Mediterranean Archaeology and Archaeometry, Vol. 15, No 3,(2015), pp. 31-44

DOI: 10.5281/zenodo.18356

ORGANIC RESIDUES IN IRON AGE II POTTERY VESSELS FROM JNENEH, JORDAN

Abdulraouf. S. Mayyas[1] and Khaled. A. Douglas[2]

[1]Department of Conservation Science, Queen Rania Institute of Tourism and Heritage, The Hashemite University, P. O. Box. 330127, Zarqa 13133, Jordan.
[2]Department of Sustainable Tourism, Queen Rania Institute of Tourism and Heritage, The Hashemite University, P. O. Box. 330127, Zarqa 13133, Jordan.
and
Department of Archaeology, Sultan Qaboos University, Oman

Received: 01/04/2015
Accepted: 01/05/2015
Corresponding author: Abdulraouf Mayyas (A_S_Mayyas@hotmail.com)

ABSTRACT

This paper discusses the analysis and occurrence of organic residues in Iron Age II pottery sherds excavated at the site of Jneneh. Gas chromatography – mass spectrometry was used for separation and identification of organic constituents. Conventional solvent extraction and alkaline hydrolysis (saponification) were used for the extraction of residues preserved in their fabrics. Three sherds among six unwashed and unhandled sherds showed significant preservation of organic constituents derived from natural materials. The results provide data on the occurrence of beeswax that could have been intentionally added on the internal surface of a collared-rim jar for storing liquid substances or dry goods. Unintentional occurrence of beeswax as a result of storing honey including beeswax fragments of the honeycomb is also possible. In addition, biomarkers of plant oil were detected in a medium jug and a small jar that might have been used for daily needs, such as food preparation. The absence of significant organic constituents in the other three vessels could be attributed to the degradation overtime during burial.

KEYWORDS: Iron Age Pottery, Jneneh, Ammonite Pottery, Ceramic, Organic Residue, Lipid, Beeswax, Biomarker, Gas Chromatography - Mass Spectrometry, Extraction.

1. INTRODUCTION

Pottery vessels were one of the ceramic materials exploited in the past for different uses. Some vessels were used for storing materials, such as seeds, oil, wine and water while others were exploited for daily uses, such as processing and serving foods. Pottery vessels vary in their porosities, and even the same vessel has heterogeneous fabric pores. Therefore the internal surfaces could have been sealed with the available suitable natural materials, in order to retain liquids, or protect commodities saved inside the vessel from harm environments. In most cases, these uses leave organic constituents "called biomarkers" of the stored material in the pores of the used pottery vessel (Eglinton and Logan, 1991; Evershed et al., 1999; Colombini et al., 2005; Evershed, 2008). Organic constituents can survive for longer periods of time, even under burial conditions, but with different degrees of alteration, depending on the material itself and on the anthropogenic, environmental and burial conditions (Evershed et al., 1995a and 1999; Regert et al., 2003b; Evershed, 2008). They are usually present as a complex mixture of organic molecules. These molecules can be extracted using organic solvents and then separated, characterized, and identified, using the common analytical organic chemical technique of gas chromatography – mass spectrometry (GC-MS). Generally, the purpose of characterization is to obtain data on ancient materials stored and/or used in these vessels, potentially shedding light on the function or use of these vessels (Heron et al., 1991; Evershed et al., 1995b; van Bergen et al., 1997; Dudd et al., 1999; Mottram et al., 1999; Regert et al., 2001; Stern et al., 2003; Evershed, 2008; Gregg et al., 2009; Baeten et al., 2010; Isaksson and Hallgren, 2012).

Jneneh is a small site (ca. 4.5 ha) located in the north-western periphery of the city of Zarqa, in north-central Jordan, ca. 26 km northeast of Amman. The site situated at the fringes of the semi-arid zone, between the Black Basalt Desert to the east, and the central Transjordanian hills to the west. It is located on a natural flat hill on the western bank of Zarqa River and surrounded by gentle slopes, except on the east side, where it is protected by a very sharp cliff looking at Wadi al-Zarqa. It is raised ca. 40 m above the bed of the Wadi.

In 2011 and 2012 two seasons of excavations were conducted by a team from The Hashemite University under the direction of Khaled Douglas. The site was occupied during two major phases: the earlier is during the Early Bronze Age I/II (EBA-I/EBA-II: late 4th millennium B.C.E.), where the site was used as an open village; while the second phase was during the Iron Age II (IA-II: second half of the 8th century B.C.E.), where the site was used as a small open town with a fortified acropolis. During the EBA-IV (late 3rd millennium B.C.E) the site was occupied in the form of a small seasonal camp.

Based on its geographical position as well as its pottery, the Iron Age II settlement at the site of Jneneh represents one of the northern towns of the Ammonite Kingdome (Hübner, 1992). In order to unveil the Ammonite Kingdom, the site of Jneneh, with its location at the northern line border of the Kingdom, was chosen to be excavated. Respectively, excavations of the first two seasons at Jneneh have focused on the latest occupational phase at the settlement and concentrated mainly on the highest area of the site (acropolis), where traces of buildings and fortification walls still appear from the surfaces on the north-eastern part of the site.

Excavations within the fortified area (acropolis) revealed three separated building complexes with narrow streets (1.2 m) in between, each composed of several buildings. The structures within the building complex were connected to each other, sharing outer walls. Each structure contains three to four rectangular rooms, and in some cases includes an open courtyard with a hearth in the corner. Some buildings had rooms with pillars that might have contained two floors, a lower one for storage and an upper living floor. In some rooms (basement) remains of large storage jars (pithos) were found in situ embedded in their floor.

The large amount of painted pottery that was found in Jneneh, including the black and white bands on red burnished slip, is representative of typical Ammonite pottery and ensures that this location was an Ammonite city. C14 dates, as well as the pottery typology, shows that the last occupational phase in Jneneh dates back to the second half of the eighth century B.C.E.

In general different pottery wares such as cocking pots, jugs, plates, storage jars and lamps were found. Loom weights, basalt grinding stones were also found. The diversity of pottery ware proved that the Iron Age II settlement was used for domestic purposes. Its location in a fertile land at the western bank of Zarqa River, and the existence of a large quantity of storage jars, as well as grinding stones, led to conclusion that the economy of the society of Jneneh was primarily based on agriculture.

A large quantity of pottery was found directly on last surface floor of most of the excavated buildings, underneath the destruction layer that ended the Iron Age II occupation at the site. The characterization of the organic residues contained therein would yield valuable information on the original content of vessels, and about the life of the Ammonites in this border town before it was demolished.

We report herein the results of the characterisation of absorbed organic residues in six Iron Age II pottery sherds excavated at the site of Jneneh, with the aim of determining the type of the materials used in the mother vessels and thereby the potential use of these vessels. These six sherds were the only unwashed and unhandled samples available for this study.

2. RESEARCH METHODOLOGY

2.1 Materials and sampling

Ultrapure or HPLC grade solvents and reagents were used in this research. N-tetratriacontane (C_{34}) was used as internal standard (IS) and N,O-bis(trimethylsilyl) trifluoroacetamide (BSTFA) present in 1% v/v trimethylchlorosilane (TMCS) was used as derivatising agent. Glassware used for sampling were thoroughly washed with soap solution and then in double distilled water. After that, they were rinsed with acetone followed by a mixture of dichloromethane/methanol (DCM/MeOH) solution (2:1 v/v), and then heated in an oven at 550 °C for 5 hours before use.

Samples: Six pottery sherds were excavated from known Iron Age II contexts (Table 1) at the site of Jneneh during the excavation season of 2011. All samples are described as common ware of coarse fabric with slip usually present on the outer surface but not on the internal one.

The six Iron Age II pottery sherds that were chosen for this study were not exposed to washing or handling. In addition, six soil samples adhering onto the internal surfaces of these sherds, and one new modern sherd used as a control sample, were also chosen for the study. All these samples were together subjected to laboratory preparation and analysis. The archaeological sherds lack visible organic residues on their internal or external surfaces. Powdered pottery samples were separately taken from the interior (from the internal surface to the core) of the archaeological sherds and the modern sherd using a *Dremel* electric drill fitted with a tungsten abrasive bit. Each powder sample was taken, after having removed the bulk of soil adhering onto the internal surface of the archaeological sherd. The remaining adhering soil was smoothly collected from the surface using a clean small stainless steel spatula, followed by cleaning the surface under a stream of clean compressed air to avoid contamination. Soil samples were also converted into fine powders using pre-cleaned mortar and pestle.

Conventional solvent extraction: About 1.0 g of each powder was transferred into a clean glass vial and extracted three times successively. Each time 2 mL mixture of DCM/MeOH solution (2:1 v/v) was added to the powder, ultrasonicated for 5 minutes and centrifuged (2000 rpm × 5 min), and then the supernatant was transferred into a small clean glass vial. The three supernatants from the successive extractions were combined in the glass vial and then were divided into two equal portions using other two small clean glass vials. The solvent in the two vials was evaporated under a stream of nitrogen gas and mild heat and the residues were left overnight in a vacuum desiccator at – 4 °C. One of the two portions was derivatised by adding BSTFA and left in a tightly closed vial for 24 hours at room temperature. After that the remaining BSTFA was evaporated as mentioned above and the derivatised residue was dissolved in 20 µL DCM. Then a measured amount of approximately 1 µL of the IS with concentration of 1mg/ml was added and the solution was gently mixed. Finally, approximately 1 µL of the residue solution was taken using a micro-syringe and injected through the injector of the GC-MS.

Table 1. General description of the pottery sherds analysed in this study

#	Code of the sherd	Codes of the square and locus	Analysed part of the vessel	Vessel description
			1st Group	
1	Jn17	Sq IB, L012	Body	Large storage jar (pithos), coarse ware, outside slip, thick, light brownish gray. Found in a small storage room (B1:R3).
			2nd Group	
2	Jn21	Sq IB, L013	Base	Large storage jar (pithos), coarse ware, outside slip, thick, light brownish gray. Found in a small storage room (B1:R3).
3	Jn23	Sq IB, L013	Base	Large storage jar (pithos), coarse ware, outside slip, thick, gray. Found in a small storage room (B1:R3).
4	Jn24	Sq IB, L013	Upper part	Medium jar with oval horizontal knob handle, coarse ware, outside slip, thin, gray. Found in a small storage room (B1:R3).
			3rd Group	
5	Jn6	Sq IA, L015	Upper part	Medium jug, coarse ware, outside slip, thin, dark reddish gray. Found inside a large fireplace in an open courtyard (B3:R1).
6	Jn15	Sq IB, L008	Base	Small jar, coarse ware, outside slip, thin, brown. Found in a small storage room (B1:R3).

Alkaline hydrolysis (saponification): The pottery residue remained after solvent extraction of sherds (Jn6, Jn15, Jn21, Jn23 and Jn24) was dried under nitrogen gas flow. This dried residue and the second portion of the solvent extract of the same sherd were separately subjected to alkaline hydrolysis. Each was saponified with 5 mL of methanolic sodium hydroxide solution (0.5 M NaOH in 9:1 (v/v) MeOH/distilled H_2O), heated for 1 hour at 70 °C in a water bath. After being allowed to cool, the saponified mixture was centrifuged (2000 r.p.m × 5 min). The liquid phase were separated and acidified with 6M hydrochloric acid (HCl) and the liberated lipid components were then extracted three times with *n*-hexane (3 × 5 mL). The *n*-hexane extracts were combined and the solvent was evaporated under a stream of nitrogen gas and mild heat. The extracts were stored in a vacuum desiccator at – 4 °C until required for derivatisation and analysis. Trimethylsilylation with BSTFA followed with the steps mentioned above were then performed as described above.

2.2 The Analytical Technique of GC-MS

Gas chromatography – mass spectrometry (GC-MS) analysis was carried out using Varian 450-GC Gas Chromatograph connected to Varian 320-MS TQ Mass Spectrometer. The GC was equipped with a split/splitless injector. Splitless mode was used. Helium was the carrier gas, with a constant head pressure of 1 psi and a flow rate of 1 ml/min at 50 °C. The injector of the GC and the interface of MS were maintained at 280 °C and 340 °C, respectively. The temperature of the oven was programmed from 50 °C (2 min isothermal) to 340 °C (20 min isothermal) at a rate of 10 °C/min. FS-Supreme-5ms/HT column of 30m length and 0.25mm internal diameter coated with a stationary phase film of 0.1µm thickness was used. The column was directly inserted into the ion source. Electron impact (EI) spectra were obtained at 70 eV with full scan from m/z 50 to 700 amu in the mass spectrometer (MS). Because there were no GC signals before the 10 min and over the 40 min retention times in the analysed samples; gas chromatograms shown in this paper were displayed in different ranges from 10 to 40 min in order to expand the chromatogram for clear showing and marking of the identified peaks.

3. RESULTS AND DISCUSSION

Organic residues retrieved from the fabrics of the six Iron Age II pottery sherds (Table 1) and their related soils and the modern pottery sherd were analysed using the GC-MS. Organic extracts of these samples were obtained via conventional solvent extraction of the soluble fraction and alkaline hydrolysis (saponification) of both the soluble fraction and the insoluble fraction of the organic residues preserved in these sherds and soils. GC-MS analysis revealed the presence of different classes of organic compounds in the archaeological sherds. These are free *n*-alkanoic fatty acids, *n*-alcohols, wax esters and dioic-, oxo- and dihydroxy- acids in addition to the glycerol and sitosterol. Three categories of organic residues were identified in the six sherds. Therefore, these sherds were divided into three groups based on the type of residues preserved in their fabrics. The results are discussed herein.

3.1 Group 1: Organic extracts of sherd Jn17:

GC-MS analysis of the conventional solvent extract of the interior of the first sherd (Jn17) revealed the presence of characteristic organic components. These are long-chain saturated wax monoesters with even-numbered carbon atoms ranging from 40 to 52 (WE_{40} - WE_{52}), long-chain *n*-alkanes with odd-numbered carbon atoms ranging from 23 to 33 (C_{23} - C_{33}) and long-chain saturated fatty acids with even-numbered carbon atoms: $C_{16:0}$ (palmitic), $C_{20:0}$ (arachidic), $C_{24:0}$ (lignoceric), $C_{26:0}$ (cerotic), $C_{32:0}$ (lacceroic) and $C_{34:0}$ (Geddic), these are shown in Fig. 1. The total amount of the lipid retrieved from the sherd was about 345 µg lipid per one gram of dry ceramic powder.

Detecting this lipid profile in the interior of this sherd informs the occurrence of beeswax. Many researchers have been reporting similar distributions of lipid profiles in various archaeological contexts and dating back to the Neolithic period. They attributed these profiles to beeswax origin (Evans and Heron, 1993; Heron et al., 1994; Charters et al., 1995; Evershed et al., 1997, 1999; Garnier et al., 2002; Regert et al., 2001, 2003a,b; Evershed et al., 2003; Regert 2004; Copley et al., 2005a; Mazar et al., 2008; Namdar et al., 2009; Parras et al., 2011; Mayyas et al., 2012).

In modern beeswax, the higher abundances of WE_{46} wax ester, $C_{24:0}$ fatty acid and C_{27} *n*-alkane compared to their counterparts are also characteristic of beeswax origin (Heron et al., 1994; Copley et al., 2001; Kimpe et al., 2002; Regert et al., 2001, 2003a, 2005; Garnier et al., 2002; Evershed et al., 2001: 334 and 2003; McGovern et al., 2004; Regert 2004; Mazar et al., 2008; Namdar et al., 2009). This pattern of the three biomarkers is still noticeable in the archaeological lipid profile detected in this study (Figs. 1 and 2); which implies that the beeswax is still well preserved in the archaeological pottery sherd. The absence of some lipid constituents and the differential depletion of others in the archaeological beeswax (Figs. 1 and 2) compared with the modern one is attributed to preferential degradation during pro-

cessing the raw material of beeswax and/or during vessel use and burial. For example, the absence of some *n*-alkanes and the differential depletion of the others could be attributed to intentional human heating during processing beeswax in that time (Regert et al., 2001). The presence of $C_{16:0}$ fatty acid together with the long-chain even-numbered alcohols (AL_{28} and AL_{30}) in the same lipid extract implies that a partial hydrolysis of wax esters was occurred through time (Charters et al., 1995; Evershed et al., 1997; Regert et al., 2001; Garnier et al., 2002). In addition, the absence of both hydroxyl wax esters and other some fatty acids, originally present in the raw beeswax, and the depleted pattern of the remaining fatty acid profile, can be attributed to their loss by leaching with water during vessel use and burial.

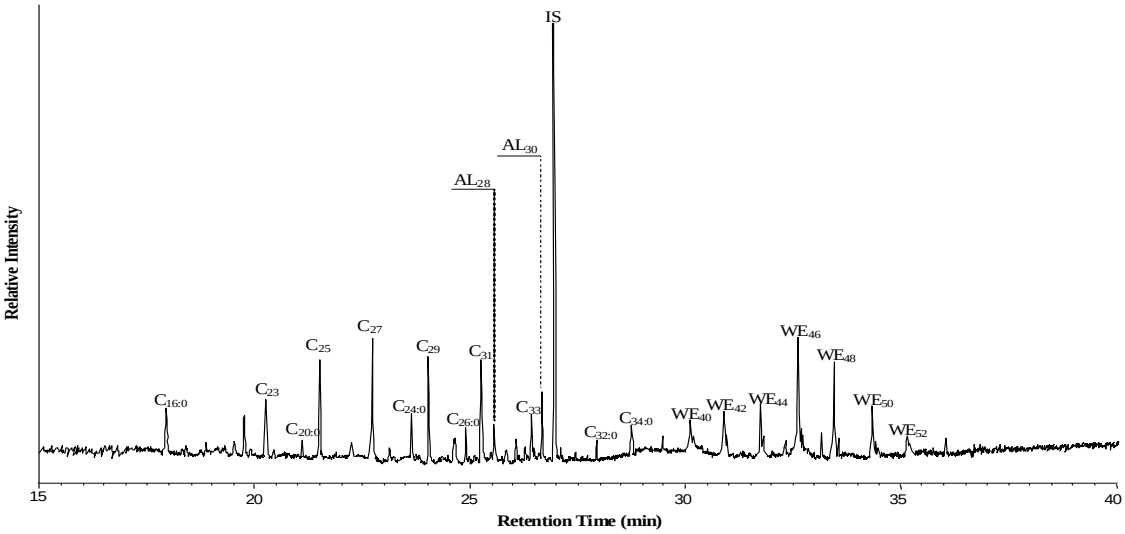

Figure 1. Partial (15-40 min) total-ion-chromatogram (TIC) of the trimethylsilylated total lipid extract of the organic residue obtained via conventional solvent extraction from the interior of sherd Jn17. ($C_{x:y}$): monocarboxylic fatty acid of carbon chain length x and degree of unsaturation y, (C_x): *n*-alkane of carbon chain length x, (WE_x): Wax ester of carbon chain length x, (AL_x): *n*-alcohol of carbon chain length x and (IS): the internal standard (*n*-tetratriacontane = C_{34}).

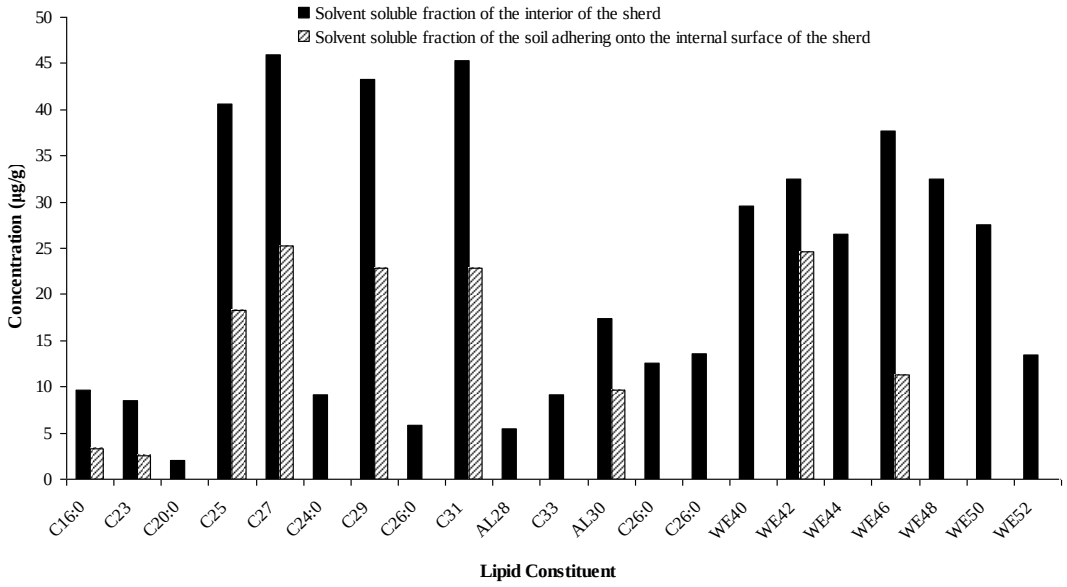

Figure 2. Histogram showing a comparison between the relative concentrations of main lipid constituents obtained via conventional solvent extraction from the interior of sherd Jn17 and its related soil. (Cx:y): monocarboxylic fatty acid of carbon chain length x and degree of unsaturation y, (Cx): n-alkane of carbon chain length x, (WEx): Wax ester of carbon chain length x and (ALx): n-alcohol of carbon chain length x.

Although of the modifications occurred to the archaeological beeswax identified in this study the detection of these distributions of lipid components in the lipid extract clearly indicates preservation of beeswax in the interior of the sherd. It is known that biomarkers of beeswax are stable and resistant to

degradative processes; therefore, they have been detected in many archaeological and art historical contexts (Heron et al., 1994; Mills and White, 1994: 49; Regert et al., 2001, 2003a, 2005; Bonaduce and Colombini, 2004; Lattuati-Derieux et al., 2009). It is known that fired clay can theoretically act as a molecular sieve or trap for organic biomolecules during vessel use and burial over a long period of time (Evershed et al., 1999). The preservation of organic materials including beeswax in pottery vessels can be attributed to the favourable environmental conditions. Jneneh site has a dry environment most of the year but cool and wet environment during winter.

Beeswax was detected in various archaeological pottery vessels found throughout the world. Its occurrence has been discussed and attributed to different purposes, such as sealing/waterproofing agent and insect repellent (Needham and Evans, 1987; Heron et al., 1994; Charters et al., 1995; Panagiotakopulu et al., 1995; Evershed et al., 1997; Aveling and Heron, 1999; Regert et al., 2001; Garnier et al., 2002; Regert et al., 2003a; Regert, 2004; Copley et al., 2005a; Mayyas et al., 2012; Namdar et al., 2009; Baeten et al., 2010). However, functions of these pottery vessels, have been attributed to the fact that the vessels were exploited in the past for different uses, such as storage (Crane, 1983: 45-47; Evershed, 1993; Heron et al., 1994; Evershed et al., 1997, 1999, 2003; Humphrey et al., 1998; Regert et al., 2001; Anderson-Stojanović and Jones, 2002; Garnier et al., 2002; Kimpe et al., 2002; Regert et al., 2003a; McGovern et al., 2004; Copley et al., 2005a,b,c; Knappett et al., 2005; Mayyas et al., 2012; Mazar and Panitz-Cohen, 2007; Mazar et al., 2008; Baeten et al., 2010).

Archaeologically, sherd Jn17 is a body part of a large storage jar with collared decoration on top of the shoulder. This jar was destroyed and found in a destruction layer, on top of the latest surface floor, of a small store room (B1:R3), adjacent to an open courtyard with a hearth. This type of jar was very common in the Iron Age II in southern Levant and was commonly used as a general storage container for multiple commodities and substances (Herr, 2001). In addition, it had been used as a transport jar and sometimes as a burial container (Killebrew, 2001). It was also suggested that collared-rim jars served two initial purposes (Ibrahim, 1978: 122–24): the first, as standard containers for olive oil, some of which would be sent off as trade goods with approximate capacity of 150-200 litres of liquid; and the second, for storage of potable water, resembling the present-day *zirs*. No any biomarker of any commodity or substance was detected in this sherd except those of beeswax.

3.2 Group 2: Organic extracts of sherds Jn21, Jn23 and Jn24

GC-MS results obtained from the analysis of the conventional solvent extracts of the interiors of the three sherds (Jn21, Jn23 and Jn24) show the preservation of lipid profiles different from that of sherd Jn17. These profiles are almost identical and composed of long-chain *n*-alkanes with odd-numbered carbon atoms ranging from 25 to 33 (C_{25} - C_{33}) and long-chain saturated fatty acids with even-numbered carbon atoms: $C_{16:0}$ (palmitic), $C_{18:0}$ (stearic), $C_{26:0}$ (cerotic), $C_{28:0}$ (montanic) and $C_{30:0}$ (melissic), in addition to long-chain even-numbered alcohols (AL_{18}, AL_{26}, AL_{28}, AL_{30} and AL_{32}) (Fig. 3). The total amount of the lipid retrieved from the three sherds were about 95, 110, and 117 µg lipid per one gram of dry ceramic powder, respectively.

Lipid components detected in the three sherds occur widely in degraded insect waxes, such as beeswax, and plant epicuticular waxes (Kolattukudy, 1976; Evershed et al., 1999; Logan et al., 2001; Feng et al., 2006). The most important diagnostic and distinguishable biomarkers between plant waxes and beeswax are the wax esters, which are absent in these lipid extracts. However, the *n*-alkane with 27 carbon atoms is the most prominent in beeswax (Regert et al., 2001, 2003a; Evershed et al., 2003), whilst that with 29 carbon atoms is the most prominent in epicuticular waxes of higher plants (Copley et al., 2001; McGovern et al., 2004). This could be discerned from both chromatographic (Fig. 3) and histogram (Fig. 4) profiles.

The trend of strong even carbon number predominance of *n*-alcohols maximizing at AL_{26} and strong odd carbon number predominance of *n*-alkanes maximizing at C_{29} imply biogenic sources, such as plant wax, and not a fossil fuel source (Logan et al., 2001; Feng et al., 2005 and 2006). It was reported that the AL_{26} or AL_{28} is often the predominant *n*-alcohol in the terrestrial vascular plant waxes (Bull et al., 2000) and AL_{26} is well-known abundant constituent in many grasses (van Bergen et al., 1998). *n*-alcohols with carbon numbers lower than 20, herein AL_{18}, could have originated from hydrolysis of wax esters of plant origin, which possibly occurs during diagenesis, but this does not rule out the microbial input as a source of these alcohols (Killops and Killops, 1993: 154; Feng et al., 2005). These lipid profiles, however, most probably originated from unknown plant remains usually retained in the soils, although, the occurrence of low abundances of $C_{16:0}$ and $C_{18:0}$ could be hydrolytic products of wax esters in beeswax.

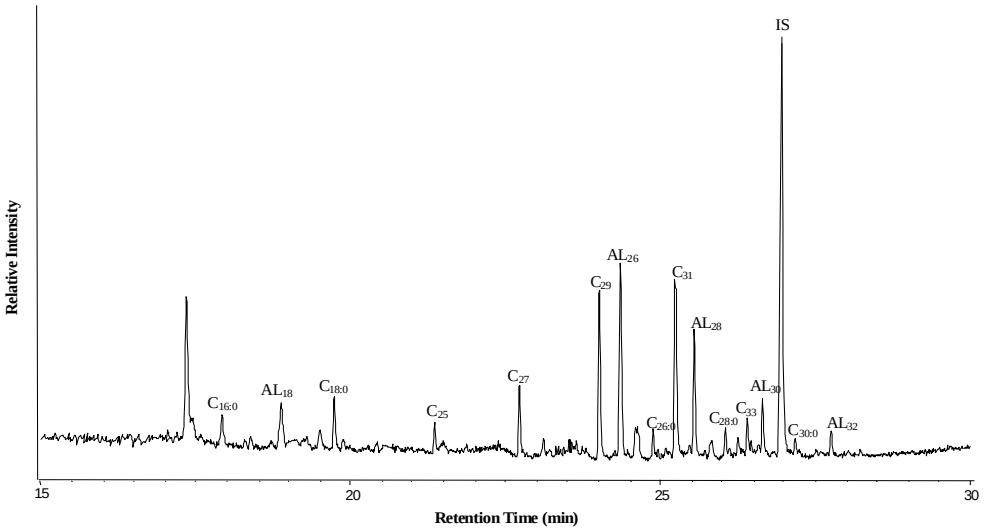

Figure 3. Partial (15-30 min) total-ion-chromatogram (TIC) of the trimethylsilylated total lipid extract of the organic residue obtained via conventional solvent extraction from the interiors of sherds Jn21, Jn23 and Jn24. ($C_{x:y}$): monocarboxylic fatty acid of carbon chain length x and degree of unsaturation y, (C_x): *n*-alkane of carbon chain length x, (AL_x): *n*-alcohol of carbon chain length x and (IS): the internal standard (*n*-tetratriacontane = C_{34}).

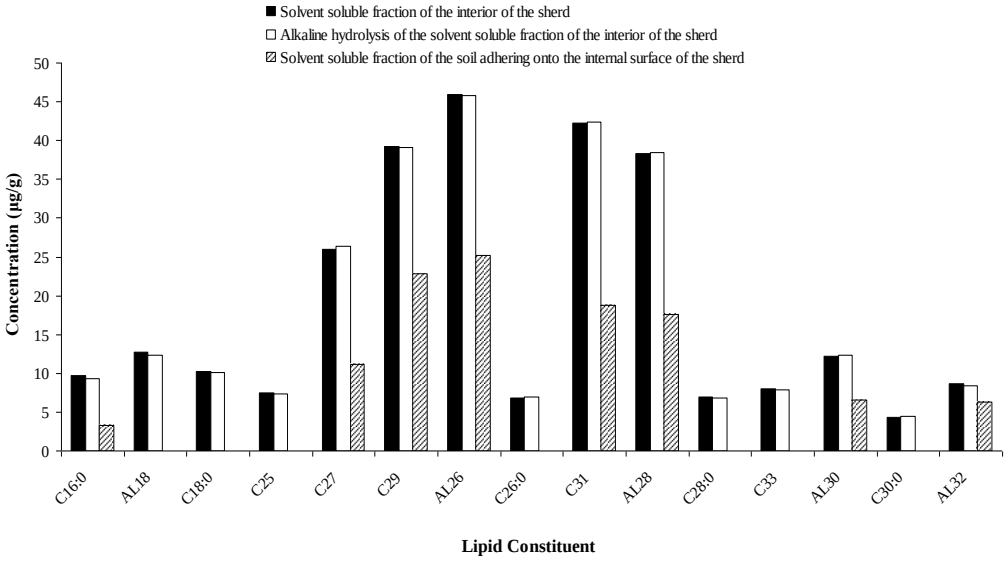

Figure 4. Histogram showing a comparison between the relative concentrations of main lipid constituents obtained via conventional solvent extraction from the interiors of sherds Jn21, Jn23 and Jn24 and their related soils. ($C_{x:y}$): monocarboxylic fatty acid of carbon chain length x and degree of unsaturation y, (C_x): *n*-alkane of carbon chain length x and (AL_x): *n*-alcohol of carbon chain length x.

GC-MS analysis of the alkaline hydrolysis extracts of the insoluble pottery residues of sherds Jn21, Jn23 and Jn24 remained after solvent extraction revealed the preservation of weakly bound lipid constituent, including $C_{16:0}$ and $C_{22:0}$ fatty acids and AL_{18}, AL_{26} and AL_{28} alcohols in addition to C_{33} *n*-alkane and sitosterol (Fig. 5). Sitosterol is the most common and abundant plant phytosterol (Rogge et al., 2006). This may support the evidence that these residues originated from a botanical source rather than beeswax or microorganisms. In addition, analysis of the lipid constituents and their abundances obtained from the alkaline hydrolysis of the solvent extract portion revealed that they were identical to those obtained from the direct analysis of solvent extract portion (Fig. 4). This informs that there are no hydrolysable constituents with ester bonds in the soluble fraction that could not be detected by the GC-MS. The phthalate plasticizers (P) with m/z 149 are modern contaminants resulting from plastics. They could also have been introduced into the samples from plastic bags during storing of the pottery sherds or during laboratory preparations.

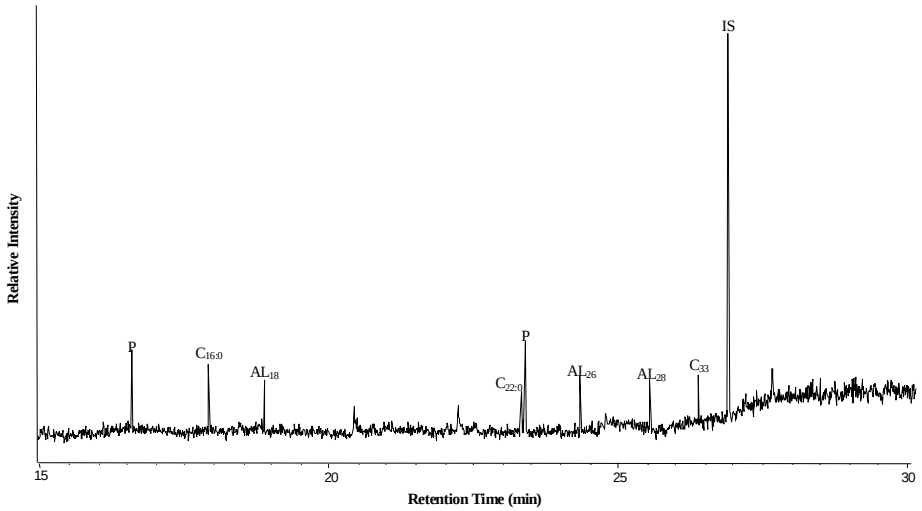

Figure 5. Partial (15-30 min) total-ion-chromatogram (TIC) of the trimethylsilylated total lipid extract of the organic residue obtained via alkaline hydrolysis (saponification) of the insoluble fraction of the interiors of sherds Jn21, Jn23 and Jn24. ($C_{x:y}$): monocarboxylic fatty acid of carbon chain length x and degree of unsaturation y, (C_x): *n*-alkane of carbon chain length x, (AL_x): *n*-alcohol of carbon chain length x, (P): plasticizer and (IS): the internal standard (*n*-tetratriacontane = C_{34}).

Archaeologically, sherds Jn21 and Jn23 are base parts from vessels similar to that of Jn17, which represents a large storage jar with its collared decoration on top of the shoulder. Sherd Jn24 is an upper part of a medium jar with oval horizontal knob handle. It was found in the same archaeological context as Jn17, Jn21 and Jn23 inside a small store room (B1:R3). This type of jar was used for storage purposes with more possibility for liquid contents.

3.3 Group 3: Organic extracts of sherds Jn6 and Jn15

GC-MS analysis of the solvent soluble extract of sherds Jn6 and Jn15 revealed the preservation of other lipid constituents (Fig. 6); most of them are different from those detected in the extracts of the other samples. The main constituents are monounsaturated $C_{18:1}$ (oleic) fatty acid followed by $C_{16:0}$ (palmitic) fatty acid. Other fatty acids: $C_{9:0}$ (pelargonic), $C_{10:0}$ (capric), $C_{14:0}$ (myristic), diunsaturated $C_{18:2}$ (linoleic) and $C_{18:0}$ (stearic) $C_{20:0}$ (arachidic) acids, oxidation products: α,ω-nonanedioic (azelaic), α,ω-decanedioic (sebacic), 10-oxo-octadecanoic and 9,10-dihydroxyoctadecanoic (9,10-diOH-$C_{18:0}$) acids, in addition to the major plant sterol biomarker, sitosterol, were also detected. The total amount of the lipids retrieved from the two sherds are about 185 and 218 μg lipid per one gram of dry ceramic powder.

Saturated $C_{16:0}$ and $C_{18:0}$ fatty acids have a wide distribution and are abundant in plant oils and animal fats (Dudd and Evershed, 1998). Monounsaturated $C_{18:1}$ fatty acid is also commonly present in most plant and animal lipids (Dey and Harborne, 1997: 238; Copley et al., 2005a; Gunstone et al., 2007: 4). Very small proportions of $C_{18:0}$ and higher contents of $C_{18:1}$ and $C_{18:2}$ are present in vegetables and their oils, cereals and fruits (Kimpe et al., 2004). The higher contents of $C_{18:1}$ acid compared with the other acids, particularly $C_{18:0}$, and the occurrence of sitosterol, suggests a plant origin, possibly plant oil. In addition, 9,10-diOH-$C_{18:0}$ acid, detected in the extracts of the two sherds, were observed in the replica olive and seed oil lamps upon burning (Regert et al., 1998).

The occurrence of $C_{9:0}$ (pelargonic), α,ω-nonanedioic, α,ω-decanedioic, 10-oxo-octadecanoic and 9,10-dihydroxyoctadecanoic acids is attributed to the oxidation of the unsaturated moieties of $C_{18:1}$ fatty acids present in the extract (Evershed et al., 1999; Reber and Evershed, 2004a,b; Colombini et al., 2005; Copley et al., 2005b; Boran et al., 2006; Spangenberg et al., 2006; Mayyas et al., 2013). The higher concentration of $C_{9:0}$, compared to those of the short chain fatty acids ($C_{10:0}$ and $C_{14:0}$) in the extracts of sherds Jn6 and Jn15 (Figs. 7), can be attributed to the oxidation degree of $C_{18:1}$ into the oxidative products, including $C_{9:0}$ during burial (Shimoyama et al., 1995). However, the oxidation has not yet been completed. The major plant sterol, sitosterol, also detected in the two extracts, supports the plant origin. Therefore, the occurrence of this lipid profile in the extracts of the interiors of sherds Jn6 and Jn15, suggests a residue of plant origin, possibly plant oil. This result can be compared and supported with the results obtained by Copley et al. (2005a), Colombini et al. (2005 and 2009), Ribechini et al. (2009) and Giorgi et al. (2010). Similar profiles of fatty acids and oxidation products were also obtained from ceramic vessels

tested by Knappett et al. (2005) and Giorgi et al. (2010). Their investigations suggest that the lipid profiles of the residues have originated from plant oil.

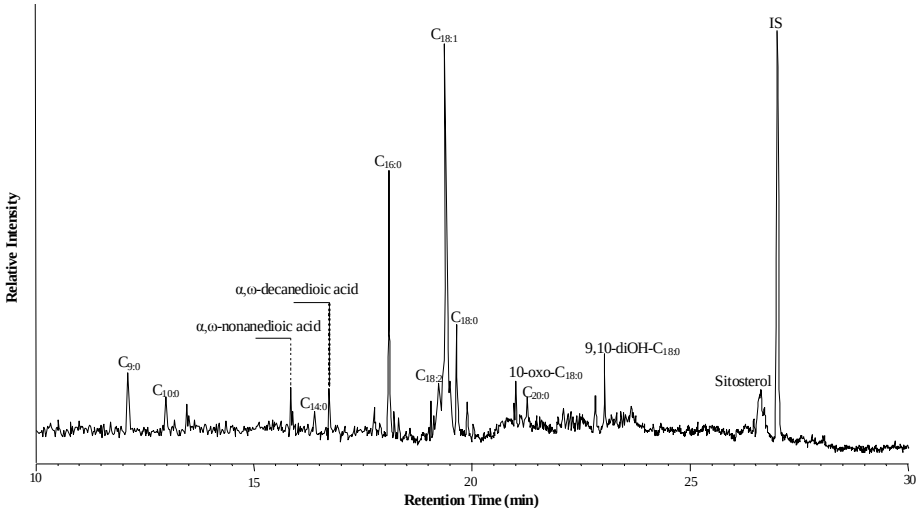

Figure 6. Partial (10-30 min) total-ion-chromatogram (TIC) of the trimethylsilylated total lipid extract of the organic residue obtained via conventional solvent extraction from the interiors of sherds Jn6 and Jn15. ($C_{x:y}$): monocarboxylic fatty acid of carbon chain length x and degree of unsaturation y and (IS): the internal standard (*n*-tetratriacontane = C_{34}).

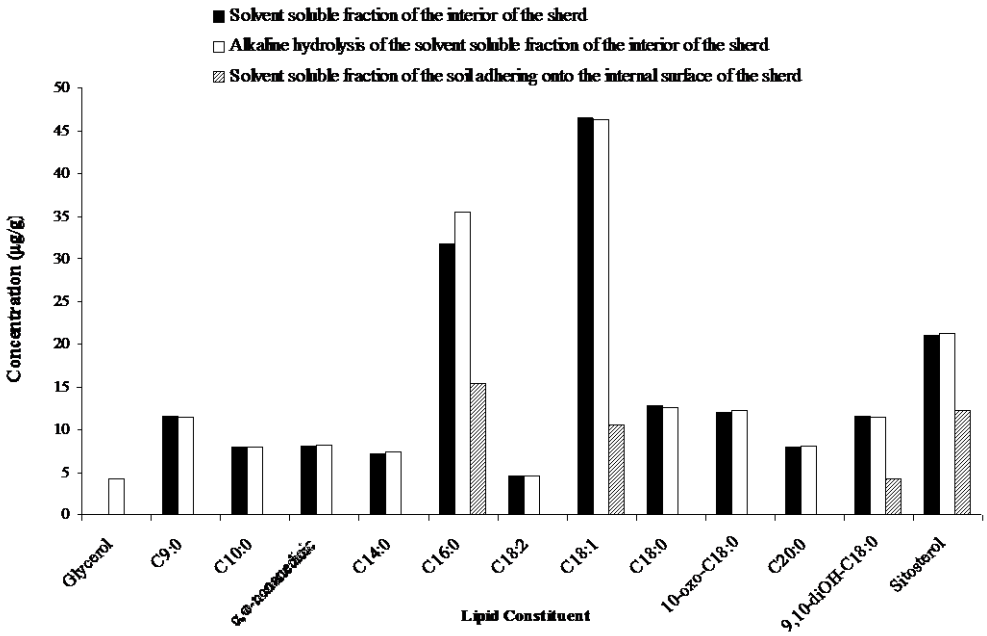

Figure 7. Histogram showing a comparison between the relative concentrations of main lipid constituents obtained via conventional solvent extraction from the interiors of Jn6 and Jn15 and their related soils. ($C_{x:y}$): monocarboxylic fatty acid of carbon chain length x and degree of unsaturation y, (C_x): *n*-alkane of carbon chain length x, (AL_x): *n*-alcohol of carbon chain length x.

GC-MS analysis of the alkaline hydrolysis extract, of the insoluble pottery residues remained after solvent extraction of the two sherds, Jn6 and Jn15, revealed the preservation of weakly bound lipid constituents, including $C_{9:0}$, $C_{16:0}$, $C_{18:1}$, $C_{18:0}$ and $C_{20:0}$ fatty acids, in addition to 9,10-diOH-$C_{18:0}$ acid with predominant $C_{16:0}$ and $C_{18:1}$ fatty acids (Fig. 8). These constituents more than likely originated from the same source of those detected in the solvent extract. The higher content of $C_{16:0}$ and $C_{18:1}$ acids, compared with $C_{18:0}$ acid and the presence of sitosterol, suggests a plant origin, possibly plant oil. Analysis of the lipid constituents obtained from the alkaline hydrolysis of the solvent extract portion revealed that they were identical to those obtained from the direct analysis of solvent extract portion, except that a new constituent (glycerol) and some increment in the concentration of $C_{16:0}$ were observed (Fig. 7). This

implies that a small amount of hydrolysable triacylglyceride of $C_{16:0}$ was present in the soluble fraction, but was not detected by the GC-MS. When alkaline hydrolysis was carried out on the soluble fraction, the triglyceride ester bonds were hydrolysed into free $C_{16:0}$ fatty acid and glycerol. It is known that $C_{16:0}$ and $C_{18:0}$ fatty acids are relatively abundant in most natural lipids and more stable to degradation processes relative to other fatty acids particularly those of unsaturated moieties. The detection of $C_{16:0}$ and $C_{18:1}$, but not $C_{18:0}$, in the alkaline extracts (Fig. 7) provides evidence of the high content of $C_{16:0}$ and very likely unsaturated fatty acids, rather than saturated $C_{18:0}$. Consequently, this indicates that the original raw material is most probably plant material and most probably plant oil and not animal fat. The occurrence of phthalate plasticizers (P) was discussed above.

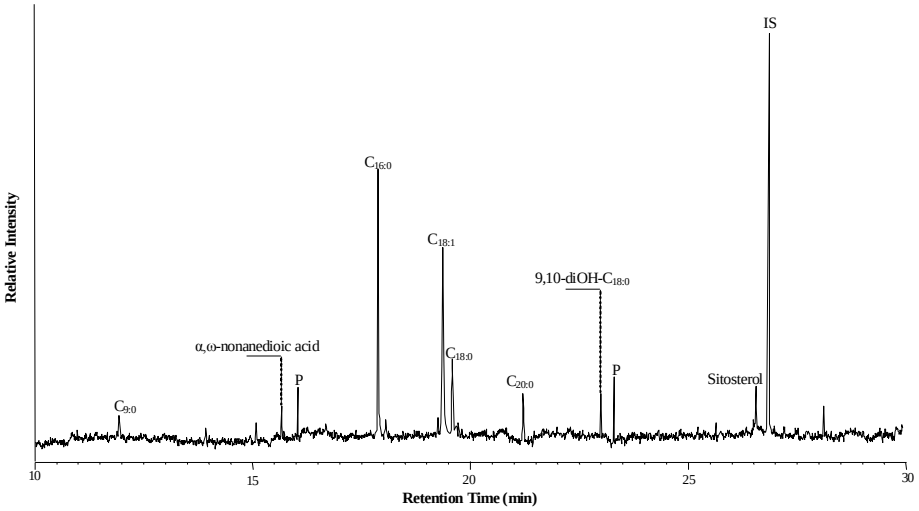

Figure 8. Partial (10-30 min) total-ion-chromatogram (TIC) of the trimethylsilylated total lipid extract of the organic residue obtained via alkaline hydrolysis (saponification) of the insoluble fraction of the interiors of sherds Jn6 and Jn15. ($C_{x:y}$): monocarboxylic fatty acid of carbon chain length x and degree of unsaturation y, (P): plasticizer and (IS): the internal standard (*n*-tetratriacontane = C_{34}).

Archaeologically, sherd Jn6 is an upper part of a medium jug. It was found inside a fireplace in an open courtyard. Since the main function of such jug is a liquid container it seems that its context is a secondary. Sherd Jn15 is the base of a small storage jar. It was uncovered in a small store room (B1:R3) together with several variant sizes storage jars. The shape and the small opining diameter of the jar indicate that this jar was used for liquid contents and more possible for precious liquid.

3.4 Organic extracts of the adhering soil

3.4.1 Adhering soil on sherd Jn17

The possible contamination from soil can be excluded and the migration of lipid residue from the interior of ceramic sherds to the attached soil on their surfaces can be proved. Soil sample adhering onto the internal surface of sherds Jn17 preserve $C_{16:0}$ fatty acid, C_{25} and C_{29} *n*-alkanes, AL_{30} *n*-alcohols and WE_{44} wax, each with abundance lower than that detected in the interior of the sherd (Fig. 2).

3.4.2 Adhering soil on sherds Jn21, Jn23 and Jn24

Soil samples adhering onto the internal surfaces of sherds Jn21, Jn23 and Jn24 preserve $C_{16:0}$ fatty acid, C_{29} and C_{31} *n*-alkanes, AL_{26} and AL_{28} *n*-alcohols. However, the concentrations of the significant lipid constituents detected in soil extracts were compared with those detected in the lipid extracts of the interior of each sherd (Fig. 4).

3.4.3 Adhering soil on sherds Jn6 and Jn15

Soil sample adhering onto the internal surfaces of sherds Jn6 and Jn15 preserve $C_{16:0}$, $C_{18:1}$, and 9,10-diOH-$C_{18:0}$ acids, in addition to the sitosterol, each having concentration in the soil lower than that in the interior of the sherd (Fig.7).

The comparison shown in Figures 2, 4 and 7 clearly inform that concentrations of the lipid constituents preserved in the interior of each sherd are higher than those preserved in the soil adhering onto the internal surface of the same sherd. This demonstrates that lipid constituents were originally present in the ceramic fabric of the sherd, mainly in the interior, and after that, they had permeated to the soil attached onto the internal surface during burial.

4. CONCLUSIONS

This work represents an initial study demonstrates on the identification and characterization of

organic biomarkers preserved in pottery sherds collected at the Iron Age II site of Jneneh in north-central part of Jordan, in order to identify the origin of organic residues and the potential use of the pottery vessels exploited in that time. The results show that two categories of organic materials of different origins were used in that time in the pottery vessels tested in this study. The first is of insect origin (beeswax), and the second is of plant origin (oil and epicuticular wax). Three of the six sherds preserved significant and informative amounts of organic residues. These are the large storage jar (Jn17), the medium jug (Jn6) and the small jar (Jn15).

Based on the chemical analysis and archaeological interpretations, the occurrence of beeswax on the internal surface of sherd Jn17 is attributed to that the vessel could have been used for storing liquid products, including water, plant oil or wine. In addition, it is known that the presence of beeswax imply the availability of honey of bees (Heron et al., 1994; McGovern et al., 2004; Copley et al., 2005a,b,c), which could have been served as a sweetener. The occurrence of beeswax in this jar may also indicate the possible storing of honey with its beeswax honeycomb or fragments of the honeycomb left unintentionally inside the jar.

Archaeologically it is known that collared-rim jars (pithos) may have been essential for water storage for inhabitants who lived away from the source of water (Hawkins, 2013: 137-158). Residents of Jneneh, during the Iron Age II, had a very close permanent source of water, which is the Zarqa River. Also the mouth of the jar is small for using such jars for daily drinking water. These factors weaken the theory that the large storage jar was used for daily water usage; therefore, beeswax most probably was not used as waterproofing to retain water.

Coating the interior of a large storage jar is time consuming and costly, which indicates that the content of the storage jar was important and valuable. It could be honey, plant oil or wine. No biomarkers of any commodities or substances, such as honey, plant oil or wine were detected in this sherd except those of beeswax. Biomarkers of honey have rarely been reported in archaeological contexts and this can be attributed to their high susceptibility to degradation under most environmental conditions (Regert et al., 2003b). The jar could be a low usage vessel after it was coated with beeswax, which does not allow the vessel fabric to absorb liquid, which might be plant oil or wine. Otherwise a dry commodity, such as seeds, was stored in the vessel and the beeswax was used to protect the commodity from possible deteriorating and harm environments such as humidity and insects. Researchers might argue that the function of collared-rim jars was not confined to only one commodity (Raban, 2001). Therefore, the occurrence of beeswax with the absence of biomarkers of other commodities can not be linked with specific use.

The existence of plant oil in smaller vessels: a medium jug (Jn6) and a small jar (Jn15), show that these vessels might have been used for daily needs, such as food preparation, but not for storage purposes. The probability of this plant oil being olive oil is high, due to the fact that olive trees were utilized in the past, and this use continues presently. The past and present environment is conducive for olive oil production.

Organic biomarkers present in sherds Jn21, Jn23 and Jn24 have originated from unknown botanical source rather than beeswax or microorganisms. The absence of diagnostic organic constituents in these sherds can be attributed to the degradation overtime during burial.

Further studies focusing on other unwashed and unhandled pottery and soil samples, which should be freshly excavated from different contexts at the same site and from other contemporary neighbouring sites, are anticipated in the near future. Although it is not available in Jordan, compound specific carbon isotope analysis (^{13}C analysis) will also be conducted abroad for further investigations using freshly excavated pottery and soil samples.

ACKNOWLEDGEMENTS

The authors would like to acknowledge the Hashemite University for providing the laboratories of the Department of Conservation Science with the chemicals, glassware and equipments required for this study and the Princess Haya Biotechnology Center at Jordan University of Science and Technology for their collaboration in using the GC-MS technique for analysing the samples.

REFERENCES

Anderson-Stojanović, V. R., Jones, J. E. (2002) Ancient Beehives from Isthmia. *Hesperia,* Vol. 71, No 4, 345-376.

Aveling, E. M., Heron, C. (1999) Chewing Tar in the Early Holocene: An Archaeological and Ethnographic Evaluation. *Antiquity*, Vol. 73, 579-584.

Baeten, J., Romanus, K., Degryse, P., De Clercq, W., Poelman, H., Verbeke, K., Luypaerts, A., Walton, M., Jacobs, P., De Vos, D., Waelkens, M. (2010) Application of a Multi-Analytical Toolset to a 16th Century Ointment: Identification as Lead Plaster Mixed with Beeswax. *Microchemical Journal*, Vol. 95, 227-234.

Bonaduce, I., Colombini, M. P. (2004) Characterisation of Beeswax in Works of Art by Gas Chromatography–Mass Spectrometry and Pyrolysis–Gas Chromatography–Mass Spectrometry Procedures. *Journal of Chromatography A,* Vol. 1028, 297-306.

Boran, G., Karaçam, H. and Boran, M. (2006) Changes in the quality of fish oils due to storage temperature and time. *Food Chemistry*, Vol. 98, 693-698.

Bull, I. D., van Bergen, P. F, Nott, C. J., Poulton, P. R. and Evershed, R. P. (2000) Organic geochemical studies of soils from the Rothamsted classical experiments–V. The fate of lipids in different long-term experiments. *Organic Geochemistry,* Vol. 31, 389–408.

Charters, S., Evershed, R. P., Blinkhorn, P. W., Denham, V. (1995) Evidence for the Mixing of Fats and Waxes in Archaeological Ceramics. *Archaeometry*, Vol. 37, 113-127.

Colombini, M. P. and Modugno, F. (2009) Organic materials in art and archaeology. In *Organic Mass Spectrometry in Art and Archaeology*, M. P. Colombini and F. Modugno (eds.), John Wiley and Sons, Ltd, Chichester, pp. 3-36.

Colombini, M. P., Giachi, G., Modugno, F. and Ribechini, E. (2005) Characterisation of organic residues in pottery vessels of the Roman age from Antinoe (Egypt). *Microchemical Journal*, Vol. 79, 83-90.

Copley, M. S., Rose, P. J., Clapham, A., Edwards, D. N., Horton, M. C. and Evershed, R. P. (2001) Detection of palm fruit lipids in archaeological pottery from Qasr Ibrim, Egyptian Nubia. *Proceedings Biological Sciences / The Royal Society London B*, Vol. 268, 593-597.

Copley, M. S., Berstan, R., Straker, V., Payne, S., Evershed R. P. (2005a) Dairying in Antiquity. II. Evidence from Absorbed Lipid Residues Dating to the British Bronze Age. *Journal of Archaeological Science*, Vol. 32, 505-521.

Copley, M. S., Berstan, R., Dudd, S. N., Straker, V., Payne, S., Evershed R. P. (2005b) Dairying in antiquity. I. Evidence from Absorbed Lipid Residues Dating to the British Iron Age. *Journal of Archaeological Science*, Vol. 32, 485-503.

Copley, M. S., Berstan, R, Mukherjee, A. J., Dudd, S. N., Straker, V., Payne, S., Evershed R. P. (2005c) Dairying in Antiquity. III. Evidence from Absorbed Lipid Residues Dating to the British Neolithic. *Journal of Archaeological Science*, Vol. 32, 523-546.

Crane, E. 1983. *The Archaeology of Beekeeping.* Cornell University Press, Ithaca. New York.

Dey, P. M. and Harborne, J. M. (eds). (1997) *Plant biochemistry*, San Diego, London: Academic Press.

Dudd, S. N. and Evershed, R. P. (1998) Direct demonstration of milk as an element of archaeological economies. *Science*, Vol. 282, 1478-1480.

Dudd, S. N., Evershed, R. P., Gibson, A. M. (1999) Evidence for Varying Patterns of Exploitation of Animal Products in Different Prehistoric Pottery Traditions Based on Lipids Preserved in Surface and Absorbed Residues. *Journal of Archaeological Science*, Vol. 26, 1473-1482.

Eglinton, G. and Logan, G. A. (1991) Molecular Preservation. *Philosophical Transactions of the Royal Society of London. Series B*, Vol. 333, 315-328.

Evans, K. and Heron, C. (1993) Glue, disinfectant and chewing gum: natural products chemistry in archaeology. *Chemistry and Industry*, Vol. 12, 446-449.

Evershed, R. P. (1993) Biomolecular Archaeology and Lipids. *World Archaeology*, Vol. 25, No 1, 74-93.

Evershed, R. P. (2008) Organic Residues Analysis in Archaeology: The Archaeological Biomarker Revolution. *Archaeometry*, Vol. 50, No 6, 895-924.

Evershed, R. P., Charters, S., Quye, A. (1995a) Interpreting Lipid Residues in Archaeological Ceramics: Preliminary Results from Laboratory Simulations of Vessel Use and Burial. *Materials Research Society Symposium Proceedings*, Vol. 352, 85-95.

Evershed, R. P., Stott, A. W., Raven, A., Dudd, S. N., Charters, S., Leyden, A. (1995b) Formation of long-chain ketones in ancient pottery vessels by pyrolysis of acyl lipids. *Tetrahedron Letters*, Vol. 36, No 48, 8875–8878.

Evershed, R. P., Vaughan, S. J., Dudd, S. N., Soles, J. S. 1997. Fuel for Thought? Beeswax in Lamps and Conical Cups from Late Minoan Crete. *Antiquity*, Vol. 71, No 274, 979-985.

Evershed, R. P., Dudd, S. N., Charters, S., Mottram, H., Stott, A. W., Raven, A., Van Bergen, P. F., Bland, H. A. (1999) Lipids as Carriers of Anthropogenic Signals from Prehistory. *Philosophical Transactions of the Royal Society London B*, Vol. 354, 19-31.

Evershed, R. P., Dudd, S. N., Lockheart, M. J., Jim, S. (2001) Lipids in archaeology. In *Handbook of Archaeological Sciences*, D. R. Brothwell and A. M. Pollard (eds.), Wiley, Chichester, pp. 331-349.

Evershed, R. P., Dudd, S. N., Anderson-Stojanovic, V. R., Gebhard, E. R. (2003) New Evidence for the Use of Combed Ware Pottery Vessels as Beehives in Ancient Greece. *Journal of Archaeological Science*, Vol. 30, 1-12.

Feng, J., Hu, M., Chan, C. K., Lau, P. S., Fang, M., He, L. and Tang, X. (2006) A comparative study of the organic matter in PM2.5 from three Chinese megacities in three different climatic zones. *Atmospheric Environment*, Vol. 40, 3983-3994.

Feng, J., Chan, C. K., Fang, M., Hu, M., He, L. and Tang, X. (2005) Impact of meteorology and energy structure on solvent extractable organic compounds of PM2.5 in Beijing, China. *Chemosphere*, Vol. 61, 623-632.

Garnier, N., Cren-Olivé C., Rolando, C., Regert, M. (2002) Characterization of Archaeological Beeswax by Electron Ionization and Elctrospray Ionization Mass Spectrometry. *Analytical Chemistry*, Vol. 74, 4868-4877.

Giorgi, G., Salvini, L., Pecci, A. (2010) The meals in a Tuscan building yard during the Middle Age. Characterization of organic residues in ceramic potsherds. *Journal of Archaeological Science*, Vol. 37, 1453-1457.

Gregg, M. W. Banning, E. B., Gibbs, K. and Slater, G. F. (2009) Subsistence practices and pottery use in Neolithic Jordan: molecular and isotopic evidence. *Journal of Archaeological Science*, Vol. 36, 937-946.

Gunstone, F. D., Harwood, J. L. and Dijkstra A. J. (eds). (2007) *The Lipid Handbook*. 3rd edition. CRC Press, Boca Raton.

Hawkins, R. K. (2013) *How Israel Became a People*. Abingdon Press, Nashville.

Heron, C., Evershed, R. P., Goad, L. J. (1991) Effects of Migration of Soil Lipids on Organic Residues Associated with Buried Potsherds. *Journal of Archaeological Science*, Vol. 18, 641-59.

Heron, C., Nemcek, N., Bonfield K. M., Dixon D., Ottaway, B. S. (1994) The Chemistry of Neolithic Beeswax. *Naturwissenschaften*, Vol. 81, 266-269.

Herr, L. G. (2001) The history of the collard rim pithos at Tell el-Umeiry, Jordan. In *Studies in the Archaeology of Israel and Neighboring Lands in Memory of Douglas L. Esse*, S. R. Wolff (ed.), The Oriental Institute of the University of Chicago, Studies in Ancient Oriental Civilization, No. 59, Chicago, Illinois, pp. 237-350.

Humphrey, J. W., Oleson, J. P., and Sherwood, A. N. (1998) *Greek and Roman Technology: A Sourcebook* 137. Routledge, London.

Hübner, U. (1992) Die Ammoniter, Untersuchungen zur Geschichte, Kulture, und Relegion eines transjordanischen Volkes im 1. Jarhtausend v. Chr. Harrasowitz Verlag, Wiesbaden, Germany.

Ibrahim, M. M. 1978. The collared-rim jar of the Early Iron Age. In *Archaeology in the Levant*, R. Moorey and P. Parr (eds). Warminster: Aris and Phillips. pp. 117-124.

Isaksson, S. and Hallgren, F. (2012) Lipid residue analyses of Early Neolithic funnel-beaker pottery from Skogsmossen, eastern Central Sweden, and the earliest evidence of dairying in Sweden. *Journal of Archaeological Science*, Vol. 39, 3600-3609.

Killebrew, A. E. (2001) The collared pithos in context: A typological, technological, and functional reassessment. In *Studies in the archaeology of Israel and neighboring lands in memory of Douglas L. Esse*, S. R. Wolff (ed.), The Oriental Institute of the University of Chicago, Studies in Ancient Oriental Civilization, No. 59, Chicago, Illinois, pp. 377-398.

Killops, S. D. and Killops, V. J. (1993) *An introduction to organic geochemistry*, Longman Scientific and Technical, New York.

Kimpe, K., Jacobs, P. A., Waelkens, M. (2002) Mass Spectrometric Methods Prove the Use of Beeswax and Ruminant Fat in Late Roman Cooking Pots. *Journal of Chromatography A*, Vol. 968, 151-160.

Kimpe, K., Drybooms, C., Schrevens, E., Jacobs, P. A., Degeest, R. and Waelkens, M. (2004) Assessing the relationship between form and use of different kinds of pottery from the archaeological site Sagalassos (southwest Turkey) with lipid analysis. *Journal of Archaeological Science*, Vol. 31, No 11, 1503-1510.

Knappett, C., Kilikoglou, V., Steele, V., Stern, B. (2005) The circulation and consumption of Red Lustrous Wheel-made ware: petrographic, chemical and residue analysis. Anatolian Studies, Vol. 55, 25-59.

Kolattukudy, P. E. (1976) *Chemistry and biochemistry of natural waxes*, Elsevier, Amsterdam/ Kurzweil, A. and Todtenhaupt, D., 1990, *Teer aus Holz*, Berlin, Museumsdorf Düppel.

Lattuati-Derieux. A., Egasse, C., Regert, M., Chung, Y., Lavédrine, B. (2009) Case Study: Characterization and Degradation Pathways of Ancient Korean Waxed Papers. *Journal of Cultural Heritage*, Vol. 10, 422-427.

Logan, G. A., Fredericks, D. J., Smith. C. and Heggie, D. T. (2001) Sources of organic matter in Wallis Lake. *AGSO Research Newsletter*, Vol. 34, 15-20.

Mayyas, A. S., Stern, B., Coningham, R., Fazeli, H. and Gillmore, G. (2012) Beeswax preserved in a Late Chalcolithic bevelled rim bowl from the Tehran Plain, Iran. *The journal of the British Institute of Persian Studies*, Vol. 50, 13-25.

Mayyas, A., Douglas, K., Hoffmann, T., Thorenz, U., Bany Yaseen, I., and El-Khalili, M. (2013) Organic residues preserved in archaeological ceramics from the Early Bronze Age site of Khirbet Al-batrawy in north-central Jordan. *Mediterranean Archaeology and Archaeometry*, Vol. 13, No 2, 189-206.

Mazar, A., Panitz-Cohen, N. (2007) It is the Land of Honey: Beekeeping at Tel Rehov. *Near Eastern Archaeology*, Vol. 70, No 4, 202-219.

Mazar, A., Namdar, D., Panitz-Cohen, N., Neumann, R., Weiner, S. (2008) The Iron Age Beehives at Tel Rehov in the Jordan Valley: Archaeological and Analytical Aspects. *Antiquity*, Vol. 82, 629-639.

McGovern, P. E., Zhang, J., Tang, J., Zhang, Z., Hall, G. R., Moreau, R. A., Alberto Nuñez, A., Eric D. Butrym,E. D., Richards, M. P., Wang, C., Cheng, G., Zhao, Z. and Wang, C. (2004) Fermented beverages of pre- and proto-historic China. *The National Academy of Sciences of the USA (PNAS)*, Vol. 101, No 51, 17593-17598.

Mills, J. S., White, R. (eds.). (1994) *Organic Chemistry of Museum Objects*, 2nd edition. London, Butterworths.

Mottram, H. R., Dudd, S. N., Lawrence, G. J., Stott, A. W., Evershed, R. P. (1999) New chromatographic, mass spectrometric and stable isotope approaches to the classification of degraded animal fats preserved in archaeological pottery. *Journal of Chromatography* A, Vol. 833, 209-221.

Namdar, D., Neumann, R., Goren, Y., Weiner, S. (2009) The Contents of Unusual Cone-Shaped Vessels (Cornets) from the Chalcolithic of the Southern Levant. *Journal of Archaeological Science*, Vol. 36, 629-636.

Needham, S., Evans, J. (1987) Honey and Dripping: Neolithic Food Residues from Runnymede Bridge. *Oxford Journal of Archaeology*, Vol. 6, No 1, 21-28.

Panagiotakopulu, E., Buckland, P. C., Day, P. M. (1995) Natural Insecticides and Insect Repellents in Antiquity: A Review of the Evidence. *Journal of Archaeological Science*, Vol. 22, 705-710.

Parras, D. J., Sánchez, A., Ramos, N., Ariza, M. O. R., Tuñón, J. A. (2011) Identification of fats and beeswax in ceramic vessels of tomb 121 of Castellón Alto (Galera, Granada). *Coalition*, Vol. 22, 7-13.

Raban, A. (2001) Standardized collared-rim pithoi and short–lived settlements. In *Studies in the Archaeology of Israel and Neighboring Lands in Memory of Douglas L. Esse*, S. R. Wolff (ed.), The Oriental Institute of the University of Chicago, Studies in Ancient Oriental Civilization, No. 59, Chicago, Illinois, pp. 493-518.

Reber, E. A. and Evershed, R. P. (2004a) How did Mississippians prepare maize? The application of compound-specific carbon isotope analysis to absorbed pottery residues from several Mississippi valley sites. *Archaeometry*, Vol. 46, No 1, 19-33.

Reber, E. A. and Evershed, R. P. (2004b) Identification of maize in absorbed organic residues: A cautionary tale. *Journal of Archaeological Science*, Vol. 31, 399-410.

Regert, M. (2004) Investigating the History of Prehistoric Glues by Gas Chromatography-Mass Spectrometry. *Journal of Separation Science*, Vol. 27, 244-254.

Regert, M., Bland, H. A., Dudd, S. N., van Bergen, P. F. and Evershed, R. P. (1998) Free and bound fatty acid oxidation products in archaeological ceramic vessels. *Proceedings of the Royal Society of London B*, Vol. 265, 2027-2032.

Regert, M., Colinart, S., Degrand L., Decavallas, O. (2001) Chemical Alteration and Use of Beeswax through Time: Accelerated Ageing Tests and Analysis of Archaeological Samples from Various Environmental Contexts. *Archaeometry*, Vol. 43, No 4, 549-569.

Regert, M., Vacher, S., Moulherat, C., Decavallas, O. (2003a) Adhesive Production and Pottery Function During the Iron Age at the Site of Grand Aunay (Sarthe, France). *Archaeometry*, Vol. 45, No 1, 101-120.

Regert, M., Garnier, N., Decavallas, O., Cren-Olivé, C., Rolando, C. (2003b) Structural Characterization of Lipid Constituents from Natural Substances Preserved in Archaeological Environments. *Measurement Science and Technology*, Vol. 14, 1620-1630.

Regert, M., Langlois, J., Colinart, S. (2005) Characterisation of Wax Works of Art by Gas Chromatographic Procedures. *Journal of Chromatography A*, Vol. 1091, 124-136.

Ribechini, E., Orsini, S., Silvano, F. and Colombini, M. P. (2009) Py-GC/MS, GC/MS and FTIR investigations on LATE Roman-Egyptian adhesives from *opus sectile*: New insights into ancient recipes and technologies. *Analytica Chimica Acta*, Vol. 638, 79-87.

Rogge, W. F., Medeiros, P. M. and Simoneit, B. R. T. (2006) Organic marker compounds for surface soil and fugitive dust from open lot dairies and cattle feedlots. *Atmospheric Environment*, Vol. 40, 27–49.

Shimoyama, A., Kisu, N., Harada, K., Wakita, S., Tsuneki, A. and Iwasaki, T. (1995) Fatty acid analysis of archaeological pottery vessels excavated in Tell Matsuma, Syria. *Bulletin of the Chemical Society of Japan*, Vol. 68, 1565-1568.

Spangenberg, J. E., Jacomet, S. and Schibler, J. (2006) Chemical analyses of organic residues in archaeological pottery from Arbon Bleiche 3, Switzerland evidence for dairying in the late Neolithic. *Journal of Archaeological Science*, Vol. 33, 1-13.

Stern, B., Heron, C., Corr, L. Serpico, M. and Bourriau, J. (2003) Compositional variations in aged and heated pistacia resin found in late Bronze Age Canaanite amphorae and bowls from Amarna, Egypt. *Archaeometry*, Vol. 45, No 3, 457-469.

Van Bergen, P. F., Peakman, T. M., Leigh-Firbank, E. C., Evershed, R. P. (1997) Chemical Evidence for Archaeological Frankincense: Boswellic Acids and Their Derivatives in Solvent Soluble and Insoluble Fractions of Resin-Like Materials. *Tetrahedron Letters*, Vol. 38, No 48, 8409-8412.

Van Bergen, P. F., Bergen, A., Nott, C. J., Bull, I. D., Poulton, R. P. and Evershed, R. P. (1998) Organic geochemical studies of soils from the Rothamsted classical experiments-IV. Preliminary results from a study of the effect of soil pH on organic matter decay. *Organic Geochemistry*, Vol. 29, No 5-7, 1779-1795.