation of specific wild plants even after agriculture replaced hunting and gathering as the primary subsistence economy. The precise role of the wild plants is unclear, but most likely these had a specific and special function and were not part of daily dietary intake.

Additional resources exploited

While not necessarily part of the consumed diet, two other key resources were noted: beeswax and resins. Beeswax was identified with confidence in the interior and exterior of very few vessels starting in the Early Chalcolithic period. It is unclear if the wax was used as a sealant (e.g.,Charters et al. 1995; Mayyas et al. 2012; Roumpou et al. 2021), or if the residues relate to the use of unfiltered honey or a different function altogether (see Chasan et al. 2021 for a wider discussion).

Resins from coniferous trees were identified in a few vessels dating to the Middle Chalcolithic (Tel Tsaf) and Late Chalcolithic (Ashqelon Agamim and Tel Bene Beraq (South)). In one vessel from Tel Bene Bereq (South), the use of resins specifically from *Cupressaceae* or *Cedrus* trees may be suggested based on its unique terpenoid signature, although sedimentary contamination must also be strongly considered. Coniferous trees including *Cupressaceae*, *Cedrus*, and others are native to Israel (Danin 2004: 20–21),

so if these markers are archeological in origin, these vessels show that resins from local trees were extracted. The specific lipid markers further suggest that the resin was sometimes transformed and used as tar. The resins could have been used as a flavoring agent (e.g., Barnard et al. 2011) or as a sealant (e.g., Colombini et al. 2003, 2005b).

Conclusion

In conclusion, while the sample size is small for some of the periods represented and issues of preservation must be considered, the results of the current study suggest, based on lipid residues and corroborating faunal and botanical data, that from the Pottery Neolithic period and through the Late Chalcolithic period, the subsistence economy and culinary practices of south Levantine communities were mostly uniform despite changing settlement patterns, new craft technologies, and socio-cultural variation. A clear preferential use of ruminants for meat, with pork consumed less frequently, was noted. Cereals and legumes also played an important dietary role, based on the botanic remains, but their lipid signatures did not preserve. The isotope analysis combined with the kill-off patterns observed within the faunal remains suggest the use of dairy products at least from the Middle Chalcolithic period onward (no evidence for dairy was documented so far from earlier contexts) in low frequencies that did not require a specialized animal management strategy (e.g., Payne 1973; Vigne and Helmer 2007). Within dairy exploitation alone, there is a potential geographic variation that may be linked to environmental conditions, with dairy products identified only in the more arid regions.

Although unique recipes and idiosyncratic behaviors linked to food preparation and serving are difficult to identify and must still be considered, the observed continuity from the Pottery Neolithic period (ca. 6,400 cal. BC; Garfinkel 2009: table 14.1) through the end of the Late Chalcolithic period (ca. 3,800 cal. BC; Garfinkel 2009: table 14.1) indicates that this timespan should be considered one long economic continuum separated by distinct cultural entities that shared a core dietary tradition. Suggestions for olive horticulture and an increased utilization of dairy products in the later half of this sequence (e.g., Dighton et al. 2017; Grigson 1987: 225, 1995: 389, 402, 2006: 225–228, 231; Meadows 2005: 240–242) are not mirrored by the results of this study. One potential explanation for this is that if foodways were still centered around self-provisioning rather than maintaining individuals not involved in food production as characteristic of more complex societies (D'Altroy and Earle 2002; Zeder 1988), then there would be no requirement to produce storable and tradeable resources such as olive oil and processed dairy products. Exploitation of olives and dairy on a need basis would not likely produce a visible lipid signature, and the animal management strategy would not be altered as in a specialized economy (Payne 1973; Vigne and Helmer 2007). Economically significant changes in foodways and the overarching economic strategy probably occurred only after the conclusion of the Chalcolithic period.

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Declarations

Conflict of interest The authors declare no competing interests.

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Chapter 6. Discussion

The results included in the three papers contribute to the discussion on changing food ways in the southern Levant during the Pottery Neolithic and Chalcolithic periods. While previous studies were one-dimensional, targeting either the faunal remains, botanical remains or the morphology of food processing tools, this study represents a novel attempt at integrating multiple lines of data, including organic residues, to create a more holistic picture of the diet and the practices revolving around food consumption. The research revealed distinct dietary patterns as well as previously unidentified herding practices and potential ceramic production techniques. The following sections review these topics, the wider implications of the study, the limitations of the research and future research directions.

6.1. Synthesis of results

6.1.1. Dietary patterns

Two economic phenomena are discussed in relation to the Chalcolithic period – the secondary product revolution and olive horticulture. Both are said to increase surplus, allowing for the increase or introduction of trade, specialization and settlement aggregation. Archaeological support for dairying, a key secondary animal product, includes ceramic churns (*e.g.* Garfinkel 1999:111, 123, 254–257, fig. 68; Kaplan 1954) and, more rarely, the animal kill-off pattern (*e.g.* Grigson 1995: table 10.8; Kolska Horwitz 2007; al-Zawahra 2008:437). Archaeological support for olive exploitation includes olive seeds identified at numerous sites (*e.g.* van den Brink *et al.* 2001; Galili *et al.* 1989; Graham 2014; Kislev 1994–1995; Kislev and Hartmann 2012: table 32:1; Liphschitz *et al.* 1996; Zaitschek 1961), although it is unclear if these olives were domesticated or processed into oil. Contrary to these suggestions, organic residue analysis and cross-comparison to the faunal and botanical remains and material culture show that the vessels were used in an unspecialized fashion typically alongside food products unrelated to dairying and horticulture.

The residue analysis results revealed that the ceramic and stone vessels tested were used indiscriminately for both animal and plant products. This is evidenced by the wide saturated fatty acid profile and the unsaturated fatty acids in most vessels, regardless of their size and form. The muddled lipid profile suggests that the vessels were used for stewing or containing various mixtures of meats and plants. Cooking in particular is supported by the identified ketones, the morphology of the vessels, dominated by various deep forms (medium-sized bowls and holemouth jars; Garfinkel 1999), and soot marks on some of the tested vessels (Appendix B). The lipid profiles

obtained could also represent subsequent use episodes as even today pots are used for multiple purposes.

The most dominant and visible residue within these mixed signatures is ruminant adipose fat. This is reflected by the high frequency of odd-numbered and branched-chain fatty acids (although these are also characteristic of milk) and the isotope results, and it is reinforced by the predominance of caprines among the faunal remains and their kill-off pattern (*e.g.* Bar *et al.* 2017; Bar-Oz 2013:408–409, table 16.1; Grigson 2006: table 6.1; Hill 2011:124–127, 164–165, table 5.1; Itach *et al.* 2019; Kolska Horwitz *et al.* 2006; Marom 2011:79, fig. 3.4). This preference traverses environmental boundaries perhaps because an emphasis on sheep and goats is an effective form of risk management due to the lower maintenance required for caprine rearing in comparison to cattle and pigs (Marston 2011). Sheep and goats may be herded across longer distances to forage and graze, allowing for the full exploitation of various topographies, including steep slopes (Marston 2011; Saleh 2003:34). Further, sheep and goats require less water than cattle and pigs (Ranjhan 1997; Saleh 2003), which is an important consideration in some of the more arid regions of Israel.

Compound specific isotope analysis suggests that pigs were reared and consumed less frequently in relation to pottery. This is reinforced by the faunal remains in which pigs usually form a small percentage of the assemblage (*e.g.* Bar *et al.* 2017; Bar-Oz 2013: table 16.1; Grigson 2006: table 6.1; Hill 2011: table 5.1; Itach *et al.* 2019; Kolska Horwitz *et al.* 2006; Marom 2011: fig. 3.4). It is however possible that pork was cooked in pit ovens as observed in ethnographic studies (*e.g.* Studer and Pillonel 2007:325; Wiessner 2001:131–137), which would not leave a testable residue signature in ceramic pots. When pork was cooked in a vessel, mixing with meat from ruminants or the subsequent vessel use for cooking ruminant meat is often suggested by comparison of the isotopic results to mixing curves

Organic residue analysis also shows some evidence for dairying. Milk residues were first identified during the Middle Chalcolithic period, with no evidence in the preceding Pottery Neolithic or Early Chalcolithic periods, although it is possible that milk was used in relation to organic containers that are unpreserved (*e.g.* Benhazera and Coleman 1908:91; von Fürer-Haimendorf 1975:390; Schapera 1930:237). There was no distinct increase in dairy residues between the Middle and Late Chalcolithic periods. The limited role of dairying is reinforced by the animal kill-off patterns at most sites, indicating animal exploitation primarily for meat (*e.g.* Bar *et al.* 2017; Hill 2011:124–127, 164–165; Itach *et al.* 2019; Marom 2011:79). This suggests that a specialized animal

management strategy targeting milk production was not required. A kill-off pattern linked to milk or wool production was observed at only a few sites, and even in these cases, the management strategy reflects a more mixed economy using meat, milk and wool (Grigson 2006:225–232).

Further, dairy residue was not found in connection to churns despite ethnographic parallels. Most churns had a low lipid yield, and two samples with sufficient abundances of palmitic and stearic acid for isotopic analysis yielded residues characteristic of a mixture of ruminant and porcine adipose. The results are also paralleled by a prior study (Gregg 2009: 227–228). This suggests that churns may not have been used in relation to milk (although lipid preservation must be considered). Therefore, archaeologists must reconsider the intensity of dairy processing, which was an assumption that was grounded primarily in the use of churns as a task specific vessel form in the dairy industry.

Among the animal resources, fish do not appear to have played a significant dietary role. While further analysis is required, key fish biomarkers, including APAAs and isoprenoids (Cramp and Evershed 2014), were not identified in the saponified acid fraction. Further, fish are often absent or found in low frequencies within the faunal remains (Bar *et al.* 2017; Bar-Oz 2013: table 16.1; van den Brink *et al.* 2021b; Galili *et al.* 2017:115–116; Grigson 2007: table 6.1; Hill 2011: table 5.1; Itach *et a.* 2019; Josien 1955; Kolska-Horwitz *et al.* 2006; Marom 2011: fig. 3.4).

Information on the vegetal portion of the diet is based primarily on botanical remains. These show that there was a dietary emphasis on cereals and legumes (*e.g.* Allen 2002:239–242, table 17.2; Be'eri *et al.* 2017, 2019; Galili *et al.* 2017:115–122; Graham 2014). However, these plants have low lipids yields, which are unlikely to preserve or remain unmasked in vessel residues (Hammann and Cramp 2018), and correspondingly, lipid organic residue analysis failed to identify conclusive evidence for cereal or legume markers in the tested vessels. Regardless, the lipid signature of nearly every vessel showed a profile characteristic in part of plant products – very long-chain even-numbered saturated fatty acids, unsaturated fatty acids, odd-numbered long-chain *n*-alkanes and even-numbered alcohols (Eglinton and Hamilton 1967; Pollard and Heron 2008:285, table 11.2).

There is no clear evidence for olive oil in the lipid signatures despite the increasing evidence for olives in the archaeological record (*e.g.* Liphschitz *et al.* 1991). Olive oil is difficult to identify using organic residue analysis because the main constituent, oleic acid, can oxidize and leave an unrecognizable signature (Dudd *et al.* 1998). Olive oil however was suggested in a previous residue analysis study on a contemporary site based on the abundance of $C_{16:0}$ and $C_{18:1}$, without the specific

isomer identified (Namdar *et al.* 2014). Despite the lack of olive residue in the current study, the increasing frequency of olives in the botanical remains and the limited organic residue analysis results suggest that olives were used during this time span. With the current evidence, it is unclear how and to what intensity olives were consumed, and they do not appear to have been used in relation to any of the vessels tested in this study.

There is some evidence for the use of wild plants. The most notable of these are mustard seed or garlic. Both plant sources may be suggested by the presence of SHFAs (Dembitsky *et al.* 2007; Eibler *et al.* 2017) in vessels from Middle Chalcolithic Tel Tsaf and Late Chalcolithic Gilat, and mustard seed oil specifically is supported in some vessels by the presence of $C_{20:1}$ and $C_{22:1}$, which are indicative of seed oils (Colombini *et al.* 2005). Mustard seed and garlic are rarely identified in the protohistoric archaeobotanical record (Kislev 1987: table 9.2; Melamed 2002; Zaitschek 1961), so the organic residue analysis results reveal a slightly wider exploitation of these resources than previously assumed.

In addition, there is evidence for the use of wild macrophytes and halophytes based on the *n*-alkane signatures. While many samples show a predominance of long-chain *n*-alkanes, a few samples show a high amount of C_{23} and C_{25} *n*-alkanes. These are characteristic of macrophytes and halophytes (Eley *et al.* 2014; Ficken *et al.* 2000). Differentiating these two plant forms is difficult, and most likely a combination of both types is represented. These signatures were identified at sites located near diverse aquatic resources (the Mediterranean Sea, the Jordan River, streams and springs), suggesting that people gathered plants from various aquatic environments. This shows that wild resources were still exploited a few millennia after the gathering of wild plants was replaced by growing domesticated crops. The precise role of these wild plants is unclear, but most likely these fulfilled a non-dietary role because as a dietary resource, they are eclipsed by the stability and nutrients of cereals and legumes. One suggestion is that the wild plants were exploited as a medicinal resource because even today, many wild halophytes and macrophytes are used globally as medication (*e.g.* Ksouri *et al.* 2012; Qasem 2015; Sanilkumar and Thomas 2007), but they could have also been used as flavoring agents or a yet unidentified function.

6.1.2. Herding strategies

Study of the animal lipid isotopic signature also elucidates on the environment and herding practices. Lipid contribution from non-ruminant adipose displays a narrow isotopic signature, while at sites in the Jordan Valley and the northern Negev, the lipid contribution from ruminants

displays a wider isotope signature, including less depleted values. This suggests that pigs were reared in one location, likely near the site, limiting variation in their isotope signature; however, ruminants were herded farther from the site and over a wide terrain, including areas with water stressed plants or saline soil that would create comparatively enriched isotopic values (Tieszen 1991: table 1). This specialized form of pastoralism may function to protect cultivated land against animal grazing (Köhler-Rollefson 1988), so it is ideal for a mixed agro-pastoral economy.

6.1.3. Ceramic production techniques

Organic residue analysis revealed the presence of post-firing vessel treatments. These used resins, beeswax and potentially other fats. Surface treatments are important as they can seal a vessel, making it more suitable for cooking and food storage (*e.g.* Drieu *et al.* 2019; Reber and Hart 2008).

Resins were identified based on the presence terpenoids, mostly abietanes. These are associated with resin from coniferous trees (Mills and White 1977). In a few samples, the presence of methyl dehydroabietate suggests specifically that sometimes the resins were used and applied as tars as methyl dehydroabietate forms during heating (Pollard and Heron 2008:243).

Beeswax was identified at multiple sites based on a combination of molecules – long-chain palmitate wax esters, even-numbered saturated fatty acids, odd-numbered alkanes and even-numbered alcohols (*e.g.* Heron *et al.* 1994; Jackson and Blomquist 1976:216–220; Regert *et al.* 2001). While beeswax may have had several functions, its application internally and externally suggests use as a sealant infrequently. This technique has a wide and long-lived history (*e.g.* Crane 1999:524), and this study marks its earliest identification in the southern Levant, albeit in low frequencies.

Post-firing vessel treatments were also suggested for a few vessels sampled from Tel Tsaf based on the pairing of ketones with alterations to the ceramic matrix. Ketones are heating markers, formed through the pyrolysis of fatty acids at temperatures over 300°C (Raven *et al.* 1997). They can form in a post-firing treatment when immediately after firing, the vessel is coated with a fatty substance to alter the vessel surface. In this process, the margins of the pottery darken as the fat seeps in (Drieu *et al.* 2019), and this alteration was observed in a few vessels. Future microscopic analysis of pottery from additional sites may identify a wider use of this practice.

6.2. Research limitations and future directions

A few trajectories can be proposed for future studies. First, future studies should increase the sample size further to combat the poor lipid preservation and target more diagnostic vessel fragments and task-specific vessels. Second, the research focused on sites located in modern day Israel; to understand the wider framework, a collaborative effort integrating these and additional sites from Israel with sites from the rest of the Levant (Jordan, Lebanon and Syria), with more emphasis on the Pottery Neolithic and Early Chalcolithic, is required. Third, the study revealed several methodological issues, which could be improved upon. Future research would benefit from a local compound specific isotope database. While this problem can be partially rectified by plotting $\delta^{13}C_{16:0}$ against $\Delta^{13}C$, a local database is preferential because it acknowledges the environmental challenges and their effects on animal diet. To create an accurate framework, a wide range of modern reference samples would be required, so this mandates a more targeted study outside the archaeological realm. Additionally, a pilot study suggests that direct acidified methanol extraction (Correa-Ascencio and Evershed 2014) cannot be applied to Chalcolithic pottery because of the calcite inclusions or clay. Instead, a prolonged technique using solvent extraction followed by saponification was employed to maximize the lipid yield; however, the published lipid yield was based solely on solvent extraction and remained low because there is no agreed upon convention to merge the TLE from solvent extraction and saponification. Future studies on pottery formed with highly alkaline clay or calcite inclusions would benefit from a modified direct acidified methanol extraction that uses an acid other than sulfuric acid or a preparatory step, which would neutralize the ceramic matrix. This would reduce the extraction time and increase the recovered TLE. To achieve this, experimental studies are required; these will be aimed at creating a streamlined approach to extract the maximum amount of lipids, while factoring into account the chemical properties of the ceramic matrix. With this enhanced methodology, a renewed study would likely identify higher lipid yields and preservation of more indicative molecules.

6.3. Research impact

Despite these limitations, the results bear significant implications, enhancing our understanding of the food ways of the Neolithic and Chalcolithic populations. Food ways are governed by a variety of external factors as well as cultural preferences. Potential regulators during this time span include changing settlement patterns (Holl 2019; Levy 1983; Winter-Livneh *et al.* 2010), climatic variation and increasing rainfall (*e.g.* Bar-Matthews *et al.* 1999; Frumkin *et al.* 1991) and the preferences of the countless cultural entities, such as the Yarmukian, Wadi Rabah, Ghassulian and other smaller

more local phenomenon (Garfinkel 1993; Gilead 2007; Gopher and Gophna 1993; Henry 1995; Rowan and Golden 2009).

Regardless of chronological or cultural variation, settlement scale, and typically environmental conditions, throughout this time span, there is negligible variation in diet or the use of vessels. Meat from domestic farm animals and the core cultivator crops were the main food sources. Wild resources, including plants and beeswax, supplemented the diet in low frequencies, indicating a targeted use of non-domesticated resources after the Neolithization process. Limited variation may be attributed to environmental factors, with dairying so far witnessed only in the more arid northern Negev and Jordan Valley.

The general dietary continuation suggests that the Pottery Neolithic and Chalcolithic represent one economic continuum separated by distinct cultural entities. The lack or limited use of intensification techniques to form storable and tradable products, such as processed dairy products and olive oil, may suggest that the economies of each site were still based on self-provisioning. Significant changes in food ways and the overarching economic strategy were only required sometime after the Chalcolithic period when economic interconnectivity increased.

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Appendix A. Overview of the sites analyzed as part of this study.

251 vessels were sampled from 16 sites (Table S1) located in modern day Israel

Site	Period	Region	N vessels sampled
Sha'ar Hagolan (SHH)	Pottery Neolithic	Jordan Valley	16
Neve Yam (NY)	Early Chalcolithic	Mediterranean coastal plain	17
Tel Hreiz (TH)	Early Chalcolithic	Mediterranean coastal plain	3
Tel Tsaf (TSF)	Middle Chalcolithic	Jordan Valley	103
Abu Matar (AM)	Late Chalcolithic	Northern Negev	3
Ashqelon Agamim (AG)	Late Chalcolithic	Mediterranean coastal plain	28
Azor	Late Chalcolithic	Mediterranean coastal plain	1
Tel Bene Beraq (South) (BB)	Late Chalcolithic	Mediterranean coastal plain	10
Bir es-Safadi	Late Chalcolithic	Northern Negev	11
Ein Gedi	Late Chalcolithic	Judean Desert	1
Fazael 7 (FZ)	Late Chalcolithic	Jordan Valley	8
Gilat (GLT)	Late Chalcolithic	Northern Negev	13
Nissim Aloni Street	Late Chalcolithic	Mediterranean coastal plain	9
Peqi'in (PQ)	Late Chalcolithic	Upper Galilee	3
Tsomet Shoket (TS)	Late Chalcolithic	Northern Negev	18
Yehud (YEH and YV)	Late Chalcolithic	Mediterranean coastal plain	7

Sha'ar Hagolan

Sha'ar Hagolan is a Yarmukian Pottery Neolithic site located in the central Joran Valley. The site is dated to 6,400–5,800 cal. BC (Garfinkel and Ben-Shlomo 2009:18). The site has large courtyard structures and streets that suggest early community planning. There is evidence of long-distance trade, including alabaster and obsidian objects (Garfinkel and Ben-Shlomo 2009:3). The faunal remains reflect a typical Neolithic diet with 51% caprine, 12% cattle and 17% pig remains in the number of identified specimens (NISP). Minor hunting was conducted, with 14% wild ungulate remains in the NISP (Marom 2011: fig. 3.4). The kill-off pattern of cattle may suggest minor use for labor (Marom 2011:92–93). Botanical remains are dominated by cereals. This includes primarily emmer wheat and barley, although einkorn wheat was also noted. Pulses include mainly lentils with trace amounts of fava beans and bitter vetch. Fig seeds and two *Pyrus* seeds were identified, suggesting the exploitation of wild fruit (Allen 2002:239–242, table 17.2).

Neve Yam

Neve Yam (South) is a submerged Wadi Rabah site on the Mediterranean coast, 0–5 m below present-day sea level. The site dates to around 7,600–6,750 cal. BP (Galili *et al.* 2017:107, table 7.1). Excavations revealed rectangular structures built from sandstone as well as unpaved pits, paved surfaces, stone slabs and postholes (Galili *et al.* 2017:117–119). Faunal remains are characterized by caprine, cattle and pigs in nearly equal frequencies. Analysis indicates a herding economy targeted at meat production. There are also some gazelle remains, suggesting minimal hunting occurred (Kolska Horwitz *et al.* 2006). Botanical remains include primarily emmer wheat, barley and lentils. There are also minor frequencies of peas, vetch, fava bean and flax (Galili *et al.* 2017:120–122).

Tel Hreiz

Tel Hreiz is a submerged Wadi Rabah site on the Mediterranean coast, 0–5 m below present-day sea level. The site dates to around 8,300–7,000 cal. BP (Galili *et al.* 2017:107, table 7.1). Surveys revealed stone structures and wooden poles, which may have supported additional structures. The faunal assemblage is small, with a predominance of cattle and low amounts of caprines and pigs. Some wild ungulates were also identified. A plethora of botanical remains was identified, including hundreds of olive pits (Galili *et al.* 2017:115–116).

Tel Tsaf

Tel Tsaf is in the central Jordan Valley near the Jordan River. The site consists of three shallow hills. The site dates to around 5,200–4,700 cal. BC (Garfinkel *et al.* 2020:29, 59–66). The Middle Chalcolithic remains are characterized by well-built mudbrick rectilinear structures, courtyards and silos (Garfinkel *et al.* 2020:117–211, 219–221; Rosenberg *et al.* 2020).

The faunal remains are dominated by sheep and goat, these accounting for nearly 40% of the NISP. Surprisingly, pigs comprise 31% of the NISP, while cattle are only 13%. There are a few wild ungulate remains, suggesting minimal hunting (Hill 2011:108). Evidence for the exploitation of animals for secondary products is minimal. Some sheep survived until four years old, with no gender bias, suggesting that sheep were kept to an older age to exploit their wool (Hill 2011:127). In addition, cattle were exploited in part for labor as evidenced by bone pathologies (Hill 2011:148–156). These however were not specialized techniques, and most animals were exploited for meat. In particular, it was suggested that the meat was used in feasting activities. This is supported by the emphasis on pigs, which provide an abundance of meat at low cost. The concentration of these remains also suggests that the pigs were butchered in a single event (Ben-Shlomo *et al.* 2009).

Botanical remains are abundant, including wood, seeds and pollen. Wood remains identified include white acacia (*Acacia albida*), olive (*Olea europaea*), Atlantic pistachio (*Pistacia atlantica*), lentisk (*Pistacia lentiscus*), Euphrates poplar (*Populus euphratica*), Mt. Tabor oak (*Quercus ithaburensis*), tamarisk (*Tamarix*) and jujube (*Ziziphus lotus*). Most of these reflect the local environment (Liphschitz 1988). In addition, over 3,000 macrobotanical remains were recovered. The most common remains were barley and wheat, most of this emmer. Lentil and pea seeds were also common (Graham 2014). These finds are reemphasized by the pollen record, which shows a high percentage of cereals (Horwitz 1988–1989). Many olive stones were also uncovered (Graham 2014; Rosenberg *et al.* 2021).

In tandem, there is evidence for intensive storage, with over a dozen silos identified. These are well-built cylindrical mudbrick structures. Each could hold 15–36 tons of grain, which is well above what one a nuclear family requires. To cultivate this much cereal, intensive and controlled labor was required (Garfinkel *et al.* 2009). There is also evidence of foreign connections within the material remains to the northern Levant and Egypt (Garfinkel *et al.* 2007). Combined, this suggests that there was increasing social complexity and perhaps the presence of a ranked society (Ben-Shlomo *et al.* 2009; Garfinkel *et al.* 2009).

Abu Matar

Abu Matar is a Chalcolithic site located on the banks of Nahal Beersheva (Gilead *et al.* 1991; Perrot 1955a). The site dates to *ca.* 4,300–4,000 cal. BC (Shugar 2000: table 3.02). The settlement is composed of complex subterranean dwellings and classical rectangular courtyard surface structures formed from mudbrick walls and stone foundations (Gilead *et al.* 1991; Perrot 1955b).

The faunal remains show a high amount of sheep, these forming 72.0% of the NISP. Goats form 12.4% and cattle form 10.5% of the NISP. Notably, no pigs were uncovered. In addition, the remains of equids, dogs and birds were recovered (Josien 1955).

Ashqelon Agamim

Ashqelon Agamim (A-8271) is located near the southern Mediterranean seashore. The site is characterized by monumental and domestic architecture. The monumental architecture consists of large rectilinear structures with thick fieldstone walls. These buildings are supplemented by smaller domestic structures. The faunal and botanical remains are presently unpublished (Abadi-Reiss and Varga 2019; Varga *et al.* 2021).

Azor

Azor is a Late Chalcolithic burial cave located near modern day Tel Aviv (Perrot and Ladiray 1980:41). Over 100 complete and fragmented ossuaries were found in addition to a rich array of other finds (Perrot and Ladiray 1980:47–53).

Tel Bene Beraq (South)

Tel Bene Beraq (South) (A-8044) is a settlement site located on the northern bank of the Ayalon River. Typical rectilinear domestic architecture and pits were identified. Botanical remains include cereals, legumes, olives and grape pips. It is unclear if the grapes are domesticated or wild (Be'eri *et al.* 2019).

Bir es-Safadi

Bir es-Safadi is located on the banks of Nahal Beer Sheva, a short distance from Abu Matar (Perrot 1959, 1984). The site dates to *ca.* 4,300–3,900 cal. BC (Joffe and Dessel 1995). The architecture is characterized by underground features and aboveground rectilinear structures (Perrot 1959, 1984).

The faunal assemblage is composed primarily of sheep – 84.8% of the NISP, while goats are less common – 9.2%. Cattle are also uncommon, forming only 2.8%. Few wild faunal remains were uncovered, and no pigs were identified. (Josien 1955). Within the faunal remains, there is evidence for secondary product exploitation. Over half of the male sheep were killed-off in their first year, after reaching maturity, while female sheep were generally killed-off after 2.5 years, suggesting use for milk and/or wool. Goats follow a similar but less extreme pattern (Grigson 1987:225, fig. 7.6).

Ein Gedi

Ein Gedi is a Late Chalcolithic temple located near the Dead Sea. Excavations revealed four structures: the main gatehouse, the secondary gatehouse, a lateral chamber and the sanctuary. These were connected by a stone wall. The wall also defined a courtyard, which contained a circular structure. The faunal remains were never fully published. However, several ibex horns were identified in addition to a few caprine bones (Ussishkin 1980).

Fazael 7

Fazael is a concentration of sites located in the central Jordan Valley along Wadi Fazael that may represent one larger site (Bar *et al.* 2013). At Fazael 7 a large architectural complex was uncovered. This comprises a stone structure with sub-rooms and three courtyards. Among the faunal remains,

sheep and goats were the most common species (*ca.* 80% of the NISP). Their kill-off pattern corresponds with meat exploitation. Cattle, pig and equid bones were found in low frequencies. Some wild species were also noted, indicating minimal hunting (Bar *et al.* 2017).

Gilat

Gilat is located on the eastern bank of Nahal Patish in the northern Negev (Levy *et al.* 2006:95). The site dates to *ca.* 4,700–3,800 cal. BC (Levy and Burton 2006: table appendix 2.2). While Gilat is a settlement with domestic remains, the site is also considered a sanctuary, renowned for its large architectural complex and 'ritualistic' artifacts (Alon and Levy 1989; Levy *et al.* 2006).

The faunal assemblage is predominated by caprines (63.0% of the NISP), with low frequencies of pigs (12.3% of the NISP) and cattle (13.8% of the NISP) in addition to a few equid remains (Grigson 2006: table 6.1). There was also minimal hunting of wild animals, with the fauna including gazelle, ox, hartebeest, caprine, boar, birds, fish and others. Aging and sexing profiles of the caprine suggest an unspecialized utilization of sheep and goats for meat, milk and wool. The latter are reemphasized by the presence of churns and spindle whorls at the site (Grigson 2006:228, 232). The aging and sexing profile of cattle indicate utilization for meat alone (Grigson 2006:228–230).

Nissim Aloni Street

Nissim Aloni Street (A-8066) is located in the heart of modern-day Tel Aviv. The site is situated south of the Yarkon River and on the west margin of the Ayalon River's flood basin. The site is dated to 4,330–4050 cal. BC. Excavations uncovered a plethora of pits and shafts. Geomorphological analysis suggests that these are wells formed to access non-saline groundwater (van den Brink *et al.* 2021b), and after they fell out of use, they were reused for waste disposal (van den Brink *et al.* 2020, 2021b).

Within the pits and shafts, a high quantity of faunal remains was uncovered, reinforcing their later use as waste pits. Pigs form 34.4% of the NISP, while cattle form 21.1% and sheep and goats form 17.2%. While age at death could only be calculated for a few specimens, some cattle, sheep and goats were kept to an older age, suggesting some exploitation of secondary animal products (van den Brink *et al.* 2021b). This pattern is paralleled at the site's extension and earlier excavation – Namir Road, although cattle was the most frequent species identified. In addition, at Namir Road, fish bones were uncovered. No botanical remains were recovered from Nissim Aloni Street, but charred wood remains of lentisk (*Pistacia lentiscus*) and carob (*Ceratonia silique*) were uncovered at Namir Road, reflecting the use of local trees (van den Brink *et al.* 2016).

Peqi'in

Peqi'in is a complex multi-chambered active karstic cave site in the Galilee used for mortuary practices (Shalem *et al.* 2013a:53–68). The burial phase is dated to *ca.* 4,500–4,200 cal. BC (Segal *et al.* 1998: table 2). A wide array of burial goods was also identified (*e.g.* Bar-Yosef Mayer and Porat 2013; Raban-Gerstel and Bar-Oz 2013; Shalem *et al.* 2013b).

Faunal remains include primarily domesticated animals with 24.1% caprine, 25.9% cattle and 14.8% pigs. In addition, there is high percentage of wild ungulates (18.5%) including primarily gazelle (Bar-Oz 2013:408–409, table 16.1).

Tsomet Shoket

Tsomet Shoket (A-7700) is located near modern day Beer Sheva. The site has four phases. Phase IV is composed of numerous cylindrical and bell-shaped pits; phase III is the fill above and in the pits; phase II represents the major subterranean occupation, with several subterranean complexes and pits dug into phase III; phase I consists of above ground rectangular broad room structures (Be'eri *et al.* 2017). The site also revealed ample botanical remains including wheat, barley, lentils and olives (Be'eri *et al.* 2017).

Yehud

In the modern town of Yehud, there is a conglomerate of Chalcolithic sites uncovered through several rescue excavations (Govrin 2015a:13; Itach *et al.* 2019: table 1). At a major excavation (YEH), deep shafts filled with trash and ash were identified (Govrin 2015b:1–10). A more recent excavation on Wienhaus St. (YV) revealed habitation levels, shafts and pits. The shafts may have functioned as refuse pits (Itach *et al.* 2019), although use as wells to reach non-saline groundwater must also be considered (van den Brink *et al.* 2020). The site dates to 4,200–4,000 cal. BC (Itach *et al.* 2019).

Only the faunal remains from Yehud Wienhaus are published. In the shafts, faunal remains reflect a near equal utilization of cattle (29.3%) and caprine (27.1%), with minimal utilization of pigs (4.3%), as well as the use of hunting with a high percentage of gazelle (14.1%). However, in the faunal assemblage from the occupational debris, gazelle is absent, and cattle is most common (50.0%) followed by caprines (34.6%) and pigs (11.5%) (Itach *et al.* 2019).

Appendix B. List and description of the vessels sampled

Vessel	Vessel type	Sherd type	Fabric	Decoration	Evidence for heating
AG-1	Unidentified	Wall	Coarse	Undecorated	-
AG-11	Unidentified	Wall	Coarse	Undecorated	-
AG-13	Unidentified	Wall	Coarse	Undecorated	-
AG-15	Unidentified	Wall	Coarse	Undecorated	-
AG-16	Unidentified	Wall	Coarse	Undecorated	-
AG-18	Unidentified	Wall	Coarse	Undecorated	-
AG-19	Unidentified	Wall	Coarse	Undecorated	-
AG-2	Unidentified	Wall	Coarse	Undecorated	-
AG-21	Unidentified	Wall	Coarse	Undecorated	-
AG-22	Unidentified	Wall	Coarse	Undecorated	-
AG-23	Unidentified	Wall	Coarse	Undecorated	-
AG-24	Unidentified	Wall	Coarse	Undecorated	-
AG-25	Unidentified	Wall	Coarse	Undecorated	-
AG-26	Unidentified	Wall	Coarse	Undecorated	-
AG-28	Unidentified	Wall	Coarse	Undecorated	-
AG-30	Cornet	-	Coarse	Undecorated	-
AG-32	Cornet	-	Coarse	Undecorated	-
AG-34	Cornet	-	Coarse	Undecorated	-
AG-36	Cornet	-	Coarse	Undecorated	-
AG-4	Bowl	Rim	Coarse	Undecorated	-
AG-40	Unidentified	Wall	Coarse	Undecorated	-
AG-41	Unidentified	Wall	Coarse	Undecorated	-
AG-43	Jar	Wall	Coarse	Undecorated	-
AG-44	Bowl	Wall	Coarse	Undecorated	-
AG-45	Unidentified	Wall	Coarse	Undecorated	-
AG-7	Unidentified	Wall	Coarse	Undecorated	-
AG-8	Unidentified	Wall	Coarse	Undecorated	Burnt
AG-9	Unidentified	Wall	Coarse	Undecorated	-
AM-1	Holemouth jar	Rim	Coarse	Undecorated	Burnt
AM-2	Churn	Wall	Coarse	Undecorated	-
AM-5	Churn	Neck	Coarse	Red paint	-
AZ-3	Strainer	Neck	Coarse	Undecorated	-
BB-1	Jar	Base	Coarse	Undecorated	-
BB-19	Jar	Base	Coarse	Undecorated	-
BB-21	Jar	Wall	Coarse	Undecorated	-
BB-25	Jar	Base	Coarse	Undecorated	-
BB-28	Jar	Wall	Coarse	Undecorated	-
BB-3	Unidentified	Base	Coarse	Undecorated	-
BB-4	Holemouth jar	Wall	Coarse	Undecorated	-
BB-5	Unidentified	Wall	Coarse	Undecorated	-

Vessel	Vessel type	Sherd type	Fabric	Decoration	Evidence for heating
BB-6	Jar	Base	Coarse	Undecorated	-
BB-7	Holemouth jar	Wall	Coarse	Undecorated	-
BES-4	Holemouth jar	Rim	Coarse	Undecorated	Burnt
BES-5	Jar	Rim	Coarse	Red paint	Burnt
BES-6	Spouted vessel	Rim	Coarse	Pie crust rim, lipstick	Burnt
BES-7	Holemouth jar	Rim	Coarse	Undecorated	-
BES-8	Jar	Rim	Coarse	Undecorated	Burnt
BES-9	Churn	Body	Coarse	Undecorated	-
BES-10	Churn	Body	Coarse	Undecorated	-
BES-12	Churn	Body	Coarse	Undecorated	-
BES-13	Churn	Neck	Coarse	Undecorated	-
BES-15	Churn	Neck	Coarse	Red paint	-
BES-16	Churn	Neck	Coarse	Undecorated	-
EGD-1	Unidentified	Wall	Coarse	Undecorated	-
FZ-10	Unidentified	Wall	Coarse	Undecorated	-
FZ-11	Unidentified	Base	Coarse	Undecorated	-
FZ-3	Jar	Base	Coarse	Undecorated	-
FZ-4	Unidentified	Base	Coarse	Undecorated	-
FZ-5	Jar	Wall	Coarse	Red paint	-
FZ-6	Holemouth jar	Rim	Coarse	Undecorated	-
FZ-7	Jar	Wall+handle	Coarse	Undecorated	-
FZ-8	Unidentified	Wall	Coarse	Undecorated	-
GLT-1	Holemouth jar	Rim	Coarse	Undecorated	Burnt
GLT-10	Unidentified	Base	Coarse	Undecorated	-
GLT-11	Jar	Wall	Coarse	Undecorated	-
GLT-12	Churn	-	Coarse	Undecorated	-
GLT-13	Jar	Rim	Coarse	Undecorated	-
GLT-2	Holemouth jar	Rim	Coarse	Undecorated	Burnt
GLT-3	Churn	-	Coarse	Undecorated	-
GLT-4	Churn	Base	Coarse	Red paint	-
GLT-5	Churn	-	Coarse	Undecorated	-
GLT-6	Cornet	-	Coarse	Undecorated	-
GLT-7	Cornet	-	Fine	Undecorated	-
GLT-8	Jar	Wall	Coarse	Red paint, impressed	-
GLT-9	Jar	Rim	Coarse	Pie crust rim	Burnt
NA-5	Jar	Rim	Coarse	Red paint	-
NA-7	Jar	Wall	Coarse	Red paint, geometric patterns	-
NA-8	Bowl	Base	Coarse	Undecorated	-
NA-11	Churn	Body	Coarse	Undecorated	-
NA-12	Churn	Body	Coarse	Undecorated	-

Vessel	Vessel type	Sherd type	Fabric	Decoration	Evidence for heating
NA-13	V-shaped bowl	Base	Coarse	Undecorated	-
NA-14	Unidentified	Base	Coarse	Undecorated	-
NA-19	V-shaped bowl	Base	Coarse	Undecorated	Burnt
NA-20	Unidentified	Wall	Coarse	Undecorated	-
NY-1	Jar	Rim	Coarse	Red paint	-
NY-10	Jar	Rim	Coarse	Rope band	-
NY-11	Unidentified	Wall+handle	Coarse	Undecorated	-
NY-12	Jar	Neck	Coarse	Red paint	-
NY-13	Unidentified	Rim	Coarse	Undecorated	-
NY-15	Unidentified	Wall	Coarse	Red paint	-
NY-16	Unidentified	Base	Coarse	Undecorated	-
NY-17	Unidentified	Base	Coarse	Undecorated	-
NY-18	Unidentified	Base	Coarse	Undecorated	-
NY-2	Unidentified	Base	Coarse	Undecorated	-
NY-3	Unidentified	Base	Coarse	Undecorated	-
NY-4	Unidentified	Base	Coarse	Undecorated	-
NY-5	Jar	Rim	Coarse	Red paint, rope band	-
NY-6	Bow-rim jar	Rim	Coarse	Undecorated	-
NY-7	Bow-rim jar	Rim	Coarse	Red paint, herringbone	-
NY-8	Spouted vessel	Spout	Coarse	Undecorated	-
NY-9	Jar	Rim	Coarse	Rope band	-
PEQ-1	V-shaped bowl	-	Coarse	Undecorated	-
PEQ-2	V-shaped bowl	Rim	Coarse	Undecorated	-
PEQ-3	V-shaped bowl	Rim	Coarse	Undecorated	-
SHH-1	Unidentified	Base	Coarse	Undecorated	-
SHH-10	Unidentified	Wall	Coarse	Undecorated	-
SHH-11	Jar	Rim	Coarse	Undecorated	-
SHH-13	Jar	Rim	Coarse	Undecorated	-
SHH-17	Unidentified	Wall	Coarse	Undecorated	-
SHH-18	Unidentified	Wall	Coarse	Undecorated	-
SHH-19	Unidentified	Wall	Coarse	Undecorated	-
SHH-2	Unidentified	Wall	Coarse	Undecorated	-
SHH-20	Bowl	Base	Coarse	Undecorated	-
SHH-21	Unidentified	Wall	Coarse	Undecorated	-
SHH-3	Unidentified	Base	Coarse	Undecorated	-
SHH-4	Unidentified	Base	Coarse	Undecorated	-
SHH-5	Unidentified	Rim	Coarse	Undecorated	-
SHH-6	Unidentified	Base	Coarse	Undecorated	-
SHH-7	Bowl	Rim	Coarse	Undecorated	-
SHH-8	Bowl	Rim	Coarse	Undecorated	-

Vessel	Vessel type	Sherd type	Fabric	Decoration	Evidence for heating
TH-1	Unidentified	Base	Coarse	Undecorated	-
TH-2	Jar	Rim	Coarse	Undecorated	-
TH-4	Unidentified	Wall+handle	Coarse	Undecorated	-
TS-1	Jar	Rim	Coarse	Pie crust rim	-
TS-10	Bowl	Rim	Coarse	Undecorated	Burnt
TS-11	Bowl	Rim	Coarse	Red paint	-
TS-12	Churn	-	Coarse	Undecorated	-
TS-13	Holemouth jar	Rim	Coarse	Undecorated	-
TS-16	V-shaped bowl	Base	Coarse	Undecorated	-
TS-18	Bowl	Rim	Coarse	Red paint	-
TS-2	Jar	Rim	Coarse	Undecorated	-
TS-20	Pithos	Rim	Coarse	Undecorated	Burnt
TS-23	Bowl	Rim	Coarse	Red paint	-
TS-26	Holemouth jar	Rim	Coarse	Undecorated	-
TS-3	Churn	-	Coarse	Undecorated	-
TS-4	Jar	Rim	Coarse	Undecorated	-
TS-5	Jar	Rim	Coarse	Undecorated	-
TS-6	Holemouth jar	Rim	Coarse	Undecorated	-
TS-7	Jar	Rim	Coarse	Undecorated	-
TS-8	Bowl	Rim	Coarse	Undecorated	-
TS-9	Bowl	Rim	Coarse	Red paint	-
TSF17-1	Holemouth jar	Wall	Coarse	Rope band	-
TSF17-10	Unidentified	Wall	Coarse	Red paint	-
TSF17-12	Bowl	Rim	Coarse	Undecorated	-
TSF17-13	Holemouth jar	Rim	Coarse	Burnished, rope band	-
TSF17-15	Bowl	Rim	Coarse	Red paint	-
TSF17-16	Holemouth jar	Wall	Coarse	Undecorated	-
TSF17-18	Unidentified	Wall	Coarse	Undecorated	-
TSF17-19	Unidentified	Wall	Coarse	Undecorated	-
TSF17-2	Unidentified	Base	Coarse	Undecorated	-
TSF17-21	Unidentified	Wall	Coarse	Red paint	-
TSF17-22	Unidentified	Wall	Coarse	Undecorated	-
TSF17-23	Unidentified	Wall	Coarse	Undecorated	-
TSF17-25	Unidentified	Wall	Coarse	Red paint	-
TSF17-26	Unidentified	Wall	Coarse	Red paint, burnished	-
TSF17-27	Holemouth jar	Rim	Coarse	Rope band, burnished	-
TSF17-28	Holemouth jar	Rim	Coarse	Undecorated	Burnt
TSF17-29	Unidentified	Wall	Coarse	Red paint	-
TSF17-3	V-shaped bowl	Base	Coarse	Red paint	-
TSF17-30	Holemouth jar	Rim	Coarse	Undecorated	-
TSF17-31	Unidentified	Wall	Coarse	Brown paint	Burnt

Vessel	Vessel type	Sherd type	Fabric	Decoration	Evidence for heating
TSF17-32	Unidentified	Wall	Coarse	Undecorated	Burnt
TSF17-4	Holemouth jar	Wall	Coarse	Rope band	-
TSF17-5	Holemouth jar	Rim	Coarse	Red paint	-
TSF17-6	Unidentified	Wall	Coarse	Undecorated	Burnt
TSF17-7	Jar	Base	Coarse	Undecorated	Burnt
TSF17-8	Unidentified	Wall	Coarse	Red paint	-
TSF17-9	Limestone pedestal bowl	Rim	-	Undecorated	-
TSF18-1	Bowl	Base	Coarse	Undecorated	-
TSF18-10	Unidentified	Wall	Coarse	Undecorated	-
TSF18-11	Unidentified	Wall	Coarse	Red paint	-
TSF18-13	Unidentified	Base	Coarse	Red paint	-
TSF18-14	Unidentified	Wall	Coarse	Red paint	Burnt
TSF18-15	Jar	Wall+handle	Coarse	Red paint	-
TSF18-16	Unidentified	Wall	Coarse	Red paint	-
TSF18-17	Unidentified	Wall	Coarse	Red paint	Burnt
TSF18-18	Unidentified	Wall	Coarse	Undecorated	-
TSF18-19	Unidentified	Base	Coarse	Red paint	-
TSF18-2	Holemouth jar	Rim	Coarse	Red paint	-
TSF18-20	Unidentified	Base	Coarse	Red paint	-
TSF18-21	Jar	Wall	Coarse	Red paint	-
TSF18-22	Unidentified	Wall	Coarse	Undecorated	-
TSF18-23	Unidentified	Wall	Coarse	Brown paint	-
TSF18-24	Unidentified	Base	Coarse	Undecorated	-
TSF18-25	Unidentified	Wall	Coarse	Red paint	-
TSF18-26	Small bowl	Wall	Coarse	Red paint, burnished	-
TSF18-27	Unidentified	Wall	Coarse	Undecorated	-
TSF18-28	Holemouth jar	Rim	Coarse	Red paint	-
TSF18-29	Unidentified	Wall+handle	Coarse	Undecorated	-
TSF18-3	bowl	Base	Coarse	Undecorated	-
TSF18-30	Unidentified	Wall	Coarse	Undecorated	-
TSF18-31	Bowl	Rim	Coarse	Red paint	-
TSF18-34	Unidentified	Wall	Coarse	Red paint	-
TSF18-35	Unidentified	Wall	Coarse	Red paint	-
TSF18-36	Unidentified	Wall	Coarse	Undecorated	-
TSF18-37	Unidentified	Wall	Coarse	Undecorated	-
TSF18-39	Unidentified	Wall	Coarse	Red paint	-
TSF18-4	V-shaped bowl	Rim	Coarse	Red paint	-
TSF18-40	Jar	Wall	Coarse	Brown paint	-
TSF18-42	Jar	Rim	Coarse	Red paint	-
TSF18-44	Unidentified	Wall	Coarse	Red paint, rope band	-
TSF18-45	Holemouth jar	Wall	Coarse	Red paint, rope band	-

Vessel	Vessel type	Sherd type	Fabric	Decoration	Evidence for heating
TSF18-46	Unidentified	Wall	Coarse	Red paint	-
TSF18-47	Unidentified	Wall	Coarse	Red paint	-
TSF18-48	Unidentified	Wall	Coarse	Undecorated	-
TSF18-49	Unidentified	Wall	Coarse	Red paint	-
TSF18-5	Unidentified	Base	Coarse	Red paint	-
TSF18-50	Unidentified	Wall	Coarse	Red paint	-
TSF18-51	Jar	Wall	Coarse	Red paint	-
TSF18-52	Bowl	Rim	Coarse	Undecorated	-
TSF18-54	Unidentified	Wall	Coarse	Red paint	-
TSF18-55	Unidentified	Wall	Coarse	Red paint	-
TSF18-56	Jar	Wall	Coarse	Rope band	-
TSF18-57	Unidentified	Wall	Coarse	Red paint	-
TSF18-58	Platter	Rim	Coarse	Red paint	-
TSF18-59	Holemouth jar	Wall	Coarse	Rope band, burnished	-
TSF18-6	Unidentified	Wall	Coarse	Red paint	-
TSF18-60	Unidentified	Wall	Coarse	Red paint	-
TSF18-62	Unidentified	Wall	Coarse	Red paint	-
TSF18-65	Unidentified	Wall	Coarse	Undecorated	-
TSF18-66	Unidentified	Base	Coarse	Undecorated	-
TSF18-67	Deep bowl	Rim	Coarse	Undecorated	Burnt
TSF18-68	Unidentified	Wall	Coarse	Red paint	-
TSF18-69	Unidentified	Wall	Coarse	Undecorated	Burnt
TSF18-7	Unidentified	Wall	Coarse	Red paint	-
TSF18-70	Unidentified	Wall	Coarse	Red paint	-
TSF18-71	Unidentified	Base	Coarse	Red paint	-
TSF18-73	Necked jar	Wall	Coarse	Red paint	-
TSF18-74	Unidentified	Wall	Coarse	Red paint	-
TSF18-75	Unidentified	Wall	Coarse	Red paint	-
TSF18-77	Unidentified	Wall	Coarse	Red paint	-
TSF18-78	V-shaped bowl	Rim	Coarse	Red paint	-
TSF18-8	Unidentified	Wall	Coarse	Red paint	-
TSF18-9	Jar	Wall+handle	Coarse	Undecorated	-
TSF19-1	Unidentified	Base	Coarse	Red paint	-
TSF19-13	Basalt bowl	Rim	-	Undecorated	-
TSF19-14	Limestone pedestal bowl	Base	-	Undecorated	-
TSF19-2	Unidentified	Wall	Coarse	Undecorated	-
TSF19-3	Necked jar	Rim	Coarse	Red paint	-
TSF19-4	Jar	Wall	Coarse	Rope band	-
TSF19-5	Necked jar	Rim	Coarse	Red paint	Burnt
TSF19-6	Necked jar	Rim	Coarse	Red paint, rope band	-
TSF19-7	Inverted bowl	Rim	Coarse	Red paint	-

Vessel	Vessel type	Sherd type	Fabric	Decoration	Evidence for heating
TSF19-9	Straight walled bowl	Rim	Fine	Biochrome geometric decoration	-
YEH-1	Strainer	Neck	Coarse	Undecorated	-
YV-10	Churn	-	Coarse	Red paint	-
YV-11	Churn	-	Coarse	Red paint	-
YV-6	Unidentified	Wall	Coarse	Undecorated	Burnt
YV-7	Churn	-	Coarse	Red paint	-
YV-8	Unidentified	Rim	Coarse	Undecorated	Burnt
YV-9	Churn	-	Coarse	Undecorated	Burnt

Appendix C. Overview of the lipid preservation (TLE=total lipid extract)

Site	Solvent extraction of internal samples				Acid fraction of saponified internal samples			
	Min. TLE (µg g^{-1})	Max TLE (µg g^{-1})	Average TLE (µg g^{-1})	% with > 5 µg g^{-1}	Min. TLE (µg g^{-1})	Max TLE (µg g^{-1})	Average TLE (µg g^{-1})	% with > 5 µg g^{-1}
Sha'ar Hagolan (SHH)	0.1	8.1	1.4	6.3	0.8	18.0	7.1	56.3
Neve Yam (NY)	0.0	5.0	1.2	5.9	0.4	58.9	5.7	11.8
Tel Hreiz (TH)	0.1	5.3	2.4	33.3	2.1	8.3	4.6	33.3
Tel Tsaf (TSF)	0.0	300.6	5.9	12.6	0.1	195.0	18.4	53.4
Azor (AZ)	-	-	1.2	0.0	-	-	5.6	100.0
Abu Matar (AM)	0.1	1.6	0.9	0.0	2.3	2.9	2.7	0.0
Ashqelon Agamim (AG)	0.0	14.8	1.1	3.6	1.1	49.3	7.2	46.4
Tel Bene Beraq (South) (BB)	0.0	55.3	6.5	10.0	4..0	86.4	17.6	60.0
Bir es-Safadi (BES)	0.1	2.2	0.6	0.0	1.2	8.7	4.8	54.5
Ein Gedi (EGD)	-	-	0.0	0.0	-	-	0.3	0.0
Fazael 7 (FZ)	0.1	3.9	1.3	0.0	3.0	28.4	9.2	62.5
Gilat (GLT)	0.1	88.1	9.1	23.1	1.4	92.2	12.3	61.5
Nissim Aloni Street (NA)	0.0	0.6	0.1	0.0	-	-	-	-
Peqi'in (PEQ)	0.0	0.5	0.3	0.0	6.7	10.7	8.0	100.0
Tsomet Shoket (TS)	0.1	2.1	0.7	0.0	0.9	30.3	6.1	38.9
Yehud (YEH and YV)	0.3	13.0	2.8	12.5	3.7	5.8	4.5	33.3
TOTAL	**0.0**	**300.6**	**3.7**	**8.8**	**0.1**	**195.0**	**12.3**	**49.4**

Appendix D. Lipid yield and the GC-c-IRMS isotope values obtained for the archaeological samples tested in this study (TLE=total lipid extract).

Sample Code	Quantification of the TLE (μg/g^{-1})		$^{13}\delta C_{16:0}$	$^{13}\delta C_{18:0}$	$\Delta^{13}C$
	Solvent extraction	Saponified acid fraction			
AG-1 In L2	0.1	1.6	-	-	-
AG-11 E	1.0	-	-	-	-
AG-11 In L2	1.1	11.3	-	-	-
AG-13 In L2	0.4	1.1	-	-	-
AG-15 In L2	0.5	1.3	-	-	-
AG-16 In L2	0.7	12.2	-	-	-
AG-18 In L2	2.3	11.4	-29.5	-29.6	-0.1
AG-19 In L2	0.7	5.2	-	-	-
AG-19 S	0.3	-	-	-	-
AG-2 In L2	0.4	1.8	-	-	-
AG-21 In L2	1.2	2.1	-	-	-
AG-22 In L2	0.1	2.0	-	-	-
AG-23 In L2	0.5	5.3	-	-	-
AG-24 In L2	0.5	3.1	-	-	-
AG-25 E	0.0	-	-	-	-
AG-25 In L2	0.5	14.1	-	-	-
AG-26 In L2	14.8	49.3	-27.2	-28.2	-1.0
AG-28 In L2	0.2	3.4	-29.4	-29.4	0.0
AG-30 In L2	0.8	16.1	-28.9	-29.7	-0.8
AG-32 In L2	2.7	18.4	-27.7	-28.8	-1.1
AG-34 In L2	0.8	6.7	-	-	-
AG-36 In L2	0.6	5.6	-27.9	-28.9	-1.0
AG-4 In L2	0.1	5.8	-	-	-
AG-40 In L2	0.5	2.3	-	-	-
AG-41 In L2	0.2	5.4	-	-	-
AG-43 In L2	0.2	1.4	-	-	-
AG-44 In L2	0.0	1.5	-	-	-
AG-45 In L2	0.1	4.3	-	-	-
AG-7 In L2	0.6	4.0	-	-	-
AG-8 In L2	1.3	2.9	-	-	-
AG-9 In L2	0.2	1.4	-	-	-
AM-1 In L2	1.6	2.3	-	-	-
AM-2 In L2	1.0	2.9	-	-	-
AM-5 E	1.3	-	-	-	-
AM-5 In L2	0.1	2.8	-	-	-
AZ-3 In L2	1.2	5.6	-	-	-
AZ-3 E	0.9	-	-	-	-
BB-1 E	5.8	-	-	-	-

Sample Code	Quantification of the TLE (µg/g^{-1})		$^{13}\delta C_{16:0}$	$^{13}\delta C_{18:0}$	$\Delta^{13}C$
	Solvent extraction	Saponified acid fraction			
BB-1 In L2	4.1	32.7	-28.4	-29.2	-0.8
BB-12 S	0.7	-	-	-	-
BB-19 In L2	2.1	8.2	-	-	-
BB-21 In L2	55.3	86.4	-	-	-
BB-25 In L2	0.3	4.8	-	-	-
BB-28 In L2	1.1	12.3	-	-	-
BB-3 E	2.2	-	-	-	-
BB-3 In L2	0.7	4.7	-	-	-
BB-4 E	1.4	-	-	-	-
BB-4 In L2	0.5	4.0	-	-	-
BB-5 E	1.0	-	-	-	-
BB-5 In L2	0.3	4.6	-	-	-
BB-6 E	1.4	-	-	-	-
BB-6 In L2	0.0	12.8	-	-	-
BB-7 E	2.3	-	-	-	-
BB-7 In L2	0.4	5.4	-	-	-
BB-8 S	0.5	-	-	-	-
BES-10 In L2	0.2	6.4	-	-	-
BES-12 In L2	0.3	2.4	-	-	-
BES-13 In L2	0.4	2.4	-	-	-
BES-15 In L2	0.3	3.8	-	-	-
BES-16 In L2	0.2	5.1	-	-	-
BES-4 In L2	0.4	7.1	-	-	-
BES-5 In L2	1.3	1.2	-	-	-
BES-6 In L2	2.2	6.2	-	-	-
BES-7 E	1.1	-	-	-	-
BES-7 In L2	0.5	7.7	-	-	-
BES-8 In L2	0.3	8.7	-	-	-
BES-9 In L2	0.1	1.8	-	-	-
EGD-1 In L2	0.0	0.3	-	-	-
EGD-1 S	3.6	-	-	-	-
FZ-10 In L2	1.4	14.0	-	-	-
FZ-11 In L2	1.5	3.9	-	-	-
FZ-3 In L2	0.9	6.0	-	-	-
FZ-4 E	4.9	-	-	-	-
FZ-4 In L2	3.9	28.4	-29.6	-29.6	0.0
FZ-5 E	1.9	-	-	-	-
FZ-5 In L2	1.1	8.7	-	-	-
FZ-6 In L2	0.1	3.0	-	-	-
FZ-6 S	0.2	-	-	-	-
FZ-7 In L2	0.6	3.4	-	-	-

Sample Code	Quantification of the TLE (µg/g^{-1})		$^{13}\delta C_{16:0}$	$^{13}\delta C_{18:0}$	$\Delta^{13}C$
	Solvent extraction	Saponified acid fraction			
FZ-8 In L2	0.8	6.1	-	-	-
GLT-1 In L2	1.6	7.9	-	-	-
GLT-10 In L2	1.3	6.0	-	-	-
GLT-11 E	1.6	-	-	-	-
GLT-11 In L2	0.7	11.2	-26.9	-30.5	-3.6
GLT-12 E	5.3	-	-	-	-
GLT-12 In L2	1.2	8.7	-28.2	-28.9	-0.7
GLT-13 In L2	0.5	5.7	-	-	-
GLT-2 In L2	4.2	5.8	-	-	-
GLT-3 In L2	0.3	3.3	-	-	-
GLT-4 E	3.1	-	-	-	-
GLT-4 In L2	5.8	3.0	-	-	-
GLT-5 In L2	0.8	4.8	-	-	-
GLT-6 In L2	10.8	1.4	-	-	-
GLT-7 E	9.1	-	-	-	-
GLT-7 In L2	3.4	6.0	-	-	-
GLT-8 In L2	88.1	92.2	-25.3	-28.6	-3.3
GLT-9 In L2	0.1	4.5	-	-	-
NA-11 In	0.1	-	-	-	-
NA-12 E	0.4	-	-	-	-
NA-12 In	0.0	-	-	-	-
NA-13 E	0.0	-	-	-	-
NA-13 In	0.6	-	-	-	-
NA-14 In	0.1	-	-	-	-
NA-19 In	0.0	-	-	-	-
NA-20 In	0.1	-	-	-	-
NA-5 In	0.0	-	-	-	-
NA-7 In	0.0	-	-	-	-
NA-8 In	0.0	-	-	-	-
NY-1 In L2	0.7	2.4	-	-	-
NY-10 In L2	0.4	1.2	-	-	-
NY-11 In L2	2.1	11.6	-26.0	-25.9	0.1
NY-12 E	1.4	-	-	-	-
NY-12 In L2	5.0	58.9	-	-	-
NY-13 In L2	0.8	0.4	-	-	-
NY-15 In L2	1.9	2.1	-	-	-
NY-16 In L2	0.5	2.3	-	-	-
NY-17 In L2	2.9	2.5	-	-	-
NY-18 In L2	0.0	0.7	-	-	-
NY-2 In L2	0.8	1.0	-	-	-
NY-3 In L2	0.8	4.1	-	-	-

Sample Code	Quantification of the TLE (µg/g^{-1})		$^{13}\delta C_{16:0}$	$^{13}\delta C_{18:0}$	$\Delta^{13}C$
	Solvent extraction	Saponified acid fraction			
NY-4 In L2	0.7	1.2	-	-	-
NY-5 In L2	0.0	1.1	-	-	-
NY-6 In L2	0.1	0.9	-	-	-
NY-7 In L2	2.1	0.9	-	-	-
NY-8 E	0.5	-	-	-	-
NY-8 In L2	1.0	3.4	-	-	-
NY-9 In L2	0.0	1.5	-	-	-
PEQ-1 In L2	0.0	6.7	-	-	-
PEQ-2 E	2.2	-	-	-	-
PEQ-2 In L2	0.5	10.7	-26.9	-26.7	0.2
PEQ-3 In L2	0.5	6.7	-	-	-
SHH-1 E	9.1	-	-	-	-
SHH-1 In L2	2.0	5.9	-	-	-
SHH-10 In L2	1.3	5.8	-	-	-
SHH-11 In L2	1.8	17.3	-	-	-
SHH-13 In L2	0.4	2.1	-	-	-
SHH-17 In L2	8.1	18.0	-29.7	-29.8	-0.1
SHH-18 In L2	1.6	5.7	-	-	-
SHH-19 In L2	0.3	2.3	-	-	-
SHH-2 In L2	0.1	3.3	-	-	-
SHH-20 In L2	1.1	4.5	-	-	-
SHH-21 In L2	0.6	0.8	-	-	-
SHH-3 In L2	0.3	1.8	-	-	-
SHH-4 In L2	0.5	6.6	-	-	-
SHH-5 E	0.9	-	-	-	-
SHH-5 In L2	1.0	16.8	-29.4	-29.3	0.2
SHH-6 In L2	2.3	9.5	-	-	-
SHH-7 In L2	1.0	10.1	-29.9	-29.3	0.6
SHH-8 In L2	0.4	3.4	-	-	-
TH-1 In L2	0.1	2.1	-	-	-
TH-2 E	3.3	-	-	-	-
TH-2 In L2	1.8	3.4	-	-	-
TH-4 In L2	5.3	8.3	-	-	-
TS-1 E	2.7	-	-	-	-
TS-1 In L2	0.8	3.3	-	-	-
TS-10 E	1.6	-	-	-	-
TS-10 In L2	1.2	21.3	-	-	-
TS-11 In L2	1.4	5.4	-27.9	-28.7	-0.8
TS-12 In L2	0.4	7.7	-29.2	-29.4	-0.2
TS-13 In L2	0.9	5.9	-28.9	-28.9	0.0
TS-16 In L2	0.2	5.6	-27.1	-28.1	-1.0

Sample Code	Quantification of the TLE (µg/g^{-1})		$^{13}\delta C_{16:0}$	$^{13}\delta C_{18:0}$	$\Delta^{13}C$
	Solvent extraction	Saponified acid fraction			
TS-18 In L2	0.4	2.6	-	-	-
TS-2 In L2	0.1	1.3	-	-	-
TS-20 In L2	0.2	1.0	-	-	-
TS-23 In L2	0.1	5.8	-26.2	-28.6	-2.4
TS-26 In L2	0.9	3.3	-	-	-
TS-2 E	0.8	-	-	-	-
TS-3 E	4.4	-	-	-	-
TS-3 In L2	0.4	2.8	-	-	-
TS-4 E	6.6	-	-	-	-
TS-4 In L2	0.6	2.8	-	-	-
TS-5 In L2	0.9	4.1	-	-	-
TS-6 In L2	2.1	3.2	-	-	-
TS-7 In L2	1.0	1.9	-	-	-
TS-8 In L2	0.9	0.9	-	-	-
TS-9 E	3.8	-	-	-	-
TS-9 In L2	0.2	30.3	-25.8	-29.1	-3.3
TSF17-1 In L2	0.8	6.0	-	-	-
TSF17-10 E	0.8	-	-	-	-
TSF17-10 In L2	0.4	51.3	-22.0	-26.5	-4.5
TSF17-12 In L2	3.3	3.3	-	-	-
TSF17-13 In L2	2.1	22.0	-	-	-
TSF17-15 E	0.8	-	-	-	-
TSF17-15 In L2	0.6	3.2	-	-	-
TSF17-16 E	0.2	-	-	-	-
TSF17-16 In L2	0.4	7.9	-	-	-
TSF17-18 In L2	1.0	13.6	-	-	-
TSF17-19 E	2.9	-	-	-	-
TSF17-19 In L2	0.4	28.1	-26.5	-26.5	0.0
TSF17-2 In L2	4.7	4.2	-	-	-
TSF17-21 In L2	0.9	2.9	-	-	-
TSF17-22 E	28.5	-	-	-	-
TSF17-22 In L2	33.3	26.7	-25.9	-29.9	0.6
TSF17-23 E	0.8	-	-	-	-
TSF17-23 In L2	1.6	32.4	-26.5	-28.7	-3.4
TSF17-25 In L2	2.7	4.6	-	-	-
TSF17-26 In L2	0.1	1.3	-	-	-
TSF17-27 E	2.7	-	-	-	-
TSF17-27 In L2	0.9	12.7	-25.7	-28.7	-3.0
TSF17-28 E	22.2	-	-	-	-
TSF17-28 In L2	1.4	22.2	-25.5	-25.0	0.5
TSF17-29 E	1.3	-	-	-	-

Sample Code	Quantification of the TLE (µg/g^{-1})		$^{13}\delta C_{16:0}$	$^{13}\delta C_{18:0}$	$\Delta^{13}C$
	Solvent extraction	Saponified acid fraction			
TSF17-29 In L2	0.5	2.9	-	-	-
TSF17-3 In L2	0.0	4.1	-	-	-
TSF17-30 E	1.1	-	-	-	-
TSF17-30 In L2	0.9	8.6	-29.7	-29.8	-0.2
TSF17-31 E	1.1	-	-	-	-
TSF17-31 In L2	0.7	8.7	-29.8	-29.7	0.1
TSF17-32 E	0.9	-	-	-	-
TSF17-32 In L2	0.4	3.9	-	-	-
TSF17-32 S	0.8	-	-	-	-
TSF17-4 E	1.0	-	-	-	-
TSF17-4 In L2	0.7	6.7	-	-	-
TSF17-5 In L2	6.3	10.6	-26.6	-29.1	-2.5
TSF17-6 In L2	13.6	18.1	-25.3	-26.9	-1.6
TSF17-7 In L2	1.2	6.5	-	-	-
TSF17-8 In L2	0.0	2.7	-	-	-
TSF17-9 In L2	0.5	2.8	-	-	-
TSF18-1 In L2	0.3	4.4	-	-	-
TSF18-10 In L2	0.3	1.0	-	-	-
TSF18-11 In L2	0.1	0.5	-	-	-
TSF18-13 In L2	5.2	5.0	-	-	-
TSF18-14 In L2	0.3	3.3	-	-	-
TSF18-15 In L2	0.0	1.5	-	-	-
TSF18-16 In L2	0.4	13.0	-	-	-
TSF18-17 In L2	0.4	4.2	-	-	-
TSF18-18 In L2	1.5	11.3	-	-	-
TSF18-19 In L2	0.4	1.9	-	-	-
TSF18-2 In L2	0.3	2.6	-	-	-
TSF18-20 E	2.9	-	-	-	-
TSF18-20 In L2	15.1	195.0	-	-	-
TSF18-21 In L2	0.1	2.1	-	-	-
TSF18-22 In L2	2.4	6.0	-	-	-
TSF18-23 In L2	0.4	2.7	-	-	-
TSF18-24 In L2	45.4	151.4	-18.9	-23.2	-4.4
TSF18-25 In L2	10.0	103.2	-25.5	-28.7	-3.2
TSF18-26 In L2	0.2	10.5	-	-	-
TSF18-27 In L2	1.9	1.9	-	-	-
TSF18-28 In L2	3.1	9.7	-	-	-
TSF18-29 In L2	2.1	13.6	-26.5	-27.4	-0.9
TSF18-3 E	1.7	-	-	-	-
TSF18-3 In L2	3.3	124.8	-23.1	-24.4	-1.4
TSF18-30 In L2	0.1	1.0	-	-	-

Sample Code	Quantification of the TLE (µg/g^{-1})		$^{13}\delta C_{16:0}$	$^{13}\delta C_{18:0}$	$\Delta^{13}C$
	Solvent extraction	Saponified acid fraction			
TSF18-31 In L2	0.4	8.7	-	-	-
TSF18-34 In L2	8.9	43.6	-	-	-
TSF18-35 In L2	1.5	5.1	-	-	-
TSF18-36 In L2	4.1	5.4	-	-	-
TSF18-37 In L2	0.8	3.8	-	-	-
TSF18-39 In L2	0.3	8.6	-	-	-
TSF18-4 E	3.0	-	-	-	-
TSF18-4 In L2	0.5	59.1	-28.0	-27.1	0.8
TSF18-40 In L2	13.8	168.1	-22.9	-25.5	-2.6
TSF18-42 In L2	0.5	6.7	-	-	-
TSF18-44 In L2	1.4	1.8	-	-	-
TSF18-45 In L2	1.0	4.0	-	-	-
TSF18-46 E	4.4	-	-	-	-
TSF18-46 In L2	0.2	25.7	-28.3	-28.6	-0.3
TSF18-47 In L2	0.2	3.6	-	-	-
TSF18-48 In L2	3.9	18.0	-	-	-
TSF18-49 In L2	0.0	0.6	-	-	-
TSF18-5 In L2	1.8	11.7	-	-	-
TSF18-50 In L2	0.9	1.8	-	-	-
TSF18-51 E	1.7	-	-	-	-
TSF18-51 In L2	22.0	33.3	-25.6	-24.6	1.0
TSF18-52 In L2	0.0	5.0	-	-	-
TSF18-54 E	0.9	-	-	-	-
TSF18-54 In L2	0.2	12.1	-	-	-
TSF18-55 In L2	0.4	3.9	-	-	-
TSF18-56 In L2	1.1	2.2	-	-	-
TSF18-57 Cr	6.8	12.8	-	-	-
TSF18-57 E	1.5	-	-	-	-
TSF18-57 In L2	3.1	5.6	-	-	-
TSF18-58 In L2	1.1	5.9	-	-	-
TSF18-59 In L2	1.8	7.6	-	-	-
TSF18-6 In L2	0.7	3.8	-	-	-
TSF18-60 In L2	0.1	0.5	-	-	-
TSF18-62 In L2	0.3	0.5	-	-	-
TSF18-65 E	1.8	-	-	-	-
TSF18-65 In L2	1.5	20.5	-	-	-
TSF18-66 E	0.5	-	-	-	-
TSF18-66 In L2	0.6	15.7	-27.2	-28.0	-0.8
TSF18-67 In L2	1.3	1.1	-	-	-
TSF18-68 In L2	0.6	2.4	-	-	-
TSF18-69 In L2	19.8	86.0	-	-	-

Sample Code	Quantification of the TLE (µg/g^{-1})		$^{13}\delta C_{16:0}$	$^{13}\delta C_{18:0}$	$\Delta^{13}C$
	Solvent extraction	Saponified acid fraction			
TSF18-7 In L2	2.0	12.6	-	-	-
TSF18-70 In L2	1.0	9.3	-	-	-
TSF18-71 In L2	3.2	4.9	-	-	-
TSF18-73 In L2	0.2	0.8	-	-	-
TSF18-74 In L2	0.5	2.2	-	-	-
TSF18-75 In L2	0.8	11.1	-	-	-
TSF18-77 In L2	0.3	0.9	-	-	-
TSF18-78 In L2	1.2	5.2	-	-	-
TSF18-8 In L2	0.0	0.8	-	-	-
TSF18-9 In L2	0.0	0.8	-	-	-
TSF19-1 In L2	0.2	0.8	-	-	-
TSF19-13 In L2	1.1	14.9	-26.4	-27.5	-1.1
TSF19-14 In L2	0.1	0.1	-	-	-
TSF19-2 In L2	1.0	19.2	-30.1	-30.0	0.1
TSF19-3 In L2	3.0	51.1	-	-	-
TSF19-4 In L2	300.6	175.9	-26.9	-26.0	0.9
TSF19-5 In L2	0.2	1.3	-	-	-
TSF19-6 In L2	23.9	4.5	-	-	-
TSF19-7 In L2	0.4	1.7	-	-	-
TSF19-9 In L2	0.2	1.4	-	-	-
YEH-1 E	2.3	-	-	-	-
YEH-1 In L2	2.5	4.0	-	-	-
YV-10 In L2	0.3	3.7	-	-	-
YV-11 In L2	1.4	-	-	-	-
YV-11 E	0.4	-	-	-	-
YV-6 E	2.2	-	-	-	-
YV-6 In L2	13.0	-	-	-	-
YV-7 In L2	0.3	5.8	-	-	-
YV-8 E	1.1	-	-	-	-
YV-8 In L2	0.3	-	-	-	-
YV-9 E	9.2	-	-	-	-
YV-9 In L2	0.9	-	-	-	-

Appendix E. Summary of the lipid profile obtained for the archaeological samples tested in this study after solvent extraction (COS=cyclic octaatomic sulfur; Cr=crust; $C_{x:y}$=fatty acid with chain length X and Y number of double bonds; D_x=diacylglycerol with X carbon atoms; E=exterior; In=interior; K_x=ketone with chain length X; L2=Layer 2; $M_{x:y}$=monoacylglycerol with chain length X and Y number of double bonds; P=plastic; S=soil; SS=sunscreen; T_x=triacylglycerol with X carbon atoms; WE_x=wax ester with chain length X).

Sample Code	Contamination	Saturated fatty acids	Unsaturated fatty acids	*n*-alkanes	Alcohols	Ketones	Wax esters	Sterols	Monoacylglycerols	Diacylglycerols	Triacylglycerols	Other
AG-1 In L2	P	$C_{16:0}$, $C_{18:0}$	-	C_{16}	C_{18}	-	-	-	-	-	-	-
AG-11 E	P	$C_{16:0}$, $C_{18:0}$	-	C_{21}, C_{23}, C_{25}-C_{29}, C_{31}	C_{18}, C_{16}, C_{28}	-	-	-	-	-	-	-
AG-11 In L2	P	$C_{16:0}$, $C_{18:0}$, $C_{20:0}$, $C_{22:0}$, $C_{24:0}$, $C_{26:0}$	-	C_{25}-C_{29}	C_{18}, C_{24}, C_{26}, C_{28}	-	-	-	-	-	-	-
AG-13 In L2	P	$C_{16:0}$, $C_{18:0}$, $C_{24:0}$, $C_{26:0}$, $C_{28:0}$	-	C_{31}	C_{18}, C_{26}	-	-	-	-	-	-	-
AG-15 In L2	P	$C_{16:0}$, $C_{18:0}$	-	-	C_{18}	-	-	-	-	-	-	-
AG-16 In L2	P	$C_{16:0}$, $C_{18:0}$	-	-	C_{18}	-	-	-	$M_{16:0}$, $M_{18:0}$	-	-	-
AG-18 In L2	P	$C_{16:0}$, $C_{18:0}$	-	C_{24}-C_{31}	C_{18}	-	-	-	$M_{16:0}$, $M_{18:0}$	-	-	-
AG-19 In L2	P	$C_{16:0}$, $C_{18:0}$	-	C_{25}-C_{29}	C_{18}	-	-	-	$M_{16:0}$, $M_{18:0}$	-	-	-
AG-19 S	P	$C_{16:0}$, $C_{18:0}$	-	C_{27}, C_{29}, C_{31}	C_{16}, C_{18}, C_{24}, C_{26}, C_{28}	-	-	β-sitosterol	$M_{16:0}$, $M_{18:0}$	-	-	-
AG-2 In L2	P	$C_{16:0}$, $C_{18:0}$	-	C_{25}-C_{27}, C_{29}, C_{31}	C_{18}, C_{26}	-	-	-	-	-	-	-
AG-21 In L2	P	$C_{16:0}$, $C_{18:0}$	-	C_{23}-C_{29}, C_{31}, C_{32}	C_{18}, C_{24}, C_{26}, C_{28}	-	-	-	-	-	-	-
AG-22 In L2	P	$C_{16:0}$, $C_{18:0}$	-	-	C_{18}	-	-	-	-	-	-	-
AG-23 In L2	P	$C_{16:0}$, $C_{18:0}$	-	-	C_{18}	-	-	-	$M_{16:0}$, $M_{18:0}$	-	-	-
AG-24 In L2	P	-	-	C_{14}	C_{18}	-	-	-	$M_{16:0}$, $M_{18:0}$	-	-	-

Sample Code	Contamination	Saturated fatty acids	Unsaturated fatty acids	*n*-alkanes	Alcohols	Ketones	Wax esters	Sterols	Monoacylglycerols	Diacylglycerols	Triacylglycerols	Other
AG-25 E	P	-	-	C_{27}	C_{18}	-	-	-	-	-	-	-
AG-25 In L2	P	$C_{16:0}$, $C_{18:0}$	-	C_{25}-C_{29}	C_{18}	-	-	-	-	-	-	-
AG-26 In L2	P	$C_{15:0}$-$C_{28:0}$	$C_{18:1}$	C_{24}-C_{31}	C_{18}, C_{26}, C_{28}, C_{30}	-	WE_{42}, WE_{44}	-	$M_{16:0}$, $M_{18:0}$	D_{32}, D_{34}	T_{46}, T_{48}, T_{50}, T_{52}, T_{54}	-
AG-28 In L2	P	$C_{16:0}$, $C_{18:0}$	-	C_{25}-C_{29}	-	-	-	-	$M_{16:0}$, $M_{18:0}$	-	-	-
AG-30 In L2	P	$C_{16:0}$, $C_{18:0}$	-	C_{25}-C_{29}	C_{18}	-	-	Cholesterol	$M_{16:0}$, $M_{18:0}$	-	-	-
AG-32 In L2	P	$C_{16:0}$, $C_{18:0}$	-	C_{24}-C_{31}	C_{18}, C_{22}, C_{24}, C_{26}, C_{28}, C_{30}	-	-	β-sitosterol, campesterol, stigmasterol	$M_{16:0}$, $M_{18:0}$, $M_{18:1}$	D_{34}	-	-
AG-34 In L2	P	$C_{16:0}$, $C_{18:0}$	-	-	C_{18}	-	-	-	-	-	-	-
AG-36 In L2	P	$C_{16:0}$, $C_{18:0}$	-	-	C_{28}	-	-	-	-	-	-	-
AG-4 In L2	P	-	-	-	C_{18}	-	-	-	$M_{16:0}$	-	-	-
AG-40 In L2	P	$C_{16:0}$, $C_{18:0}$	$C_{18:1}$	C_{25}-C_{29}	C_{16}, C_{18}, C_{24}, C_{28}	-	-	β-sitosterol, campesterol, cholesterol, stigmasterol	$M_{16:0}$	-	-	-
AG-41 In L2	P	$C_{16:0}$, $C_{18:0}$	-	C_{27}	C_{18}	-	-	-	-	-	-	-
AG-43 In L2	P	$C_{16:0}$, $C_{18:0}$	-	-	C_{18}	-	-	-	-	-	-	-
AG-44 In L2	P	-	-	C_{31}	C_{18}	-	-	-	-	-	-	-
AG-45 In L2	P	$C_{16:0}$, $C_{18:0}$	-	-	-	-	-	-	-	-	-	-
AG-7 In L2	P	-	-	C_{25}, C_{27}, C_{29}, C_{31}	C_{18}	-	-	-	$M_{16:0}$, $M_{18:0}$	-	-	-
AG-8 In L2	P	$C_{16:0}$, $C_{18:0}$	-	C_{27}	C_{18}	-	-	-	-	-	-	-
AG-9 In L2	P	$C_{16:0}$, $C_{18:0}$	-	C_{27}	-	-	-	-	-	-	-	-
AM-1 In L2	P	-	-	-	-	-	-	-	-	-	-	-

Sample Code	Contamination	Saturated fatty acids	Unsaturated fatty acids	*n*-alkanes	Alcohols	Ketones	Wax esters	Sterols	Monoacylglycerols	Diacylglycerols	Triacylglycerols	Other
AM-2 In L2	P	$C_{8:0}$, $C_{9:0}$	-	-	C_{16}	-	-	-	-	-	-	-
AM-5 E	P	$C_{16:0}$, $C_{18:0}$	-	C_{22}-C_{25}, C_{29}, C_{31}	C_{16}, C_{18}, C_{26}, C_{28}	-	-	-	-	-	-	-
AM-5 In L2	P	-	-	-	C_{18}	-	-	-	-	-	-	-
AZ-3 In L2	P	$C_{16:0}$-$C_{18:0}$	-	-	C_{18}, C_{20}-C_{27}, C_{29}	C16, C_{18}	-	-	$M_{16:0}$	-	-	-
AZ-3 E	P	$C_{16:0}$, $C_{18:0}$	-	-	C_{20}, C_{29}	C_{14}, C_{16}, C_{18}	-	-	$M_{16:0}$	-	-	-
BB-1 E	P	$C_{16:0}$	$C_{16:1}$, $C_{18:1}$	-	C_{18}, C_{22}, C_{28}	-	-	β-sitosterol, campesterol, Stigmasterol	-	-	-	Ferruginol, 2,3-dehydroferruginol, sempervirol, totarol
BB-1 In L2	P	$C_{16:0}$, $C_{18:0}$	$C_{16:1}$, $C_{18:1}$	-	C_{18}, C_{22}	-	-	β-sitosterol, campesterol, cholesterol, stigmasterol	-	-	-	Ferruginol, 2,3-dehydroferruginol, sempervirol, totarol
BB-12 S	P	$C_{16:0}$, $C_{18:0}$, $C_{22:0}$, $C_{24:0}$, $C_{26:0}$	-	C_{27}, C_{29}	C_{16}, C_{18}, C_{22}, C_{24}, C_{26}, C_{28}, C_{30}	-	-	β-sitosterol, campesterol, stigmasterol	$M_{16:0}$	-	-	Ferruginol, 2,3-dehydroferruginol, sempervirol, totarol
BB-19 In L2	P	$C_{15:0}$-$C_{18:0}$	$C_{18:1}$	C_{25}-C_{30}	C_{18}	-	-	-	-	-	-	-
BB-21 In L2	P	$C_{16:0}$, $C_{18:0}$, $C_{24:0}$	-	C_{23}-C_{33}, C_{35}, C_{37}	C_{24}, C_{26}, C_{28}, C_{30}, C_{32}	K_{33}	WE_{40}, WE_{42}, WE_{44}, WE_{46}, WE_{48}, WE_{50}	-	-	-	-	-
BB-25 In L2	P	-	-	C_{25}-C_{29}, C_{31}	C_{28}	-	-	-	-	-	-	-
BB-28 In L2	P	$C_{16:0}$, $C_{18:0}$	$C_{18:1}$	C_{24}-C_{32}	C_{18}	-	-	β-sitosterol, campesterol, stigmasterol	$M_{16:0}$, $M_{18:1}$	D_{32}	-	Dehydroabietic acid
BB-3 E	P	$C_{15:0}$-$C_{18:0}$	$C_{16:1}$, $C_{18:1}$	-	C_{16}, C_{18}, C_{22}	-	-	β-sitosterol, cholesterol	-	-	-	-
BB-3 In L2	P	$C_{15:0}$-$C_{18:0}$	$C_{16:1}$, $C_{18:1}$	-	C_{18}	-	-	Cholesterol	-	-	-	Dehydroabietic acid
BB-4 E	P, SS	$C_{16:0}$-$C_{18:0}$, $C_{26:0}$	-	-	C_{16}, C_{18}, C_{20}, C_{22}, C_{26}-C_{28}, C_{30}	-	-	-	-	-	-	-
BB-4 In L2	P	$C_{16:0}$-$C_{18:0}$	$C_{18:1}$	-	C_{16}, C_{22}, C_{24}, C_{26}, C_{28}	-	-	β-sitosterol, cholesterol, stigmasterol	-	-	-	-

Sample Code	Contamination	Saturated fatty acids	Unsaturated fatty acids	*n*-alkanes	Alcohols	Ketones	Wax esters	Sterols	Monoacylglycerols	Diacylglycerols	Triacylglycerols	Other
BB-5 E	P, SS	$C_{16:0}$-$C_{18:0}$	-	-	C_{16}, C_{18}, C_{22}, C_{24}, C_{26}-C_{28}	-	-	-	-	-	-	-
BB-5 In L2	P	$C_{16:0}$-$C_{18:0}$	-	-	C_{18}	-	-	-	-	-	-	-
BB-6 E	P	-	-	-	-	-	-	-	-	-	-	-
BB-6 In L2	-	-	-	-	-	-	-	-	-	-	-	-
BB-7 E	P	$C_{16:0}$-$C_{18:0}$, $C_{26:0}$	$C_{16:1}$, $C_{20:1}$	-	C_{18}, C_{24}, C_{26}, C_{28}, C_{30}	-	-	-	-	-	-	-
BB-7 In L2	P	$C_{16:0}$-$C_{18:0}$	-	-	C_{18}, C_{28}	-	-	Cholesterol	-	-	-	-
BB-8 S	P	$C_{15:0}$-$C_{18:0}$, $C_{23:0}$, $C_{24:0}$, $C_{26:0}$	$C_{18:1}$	C_{23}-C_{31}	C_{18}, C_{22}, C_{24}, C_{26}, C_{28}, C_{30}	-	-	β-sitosterol, campesterol, stigmasterol	$M_{16:0}$, $M_{18:0}$	-	-	-
BES-9 In L2	P	-	-	-	C_{16}, C_{18}	-	-	-	-	-	-	-
BES-16 In L2	P	$C_{8:0}$-$C_{9:0}$	-	-	C_{14}, C_{16}, C_{18}	-	-	-	-	-	-	-
BES-10 In L2	P	$C_{7:0}$-$C_{8:0}$	-	-	C_{16}, C_{18}	-	-	-	-	-	-	-
BES-15 In L2	P	-	-	-	C_{16}, C_{18}	-	-	-	-	-	-	-
BES-12 In L2	P	$C_{7:0}$-$C_{9:0}$	-	-	C_{16}, C_{18}	-	-	-	-	-	-	-
BES-8 In L2	P	-	-	-	C_{18}	-	-	-	-	-	-	-
BES-4 In L2	P	-	-	-	C_{16}, C_{18}, C_{26}	-	-	-	-	-	-	-
BES-13 In L2	P	$C_{8:0}$, $C_{9:0}$, $C_{16:0}$, $C_{18:0}$	-	-	C_{16}, C_{18}	-	-	-	-	-	-	-
BES-7 E	P	$C_{12:0}$, $C_{14:0}$	-	C_{18}-C_{22}, C_{24}	C_{16}, C_{18}	-	-	-	$M_{16:0}$, $M_{18:0}$	-	-	-
BES-7 In L2	P	-	-	-	C_{16}, C_{18}	-	-	-	-	-	-	-

Sample Code	Contamination	Saturated fatty acids	Unsaturated fatty acids	*n*-alkanes	Alcohols	Ketones	Wax esters	Sterols	Monoacylglycerols	Diacylglycerols	Triacylglycerols	Other
BES-5 In L2	P	$C_{8:0}$-$C_{9:0}$	-	-	C_{12}, C_{16}, C_{18}	-	-	-	-	-	-	-
BES-6 In L2	P	$C_{7:0}$-$C_{18:0}$	-	C_{26}-C_{39}	C_{16}, C_{18}	-	-	-	-	-	-	-
EGD-1 In L2	SS	-	-	-	-	-	-	-	$M_{18:0}$	-	-	-
EGD-1 S	-	$C_{16:0}$-$C_{34:0}$	$C_{18:1}$	C_{23}, C_{25}-C_{31}	C_{20}, C_{24}, C_{26}, C_{27}, C_{28}, C_{30}	-	-	-	$M_{18:0}$	-	-	-
FZ-10 In L2	P	$C_{16:0,}$ $C_{18:0}$	$C_{18:1}$	C_{23}, C_{25}, C_{27}-C_{28}, C_{31}	C_{18}, C_{24}, C_{26}, C_{28}	-	-	-	-	-	-	-
FZ-11 In L2	P	-	-	C_{24}-C_{32}	C_{18}, C_{26}, C_{28}, C_{30}	-	-	-	-	-	-	-
FZ-3 In L2	P	$C_{16:0,}$ $C_{18:0}$	-	C_{27}, C_{29}, C_{31}, C_{33}	C_{18}, C_{26}, C_{28}, C_{30}	-	-	-	-	-	-	-
FZ-4 E	P	$C_{28:0}$, $C_{30:0}$	-	C_{21}, C_{23}, C_{25}-C_{33}	C_{18}, C_{26}, C_{28}, C_{30}	-	-	-	-	-	-	-
FZ-4 In L2	P	$C_{9:0}$, $C_{16:0}$, $C_{18:0}$	$C_{18:1}$	C_{23}-C_{31}	C_{18}, C_{26}, C_{28}	-	-	Cholesterol	-	-	-	-
FZ-5 E	P	$C_{6:0}$-$C_{10:0}$, $C_{16:0}$, $C_{18:0}$	-	C_{21}, C_{25}-C_{31}, C_{33}	C_{18}, C_{26}, C_{28}, C_{30}	-	-	-	-	-	-	-
FZ-5 In L2	P	-	-	C_{25}-C_{31}	C_{18}, C_{26}, C_{28}	-	-	-	$M_{16:0}$, $M_{18:0}$	-	-	-
FZ-6 In L2	P	$C_{16:0,}$ $C_{18:0}$	-	-	C_{18}	-	-	β-sitosterol	-	-	-	-
FZ-6 S	P	$C_{16:0,}$ $C_{18:0}$	-	C_{29}, C_{31}	C_{18}, C_{24}, C_{26}, C_{28}	-	-	β-sitosterol, stigmasterol	$M_{18:0}$	-	-	-
FZ-7 In L2	P	$C_{16:0,}$ $C_{18:0}$	-	C_{24}-C_{32}	C_{18}, C_{26}, C_{28}, C_{30}	-	-	-	-	-	-	-
FZ-8 In L2	P	$C_{16:0,}$ $C_{18:0}$	-	C_{25}, C_{27}, C_{29}, C_{31}	C_{18}, C_{26}, C_{28}, C_{30}	-	-	-	-	-	-	-
GLT-1 In L2	P	$C_{7:0}$-$C_{10:0}$	-	C_{20}, C_{21}, C_{29}	C_{14}, C_{16}, C_{18}, C_{26}, C_{28}, C_{30}	K_{33}	-	β-sitosterol, campesterol, cholesterol	-	D_{32}, D_{34}	-	-
GLT-10 In L2	P	$C_{9:0}$, $C_{10:0}$	-	-	C_{14}, C_{16}, C_{28}	-	-	-	-	-	-	-
GLT-11 E	P	$C_{14:0}$, $C_{16:0}$, $C_{18:0}$	$C_{18:1}$	C_{29}	C_{16}, C_{18}	-	-	-	$M_{16:0}$	-	-	-

Sample Code	Contamination	Saturated fatty acids	Unsaturated fatty acids	*n*-alkanes	Alcohols	Ketones	Wax esters	Sterols	Monoacylglycerols	Diacylglycerols	Triacylglycerols	Other
GLT-11 In L2	P	-	-	-	C_{14}, C_{16}, C_{18}	-	-	-	-	-	-	-
GLT-12 E	P	$C_{7:0}$-$C_{14:0}$, $C_{16:0}$, $C_{18:0}$	$C_{18:1}$	-	C_{16}, C_{18}	-	-	-	$M_{16:0}$, $M_{18:0}$	-	-	-
GLT-12 In L2	P	$C_{7:0}$-$C_{10:0}$	-	-	-	-	-	-	-	-	-	-
GLT-13 In L2	P	$C_{9:0}$, $C_{10:0}$	-	-	C_{18}	-	-	-	-	-	-	-
GLT-2 In L2	P	-	-	C_{22}-C_{25}, C_{27}, C_{29}-C_{31}	C_{16}, C_{18}, C_{24}, C_{26}	-	-	-	-	-	-	C_7-C_{12} γ lactone
GLT-3 In L2	P	$C_{9:0}$	-	C_{21}, C_{29}	C_{16}, C_{26}, C_{28}	-	WE_{36}, WE_{38}	Cholesterol	$M_{16:0}$, $M_{18:0}$	D_{32}	-	C_7-C_{12} γ lactone
GLT-4 E	P	$C_{9:0}$-$C_{12:0}$, $C_{14:0}$, $C_{16:0}$, $C_{18:0}$	-	C_{20}-C_{23}, C_{25}-C_{27}, C_{29}, C_{31}	C_{14}, C_{16}, C_{18}, C_{24}, C_{26}, C_{28}	-	-	Cholesterol	$M_{16:0}$, $M_{18:0}$	D_{26}, D_{32}	-	-
GLT-4 In L2	P	$C_{12:0}$, $C_{13:0}$, $C_{16:0}$	-	C_{20}-C_{23}, C_{25}, C_{27}-C_{29}, C_{31}, C_{33}	C_{12}, C_{14}, C_{16}, C_{18}, C_{22}, C_{24}, C_{26}, C_{28}	-	-	-	$M_{16:0}$	D_{32}	-	-
GLT-5 In L2	P	$C_{16:0,}$ $C_{18:0}$	-	C_{21}, C_{23}, C_{25}, C_{29}	C_{14}, C_{16}, C_{18}, C_{26}, C_{28}	-	-	β-sitosterol, campesterol, stigmasterol	$M_{16:0}$, $M_{18:0}$	D_{32}	T_{48}	-
GLT-6 In L2	P	$C_{7:0}$-$C_{14:0}$, $C_{16:0}$, $C_{18:0}$	$C_{16:1}$	C_{20}-C_{25}	C_{16}, C_{18}	-	-	-	$M_{16:0}$	-	-	-
GLT-7 E	P	$C_{9:0}$, $C_{10:0}$, $C_{12:0}$, $C_{14:0}$, $C_{16:0}$, $C_{18:0}$	-	C_{19}-C_{25}, C_{27}, C_{29}	C_{12}, C_{14}, C_{16}, C_{18}	-	WE_{36}	Cholesterol	$M_{16:0}$	D_{30}, D_{32}	-	-
GLT-7 In L2	P, COS	$C_{12:0}$, $C_{16:0}$, $C_{18:0}$	-	C_{20}-C_{26}	C_{14}, C_{16}, C_{18}	-	-	-	$M_{16:0}$, $M_{18:0}$	D_{32}	-	-
GLT-8 In L2	P	$C_{10:0}$, $C_{12:0}$-$C_{14:0}$, $C_{16:0}$, $C_{18:0}$	-	-	C_{14}	-	-	-	-	-	-	-
GLT-9 In L2	P	-	-	-	C_{14}, C_{16}	-	-	-	-	-	-	-
NA-11 In	P	-	-	C_{32}	-	-	-	-	-	-	-	-
NA-12 E	P	-	-	C_{27}, C_{29}, C_31, C_{32}	-	-	-	-	-	-	-	-
NA-12 In	P	-	-	-	-	-	-	-	-	-	-	-
NA-13 E	P	-	-	-	-	-	-	-	$M_{16:0}$	-	-	-

Sample Code	Contamination	Saturated fatty acids	Unsaturated fatty acids	*n*-alkanes	Alcohols	Ketones	Wax esters	Sterols	Monoacylglycerols	Diacylglycerols	Triacylglycerols	Other
NA-13 In	P	$C_{16:0}$, $C_{18:0}$	-	C_{21}, C_{25}, C_{27}, C_{29}, C_{31}, C_{32}	C_{26}	-	-	-	$M_{16:0}$, $M_{18:0}$	-	-	-
NA-14 In	P	$C_{16:0}$, $C_{18:0}$	-	C_{32}	-	-	-	-	-	-	-	-
NA-19 In	P	$C_{16:0}$, $C_{18:0}$	-	-	-	-	-	-	-	-	-	-
NA-20 In	P	$C_{16:0}$	-	C_{32}		-	-	-	-	-	-	-
NA-5 In	P	-	-	C_{31}	C_{24}	-	-	-	-	-	-	-
NA-7 In	P	$C_{16:0}$, $C_{18:0}$	-	-	-	-	-	-	$M_{18:0}$	-	-	-
NA-8 In	P	-	-	-	-	-	-	-	-	-	-	-
NY-1 In L2	P	$C_{16:0}$, $C_{18:0}$	-	C_{18}	C_{14}, C_{16}, C_{18}	-	-	Cholesterol	$M_{16:0}$	-	-	-
NY-10 In L2	P	$C_{16:0}$, $C_{18:0}$	-	C_{23}-C_{27}, C_{29}	C_{14}, C_{16}, C_{18}, C_{26}, C_{28}	-	-	Cholesterol	$M_{16:0}$, $M_{18:0}$	-	-	-
NY-11 In L2	P	$C_{12:0}$, $C_{15:0}$, $C_{16:0}$, $C1_{8:0}$, $C_{20:0}$	$C_{18:1}$	C_{18}, C_{20}-C_{22}, C_{24}-C_{26}	C_{14}-C_{16}, C_{18}	-	-	Cholesterol	$M_{14:0}$, $M_{16:0}$, $M_{18:0}$	D_{32}, D_{34}, D_{36}	-	-
NY-12 E	P	$C_{16:0}$, $C_{18:0}$	-	C_{20}-C_{29}, C_{31}	C_{14}, C_{16}, C_{18}, C_{20}, C_{22}, C_{24}, C_{26}, C_{28}	-	WE_{42}, WE_{44}	-	$M_{16:0}$	-	-	-
NY-12 In L2	P	$C_{9:0}$, $C_{15:0}$-$C_{18:0}$	$C_{18:1}$	C_{20}, C_{21}, C_{23}-C_{25}, C_{27}, C_{29}	C_{12}, C_{14}, C_{16}, C_{18}, C_{24}, C_{26}, C_{28}, C_{30}	K_{31}, K_{33}	WE_{42}, WE_{44}, WE_{46}	Cholesterol	$M_{16:0}$	-	-	-
NY-13 In L2	P	$C_{10:0}$, $C_{12:0}$, $C_{16:0}$, $C_{18:0}$	-	C_{18}, C_{20}, C_{21}, C_{27}	C_{12}, C_{14}, C_{16}, C_{18}, C_{28}, C_{30}	-	-	β-sitosterol, cholesterol	-	-	-	-
NY-15 In L2	P	$C_{7:0}$-$C_{14:0}$, $C_{16:0}$, $C_{18:0}$	-	-	C_{14}, C_{16}, C_{18}	-	-	-	$M_{16:0}$	-	-	-
NY-16 In L2	P	$C_{16:0}$, $C_{18:0}$	-	-	C_{14}-C_{16}, C_{18}, C_{20}, C_{26}, C_{28}, C_{30}	-	-	β-sitosterol, campesterol	$M_{14:0}$, $M_{16:0}$, $M_{18:0}$	-	-	-
NY-17 In L2	P	$C_{16:0}$, $C_{18:0}$, $C_{20:0}$, $C_{22:0}$, $C_{24:0}$	$C_{18:1}$	C_{18}, C_{20}, C_{21}, C_{25}, C_{27}, C_{29}, C_{31}	C_{12}-C_{16}, C_{18}, C_{24}, C_{26}, C_{28}, C_{30}	-	-	β-sitosterol, cholesterol	$M_{16:0}$, $M_{18:0}$	-	-	-
NY-18 In L2	P	-	-	-	C_{18}	-	-	-	-	-	-	-
NY-2 In L2	P	$C_{9:0}$, $C_{12:0}$, $C_{16:0}$, $C_{18:0}$	-	C_{27}	C_{13}, C_{14}, C_{16}	-	-	Cholesterol	$M_{16:0}$	-	-	-
NY-3 In L2	P	$C_{14:0}$-$C_{16:0}$, $C_{18:0}$, $C_{20:0}$	$C_{18:1}$	C_{20}, C_{23}, C_{25}, C_{27}, C_{29}	C_{14}, C_{16}, C_{18}, C_{24}, C_{26}, C_{28}, C_{30}	-	-	β-sitosterol, cholesterol	$M_{16:0}$	-	-	-

Sample Code	Contamination	Saturated fatty acids	Unsaturated fatty acids	*n*-alkanes	Alcohols	Ketones	Wax esters	Sterols	Monoacylglycerols	Diacylglycerols	Triacylglycerols	Other
NY-4 In L2	P	$C_{6:0}$-$C_{10:0}$, $C_{16:0}$, $C_{18:0}$	$C_{16:1}$, $C_{18:1}$	C_{23}, C_{25}, C_{27}, C_{28}	C_{13}, C_{14}, C_{16}, C_{18}	-	-	β-sitosterol, cholesterol	-	-	-	-
NY-5 In L2	P	-	-	-	C_{18}	-	-	-	-	-	-	-
NY-6 In L2	P	$C_{16:0,}$ $C_{18:0}$	-	-	C_{16}, C_{18}	-	-	Cholesterol	-	-	-	-
NY-7 In L2	P	$C_{8:0}$-$C_{10:0}$, $C_{16:0}$, $C_{18:0}$	-	-	C_{12}-C_{14}, C_{16}, C_{18}	-	-	-	-	-	-	-
NY-8 E	P	$C_{14:0}$, $C_{16:0}$, $C_{18:0}$	-	C_{18}, C_{20}	C_{14}, C_{16}, C_{18}	-	-	Cholesterol	$M_{16:0}$	-	-	-
NY-8 In L2	P	$C_{16:0,}$ $C_{18:0}$	-	-	C_{16}, C_{18}, C_{28}	-	-	Cholesterol	$M_{14:0}$, $M_{16:0}$	D_{32}	-	-
NY-9 In L2	P	-	-	-	C_{16}, C_{18}	-	-	-	-	-	-	-
PEQ-1 In L2	P	-	-	-	-	-	-	-	-	-	-	-
PEQ-2 E	P	$C_{8:0}$-$C_{12:0}$, $C_{16:0}$, $C_{16:0,}$ $C_{18:0}$	-	-	C_{16}, C_{18}, C_{28}	-	-	-	-	-	-	-
PEQ-2 In L2	P	-	-	-	C_{16}, C_{18}	-	-	-	-	-	-	-
PEQ-3 In L2	P	$C_{9:0}$, $C_{16:0}$, $C_{18:0}$	$C_{18:1}$	-	C_{18}, C_{28}	-	-	-	-	-	-	-
SHH-1 E	P	$C_{7:0}$-$C_{12:0}$, $C_{14:0}$, $C_{16:0}$, $C_{18:0}$	$C_{16:1}$, $C_{18:1}$	C_{16}	C_{16}, C_{18}, C_{20}, C_{22}, C_{24}	-	WE_{36}	Cholesterol	$M_{14:0}$, $M_{15:0}$, $M_{16:0}$, $M_{18:0}$	D_{30}, D_{32}	T_{24}, T_{26}	-
SHH-1 In L2	P	$C_{14:0}$, $C_{16:0}$, $C_{18:0}$	-	C_{17}-C_{20}, C_{22}, C_{25}, C_{27}, C_{29}, C_{31}	C_{14}-C_{24}, C_{28}, C_{30}	-	-	β-sitosterol, campesterol, cholesterol, 7-ketocholesterol	$M_{14:0}$, $M_{15:0}$, $M_{16:0}$, $M_{18:0}$	D_{30}, D_{34}	T_{26}	-
SHH-10 In L2	P	$C_{9:0}$, $C_{14:0}$, $C_{16:0}$, $C_{18:0}$	-	-	C_{16}, C_{18}	-	-	-	$M_{16:0}$, $M_{18:0}$	-	-	-
SHH-11 In L2	P, COS	$C_{14:0}$, $C_{16:0}$, $C_{18:0}$	-	C_{29}	C_{16}, C_{18}, C_{20}, C_{24}, C_{28}	-	-	Cholesterol	$M_{14:0}$, $M_{15:0}$, $M_{16:0}$, $M_{18:0}$	D_{32}, D_{34}	-	-
SHH-13 In L2	P	$C_{9:0}$, $C_{16:0}$, $C_{18:0}$	$C_{18:1}$	C_{20}, C_{27}, C_{28}	C_{14}, C_{16}, C_{18}	-	-	-	$M_{18:0}$	-	-	-
SHH-17 In L2	P	$C_{16:0,}$ $C_{18:0}$	-	-	C_{16}, C_{18}	-	-	Cholesterol	$M_{8:0}$, $M_{16:0}$, $M_{18:0}$	-	T_{24}, T_{26}, T_{28}, T_{30}	-

Sample Code	Contamination	Saturated fatty acids	Unsaturated fatty acids	*n*-alkanes	Alcohols	Ketones	Wax esters	Sterols	Monoacylglycerols	Diacylglycerols	Triacylglycerols	Other
SHH-18 In L2	P, COS	$C_{16:0}$, $C_{18:0}$	-	C_{18}	C_{14}, C_{16}, C_{18}, C_{28}	-	WE_{24}	Cholesterol	$M_{16:0}$, $M_{18:0}$	-	-	-
SHH-19 In L2	P	$C_{16:0}$	-	-	C_{14}, C_{16}, C_{18}	-	-	Cholesterol	$M_{16:0}$, $M_{18:0}$	-	T_{24}, T_{26}, T_{28}, T_{30}	-
SHH-2 In L2	P	$C_{16:0}$, $C_{18:0}$	-	-	C_{16}, C_{18}	-	-	-	$M_{16:0}$, $M_{18:0}$	-	-	-
SHH-20 In L2	P	$C_{16:0}$, $C_{18:0}$	$C_{18:1}$	C_{20}, C_{27}	C_{16}, C_{18}	-	-	-	$M_{16:0}$, $M_{18:0}$	-	T_{26}, T_{32}, T_{34}, T_{36}, T_{38}, T_{40}	-
SHH-21 In L2	P, COS	$C_{16:0}$, $C_{18:0}$	-	C_{16}-C_{18}, C_{20}	C_{14}, C_{16}, C_{18}	-	-	-	$M_{16:0}$, $M_{18:0}$	-	-	-
SHH-3 In L2	P	$C_{9:0}$, $C_{10:0}$, $C_{12:0}$, $C_{16:0}$, $C_{18:0}$	-	C_{17}, C_{18}, C_{20}, C_{27}	C_{12}, C_{14}, C_{16}, C_{18}, C_{20}	-	-	Cholesterol	$M_{16:0}$, $M_{18:0}$	-	-	-
SHH-4 In L2	P, COS	$C_{16:0}$, $C_{18:0}$	-	C_{18}, C_{20}	C_{16}, C_{18}	-	WE_{36}	β-sitosterol, cholesterol	$M_{16:0}$, $M_{18:0}$	-	T_{24}, T_{26}	-
SHH-5 E	P	$C_{16:0}$, $C_{18:0}$	-	C_{14}, C_{16}-C_{18}	C_{16}, C_{18}	-	-	Cholesterol	$M_{16:0}$, $M_{18:0}$	D_{32}	-	Dehydroabiatic acid
SHH-5 In L2	P	$C_{16:0}$, $C_{18:0}$	-	C_{18}-C_{20}, C_{22}, C_{29}	C_{16}, C_{18}, C_{20}, C_{26}, C_{28}	-	-	β-sitosterol, cholesterol	$M_{16:0}$, $M_{18:0}$	-	-	-
SHH-6 In L2	P	$C_{9:0}$, $C_{13:0}$, $C_{14:0}$, $C_{16:0}$, $C_{18:0}$	$C_{18:1}$	C_{18}, C_{20}, C_{22}, C_{29}	C_{24}, C_{16}, C_{18}, C_{20}, C_{28}	-	-	Cholesterol	$M_{16:0}$, $M_{18:0}$	-	-	-
SHH-7 In L2	P, COS	$C_{14:0}$, $C_{16:0}$, $C_{18:0}$	$C_{18:1}$	C_{18}, C_{20}, C_{21}, C_{25}	C_{16}, C_{18}, C_{20}	-	-	Cholesterol	$M_{16:0}$, $M_{18:0}$	-	T_{24}, T_{26}, T_{28}, T_{30}	-
SHH-8 In L2	P, COS	$C_{16:0}$, $C_{18:0}$	-	C_{20}	C_{14}, C_{16}, C_{18}	-	-	Cholesterol	$M_{16:0}$, $M_{18:0}$	-	-	-
TH-1 In L2	P	-	-	-	C_{14}, C_{16}, C_{18}, C_{26}, C_{28}	-	-	Cholesterol	$M_{16:0}$	-	-	-
TH-2 E	P	$C_{9:0}$, $C_{14:0}$, $C_{16:0}$, $C_{18:0}$	-	C_{21}, C_{23}, C_{25}, C_{26}, C_{29}-C_{32}	C_{12}, C_{14}, C_{16}, C_{18}, C_{20}, C_{26}, C_{28}	-	-	-	$M_{14:0}$, $M_{16:0}$, $M_{18:0}$	-	-	-

Sample Code	Contamination	Saturated fatty acids	Unsaturated fatty acids	*n*-alkanes	Alcohols	Ketones	Wax esters	Sterols	Monoacylglycerols	Diacylglycerols	Triacylglycerols	Other
TH-2 In L2	P	-	-	C_{21}-C_{23}, C_{25}-C_{29}, C_{31}	C_{12}-C_{14}, C_{16}, C_{18}, C_{20}, C_{22}, C_{24}, C_{26}, C_{28}, C_{30}	K_{31}, K_{33}	-	-	$M_{16:0}$	-	-	C_7-C_{12} γ lactone
TH-4 In L2	P	$C_{9:0}$, $C_{10:0}$, $C_{12:0}$, $C_{15:0}$, $C_{16:0}$, $C_{18:0}$	$C_{18:1}$	C_{29}	C_{12}, C_{14}, C_{16}, C_{18}, C_{28}	-	-	β-sitosterol, cholesterol, stigmasterol	$M_{14:0}$, $M_{16:0}$, $M_{18:0}$, $M_{18:1}$	-	-	-
TS-1 E	P	$C_{6:0}$, $C_{8:0}$, $C_{9:0}$, $C_{15:0}$, $C_{16:0}$, $C_{18:0}$	$C_{16:1}$, $C_{18:1}$	C_{23}, C_{25}-C_{29}	C_{14}-C_{22}, C_{24}, C_{26}, C_{28}	-	-	Cholesterol	$M_{16:0}$	D_{30}, D_{32}	-	-
TS-1 In L2	P	$C_{16:0}$, $C_{18:0}$	-	C_{22}-C_{30}, C_{33}, C_{35}	C_{16}, C_{18}, C_{20}, C_{22}, C_{24}, C_{28}	-	-	Cholesterol	$M_{16:0}$	-	-	-
TS-10 E	P	$C_{6:0}$, $C_{8:0}$-$C_{10:0}$, $C_{12:0}$, $C_{14:0}$, $C_{16:0}$, $C_{18:0}$	$C_{16:1}$	C_{23}, C_{25}, C_{27}, C_{29}, C_{31}	C_{18}, C_{22}, C_{24}, C_{26}, C_{28}, C_{30}	-	WE_{40}, WE_{42}, WE_{44}	Cholesterol	$M_{16:0}$, $M_{18:0}$	-	-	-
TS-10 In L2	P	$C_{8:0}$-$C_{18:0}$	$C_{18:1}$	C_{23}-C_{25}, C_{27}-C_{29}, C_{33}	C_{18}, C_{24}, C_{26}, C_{28}	-	WE_{42}, WE_{44}	Cholesterol	-	-	-	-
TS-11 In L2	P, SS	$C_{16:0}$, $C_{18:0}$	-	C_{25}-C_{29}, C_{31}	C_{18}, C_{26}, C_{28}	-	-	-	-	-	-	-
TS-12 In L2	P	$C_{16:0}$, $C_{18:0}$	-	-	C_{18}	-	-	Cholesterol	-	-	-	-
TS-13 In L2	P	$C_{16:0}$, $C_{18:0}$	-	C_{24}-C_{27}, C_{29}, C_{31}, C_{33}	C_{16}, C_{18}, C_{26}, C_{28}	-	-	-	$M_{16:0}$, $M_{18:0}$	-	-	-
TS-16 In L2	P, SS	-	-	C_{22}, C_{25}, C_{27}, C_{29}, C_{31}, C_{33}	-	-	-	-	$M_{16:0}$	-	-	-
TS-18 In L2	P	$C_{6:0}$-$C_{10:0}$, $C_{14:0}$, $C_{16:0}$, $C_{18:0}$	-	C_{25}, C_{27}	C_{14}, C_{16}, C_{18}, C_{26}	-	-	Cholesterol	$M_{16:0}$	-	-	-
TS-2 In L2	P	$C_{16:0}$	-	C_{27}, C_{29}	C_{18}	-	-	Cholesterol	$M_{16:0}$, $M_{18:0}$	-	-	-
TS-20 In L2	P	$C_{7:0}$-$C_{10:0}$	-	C_{25}, C_{27}, C_{29}, C_{31}	C_{16}, C_{18}, C_{26}	-	-	-	-	-	-	-
TS-23 In L2	P	$C_{16:0}$, $C_{18:0}$	-	-	C_{18}	-	-	-	-	-	-	-
TS-26 In L2	P	-	-	C_{23}, C_{25}, C_{27}, C_{29}, C_{31}	C_{16}, C_{18}, C_{26}, C_{28}	-	-	-	$M_{16:0}$	-	-	-
TS-2E	P	$C_{7:0}$-$C_{9:0}$, $C_{14:0}$, $C_{16:0}$, $C_{18:0}$	$C_{16:1}$, $C_{18:1}$	C_{27}	C_{18}, C_{22}, C_{26}	-	-	-	$M_{16:0}$, $M_{18:0}$	-	-	-
TS-3 E	P	$C_{6:0}$-$C_{12:0}$, $C_{16:0}$, $C_{18:0}$	$C_{18:1}$	C_{14}, C_{16}, C_{18}, C_{24}-C_{27}, C_{29}, C_{31}, C_{33}	C_{12}-C_{14}, C_{16}, C_{18}, C_{26}, C_{28}	-	-	-	$M_{16:0}$	-	-	-
TS-3 In L2	P	$C_{16:0}$	-	C_{22}, C_{29}	C_{16}, C_{18}, C_{26}	-	-	Cholesterol	$M_{16:0}$	-	-	-

Sample Code	Contamination	Saturated fatty acids	Unsaturated fatty acids	*n*-alkanes	Alcohols	Ketones	Wax esters	Sterols	Monoacylglycerols	Diacylglycerols	Triacylglycerols	Other
TS-4 E	P	-	-	C_{25}-C_{31}, C_{33}	C_{18}, C_{24}, C_{26}, C_{28}, C_{30}, C_{32}	-	WE_{40}, WE_{42}, WE_{44}, WE_{46}	-	-	-	-	-
TS-4 In L2	P	$C_{16:0}$	-	C_{27}, C_{29}	C_{16}, C_{18}	-	-	β-sitosterol, cholesterol	$M_{16:0}$	-	-	-
TS-5 In L2	P	$C_{16:0,}$ $C_{18:0}$	-	C_{25}, C_{27}, C_{29}	C_{14}, C_{18}, C_{22}, C_{26}, C_{28}	-	-	-	-	-	-	-
TS-6 In L2	P	$C_{7:0}$-$C_{10:0}$, $C_{12:0}$, $C_{14:0}$-$C_{16:0}$, $C_{18:0}$, $C_{20:0}$	$C_{16:1}$, $C_{18:1}$	C_{20}, C_{27}, C_{29}	C_{14}, C_{16}, C_{18}, C_{20}, C_{22}	-	-	Cholesterol	$M_{16:0}$	D_{32}	-	-
TS-7 In L2	P	$C_{9:0}$, $C_{16:0}$, $C_{18:0}$	-	C_{14}, C_{24}, C_{27}, C_{31}	C_{16}, C_{18}, C_{20}, C_{26}, C_{28}, C_{30}	-	-	-	$M_{16:0}$, $M_{18:0}$, $M_{18:1}$	-	-	-
TS-8 In L2	P	$C_{16:0}$	-	C_{14}, C_{16}, C_{20}, C_{24}, C_{27}, C_{29}	C_{14}, C_{16}, C_{18}, C_{26}, C_{28}	-	-	-	$M_{16:0}$, $M_{18:0}$	D_{34}	-	-
TS-9 E	P, COS, SS	$C_{6:0}$, $C_{8:0}$-$C_{10:0}$, $C_{12:0}$, $C_{14:0}$-$C_{18:0}$, $C_{20:0}$	$C_{16:1}$, $C_{18:1}$	C_{17}, C_{18}, C_{23}, C_{25}, C_{27}, C_{29}	C_{12}, C_{14}, C_{16}, C_{18}, C_{20}, C_{22}, C_{26}, C_{28}	-	-	β-sitosterol, cholesterol	$M_{16:0}$, $M_{18:0}$	D_{30}, D_{32}, D_{34}, D_{36}	-	-
TS-9 In L2	P	$C_{16:0}$	-	C_{23}, C_{25}, C_{29}	C_{18}	-	-	-	-	-	-	-
TSF17-1 In L2	P	$C_{16:0,}$ $C_{18:0}$	$C_{18:1}$	C_{27}, C_{29}	C_{18}	-	-	-	-	-	-	-
TSF17-10 E	P	$C_{6:0}$-$C_{9:0}$, $C_{16:0}$, $C_{18:0}$	-	-	C_{18}	-	-	-	$M_{16:0}$, $M_{18:0}$	-	-	-
TSF17-10 In L2	P	$C_{16:0,}$ $C_{18:0}$	$C_{18:1}$	C_{32}	C_{18}	-	-	-	-	-	-	-
TSF17-12 In L2	P	$C_{6:0}$-$C_{18:0}$	$C_{18:1}$	-	C_{18}	-	-	-	-	-	-	-
TSF17-13 In L2	P	$C_{6:0}$-$C_{18:0}$	-	-	C_{18}	-	-	-	-	-	-	-
TSF17-15 E	P	-	-	-	C_{18}	-	-	-	-	-	-	-
TSF17-15 In L2	P	-	-	-	C_{18}	-	-	Cholesterol	-	-	-	-
TSF17-16 E	P	-	-	-	C_{18}, C_{26}, C_{28}	-	-	-	-	-	-	-

Sample Code	Contamination	Saturated fatty acids	Unsaturated fatty acids	*n*-alkanes	Alcohols	Ketones	Wax esters	Sterols	Monoacylglycerols	Diacylglycerols	Triacylglycerols	Other
TSF17-16 In L2	P	-	-	-	C_{18}, C_{24}, C_{26}, C_{28}, C_{30}	-	-	Cholesterol	-	-	-	-
TSF17-18 In L2	P	$C_{16:0}$, $C_{18:0}$	-	C_{23}-C_{30}	C_{18}	-	-	Cholesterol	-	-	-	-
TSF17-19 E	COS	$C_{7:0}$-$C_{9:0}$, $C_{16:0}$, $C_{18:0}$	$C_{18:1}$	C_{29}, C_{31}	C_{18}, C_{28}	-	-	-	-	-	-	-
TSF17-19 In L2	P, COS	$C_{16:0}$, $C_{18:0}$	-	-	C_{18}	-	-	Cholesterol	-	-	-	-
TSF17-2 In L2	P	$C_{16:0}$, $C_{18:0}$	-	C_{23}-C_{27}, C_{29}-C_{31}	C_{18}, C_{28}, C_{30}	-	-	β-sitosterol	-	-	-	-
TSF17-21 In L2	P	$C_{7:0}$-$C_{18:0}$	-	-	C_{18}, C_{26}, C_{28}, C_{30}	-	-	-	-	-	-	-
TSF17-22 E	P	$C_{8:0}$-$C_{10:0}$, $C_{12:0}$, $C_{14:0}$-$C_{24:0}$, $C_{26:0}$	$C_{18:1}$	C_{16}, C_{29}	C_{18}, C_{26}, C_{28}, C_{30}	-	-	Cholesterol	$M_{16:0}$, $M_{18:0}$	-	-	-
TSF17-22 In L2	P	$C_{8:0}$-$C_{20:0}$	$C_{16:1}$, $C_{18:1}$, $C_{20:1}$	-	C_{18}	K_{31}, K_{33}, K_{35}	-	Cholesterol	-	-	-	-
TSF17-23 E	P	$C_{16:0}$, $C_{18:0}$	-	-	C_{18}, C_{22}, C_{24}, C_{26}-C_{28}, C_{30}	-	-	-	-	-	-	-
TSF17-23 In L2	P	$C_{16:0}$, $C_{18:0}$	-	-	C_{16}, C_{18}, C_{24}, C_{26}-C_{28}, C_{30}	K_{31}, K_{33}, K_{35}	-	Cholesterol	-	-	-	-
TSF17-25 In L2	P	$C_{16:0}$, $C_{18:0}$	-	C_{25}, C_{27}, C_{29}-C_{31}	C_{18}, C_{22}, C_{24}, C_{26}-C_{28}, C_{30}	-	-	-	-	-	-	-
TSF17-26 In L2	P	$C_{16:0}$	-	-	C_{18}	-	-	-	-	-	-	-
TSF17-27 E	P	$C_{6:0}$, $C_{8:0}$, $C_{9:0}$, $C_{16:0}$, $C_{18:0}$, $C_{20:0}$	-	C_{29}, C_{31}	C_{12}, C_{14}, C_{16}, C_{18}, C_{20}, C_{22}, C_{26}, C_{28}, C_{30}	-	-	-	$M_{16:0}$, $M_{18:0}$	-	-	-
TSF17-27 In L2	P	$C_{16:0}$, $C_{18:0}$	-	C_{29}, C_{31}, C_{32}	C_{18}, C_{26}, C_{28}, C_{30}	-	-	-	-	D_{28}, D_{32}, D_{34}	-	-

Sample Code	Contamination	Saturated fatty acids	Unsaturated fatty acids	*n*-alkanes	Alcohols	Ketones	Wax esters	Sterols	Monoacylglycerols	Diacylglycerols	Triacylglycerols	Other
TSF17-28 E	P	$C_{14:0}$-$C_{22:0}$	$C_{16:1}$, $C_{18:1}$, $C_{20:1}$	-	C_{24}, C_{26}-C_{28}, C_{30}	K_{31}, K_{33}, K_{35}	-	-	-	-	-	-
TSF17-28 In L2	P	$C_{16:0}$-$C_{18:0}$	$C_{18:1}$	-	-	K_{31}, K_{33}, K_{35}	-	-	-	-	-	-
TSF17-29 E	P	$C_{16:0,}$ $C_{18:0}$	-	-	C_{18}, C_{28}	K_{31}, K_{33}, K_{35}	-	-	-	-	-	-
TSF17-29 In L2	P	$C_{16:0,}$ $C_{18:0}$	-	-	C_{18}, C_{28}	K_{33}, K_{35}	-	Cholesterol	-	-	-	-
TSF17-3 In L2	P	-	-	-	-	-	-	-	-	-	-	-
TSF17-30 E	P	-	-	-	C_{16}, C_{18}, C_{22}, C_{24}, C_{26}-C_{28}, C_{30}	-	-	-	-	-	-	-
TSF17-30 In L2	P	-	-	-	C_{28}, C_{30}	-	-	Cholesterol	-	-	-	-
TSF17-31 E	P	-	-	-	C_{24}, C_{26}, C_{28}, C_{30}	-	-	-	-	-	-	-
TSF17-31 In L2	P	-	-	-	C_{28}, C_{30}	-	-	Cholesterol	-	-	-	-
TSF17-32 E	P	-	-	-	C_{18}, C_{28}	-	-	-	-	-	-	-
TSF17-32 In L2	P	$C_{16:0,}$ $C_{18:0}$	-	-	C_{28}	K_{31}, K_{33}, K_{35}	-	Cholesterol	-	-	-	-
TSF17-32 S	P	$C_{16:0,}$ $C_{18:0}$	-	C_{25}, C_{27}-C_{29}, C_{31}	C_{26}, C_{28}, C_{30}	-	-	Stigmasterol	-	-	-	-
TSF17-4 E	P	-	-	-	C_{18}, C_{26}, C_{28}, C_{30}	-	-	-	-	-	-	-
TSF17-4 In L2	P	$C_{16:0}$	-	-	C_{28}	-	-	Cholesterol	-	-	-	-
TSF17-5 In L2	P, COS	$C_{7:0}$-$C_{18:0}$	$C_{18:1}$	C_{25}, C_{27}, C_{29}	C_{18}, C_{28}	-	-	-	-	-	-	-

Sample Code	Contamination	Saturated fatty acids	Unsaturated fatty acids	*n*-alkanes	Alcohols	Ketones	Wax esters	Sterols	Monoacylglycerols	Diacylglycerols	Triacylglycerols	Other
TSF17-6 In L2	P	$C_{8:0}$-$C_{26:0}$	$C_{16:1}$, $C_{18:1}$	C_{25}-C_{30}	C_{18}, C_{28}	K_{29}, K_{31}, K_{33}, K_{35}	-	-	-	-	-	-
TSF17-7 In L2	P, COS	$C_{16:0}$, $C_{18:0}$	-	C_{24}-C_{33}	C_{18}, C_{22}, C_{24}, C_{26}, C_{28}, C_{30}	-	-	-	-	-	-	-
TSF17-8 In L2	P	-	-	-	-	-	-	-	-	-	-	-
TSF17-9 In L2	P, SS	$C_{16:0}$, $C_{18:0}$	-	C_{25}-C_{29}	C_{18}	-	-	Cholesterol	-	-	-	-
TSF18-1 In L2	P	$C_{16:0}$, $C_{18:0}$	-	C_{32}	C_{26}, C_{28}	-	-	-	$M_{16:0}$, $M_{18:0}$	-	-	-
TSF18-10 In L2	P	$C_{16:0}$, $C_{18:0}$	-	C_{29}, C_{31}, C_{32}	C_{18}, C_{26}, C_{28}	-	-	-	-	-	-	-
TSF18-11 In L2	P	-	-	C_{32}	-	-	-	-	$M_{16:0}$, $M_{18:0}$	-	-	-
TSF18-13 In L2	P	$C_{14:0}$-$C_{18:0}$, $C_{20:0}$	$C_{18:1}$	-	C_{28}	K_{33}, K_{35}	-	-	-	-	-	-
TSF18-14 In L2	P	$C_{16:0}$, $C_{18:0}$	-	C_{27}, C_{29}, C_{31}	C_{26}, C_{28}	-	-	-	-	-	-	-
TSF18-15 In L2	P	-	-	-	-	-	-	-	-	-	-	-
TSF18-16 In L2	P	$C_{16:0}$, $C_{18:0}$	-	C_{27}, C_{29}, C_{31}	C_{26}, C_{28}, C_{30}	-	-	-	-	-	-	-
TSF18-17 In L2	P	$C_{16:0}$, $C_{18:0}$	$C_{18:1}$	-	C_{18}, C_{28}	-	-	-	-	-	-	-
TSF18-18 In L2	P	$C_{16:0}$, $C_{18:0}$	-	C_{27}, C_{29}, C_{31}	C_{18}, C_{26}, C_{28}	-	-	-	-	-	-	-

Sample Code	Contamination	Saturated fatty acids	Unsaturated fatty acids	*n*-alkanes	Alcohols	Ketones	Wax esters	Sterols	Monoacylglycerols	Diacylglycerols	Triacylglycerols	Other
TSF18-19 In L2	P	$C_{16:0}$, $C_{18:0}$	-	C_{32}	C_{26}, C_{28}	-	-	Cholesterol	$M_{16:0}$, $M_{18:0}$	-	-	-
TSF18-2 In L2	P	$C_{16:0}$, $C_{18:0}$, $C_{20:0}$	$C_{18:1}$	-	C_{18}	-	-	-	-	-	-	-
TSF18-20 E	P	$C_{15:0}$, $C_{16:0}$, $C_{18:0}$	-	C_{21}, C_{27}, C_{29}, C_{31}, C_{33}	C_{18}, C_{16}, C_{28}, C_{30}	-	-	-	$M_{16:0}$, $M_{18:0}$	-	-	-
TSF18-20 In L2	P	$C_{12:0}$-$C_{22:0}$	$C_{16:1}$, $C_{18:1}$	C_{27}-C_{29}, C_{31}	C_{24}, C_{26}-C_{28}, C_{30}	-	-	Cholesterol	-	-	-	-
TSF18-21 In L2	P	-	-	C_{29}, C_{31}, C_{32}	C_{24}, C_{26}, C_{28}, C_{30}	-	-	-	-	-	-	-
TSF18-22 In L2	P	$C_{14:0}$, $C_{16:0}$-$C_{18:0}$, $C_{20:0}$	$C_{18:1}$	-	C_{18}	-	-	-	-	-	-	-
TSF18-23 In L2	P	$C_{16:0}$, $C_{18:0}$	$C_{18:1}$	-	C_{18}	-	-	-	-	-	-	-
TSF18-24 In L2	-	$C_{10:0}$-$C_{23:0}$	$C_{16:1}$, $C_{18:1}$	C_{27}, C_{29}, C_{31}	C_{18}, C_{24}, C_{26}, C_{28}, C_{30}	-	-	-	-	-	-	-
TSF18-25 In L2	P	$C_{12:0}$, $C_{14:0}$-$C_{22:0}$	$C_{18:1}$	C_{27}, C_{29}, C_{31}	C_{18}, C_{24}, C_{26}, C_{28}	-	-	-	-	-	-	-
TSF18-26 In L2	P	$C_{16:0}$, $C_{18:0}$	-	-	C_{18}	-	-	-	$M_{16:0}$, $M_{18:0}$	-	-	-
TSF18-27 In L2	P	$C_{16:0}$-$C_{20:0}$	$C_{18:1}$	-	-	-	-	-	$M_{16:0}$, $M_{18:0}$	D_{32}	-	-
TSF18-28 In L2	P	$C_{14:0}$, $C_{16:0}$-$C_{18:0}$	$C_{18:1}$	-	C_{18}	K_{31}, K_{33}, K_{35}	-	-	-	-	-	-
TSF18-29 In L2	P	$C_{14:0}$-$C_{18:0}$	$C_{18:1}$	-	C_{18}, C_{28}	K_{31}, K_{33}, K_{35}	-	-	-	-	-	-
TSF18-3 E	P	$C_{14:0}$-$C_{16:0}$	-	C_{14}, C_{16}, C_{27}, C_{29}, C_{31}, C_{33}	C_{18}, C_{26}, C_{28}, C_{30}	-	-	-	$M_{16:0}$, $M_{18:0}$	-	-	-

Sample Code	Contamination	Saturated fatty acids	Unsaturated fatty acids	*n*-alkanes	Alcohols	Ketones	Wax esters	Sterols	Monoacylglycerols	Diacylglycerols	Triacylglycerols	Other
TSF18-3 In L2	P	$C_{6:0}$-$C_{18:0}$	$C_{18:1}$	C_{29}, C_{31}	C_{18}, C_{26}, C_{28}, C_{30}	-	-	Cholesterol	-	-	-	-
TSF18-30 In L2	P	$C_{16:0,}$ $C_{18:0}$	-	-	-	-	-	-	$M_{16:0}$, $M_{18:0}$	-	-	-
TSF18-31 In L2	P	$C_{16:0,}$ $C_{18:0}$	$C_{18:1}$	-	C_{18}	-	-	-	-	-	-	-
TSF18-34 In L2	P	$C_{14:0}$-$C_{20:0}$	$C_{16:1}$, $C_{18:1}$	C_{29}	C_{18}, C_{28}	K_{31}, K_{33}, K_{35}	WE_{42}, WE_{44}	-	$M_{16:0}$	-	-	-
TSF18-35 In L2	-	$C_{14:0}$-$C_{18:0}$, $C_{20:0}$, $C_{22:0}$, $C_{24:0}$, $C_{26:0}$, $C_{28:0}$	$C_{18:1}$	C_{27}, C_{29}, C_{31}	C_{24}, C_{26}, C_{28}, C_{30}	-	WE_{42}, WE_{44}, WE_{46}, WE_{48}, WE_{50}	-	-	-	-	-
TSF18-36 In L2	P, COS	$C_{13:0}$-$C_{18:0}$, $C_{20:0}$	$C_{16:1}$, $C_{18:1}$	-	C_{18}, C_{28}, C_{30}	K_{29}, K_{31}, K_{33}, $K_{33:1}$, K_{34}, K_{35}, $K_{35:1}$	-	Cholesterol	-	-	-	-
TSF18-37 In L2	P	$C_{16:0}$, $C_{18:0}$, $C_{20:0}$	$C_{18:1}$	C_{29}, C_{31}	C_{18}, C_{28}	-	-	-	$M_{16:0}$	-	-	-
TSF18-39 In L2	P	$C_{16:0}$-$C_{18:0}$, $C_{20:0}$	$C_{18:1}$	-	C_{24}, C_{26}, C_{28}	-	-	-	-	-	-	-
TSF18-4 E	P	$C_{16:0,}$ $C_{18:0}$	-	C_{12}, C_{14}, C_{16}, C_{18}, C_{23}, C_{27}, C_{29}, C_{31}, C_{33}	C_{18}, C_{22}, C_{26}, C_{28}	-	WE_{42}, WE_{44}	-	$M_{16:0}$, $M_{18:0}$	-	-	-
TSF18-4 In L2	P	$C_{16:0,}$ $C_{18:0}$	$C_{18:1}$	C_{32}	C_{18}, C_{24}, C_{26}, C_{28}	-	-	Cholesterol	-	-	-	-
TSF18-40 In L2	P	$C_{14:0}$-$C_{20:0}$, $C_{22:0}$	$C_{16:1}$, $C_{18:1}$	C_{29}	C_{18}, C_{26}, C_{28}, C_{30}	-	-	Cholesterol	-	-	-	-
TSF18-42 In L2	P	$C_{16:0,}$ $C_{18:0}$	-	C_{21}	C_{18}, C_{28}	-	-	-	$M_{16:0}$	-	-	-
TSF18-44 In L2	P	$C_{14:0}$, $C_{16:0}$-$C_{18:0}$, $C_{20:0}$, $C_{26:0}$, $C_{28:0}$, $C_{30:0}$	$C_{18:1}$	C_{27}, C_{29}, C_{31}, C_{33}	C_{18}, C_{26}, C_{28}, C_{30}	-	-	-	-	-	-	-

Sample Code	Contamination	Saturated fatty acids	Unsaturated fatty acids	*n*-alkanes	Alcohols	Ketones	Wax esters	Sterols	Monoacylglycerols	Diacylglycerols	Triacylglycerols	Other
TSF18-45 In L2	P	$C_{16:0}$, $C_{18:0}$, $C_{20:0}$	-	C_{27}, C_{29}, C_{31}, C_{33}	C_{18}, C_{26}, C_{28}, C_{30}	-	-	-	-	-	-	-
TSF18-46 E	P	$C_{14:0}$, $C_{16:0}$-$C_{18:0}$	$C_{18:1}$	C_{27}, C_{29}, C_{31}	C_{18}, C_{24}, C_{26}, C_{28}, C_{30}	-	WE_{44}, WE_{46}	-	-	-	-	-
TSF18-46 In L2	P	$C_{16:0,}$ $C_{18:0}$	-	C_{29}, C_{31}	C_{18}, C_{28}	-	-	-	-	-	-	-
TSF18-47 In L2	P	$C_{16:0,}$ $C_{18:0}$	-	C_{29}, C_{31}-C_{33}	C_{18}	-	-	-	-	-	-	-
TSF18-48 In L2	P	$C_{14:0}$, $C_{16:0}$, $C_{18:0}$, $C_{20:0}$	$C_{18:1}$	-	C_{18}, C_{28}	-	-	-	-	-	-	-
TSF18-49 In L2	P	-	-	-	C_{18}	-	-	-	-	-	-	-
TSF18-5 In L2	P	$C_{16:0,}$ $C_{18:0}$	-	C_{29}, C_{31}	C_{18}, C_{26}, C_{28}, C_{30}	-	-	β-sitosterol	-	-	-	-
TSF18-50 In L2	P	$C_{16:0}$, $C_{18:0}$, $C_{24:0}$, $C_{26:0}$, $C_{28:0}$, $C_{30:0}$	-	C_{25}, C_{27}, C_{29}, C_{31}, C_{33}	C_{24}, C_{26}, C_{28}, C_{30}	-	-	-	-	-	-	-
TSF18-51 E	P	$C_{12:0}$, $C_{14:0}$-$C_{18:0}$	-	C_{14}, C_{23}, C_{27}, C_{29}, C_{31}, C_{33}	C_{18}, C_{26}, C_{28}, C_{30}	K_{31}, K_{33}, K_{35}	-	-	-	-	-	-
TSF18-51 In L2	P	$C_{12:0}$, $C_{14:0}$-$C_{20:0}$	$C_{18:1}$, $C_{20:1}$	C_{31}	C_{18}, C_{26}, C_{28}	-	-	-	$M_{16:0}$, $M_{18:0}$	-	-	-
TSF18-52 In L2	P	$C_{16:0,}$ $C_{18:0}$	-	-	C_{18}	-	-	-	-	-	-	-
TSF18-54 E	P	$C_{8:0}$, $C_{9:0}$, $C_{16:0}$, $C_{18:0}$	$C_{18:1}$	C_{14}, C_{16}, C_{27}, C_{29}	C_{18}, C_{26}, C_{28}	-	-	Cholesterol	-	-	-	-
TSF18-54 In L2	P, COS	-	-	C_{27}, C_{29}, C_{31}	C_{18}, C_{26}, C_{28}	-	-	-	-	-	-	-
TSF18-55 In L2	P	$C_{9:0}$, $C_{16:0}$, $C_{18:0}$	-	C_{29}, C_{31}	C_{18}, C_{26}, C_{28}	-	-	-	$M_{16:0}$	-	-	-

Sample Code	Contamination	Saturated fatty acids	Unsaturated fatty acids	*n*-alkanes	Alcohols	Ketones	Wax esters	Sterols	Monoacylglycerols	Diacylglycerols	Triacylglycerols	Other
TSF18-56 In L2	P	$C_{16:0}$, $C_{18:0}$	$C_{18:1}$	C_{29}	C_{28}, C_{30}	-	-	-	-	-	-	-
TSF18-57 Cr	P	$C_{16:0}$, $C_{18:0}$	-	C_{23}, C_{25}-C_{33}	C_{20}-C_{28}, C_{30}, C_{32}, C_{34}	K_{31}	WE_{40}, WE_{42}, WE_{44}, WE_{46}, WE_{48}	-	-	-	-	-
TSF18-57 E	P	$C_{16:0}$, $C_{18:0}$, $C_{28:0}$	-	C_{14}, C_{23}, C_{25}-C_{27}, C_{29}-C_{31}, C_{33}	C_{22}, C_{24}, C_{26}-C_{28}, C_{30}	-	-	Stigmasterol	-	-	-	-
TSF18-57 In L2	P	$C_{9:0}$, $C_{10:0}$, $C_{15:0}$, $C_{16:0}$, $C_{18:0}$, $C_{20:0}$, $C_{22:0}$, $C_{24:0}$, $C_{26:0}$, $C_{28:0}$	$C_{18:1}$	C_{23}, C_{25}, C_{27}, C_{29}, C_{31}	C_{18}, C_{20}, C_{22}-C_{24}, C_{26}-C_{28}, C_{30}	K_{31}	WE_{40}, WE_{42}, WE_{44}, WE_{46}	Stigmasterol	$M_{16:0}$, $M_{18:0}$	-	-	-
TSF18-58 In L2	P	$C_{16:0}$, $C_{18:0}$	$C_{18:1}$	-	C_{18}, C_{28}	K_{31}, K_{33}, K_{35}	-	-	-	-	-	-
TSF18-59 In L2	P	$C_{14:0}$, $C_{16:0}$-$C_{18:0}$, $C_{20:0}$	$C_{18:1}$	-	C_{18}, C_{28}	-	-	-	-	-	-	-
TSF18-6 In L2	P	$C_{16:0}$, $C_{18:0}$, $C_{20:0}$	$C_{18:1}$	C_{32}	C_{26}, C_{28}	-	-	-	$M_{16:0}$, $M_{18:0}$	-	-	-
TSF18-60 In L2	P	$C_{16:0}$, $C_{18:0}$	-	-	-	-	-	-	-	-	-	-
TSF18-62 In L2	P	$C_{16:0}$, $C_{18:0}$	-	C_{29}, C_{31}	C_{28}, C_{30}	-	-	-	-	-	-	-
TSF18-65 E	P	$C_{16:0}$, $C_{18:0}$	-	C_{27}, C_{29}, C_{31}, C_{33}	C_{18}, C_{26}, C_{28}, C_{30}	-	WE_{44}, WE_{46}, WE_{48}	-	$M_{16:0}$, $M_{18:0}$	-	-	-
TSF18-65 In L2	P	$C_{16:0}$, $C_{18:0}$	-	C_{25}, C_{27}, C_{29}	C_{18}, C_{24}, C_{26}-C_{28}, C_{30}	-	-	Cholesterol	-	-	-	-
TSF18-66 E	P	-	-	C_{23}, C_{27}, C_{29}, C_{31}	C_{18}, C_{26}, C_{28}, C_{30}	-	-	-	$M_{16:0}$, $M_{18:0}$	-	-	-
TSF18-66 In L2	P	-	-	C_{23}, C_{25}, C_{27}, C_{29}, C_{31}	C_{18}, C_{24}, C_{26}, C_{28}, C_{30}	-	-	-	$M_{16:0}$, $M_{18:0}$	-	-	-
TSF18-67 In L2	P	$C_{9:0}$, $C_{10:0}$, $C_{14:0}$, $C_{16:0}$, $C_{18:0}$, $C_{20:0}$-$C_{22:0}$, $C_{24:0}$, $C_{26:0}$	$C_{18:1}$	C_{29}, C_{31}	C_{18}, C_{26}, C_{28}	-	-	-	-	-	-	-

Sample Code	Contamination	Saturated fatty acids	Unsaturated fatty acids	*n*-alkanes	Alcohols	Ketones	Wax esters	Sterols	Monoacylglycerols	Diacylglycerols	Triacylglycerols	Other
TSF18-68 In L2	P, COS	$C_{16:0}$, $C_{18:0}$	$C_{18:1}$	-	C_{18}, C_{28}	-	-	-	-	-	-	-
TSF18-69 In L2	P	$C_{9:0}$-$C_{19:0}$	$C_{16:1}$, $C_{18:1}$	-	C_{16}, C_{28}, C_{30}	K_{31}, K_{33}, K_{34}, K_{35}, $K_{35:1}$	-	-	$M_{14:0}$, $M_{15:0}$, $M_{16:0}$, $M_{17:0}$, $M_{18:0}$, $M_{18:1}$	-	-	-
TSF18-7 In L2	P	$C_{9:0}$, $C_{16:0}$, $C_{18:0}$	$C_{18:1}$	C_{27}, C_{29}	C_{18}, C_{24}, C_{26}-C_{28}, C_{30}	-	WE_{38}, WE_{40}, WE_{42}, WE_{44}, WE_{46}	Cholesterol	-	-	-	-
TSF18-70 In L2	P	$C_{16:0}$, $C_{18:0}$	-	C_{29}, C_{31}	C_{18}, C_{24}, C_{26}, C_{28}, C_{30}	-	-	-	-	-	-	-
TSF18-71 In L2	P	$C_{9:0}$, $C_{10:0}$, $C_{12:0}$, $C_{14:0}$-$C_{24:0}$	$C_{18:1}$	C_{21}, C_{25}, C_{27}, C_{29}, C_{31}	C_{18}, C_{22}, C_{24}, C_{26}, C_{28}, C_{30}	-	-	Stigmasterol	$M_{16:0}$, $M_{18:0}$	-	-	-
TSF18-73 In L2	P	-	-	C_{27}, C_{29}, C_{31}	C_{28}, C_{30}	-	-	-	-	-	-	-
TSF18-74 In L2	P	$C_{6:0}$, $C_{8:0}$-$C_{10:0}$, $C_{16:0}$, $C_{18:0}$	-	C_{29}, C_{31}	C_{26}, C_{28}, C_{30}	-	-	-	-	-	-	-
TSF18-75 In L2	P	$C_{16:0}$	-	C_{27}, C_{29}	C_{18}, C_{24}, C_{26}, C_{28}, C_{30}	-	-	Cholesterol	$M_{16:0}$, $M_{18:0}$	-	-	-
TSF18-77 In L2	P	$C_{16:0}$, $C_{18:0}$	-	-	C_{28}, C_{30}	-	-	-	-	-	-	-
TSF18-78 In L2	P	$C_{16:0}$, $C_{18:0}$, $C_{20:0}$, $C_{22:0}$	-	C_{27}, C_{29}, C_{31}	C_{28}, C_{30}	-	-	-	-	-	-	-
TSF18-8 In L2	P	$C_{16:0}$, $C_{18:0}$	-	C_{32}	C_{18}, C_{28}	-	-	-	-	-	-	-
TSF18-9 In L2	P	$C_{18:0}$	-	-	C_{18}	-	-	-	-	-	-	-
TSF19-1 In L2	P	$C_{16:0}$, $C_{18:0}$	-	-	-	-	-	-	-	-	-	-

Sample Code	Contamination	Saturated fatty acids	Unsaturated fatty acids	*n*-alkanes	Alcohols	Ketones	Wax esters	Sterols	Monoacylglycerols	Diacylglycerols	Triacylglycerols	Other
TSF19-13 In L2	P, COS	$C_{14:0}$, $C_{16:0}$, $C_{18:0}$	$C_{18:1}$	C_{23}, C_{25}, C_{27}-C_{29}, C_{31}	C_{18}, C_{26}, C_{28}, C_{30}	K_{31}, K_{33}, K_{35}	-	-	$M_{16:0}$	-	-	-
TSF19-14 In L2	-	$C_{16:0}$	-	-	-	-	-	-	$M_{16:0}$, $M_{18:0}$	-	-	-
TSF19-2 In L2	P	$C_{16:0}$, $C_{18:0}$, $C_{20:0}$	$C_{18:1}$	C_{25}, C_{27}, C_{29}, C_{31}	C_{18}, C_{22}, C_{24}, C_{26}, C_{28}	-	-	-	-	-	-	-
TSF19-3 In L2	P	$C_{14:0}$, $C_{16:0}$-$C_{18:0}$	$C_{18:1}$	C_{23}, C_{29}, C_{31}	C_{18}, C_{24}, C_{26}, C_{28}, C_{30}	K_{31}, K_{33}, K_{35}	WE_{42}, WE_{44}, WE_{46}, WE_{48}	-	-	-	-	Dehydroabiatic acid
TSF19-4 In L2	P	$C_{7:0}$-$C_{18:0}$	$C_{18:1}$	C_{27}, C_{29}, C_{31}	C_{24}, C_{26}, C_{28}	-	-	-	-	-	-	Sebacic acid
TSF19-5 In L2	P	$C_{18:0}$	-	C_{27}, C_{29}, C_{31}	C_{28}	-	-	-	-	-	-	-
TSF19-6 In L2	P	$C_{9:0}$, $C_{16:0}$	-	C_{17}, C_{24}-C_{31}	C_{12}, C_{14}, C_{16}, C_{18}, C_{20}, C_{22}, C_{26}, C_{28}, C_{30}	K_{31}, K_{33}, K_{35}	-	β-sitosterol	$M_{16:0}$	-	-	Dehydroabiatic acid
TSF19-7 In L2	P	$C_{16:0}$, $C_{18:0}$	-	C_{14}, C_{16}, C_{18}, C_{20}, C_{27}, C_{30}	C_{26}, C_{28}, C_{30}	-	-	-	-	-	-	-
TSF19-9 In L2	P, SS	$C_{16:0}$, $C_{18:0}$	-	-	C_{28}, C_{30}	-	-	-	-	-	-	-
YEH-1 E	P, SS	$C_{16:0}$, $C_{18:0}$	-	C_{18}, C_{20}-C_{24}, C_{26}, C_{29}	C_{14}, C_{16}, C_{18}, C_{20}, C_{26}, C_{28}	-	WE_{34}	-	$M_{14:0}$, $M_{16:0}$, $M_{18:0}$	-	-	-
YEH-1 In L2	P, SS	$C_{12:0}$, $C_{14:0}$, $C_{16:0}$, $C_{18:0}$	-	C_{17}, C_{18}, C_{20}, C_{24}-C_{31}	C_{12}-C_{16}, C_{18}, C_{20}, C_{22}	-	WE_{36}	Cholesterol	$M_{14:0}$, $M_{16:0}$, $M_{18:0}$	D_{32}	T_{26}	-
YV-10 In L2	P, SS	$C_{16:0}$	-	-	C_{18}	-	-	Cholesterol	-	-	-	-
YV-11 In L2	P	$C_{16:0}$, $C_{18:0}$	-	-	C_{18}, C_{24}, C_{26}, C_{28}	K_{31}, K_{33}, K_{35}	-	Cholesterol	-	-	-	-
YV-11e	P, SS	$C_{16:0}$, $C_{18:0}$	-	-	C_{26}, C_{28}	-	-	Cholesterol	-	-	-	-
YV-6 E	P	$C_{15:0}$-$C_{18:0}$	-	-	C_{18}	-	-	Cholesterol	-	-	-	-

Sample Code	Contamination	Saturated fatty acids	Unsaturated fatty acids	*n*-alkanes	Alcohols	Ketones	Wax esters	Sterols	Monoacylglycerols	Diacylglycerols	Triacylglycerols	Other
YV-6 In L2	P	$C_{6:0}$-$C_{22:0}$	-	-	-	-	-	β-sitosterol	-	-	-	-
YV-7 In L2	P	$C_{16:0,}$ $C_{18:0}$	-	C_{24}, C_{25}, C_{27}-C_{29}	C_{18}	-	-	Cholesterol	-	-	-	-
YV-8 E	P	$C_{6:0}$-$C_{18:0}$	-	-	C_{18}, C_{22}, C_{28}	-	-	Cholesterol, 7-ketocholesterol	-	-	-	-
YV-8 In L2	P	$C_{16:0}$	-	-	C_{16}, C_{18}, C_{22}, C_{24}	-	-	Cholesterol	-	-	-	-
YV-9 E	P	$C_{9:0}$-$C_{18:0}$	-	-	C_{16}, C_{18}, C_{22}, C_{24}	-	-	Cholesterol	-	-	-	-
YV-9 In L2	P	$C_{16:0,}$ $C_{18:0}$	-	-	C_{16}, C_{18}	-	-	Cholesterol	-	-	-	-

Appendix F. Summary of the lipid profile in the acid fraction obtained for the archaeological samples tested in this study after saponification of the ceramic powder (COS=cyclic octaatomic sulfur; Cr=crust; $C_{x:y}$=fatty acid with chain length X and Y number of double bonds; E=exterior; In=interior; L2=Layer 2; P=plastic; S=soil).

Sample Code	Contamination	Saturated fatty acids	Unsaturated fatty acids	Branched-chain fatty acids	Dicarboxylic acids	Sulfur-heterocyclic fatty acids	Terpenoids
AG-1 In L2	P, COS	$C_{16:0}$, $C_{18:0}$	$C_{18:1}$	-	-	-	-
AG-11 In L2	P	$C_{16:0}$-$C_{30:0}$	$C_{18:1}$, $C_{20:1}$, $C_{22:1}$	$C_{17:0}$	-	-	-
AG-13 In L2	P	$C_{16:0}$, $C_{18:0}$, $C_{20:0}$, $C_{22:0}$, $C_{24:0}$, $C_{26:0}$	$C_{18:1}$	-	-	-	-
AG-15 In L2	P, COS	$C_{16:0}$, $C_{18:0}$	$C_{18:1}$	-	-	-	-
AG-16 In L2	P	$C_{14:0}$-$C_{18:0}$, $C_{20:0}$, $C_{22:0}$, $C_{24:0}$, $C_{26:0}$, $C_{28:0}$	$C_{16:1}$, $C_{18:1}$	$C_{15:0}$, $C_{16:0}$	-	-	-
AG-18 In L2	P, COS	$C_{12:0}$, $C_{14:0}$-$C_{18:0}$, $C_{20:0}$, $C_{22:0}$, $C_{24:0}$, $C_{26:0}$, $C_{28:0}$	$C_{16:1}$, $C_{18:1}$	$C_{15:0}$, $C_{16:0}$	-	-	-
AG-19 In L2	P	$C_{15:0}$-$C_{18:0}$, $C_{20:0}$-$C_{26:0}$, $C_{28:0}$	$C_{16:1}$, $C_{18:1}$	$C_{16:0}$	-	-	-
AG-2 In L2	P, COS	$C_{14:0}$-$C_{18:0}$	$C_{18:1}$, $C_{18:2}$	$C_{15:0}$	-	-	-
AG-21 In L2	P, COS	$C_{16:0}$, $C_{18:0}$, $C_{24:0}$, $C_{26:0}$	$C_{18:1}$	-	-	-	-
AG-22 In L2	P, COS	$C_{16:0}$, $C_{18:0}$, $C_{20:0}$, $C_{22:0}$, $C_{24:0}$	$C_{18:1}$	-	-	-	-
AG-23 In L2	P, COS	$C_{14:0}$-$C_{18:0}$, $C_{20:0}$, $C_{22:0}$, $C_{24:0}$, $C_{26:0}$, $C_{28:0}$	$C_{16:1}$, $C_{18:1}$	$C_{15:0}$-$C_{18:0}$	-	-	-
AG-24 In L2	P, COS	$C_{16:0}$, $C_{18:0}$, $C_{20:0}$, $C_{22:0}$	$C_{18:1}$	-	-	-	-
AG-25 In L2	P, COS	$C_{14:0}$-$C_{26:0}$	$C_{18:1}$	-	-	-	Dehydroabiatic acid
AG-26 In L2	P	$C_{12:0}$, $C_{14:0}$-$C_{26:0}$, $C_{28:0}$	$C_{16:1}$, $C_{18:1}$	$C_{15:0}$-$C_{17:0}$	-	-	-
AG-28 In L2	P, COS	$C_{8:0}$, $C_{9:0}$, $C_{12:0}$, $C_{14:0}$-$C_{18:0}$, $C_{20:0}$	$C_{16:1}$, $C_{18:1}$	$C_{15:0}$, $C_{16:0}$	-	-	-
AG-30 In L2	P	$C_{13:0}$-$C_{18:0}$, $C_{20:0}$, $C_{24:0}$, $C_{26:0}$	$C_{18:1}$	$C_{15:0}$, $C_{16:0}$	-	-	β-pimaric acid, pimaric acid, methyl abietate, methyl dehydroabietate, dehydroabiatic acid, 7-oxodehydroabietic acid
AG-32 In L2	P	$C_{13:0}$-$C_{18:0}$, $C_{20:0}$, $C_{22:0}$-$C_{28:0}$, $C_{30:0}$	$C_{16:1}$, $C_{18:1}$, $C_{20:1}$	$C_{15:0}$, $C_{16:0}$	C_{22}, C_{24}, C_{26}	-	-

Sample Code	Contamination	Saturated fatty acids	Unsaturated fatty acids	Branched-chain fatty acids	Dicarboxylic acids	Sulfur-heterocyclic fatty acids	Terpenoids
AG-34 In L2	P, COS	$C_{14:0}$-$C_{24:0}$	$C_{18:1}$, $C_{18:2}$, $C_{22:1}$	$C_{15:0}$-$C_{17:0}$	-	-	-
AG-36 In L2	P, COS	$C_{9:0}$-$C_{28:0}$	$C_{18:1}$, $C_{18:2}$	$C_{15:0}$-$C_{18:0}$	-	-	-
AG-4 In L2	P, COS	$C_{9:0}$, $C_{10:0}$, $C_{12:0}$, $C_{14:0}$-$C_{18:0}$, $C_{20:0}$, $C_{22:0}$, $C_{24:0}$	$C_{16:1}$, $C_{18:1}$	$C_{18:0}$	-	-	-
AG-40 In L2	P	$C_{16:0}$-$C_{26:0}$	$C_{18:1}$, $C_{18:2}$, $C_{20:1}$, $C_{22:1}$	-	-	-	-
AG-41 In L2	P, COS	$C_{14:0}$-$C_{20:0}$	$C_{18:1}$, $C_{20:1}$	$C_{15:0}$-$C_{17:0}$	-	-	-
AG-43 In L2	P, COS	$C_{14:0}$, $C_{16:0}$, $C_{18:0}$, $C_{20:0}$, $C_{22:0}$, $C_{24:0}$, $C_{26:0}$	$C_{18:1}$	$C_{15:0}$-$C_{17:0}$	-	-	-
AG-44 In L2	P, COS	$C_{15:0}$-$C_{26:0}$	-	-	-	-	-
AG-45 In L2	P	$C_{14:0}$-$C_{30:0}$	$C_{16:1}$, $C_{18:1}$	$C_{15:0}$, $C_{16:0}$	-	-	-
AG-7 In L2	P, COS	$C_{15:0}$-$C_{18:0}$, $C_{20:0}$, $C_{22:0}$, $C_{24:0}$, $C_{26:0}$, $C_{28:0}$	$C_{16:1}$, $C_{18:1}$, $C_{18:2}$	-	-	-	-
AG-8 In L2	P, COS	$C_{14:0}$-$C_{16:0}$, $C_{18:0}$, $C_{20:0}$, $C_{22:0}$, $C_{24:0}$	$C_{18:1}$	$C_{15:0}$, $C_{17:0}$	-	-	-
AG-9 In L2	P, COS	$C_{16:0,}$ $C_{18:0}$	-	-	-	-	-
AM-1 In L2	P, COS	$C_{14:0}$-$C_{20:0}$, $C_{22:0}$, $C_{24:0}$	$C_{18:1}$	$C_{15:0}$	-	-	-
AM-2 In L2	P, COS	$C_{12:0}$-$C_{18:0}$	$C_{18:1}$, $C_{18:2}$	-	-	-	-
AM-5 In L2	P	$C_{14:0}$-$C_{18:0}$	$C_{18:1}$, $C_{18:2}$	-	-	-	-
AZ-3 In L2	P, COS	$C_{12:0}$-$C_{20:0}$	$C_{18:1}$	-	-	-	-
BB-1 In L2	P	$C_{12:0}$-$C_{20:0}$, $C_{22:0}$-$C_{26:0}$, $C_{28:0}$	$C_{16:1}$, $C_{18:1}$, $C_{18:2}$	$C_{15:0}$, $C_{16:0}$, $C_{18:0}$	C_{16}, C_{18}, C_{20}, C_{22}-C_{24}, C_{26}, C_{28}	-	-
BB-19 In L2	P	$C_{14:0}$-$C_{18:0}$, $C_{20:0}$, $C_{22:0}$-$C_{24:0}$, $C_{26:0}$	$C_{16:1}$, $C_{18:1}$	-	-	-	Methyl abietate, methyl dehydroabietate, 7-oxodehydroabiatic acid
BB-21 In L2	P	$C_{9:0}$-$C_{28:0}$, $C_{30:0}$	$C_{18:1}$	$C_{15:0}$, $C_{18:0}$	C_{14}, C_{16}, C_{18}-C_{22}, C_{24}-C_{30}	-	-
BB-25 In L2	P	$C_{13:0}$-$C_{18:0}$, $C_{20:0}$-$C_{26:0}$, $C_{28:0}$	$C_{16:1}$, $C_{18:1}$	$C_{15:0}$, $C_{16:0}$	-	-	-
BB-28 In L2	P	$C_{13:0}$-$C_{18:0}$, $C_{22:0}$-$C_{26:0}$, $C_{28:0}$	$C_{16:1}$, $C_{18:1}$	$C_{15:0}$, $C_{16:0}$, $C_{18:0}$	-	-	-

Sample Code	Contamination	Saturated fatty acids	Unsaturated fatty acids	Branched-chain fatty acids	Dicarboxylic acids	Sulfur-heterocyclic fatty acids	Terpenoids
BB-3 In L2	P	$C_{14:0}$-$C_{26:0}$, $C_{28:0}$	$C_{16:1}$, $C_{18:1}$, $C_{20:1}$, $C_{22:1}$	$C_{15:0}$-$C_{17:0}$	-	-	-
BB-4 In L2	P	$C_{14:0}$-$C_{20:0}$, $C_{22:0}$-$C_{28:0}$	$C_{16:1}$, $C_{18:1}$, $C_{20:1}$	$C_{15:0}$-$C_{18:0}$	C_{18}, C_{20}, C_{22}-C_{26}, C_{30}	-	-
BB-5 In L2	P	$C_{14:0}$-$C_{18:0}$, $C_{20:0}$, $C_{22:0}$, $C_{24:0}$	$C_{16:1}$, $C_{18:1}$	-	-	-	-
BB-6 In L2	P	$C_{14:0}$-$C_{18:0}$, $C_{20:0}$, $C_{22:0}$-$C_{26:0}$, $C_{28:0}$	$C_{16:1}$, $C_{18:1}$	$C_{15:0}$, $C_{16:0}$	-	-	-
BB-7 In L2	P	$C_{14:0}$-$C_{18:0}$, $C_{20:0}$, $C_{24:0}$	$C_{16:1}$, $C_{18:1}$, $C_{18:2}$	$C_{15:0}$-$C_{17:0}$	-	-	-
BES-5 In L2	P, COS	$C_{12:0}$-$C_{18:0}$, $C_{20:0}$, $C_{24:0}$	$C_{16:1}$, $C_{18:1}$	-	-	-	-
BES-9 In L2	P, COS	$C_{12:0}$-$C_{18:0}$, $C_{20:0}$, $C_{22:0}$-$C_{24:0}$, $C_{26:0}$	$C_{16:1}$, $C_{18:1}$, $C_{18:2}$	-	-	-	-
BES-12 In L2	P, COS	$C_{14:0}$-$C_{20:0}$, $C_{22:0}$-$C_{24:0}$, $C_{26:0}$	$C_{16:1}$, $C_{18:1}$, $C_{18:2}$	-	-	-	-
BES-13 In L2	P, COS	$C_{14:0}$-$C_{20:0}$, $C_{22:0}$-$C_{26:0}$	$C_{16:1}$, $C_{18:1}$, $C_{18:2}$	-	-	-	-
BES-15 In L2	P, COS	$C_{13:0}$-$C_{18:0}$, $C_{20:0}$, $C_{22:0}$, $C_{24:0}$, $C_{26:0}$	$C_{16:1}$, $C_{18:1}$	$C_{15:0}$, $C_{16:0}$	-	-	-
BES-16 In L2	P, COS	$C_{12:0}$-$C_{18:0}$, $C_{20:0}$, $C_{22:0}$, $C_{24:0}$	$C_{16:1}$, $C_{18:1}$	-	-	-	-
BES-6 In L2	P, COS	$C_{12:0}$-$C_{20:0}$, $C_{22:0}$-$C_{26:0}$, $C_{28:0}$	$C_{16:1}$, $C_{18:1}$, $C_{18:2}$	$C_{15:0}$, $C_{16:0}$	-	-	-
BES-10 In L2	P, COS	$C_{8:0}$-$C_{20:0}$, $C_{22:0}$, $C_{24:0}$, $C_{26:0}$	$C_{16:1}$, $C_{18:1}$, $C_{18:2}$	-	-	-	-
BES-4 In L2	P, COS	$C_{10:0}$, $C_{12:0}$-$C_{18:0}$, $C_{20:0}$, $C_{22:0}$-$C_{26:0}$, $C_{28:0}$	$C_{16:1}$, $C_{18:1}$, $C_{18:2}$	$C_{15:0}$	-	-	-
BES-7 In L2	P, COS	$C_{12:0}$-$C_{20:0}$, $C_{22:0}$-$C_{26:0}$, $C_{28:0}$	$C_{16:1}$, $C_{18:1}$	$C_{15:0}$, $C_{16:0}$	-	-	-
BES-8 In L2	P, COS	$C_{12:0}$-$C_{20:0}$, $C_{22:0}$-$C_{26:0}$, $C_{28:0}$	$C_{16:1}$, $C_{18:1}$	$C_{15:0}$, $C_{16:0}$	C_{14}, C_{16}, C_{18}	-	-
EGD-1 In L2	P	$C_{16:0}$-$C_{18:0}$	$C_{18:1}$	-	-	-	-
FZ-10 In L2	P	$C_{14:0}$-$C_{24:0}$	$C_{18:1}$	-	-	-	-
FZ-11 In L2	P	$C_{14:0}$-$C_{18:0}$, $C_{20:0}$, $C_{22:0}$-$C_{26:0}$, $C_{28:0}$	$C_{16:1}$, $C_{18:1}$	$C_{15:0}$, $C_{16:0}$, $C_{18:0}$	-	-	-
FZ-3 In L2	P, COS	$C_{14:0}$-$C_{18:0}$, $C_{20:0}$	$C_{18:1}$	$C_{17:0}$	-	-	-

Sample Code	Contamination	Saturated fatty acids	Unsaturated fatty acids	Branched-chain fatty acids	Dicarboxylic acids	Sulfur-heterocyclic fatty acids	Terpenoids
FZ-4 In L2	P	$C_{14:0}$-$C_{18:0}$, $C_{20:0}$, $C_{22:0}$-$C_{26:0}$, $C_{28:0}$	$C_{18:1}$, $C_{18:2}$	$C_{16:0}$	-	-	-
FZ-5 In L2	P, COS	$C_{14:0}$-$C_{18:0}$, $C_{20:0}$	$C_{18:1}$	$C_{15:0}$, $C_{17:0}$	-	-	-
FZ-6 In L2	P	$C_{14:0}$-$C_{16:0}$, $C_{18:0}$, $C_{22:0}$, $C_{24:0}$	$C_{16:1}$, $C_{18:1}$	-	-	-	-
FZ-7 In L2	P	$C_{14:0}$-$C_{16:0}$, $C_{18:0}$, $C_{20:0}$, $C_{22:0}$, $C_{24:0}$, $C_{26:0}$	$C_{16:1}$, $C_{18:1}$	-	-	-	-
FZ-8 In L2	P, COS	$C_{14:0}$-$C_{18:0}$, $C_{20:0}$, $C_{22:0}$, $C_{24:0}$, $C_{26:0}$	$C_{18:1}$, $C_{18:2}$	$C_{17:0}$	-	-	-
GLT-1 In L2	P	$C_{9:0}$-$C_{26:0}$, $C_{28:0}$	$C_{16:1}$, $C_{18:1}$, $C_{22:1}$	$C_{15:0}$, $C_{16:0}$, $C_{18:0}$	-	-	-
GLT-10 In L2	P	$C_{10:0}$-$C_{18:0}$, $C_{20:0}$, $C_{22:0}$-$C_{24:0}$	$C_{18:1}$, $C_{18:2}$	$C_{15:0}$-$C_{17:0}$	-	-	-
GLT-11 In L2	P	$C_{12:0}$-$C_{26:0}$, $C_{28:0}$	$C_{18:1}$	$C_{15:0}$, $C_{17:0}$	-	-	-
GLT-12 In L2	P	$C_{9:0}$-$C_{18:0}$, $C_{20:0}$, $C_{22:0}$, $C_{24:0}$, $C_{26:0}$	$C_{16:1}$, $C_{18:1}$, $C_{18:2}$	$C_{15:0}$	-	-	-
GLT-13 In L2	P	$C_{11:0}$-$C_{18:0}$, $C_{20:0}$, $C_{22:0}$, $C_{24:0}$, $C_{26:0}$	$C_{16:1}$, $C_{18:1}$, $C_{18:2}$, $C_{20:1}$, $C_{22:1}$	$C_{15:0}$, $C_{17:0}$	-	-	-
GLT-2 In L2	P	$C_{8:0}$-$C_{20:0}$, $C_{22:0}$-$C_{28:0}$	$C_{16:1}$, $C_{18:1}$, $C_{18:2}$	-	-	-	-
GLT-3 In L2	P	$C_{8:0}$-$C_{26:0}$	$C_{16:1}$, $C_{18:1}$, $C_{18:2}$	$C_{15:0}$, $C_{16:0}$	-	-	-
GLT-4 In L2	P, COS	$C_{12:0}$-$C_{20:0}$, $C_{22:0}$-$C_{26:0}$, $C_{28:0}$	$C_{16:1}$, $C_{18:1}$, $C_{18:2}$	$C_{15:0}$, $C_{16:0}$	-	-	-
GLT-5 In L2	P	$C_{10:0}$-$C_{26:0}$, $C_{28:0}$	$C_{16:1}$, $C_{18:1}$, $C_{20:1}$, $C_{22:1}$	$C_{15:0}$, $C_{16:0}$, $C_{18:0}$	-	8THTph6, 7THTph7, 5THTph9, 4THTph10	-
GLT-6 In L2	P, COS	$C_{12:0}$-$C_{18:0}$, $C_{20:0}$, $C_{22:0}$, $C_{24:0}$	$C_{16:1}$, $C_{18:1}$, $C_{18:2}$	$C_{15:0}$, $C_{16:0}$	-	-	-
GLT-7 In L2	P, COS	$C_{8:0}$-$C_{18:0}$, $C_{20:0}$, $C_{22:0}$, $C_{24:0}$	$C_{16:1}$, $C_{18:1}$, $C_{18:2}$	$C_{15:0}$, $C_{16:0}$	-	-	-
GLT-8 In L2	P, COS	$C_{8:0}$-$C_{24:0}$, $C_{26:0}$	$C_{18:1}$	$C_{17:0}$	-	-	-
GLT-9 In L2	P, COS	$C_{12:0}$-$C_{18:0}$, $C_{22:0}$, $C_{24:0}$	$C_{18:1}$, $C_{18:2}$	$C_{15:0}$	-	-	-
NY-1 In L2	P	$C_{14:0}$-$C_{22:0}$	$C_{16:1}$, $C_{18:1}$	-	-	-	-
NY-10 In L2	P, COS	$C_{14:0}$-$C_{24:0}$	$C_{16:1}$, $C_{18:1}$	-	-	-	-

Sample Code	Contamination	Saturated fatty acids	Unsaturated fatty acids	Branched-chain fatty acids	Dicarboxylic acids	Sulfur-heterocyclic fatty acids	Terpenoids
NY-11 In L2	P, COS	$C_{12:0}$-$C_{26:0}$	$C_{16:1}$, $C_{18:1}$, $C_{20:1}$	$C_{15:0}$, $C_{16:0}$	-	-	-
NY-12 In L2	P	$C_{12:0}$-$C_{30:0}$	$C_{16:1}$, $C_{18:1}$, $C_{20:1}$	$C_{15:0}$-$C_{17:0}$	-	-	-
NY-13 In L2	P, COS	$C_{14:0}$-$C_{20:0}$	$C_{16:1}$, $C_{18:1}$, $C_{18:2}$	-	-	-	-
NY-15 In L2	P	$C_{10:0}$-$C_{20:0}$	$C_{16:1}$, $C_{18:1}$	-	-	-	-
NY-16 In L2	P, COS	$C_{12:0}$-$C_{26:0}$	$C_{16:1}$, $C_{18:1}$, $C_{18:2}$, $C_{18:3}$	-	-	-	-
NY-17 In L2	P, COS	$C_{12:0}$-$C_{28:0}$	$C_{16:1}$, $C_{18:1}$	$C_{15:0}$	-	-	-
NY-18 In L2	P	$C_{14:0}$-$C_{28:0}$	$C_{16:1}$, $C_{18:1}$	$C_{15:0}$	-	-	-
NY-2 In L2	P	$C_{8:0}$-$C_{20:0}$	$C_{16:1}$, $C_{18:1}$	$C_{15:0}$, $C_{16:0}$	-	-	-
NY-3 In L2	P	$C_{10:0}$-$C_{26:0}$	$C_{16:1}$, $C_{18:1}$, $C_{18:2}$	$C_{15:0}$, $C_{16:0}$	-	-	-
NY-4 In L2	P, COS	$C_{14:0}$-$C_{20:0}$	$C_{16:1}$, $C_{18:1}$	-	-	-	-
NY-5 In L2	P	$C_{14:0}$-$C_{20:0}$	$C_{16:1}$, $C_{18:1}$	$C_{15:0}$	-	-	-
NY-6 In L2	P, COS	$C_{14:0}$-$C_{16:0}$, $C_{18:0}$	$C_{16:1}$, $C_{18:1}$	-	-	-	-
NY-7 In L2	P, COS	$C_{14:0}$-$C_{16:0}$, $C_{18:0}$	$C_{16:1}$, $C_{18:1}$	-	-	-	-
NY-8 In L2	P, COS	$C_{14:0}$-$C_{18:0}$, $C_{20:0}$, $C_{22:0}$, $C_{24:0}$, $C_{26:0}$, $C_{28:0}$	$C_{16:1}$, $C_{18:1}$	-	-	-	-
NY-9 In L2	P, COS	$C_{12:0}$-$C_{24:0}$	$C_{16:1}$, $C_{18:1}$, $C_{18:2}$	-	-	-	-
PEQ-1 In L2	P	$C_{12:0}$-$C_{26:0}$	$C_{18:1}$	$C_{15:0}$, $C_{16:0}$	-	-	-
PEQ-2 In L2	P	$C_{14:0}$-$C_{28:0}$	$C_{18:1}$	$C_{15:0}$-$C_{17:0}$	-	-	-
PEQ-3 In L2	P	$C_{10:0}$-$C_{28:0}$	$C_{16:1}$, $C_{18:1}$	$C_{15:0}$	-	-	-
SHH-1 In L2	P, COS	$C_{10:0}$-$C_{30:0}$	$C_{18:1}$, $C_{18:2}$	-	-	-	-
SHH-10 In L2	P	$C_{11:0}$-$C_{16:0}$, $C_{18:0}$, $C_{20:0}$	$C_{18:1}$	-	-	-	-

Sample Code	Contamination	Saturated fatty acids	Unsaturated fatty acids	Branched-chain fatty acids	Dicarboxylic acids	Sulfur-heterocyclic fatty acids	Terpenoids
SHH-11 In L2	P	$C_{10:0}$, $C_{12:0}$-$C_{18:0}$, $C_{20:0}$, $C_{22:0}$, $C_{24:0}$, $C_{26:0}$, $C_{28:0}$	$C_{16:1}$, $C_{18:1}$	$C_{15:0}$	-	-	-
SHH-13 In L2	P	$C_{14:0}$-$C_{18:0}$, $C_{20:0}$	$C_{16:1}$, $C_{18:1}$	-	-	-	-
SHH-17 In L2	P, COS	$C_{10:0}$-$C_{20:0}$	$C_{18:1}$	$C_{15:0}$, $C_{16:0}$	-	-	-
SHH-18 In L2	P, COS	$C_{12:0}$-$C_{28:0}$	$C_{18:1}$, $C_{18:2}$	$C_{15:0}$	-	-	-
SHH-19 In L2	P	$C_{14:0}$, $C_{16:0}$-$C_{18:0}$, $C_{20:0}$, $C_{22:0}$, $C_{24:0}$	$C_{16:1}$, $C_{18:1}$	$C_{15:0}$	-	-	-
SHH-2 In L2	P, COS	$C_{14:0}$-$C_{20:0}$	$C_{18:1}$	$C_{15:0}$, $C_{17:0}$	-	-	-
SHH-20 In L2	P	$C_{14:0}$, $C_{16:0}$, $C_{18:0}$, $C_{20:0}$	$C_{16:1}$, $C_{18:1}$	-	-	-	-
SHH-21 In L2	P, COS	$C_{13:0}$-$C_{24:0}$	$C_{18:1}$	-	-	-	-
SHH-3 In L2	P	$C_{12:0}$-$C_{18:0}$, $C_{20:0}$, $C_{22:0}$, $C_{24:0}$, $C_{26:0}$, $C_{28:0}$	$C_{16:1}$, $C_{18:1}$	$C_{14:0}$-$C_{18:0}$	-	-	-
SHH-4 In L2	P, COS	$C_{14:0}$-$C_{20:0}$	$C_{18:1}$	$C_{15:0}$, $C_{16:0}$	-	-	-
SHH-5 In L2	P, COS	$C_{13:0}$-$C_{28:0}$	$C_{18:1}$, $C_{18:2}$	$C_{15:0}$	-	-	-
SHH-6 In L2	P	$C_{10:0}$-$C_{18:0}$, $C_{20:0}$, $C_{22:0}$, $C_{24:0}$, $C_{26:0}$, $C_{28:0}$	$C_{16:1}$, $C_{18:1}$, $C_{18:2}$	$C_{15:0}$	-	-	-
SHH-7 In L2	P	$C_{12:0}$-$C_{18:0}$, $C_{22:0}$-$C_{28:0}$	$C_{16:1}$, $C_{18:1}$, $C_{18:2}$	$C_{15:0}$, $C_{16:0}$	-	-	-
SHH-8 In L2	P	$C_{14:0}$, $C_{16:0}$, $C_{18:0}$, $C_{20:0}$	$C_{16:1}$, $C_{18:1}$	-	-	-	-
TH-1 In L2	P	$C_{12:0}$-$C_{24:0}$	$C_{16:1}$, $C_{18:1}$, $C_{18:2}$	-	-	-	-
TH-2 In L2	P	$C_{10:0}$-$C_{28:0}$	$C_{16:1}$, $C_{18:1}$	$C_{15:0}$	-	-	-
TH-4 In L2	P	$C_{12:0}$-$C_{28:0}$	$C_{16:1}$, $C_{18:1}$, $C_{18:2}$	$C_{15:0}$	-	-	-
TS-1 In L2	P, COS	$C_{14:0}$-$C_{18:0}$, $C_{20:0}$, $C_{22:0}$-$C_{26:0}$	$C_{16:1}$, $C_{18:1}$, $C_{18:2}$, $C_{20:1}$, $C_{22:1}$	-	-	-	-
TS-10 In L2	P	$C_{9:0}$-$C_{28:0}$	$C_{18:1}$, $C_{20:1}$, $C_{24:1}$	$C_{15:0}$-$C_{17:0}$	-	-	-
TS-11 In L2	P, COS	$C_{14:0}$-$C_{26:0}$	$C_{18:1}$, $C_{18:2}$, $C_{22:1}$	$C_{15:0}$-$C_{17:0}$	-	-	-

Sample Code	Contamination	Saturated fatty acids	Unsaturated fatty acids	Branched-chain fatty acids	Dicarboxylic acids	Sulfur-heterocyclic fatty acids	Terpenoids
TS-12 In L2	P, COS	$C_{14:0}$-$C_{20:0}$	$C_{18:1}$, $C_{18:2}$	$C_{15:0}$, $C_{16:0}$	-	-	-
TS-13 In L2	P	$C_{14:0}$-$C_{26:0}$	$C_{18:1}$, $C_{18:2}$, $C_{20:1}$, $C_{22:1}$	$C_{15:0}$, $C_{16:0}$	-	-	-
TS-16 In L2	P	$C_{12:0}$-$C_{28:0}$	$C_{18:1}$, $C_{20:1}$, $C_{24:1}$	$C_{15:0}$, $C_{16:0}$	-	-	-
TS-18 In L2	P	$C_{12:0}$-$C_{26:0}$	$C_{16:1}$, $C_{18:1}$, $C_{18:2}$	$C_{14:0}$-$C_{16:0}$	-	-	-
TS-2 In L2	P, COS	$C_{14:0}$-$C_{18:0}$	$C_{18:1}$, $C_{18:2}$	-	-	-	-
TS-20 In L2	P	$C_{12:0}$-$C_{26:0}$	$C_{16:1}$, $C_{18:1}$	-	-	-	-
TS-23 In L2	P, COS	$C_{12:0}$-$C_{26:0}$	$C_{18:1}$, $C_{18:2}$	$C_{15:0}$-$C_{17:0}$	-	-	-
TS-26 In L2	P, COS	$C_{14:0}$-$C_{24:0}$	$C_{18:1}$, $C_{18:2}$	$C_{15:0}$, $C_{16:0}$, $C_{18:0}$	-	-	-
TS-3 In L2	P, COS	$C_{12:0}$-$C_{18:0}$, $C_{20:0}$, $C_{22:0}$, $C_{24:0}$, $C_{26:0}$	$C_{16:1}$, $C_{18:1}$	$C_{15:0}$	-	-	-
TS-4 In L2	P, COS	$C_{14:0}$-$C_{16:0}$, $C_{18:0}$, $C_{20:0}$, $C_{22:0}$-$C_{24:0}$	$C_{16:1}$, $C_{18:1}$, $C_{18:2}$, $C_{20:1}$, $C_{22:1}$	$C_{15:0}$, $C_{16:0}$	-	-	-
TS-5 In L2	P	$C_{14:0}$-$C_{30:0}$	$C_{18:1}$, $C_{18:2}$	$C_{15:0}$-$C_{17:0}$	-	-	-
TS-6 In L2	P	$C_{12:0}$-$C_{26:0}$	$C_{16:1}$, $C_{18:1}$	$C_{15:0}$-$C_{17:0}$	-	-	-
TS-7 In L2	P	$C_{12:0}$-$C_{30:0}$	$C_{16:1}$, $C_{18:1}$	$C_{15:0}$, $C_{16:0}$	-	-	-
TS-8 In L2	P	$C_{12:0}$-$C_{28:0}$	$C_{16:1}$, $C_{18:1}$, $C_{18:2}$, $C_{20:1}$, $C_{22:1}$	$C_{15:0}$, $C_{16:0}$, $C_{18:0}$	-	-	-
TS-9 In L2	P	$C_{12:0}$-$C_{28:0}$	$C_{18:1}$	$C_{15:0}$, $C_{17:0}$	-	-	-
TSF17-1 In L2	P, COS	$C_{14:0}$-$C_{22:0}$	$C_{18:1}$	$C_{15:0}$, $C_{16:0}$	-	-	-
TSF17-10 In L2	P	$C_{12:0}$-$C_{30:0}$	$C_{18:1}$	$C_{15:0}$-$C_{17:0}$	-	-	-
TSF17-12 In L2	P, COS	$C_{16:0,}$ $C_{18:0}$	$C_{18:1}$	-	-	-	-
TSF17-13 In L2	P, COS	$C_{14:0}$-$C_{20:0}$	$C_{18:1}$	$C_{15:0}$, $C_{17:0}$	-	-	-
TSF17-15 In L2	P	$C_{14:0}$-$C_{18:0}$, $C_{20:0}$	$C_{18:1}$	-	-	-	-

Sample Code	Contamination	Saturated fatty acids	Unsaturated fatty acids	Branched-chain fatty acids	Dicarboxylic acids	Sulfur-heterocyclic fatty acids	Terpenoids
TSF17-16 In L2	P	$C_{12:0}$, $C_{14:0}$-$C_{18:0}$, $C_{20:0}$, $C_{22:0}$-$C_{26:0}$, $C_{28:0}$	$C_{16:1}$, $C_{18:1}$, $C_{18:2}$	$C_{15:0}$, $C_{16:0}$	-	-	-
TSF17-18 In L2	P	$C_{14:0}$-$C_{22:0}$	$C_{16:1}$, $C_{18:1}$, $C_{18:2}$	-	-	-	-
TSF17-19 In L2	P	$C_{14:0}$-$C_{24:0}$	$C_{18:1}$	$C_{17:0}$	-	-	-
TSF17-2 In L2	P, COS	$C_{14:0}$-$C_{20:0}$	$C_{18:1}$	-	-	-	-
TSF17-21 In L2	P, COS	$C_{14:0}$-$C_{24:0}$	$C_{18:1}$	-	-	-	-
TSF17-22 In L2	P	$C_{12:0}$-$C_{26:0}$	$C_{18:1}$, $C_{22:1}$	-	-	-	-
TSF17-23 In L2	P	$C_{12:0}$-$C_{28:0}$	$C_{16:1}$, $C_{18:1}$, $C_{20:1}$	$C_{15:0}$-$C_{17:0}$	C_9, C_{16}, C_{18}, C_{20}, C_{22}, C_{24}	-	-
TSF17-25 In L2	P, COS	$C_{16:0}$-$C_{28:0}$	$C_{18:1}$	-	-	-	-
TSF17-26 In L2	P	$C_{16:0,}$ $C_{18:0}$	$C_{18:1}$	-	-	-	-
TSF17-27 In L2	P, COS	$C_{10:0}$, $C_{12:0}$-$C_{20:0}$, $C_{22:0}$-$C_{24:0}$, $C_{26:0}$	$C_{18:1}$, $C_{18:2}$	$C_{15:0}$	-	-	-
TSF17-28 In L2	P	$C_{14:0}$-$C_{24:0}$, $C_{26:0}$	$C_{16:1}$, $C_{18:1}$, $C_{20:1}$	$C_{17:0}$	C_{16}, C_{18}	-	-
TSF17-29 In L2	P	$C_{14:0}$-$C_{18:0}$, $C_{20:0}$, $C_{22:0}$-$C_{24:0}$, $C_{26:0}$	$C_{18:1}$	-	-	-	-
TSF17-3 In L2	P, COS	$C_{14:0}$-$C_{28:0}$	$C_{18:1}$, $C_{18:2}$	$C_{17:0}$	-	-	-
TSF17-30 In L2	P	$C_{14:0}$-$C_{18:0}$, $C_{20:0}$, $C_{22:0}$-$C_{28:0}$	$C_{16:1}$, $C_{18:1}$	$C_{15:0}$	-	-	-
TSF17-31 In L2	P	$C_{14:0}$-$C_{18:0}$, $C_{20:0}$, $C_{22:0}$, $C_{24:0}$	$C_{16:1}$, $C_{18:1}$	$C_{15:0}$, $C_{16:0}$	C_{22}	-	-
TSF17-32 In L2	P	$C_{14:0}$-$C_{18:0}$, $C_{20:0}$, $C_{22:0}$, $C_{24:0}$	$C_{16:1}$, $C_{18:1}$	-	-	-	-
TSF17-4 In L2	P	$C_{14:0}$-$C_{18:0}$, $C_{20:0}$, $C_{22:0}$-$C_{26:0}$, $C_{28:0}$	$C_{16:1}$, $C_{18:1}$	-	-	-	-
TSF17-5 In L2	P	$C_{14:0}$-$C_{28:0}$	$C_{18:1}$, $C_{18:2}$	$C_{15:0}$-$C_{17:0}$	-	-	-
TSF17-6 In L2	P	$C_{10:0}$-$C_{26:0}$	$C_{18:1}$	$C_{17:0}$	-	-	-
TSF17-7 In L2	P	$C_{14:0}$-$C_{18:0}$, $C_{20:0}$, $C_{22:0}$-$C_{26:0}$, $C_{28:0}$	$C_{16:1}$, $C_{18:1}$, $C_{18:2}$	-	C_9	-	-

Sample Code	Contamination	Saturated fatty acids	Unsaturated fatty acids	Branched-chain fatty acids	Dicarboxylic acids	Sulfur-heterocyclic fatty acids	Terpenoids
TSF17-8 In L2	P, COS	$C_{14:0}$-$C_{30:0}$	$C_{18:1}$, $C_{18:2}$, $C_{18:3}$	-	-	-	-
TSF17-9 In L2	P	$C_{14:0}$-$C_{16:0}$, $C_{18:0}$, $C_{20:0}$, $C_{22:0}$, $C_{24:0}$	$C_{16:1}$, $C_{18:1}$	-	-	-	-
TSF18-1 In L2	P	$C_{9:0}$-$C_{28:0}$	$C_{18:1}$, $C_{18:2}$	-	-	-	Methyl dehydroabietate
TSF18-10 In L2	P	$C_{14:0}$-$C_{16:0}$, $C_{18:0}$, $C_{20:0}$	$C_{18:1}$	-	-	-	-
TSF18-11 In L2	P	$C_{16:0}$, $C_{18:0}$	$C_{18:1}$	-	-	-	-
TSF18-13 In L2	P	$C_{14:0}$-$C_{18:0}$, $C_{20:0}$, $C_{22:0}$	$C_{16:1}$, $C_{18:1}$	-	C_9	-	-
TSF18-14 In L2	P	$C_{9:0}$, $C_{12:0}$-$C_{18:0}$, $C_{20:0}$, $C_{22:0}$-$C_{24:0}$, $C_{26:0}$, $C_{28:0}$	$C_{16:1}$, $C_{18:1}$	$C_{15:0}$, $C_{16:0}$	-	-	-
TSF18-15 In L2	P	$C_{14:0}$-$C_{22:0}$	$C_{18:1}$	$C_{18:0}$	-	-	-
TSF18-16 In L2	P	$C_{14:0}$-$C_{28:0}$	$C_{18:1}$, $C_{18:2}$, $C_{20:1}$, $C_{22:1}$	$C_{16:0}$	-	7THTph7, 6THTph8, 5THTph9, 4THTph10	-
TSF18-17 In L2	P	$C_{13:0}$-$C_{18:0}$, $C_{22:0}$-$C_{28:0}$	$C_{16:1}$, $C_{18:1}$	$C_{15:0}$	-	-	-
TSF18-18 In L2	P	$C_{14:0}$-$C_{20:0}$, $C_{23:0}$-$C_{28:0}$, $C_{30:0}$	$C_{16:1}$, $C_{18:1}$	-	-	-	-
TSF18-19 In L2	P	$C_{14:0}$-$C_{24:0}$	$C_{18:1}$, $C_{18:2}$	-	-	-	-
TSF18-2 In L2	P	$C_{14:0}$, $C_{16:0}$, $C_{18:0}$, $C_{20:0}$, $C_{22:0}$-$C_{26:0}$, $C_{28:0}$	$C_{16:1}$, $C_{18:1}$, $C_{18:2}$	-	-	-	-
TSF18-20 In L2	P	$C_{12:0}$-$C_{30:0}$	$C_{18:1}$, $C_{20:1}$	$C_{15:0}$-$C_{17:0}$	-	-	-
TSF18-21 In L2	P	$C_{14:0}$-$C_{18:0}$, $C_{20:0}$, $C_{22:0}$-$C_{26:0}$	$C_{16:1}$, $C_{18:1}$	$C_{16:0}$	-	-	-
TSF18-22 In L2	P	$C_{14:0}$-$C_{18:0}$, $C_{22:0}$	$C_{16:1}$, $C_{18:1}$	-	-	-	-
TSF18-23 In L2	P	$C_{13:0}$-$C_{18:0}$, $C_{20:0}$, $C_{24:0}$	$C_{16:1}$, $C_{18:1}$	$C_{15:0}$, $C_{16:0}$	-	-	-
TSF18-24 In L2	P	$C_{10:0}$-$C_{20:0}$, $C_{22:0}$-$C_{26:0}$, $C_{28:0}$, $C_{30:0}$	$C_{16:1}$, $C_{18:1}$, $C_{20:1}$	$C_{14:0}$, $C_{15:0}$	C_9	-	-
TSF18-25 In L2	P	$C_{10:0}$, $C_{12:0}$-$C_{20:0}$, $C_{22:0}$-$C_{26:0}$, $C_{28:0}$	$C_{18:1}$, $C_{20:1}$	$C_{15:0}$, $C_{17:0}$	-	-	-
TSF18-26 In L2	P	$C_{12:0}$, $C_{14:0}$-$C_{20:0}$, $C_{22:0}$-$C_{24:0}$, $C_{26:0}$	$C_{16:1}$, $C_{18:1}$	$C_{15:0}$	-	-	-

Sample Code	Contamination	Saturated fatty acids	Unsaturated fatty acids	Branched-chain fatty acids	Dicarboxylic acids	Sulfur-heterocyclic fatty acids	Terpenoids
TSF18-27 In L2	P	$C_{14:0}$-$C_{20:0}$	$C_{18:1}$	-	-	-	-
TSF18-28 In L2	P	$C_{14:0}$-$C_{20:0}$, $C_{22:0}$-$C_{24:0}$, $C_{26:0}$, $C_{28:0}$	$C_{16:1}$, $C_{18:1}$, $C_{18:2}$	-	-	-	-
TSF18-29 In L2	P	$C_{14:0}$-$C_{20:0}$, $C_{22:0}$-$C_{28:0}$	$C_{16:1}$, $C_{18:1}$	$C_{17:0}$	-	-	-
TSF18-3 In L2	P	$C_{10:0}$-$C_{30:0}$	$C_{18:1}$, $C_{20:1}$	$C_{15:0}$, $C_{17:0}$	-	-	-
TSF18-30 In L2	P, COS	$C_{14:0}$-$C_{18:0}$	$C_{16:1}$, $C_{18:1}$	$C_{15:0}$, $C_{16:0}$	-	-	-
TSF18-31 In L2	P	$C_{9:0}$, $C_{10:0}$, $C_{12:0}$, $C_{14:0}$-$C_{18:0}$, $C_{20:0}$	$C_{16:1}$, $C_{18:1}$	$C_{15:0}$, $C_{16:0}$	-	-	-
TSF18-34 In L2	P	$C_{12:0}$-$C_{28:0}$	$C_{16:1}$, $C_{18:1}$, $C_{20:1}$	$C_{15:0}$, $C_{16:0}$	-	-	-
TSF18-35 In L2	P	$C_{14:0}$-$C_{20:0}$, $C_{22:0}$-$C_{28:0}$, $C_{30:0}$	$C_{16:1}$, $C_{18:1}$, $C_{20:1}$	$C_{15:0}$, $C_{16:0}$, $C_{18:0}$	-	-	-
TSF18-36 In L2	P	$C_{12:0}$-$C_{18:0}$, $C_{20:0}$, $C_{22:0}$, $C_{24:0}$	$C_{16:1}$, $C_{18:1}$	$C_{17:0}$	-	-	-
TSF18-37 In L2	P	$C_{14:0}$-$C_{26:0}$, $C_{28:0}$	$C_{16:1}$, $C_{18:1}$	-	-	-	-
TSF18-39 In L2	P	$C_{14:0}$-$C_{28:0}$, $C_{30:0}$	$C_{16:1}$, $C_{18:1}$	$C_{16:0}$, $C_{18:0}$	-	-	-
TSF18-4 In L2	P	$C_{12:0}$-$C_{28:0}$	$C_{18:1}$, $C_{18:2}$, $C_{20:1}$, $C_{22:1}$	-	-	6THTph8, 5THTph9	-
TSF18-40 In L2	P	$C_{12:0}$-$C_{28:0}$, $C_{30:0}$	$C_{16:1}$, $C_{18:1}$, $C_{20:1}$	$C_{15:0}$, $C_{16:0}$, $C_{17:0}$	-	-	-
TSF18-42 In L2	P	$C_{13:0}$-$C_{18:0}$, $C_{20:0}$, $C_{22:0}$, $C_{24:0}$	$C_{16:1}$, $C_{18:1}$	$C_{15:0}$, $C_{16:0}$	-	-	-
TSF18-44 In L2	P, COS	$C_{12:0}$-$C_{20:0}$	$C_{16:1}$, $C_{18:1}$	-	-	-	-
TSF18-45 In L2	P, COS	$C_{12:0}$-$C_{30:0}$	$C_{16:1}$, $C_{18:1}$, $C_{18:2}$	$C_{15:0}$, $C_{16:0}$	-	-	-
TSF18-46 In L2	P, COS	$C_{12:0}$-$C_{26:0}$	$C_{16:1}$, $C_{18:1}$, $C_{18:2}$, $C_{20:1}$	$C_{15:0}$, $C_{16:0}$	-	-	-
TSF18-47 In L2	P, COS	$C_{12:0}$-$C_{24:0}$	$C_{16:1}$, $C_{18:1}$, $C_{18:2}$	$C_{15:0}$, $C_{16:0}$	-	-	-
TSF18-48 In L2	P	$C_{14:0}$-$C_{16:0}$, $C_{18:0}$, $C_{20:0}$, $C_{22:0}$, $C_{24:0}$	$C_{16:1}$, $C_{18:1}$	-	-	-	-
TSF18-49 In L2	P, COS	$C_{14:0}$-$C_{18:0}$	$C_{16:1}$, $C_{18:1}$	$C_{15:0}$, $C_{16:0}$	-	-	-

Sample Code	Contamination	Saturated fatty acids	Unsaturated fatty acids	Branched-chain fatty acids	Dicarboxylic acids	Sulfur-heterocyclic fatty acids	Terpenoids
TSF18-5 In L2	P	$C_{14:0}$-$C_{24:0}$	$C_{18:1}$	-	-	-	-
TSF18-50 In L2	P, COS	$C_{10:0}$-$C_{30:0}$	$C_{16:1}$, $C_{18:1}$	-	-	-	-
TSF18-51 In L2	P, COS	$C_{8:0}$-$C_{26:0}$	$C_{16:1}$, $C_{18:1}$, $C_{20:1}$	$C_{15:0}$-$C_{17:0}$	-	-	-
TSF18-52 In L2	P, COS	$C_{8:0}$-$C_{26:0}$	$C_{16:1}$, $C_{18:1}$	$C_{15:0}$, $C_{16:0}$	-	-	-
TSF18-54 In L2	P, COS	$C_{14:0}$-$C_{26:0}$	$C_{16:1}$, $C_{18:1}$, $C_{18:2}$, $C_{20:1}$	$C_{15:0}$, $C_{16:0}$	-	-	-
TSF18-55 In L2	P, COS	$C_{14:0}$-$C_{24:0}$	$C_{16:1}$, $C_{18:1}$, $C_{18:2}$	-	-	6THTph8, 5THTph9	-
TSF18-56 In L2	P	$C_{16:0}$, $C_{18:0}$-$C_{20:0}$, $C_{24:0}$, $C_{28:0}$	$C_{18:1}$, $C_{18:2}$	$C_{17:0}$	-	-	-
TSF18-57 Cr	P	$C_{14:0}$-$C_{30:0}$	$C_{18:1}$, $C_{18:2}$	$C_{15:0}$-$C_{17:0}$	-	-	-
TSF18-57 In L2	P	$C_{14:0}$-$C_{28:0}$	$C_{18:1}$, $C_{18:2}$	-	-	-	-
TSF18-58 In L2	P	$C_{14:0}$-$C_{24:0}$	$C_{18:1}$	-	-	-	-
TSF18-59 In L2	P	$C_{14:0}$-$C_{24:0}$	$C_{18:1}$	-	-	-	-
TSF18-6 In L2	P	$C_{14:0}$-$C_{28:0}$	$C_{18:1}$	-	-	6THTph8, 5THTph9	Dehydroabiatic acid
TSF18-60 In L2	P	$C_{16:0,}$ $C_{18:0}$	$C_{18:1}$	-	-	-	-
TSF18-62 In L2	P	$C_{16:0,}$ $C_{18:0}$	$C_{18:1}$	-	-	-	-
TSF18-65 In L2	P, COS	$C_{12:0}$-$C_{20:0}$, $C_{22:0}$-$C_{28:0}$	$C_{16:1}$, $C_{18:1}$	$C_{15:0}$-$C_{17:0}$	-	-	-
TSF18-66 In L2	P, COS	$C_{8:0}$-$C_{28:0}$	$C_{18:1}$, $C_{20:1}$	$C_{15:0}$, $C_{17:0}$	-	-	-
TSF18-67 In L2	P	$C_{14:0}$-$C_{26:0}$	$C_{18:1}$, $C_{18:2}$, $C_{20:1}$	-	-	-	-
TSF18-68 In L2	P	$C_{16:0,}$ $C_{18:0}$	$C_{18:1}$	-	-	-	-
TSF18-69 In L2	P	$C_{12:0}$-$C_{28:0}$	$C_{16:1}$, $C_{18:1}$, $C_{20:1}$	$C_{15:0}$, $C_{16:0}$	-	-	-
TSF18-7 In L2	P	$C_{14:0}$-$C_{28:0}$	$C_{18:1}$	$C_{16:0}$	-	-	-

Sample Code	Contamination	Saturated fatty acids	Unsaturated fatty acids	Branched-chain fatty acids	Dicarboxylic acids	Sulfur-heterocyclic fatty acids	Terpenoids
TSF18-70 In L2	P	$C_{14:0}$, $C_{16:0}$, $C_{18:0}$, $C_{20:0}$, $C_{22:0}$	$C_{18:1}$	-	-	-	-
TSF18-71 In L2	P	$C_{14:0}$-$C_{24:0}$	$C_{18:1}$	-	-	-	-
TSF18-73 In L2	P	$C_{12:0}$-$C_{28:0}$	$C_{16:1}$, $C_{18:1}$	-	-	-	-
TSF18-74 In L2	P	$C_{14:0}$-$C_{28:0}$	$C_{16:1}$, $C_{18:1}$, $C_{18:2}$	-	-	-	-
TSF18-75 In L2	P	$C_{13:0}$-$C_{28:0}$	$C_{16:1}$, $C_{18:1}$	$C_{16:0}$	-	-	-
TSF18-77 In L2	P	$C_{16:0,}$ $C_{18:0}$	$C_{16:1}$, $C_{18:1}$, $C_{18:2}$	-	-	-	-
TSF18-78 In L2	P	$C_{14:0}$-$C_{30:0}$	$C_{16:1}$, $C_{18:1}$, $C_{20:1}$	$C_{15:0}$, $C_{16:0}$	-	-	-
TSF18-8 In L2	P	$C_{14:0}$-$C_{24:0}$	$C_{18:1}$	-	-	-	-
TSF18-9 In L2	P	$C_{14:0}$-$C_{20:0}$	$C_{18:1}$	-	-	-	-
TSF19-1 In L2	-	$C_{16:0}$-$C_{18:0}$	$C_{18:1}$	-	-	-	-
TSF19-13 In L2	-	$C_{14:0}$-$C_{24:0}$, $C_{26:0}$	$C_{16:1}$, $C_{18:1}$, $C_{18:2}$	-	-	-	-
TSF19-14 In L2	P, COS	$C_{16:0,}$ $C_{18:0}$	-	-	-	-	-
TSF19-2 In L2	-	$C_{12:0}$, $C_{14:0}$-$C_{28:0}$	$C_{16:1}$, $C_{18:1}$, $C_{18:2}$, $C_{20:1}$, $C_{22:1}$	$C_{15:0}$, $C_{16:0}$, $C_{18:0}$	C_{18}, C_{20}, C_{22}, C_{24}, C_{26}	-	-
TSF19-3 In L2	-	$C_{9:0}$, $C_{12:0}$-$C_{28:0}$	$C_{16:1}$, $C_{18:1}$, $C_{20:1}$, $C_{22:1}$	$C_{15:0}$-$C_{18:0}$	C_{16}-C_{18}, C_{20}, C_{22}, C_{24}, C_{26}	-	-
TSF19-4 In L2	-	$C_{12:0}$-$C_{24:0}$, $C_{26:0}$, $C_{28:0}$	$C_{18:1}$, $C_{20:1}$	-	-	-	-
TSF19-5 In L2	P	$C_{15:0}$-$C_{18:0}$, $C_{20:0}$	$C_{16:1}$, $C_{18:1}$, $C_{18:2}$	$C_{18:0}$	-	-	-
TSF19-6 In L2	P, COS	$C_{14:0}$-$C_{28:0}$	$C_{18:1}$, $C_{18:2}$, $C_{22:1}$	$C_{15:0}$, $C_{16:0}$	-	-	-
TSF19-7 In L2	P, COS	$C_{14:0}$-$C_{28:0}$	$C_{18:1}$, $C_{18:2}$	$C_{15:0}$, $C_{16:0}$	-	-	-
TSF19-9 In L2	P	$C_{14:0}$-$C_{18:0}$, $C_{20:0}$, $C_{22:0}$-$C_{24:0}$, $C_{26:0}$, $C_{28:0}$	$C_{16:1}$, $C_{18:1}$, $C_{18:2}$	$C_{15:0}$, $C_{16:0}$, $C_{18:0}$	-	-	-
YEH-1 In L2	P	$C_{12:0}$-$C_{26:0}$	$C_{18:1}$, $C_{18:2}$	$C_{15:0}$-$C_{17:0}$	-	-	-

Sample Code	Contamination	Saturated fatty acids	Unsaturated fatty acids	Branched-chain fatty acids	Dicarboxylic acids	Sulfur-heterocyclic fatty acids	Terpenoids
YV-10 In L2	P, COS	$C_{14:0}$, $C_{16:0}$, $C_{18:0}$, $C_{20:0}$, $C_{22:0}$, $C_{24:0}$, $C_{26:0}$, $C_{28:0}$	$C_{18:1}$	$C_{15:0}$, $C_{16:0}$	-	-	-
YV-7 In L2	P, COS	$C_{14:0}$-$C_{26:0}$	$C_{18:1}$, $C_{18:2}$, $C_{22:1}$	$C_{15:0}$	-	-	-

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רבקה חזן

תקציר

דרום הלבאנט הפרוטו-היסטורי (3,800–6,400 לפמנ"ס, תיארון מכויל) מסמל את שלהי הנאוליטיזציה. אנשי התקופה גרו בכפרים קטנים וכלכלת קיומם התבססה על חקלאות ומרעה, בדגש על כבשים, עיזים, בקר וחזירים וכן דגנים וקטניות. תקופה זו מאופיינת גם בחידושים טכנולוגיים רבים, החל בכלי חרס וכלה בנחושת. מחקרים קודמים מצביעים על התפתחויות במורכבות החברתית-כלכלית לאורך התקופה, שאת תוצאותיהן ניתן לראות בתופעות כגון הופעת בתי מלאכה והיווצרות כלכלות קיום המושתתות על יצור חקלאי מתמחה, במיוחד של זיתים וחלב.

מחקר זה מתמקד בבדיקת ההתפתחויות שהוצעו לעיל, ובמהלכו אותרו דפוסי תזונה ודפוסים כלכליים יחודיים בעזרת אנליזה של שרידים אורגנים (שומנים) ותצרופת של תוצאותיה עם המכלולים הארכיאו-זואולוגיים ,הארכיאובוטניים ומכלולי כלי החרס והאבן. כחלק ממתודולוגיית המחקר, נעשה שימוש בטכניקות מתקדמות של כימיה אורגנית על מנת לזהות שומנים וחתימות איזוטופיות של פחמן ממאות כלי חרס ושלושה כלי אבן מאתרים ארכיאולוגיים בישראל, המתוארכים לתקופות הניאולית והכלקולית. הזיהוי נעשה באמצעות שיטות ה- Gas-Chromatography-Mass Spectrometry ו-Gas-Chromatography-combustion-Isotope Ratio Mass Spectrometry.

התוצאות מצביעות על המשכיות בכלכלה ובמסורות התזונה לאורך התקופות שאינה תלויה בגודלם וסגנונם של הכפרים, ולרוב, גם בתנאים הסביבתיים והאקלימיים מסביב. לאורך כל התקופה הנחקרת, נצרך בעיקר בשר חיות מקנה (בדגש על כבשים ועזים) בשילוב עם דגנים וקטניות. הבשר, הדגנים והקטניות בושלו בצורות מגוונות ואין אינדיקציה לכך שכלים מסוימים שומשו באופן מיוחד למטרות מסוימות. כאשר נתגלו חותמות איזוטופיות של בשר חזיר, השומנים תמיד נמצאו בערבוב עם שומנים של כבשים, עיזים או פרות. על פי כך ניתן לשער שגם בשרים שונים בושלו יחד.

בהשוואת הממצאים הכימיים מכל התקופות שנחקרו, ניתן לראות הבדל מרכזי, וזה הוא השימוש בחלב. נראה ששימוש בחלב (בתדירות נמוכה) החל בתקופת הכלקוליתית התיכונה, ספציפית באזורים מדבריים או צחיחים-למחצה. זו הדוגמה היחידה של כלכלת קיום התלויה באקלים ובמיקום גיאוגרפי. בתקופת הכלקולית המאוחר, אין אינדיקציה לעלייה בשימוש בחלב או לקשר בין חלב לבין מחבצות לפי השרידים האורגנים שחולצו מהן. תוצאות אלה מציעות להעריך מחדש את רעיון מהפכת מוצרי המשנה, הנחשב לאחד המאפיינים המרכזיים של התקופה הכלקולית המאוחר. בנוסף, לא נמצאו סימנים של שמן זית בשרידים האורגנים, למרות שנמצאו גלעיני זיתים במכלולים הארכיאובוטניים. המשמעות לכך היא ששומנים אלה לא שרדו, או שכמות שמן הזית לא הייתה רבה דיה על מנת להשאיר שרידי שומן.

האחידות בדפוסי התזונה מצביעה על כך שב ראשיתה של התקופה הניאולתית התבססה כלכלת קיום מעורבת אגרו-פסטורלית אשר הפכה לחלק בלתי נפרד במערכת החברתית-כלכלית של הכפריים בתקופת הניאולית הקרמי. לתהליכים

החברתיים-כלכליים שהתרחשו לאורך התקופה הכלקוליתית לא הייתה השפעה משמעותית על דפוסי התזונה של החברות האנושיות באזור, או על אופן שימושם בכלים להכנת ואכילת מזון.

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מאת: רבקה חזן

בהנחיית: פרופ' דני רוזנברג

פרופ' סינטיאן ספיתרי

חיבור לשם קבלת התואר

"דוקטור לפילוסופיה"

דוקטורט פרסומים

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