Received: 22 February 2022 | Accepted: 1 February 2023

DOI: 10.1111/arcm.12852

ORIGINAL ARTICLE

Toward an understanding of the exchange in ancient scented oils through organic residue analysis of Bronze Age Near Eastern ceramic bottles by GC-MS

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Abstract

This paper presents a gas chromatography-mass spectrometry (GC-MS) organic residue analysis (ORA) of samples extracted from five Early Bronze Age ceramic bottles excavated at the archaeological site of Küllüoba in Anatolia (modern Turkey), and the first attempt at directly analysing the content of this category of products. Our results show that various types of liquid have been contained in different bottles and identify the presence of dicarboxylic and oleic acids with a large amount of palmitic acid in most samples, suggesting that they may have mostly contained a plant-based oil. The presence of diterpenoids further shows the addition of ingredients such as conifer resin and other plant-derived products. Overall, the analytical results presented here indicate the exchange of scented oils in Anatolia already during the late third millennium BCE. The different organic residue profiles contained in different samples also suggest a range of different recipes for these products.

KEYWORDS

Anatolia, ceramic containers, Early Bronze Age, gas chromatography-mass spectrometry, oil exchange, organic residue analysis

Funding information The authors are grateful to the Scientific Research Projects Foundation of Selçuk University for the financial support of this work; and to the Bilecik Şeyh Edebali University Research Foundation for supporting Küllüoba Excavation Project Numbers 2019-01.BŞEÜ.04-03, and 2018-01.BŞEÜ.04-06.

INTRODUCTION

This article presents the results of the gas chromatography-mass spectrometry (GC-MS) organic residue analysis (ORA) extracted from ceramic bottles found at the archaeological site of Küllüoba in central Anatolia (ancient Turkey) (Figure 1). The size and shape of these vessels (cf. Figure 2)—in particular their narrow necks and mouths—indicates they contained liquids of some kind and belonged to a category of bottles/flasks produced between *c*.2700–2200 BCE. They have been ascribed in earlier publications to a broad and ill-defined class of Early Bronze Age ceramic vessels often referred to as 'Syrian bottles' in the literature (Efe, 2007). As described in more detail below, our typological analysis of ceramic bottles found at Küllüoba suggests that they are all imported from areas several hundred kilometres away. However, only some can be described as true 'Syrian bottles', while others were likely produced in Cilicia/northern Levant (modern southern Turkey).

These 'Syrian bottles' and related products are among the most recognizable archaeological markers of a series of interlocking exchange networks that existed between Anatolia, the northern Levant and northern Mesopotamia during the mid- to late Early Bronze Age (*c*.2700–2200 BCE) (Efe, 2007; Massa & Palmisano, 2018; Rahmstorf, 2006; Şahoğlu, 2005). Over the last 20 years, an increasingly large body of evidence indicates that such long-distance (up to 5000 km in size) networks existed at least since the fourth millennium BCE and involved the trade in raw or semi-worked products such as ivory, metals, semiprecious stones and other easily transportable, high-value/low-bulk commodities (Massa & Palmisano, 2018; Rahmstorf, 2006, 2011; Wilkinson, 2014). Specifically, it has been suggested that the 'Syrian bottles' may have contained scented oils or medicinal unguents (Mellink, 1989; Zimmermann, 2005), but this hypothesis has not been tested so far through an ORA.

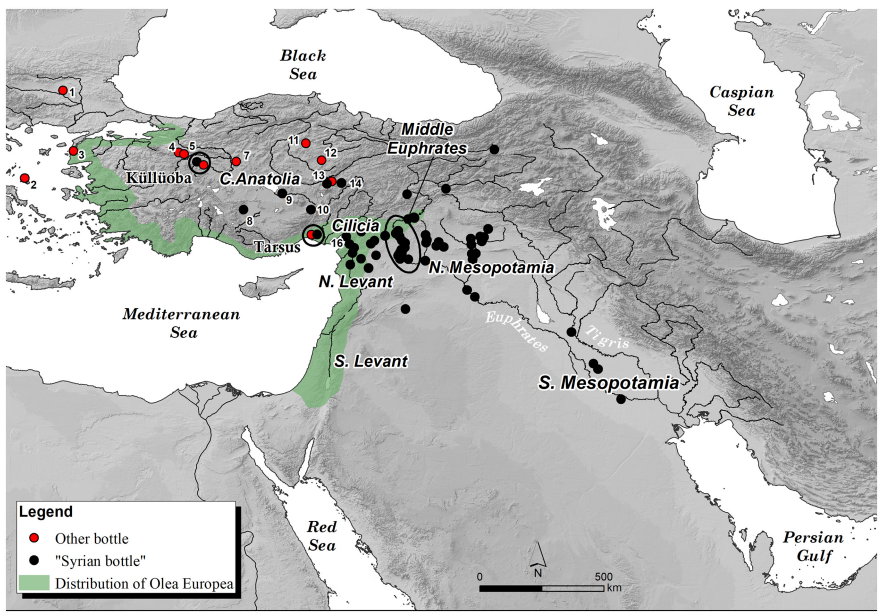

FIGURE 1 Distribution of 'Syrian bottles' and related bottle/flasks in the Near East during the period 2700–2200 BCE. Major cultural regions and archaeological sites mentioned in the text are also shown, together with the distribution of domesticated olive tree in the Middle to Late Holocene (for the latter, see Oflaz et al., 2019). Anatolian findspots of 'Syrian bottles': 6, Küllüoba; 8, Konya-Karahöyük; 9, Acemhöyük; 10, Kestel; 13, Kültepe; 14, Hazarşah; 15, Tarsus-Gözlükule; and 16, Kinet Höyük (data from Massa 2016). Anatolian findspots of other bottle types: 1, Galabovo; 2, Palamari; 3, Troy; 4, Küçükhöyük; 5, Demircihüyük; 6, Küllüoba; 7, Polatlı; 11, Eskiyapar; 12, Alişar Höyük; 13, Kültepe; and 15, Tarsus-Gözlükule (data from Massa 2016). For other findspots, see Massa and Palmisano (2018); and Rahmstorf (2006).

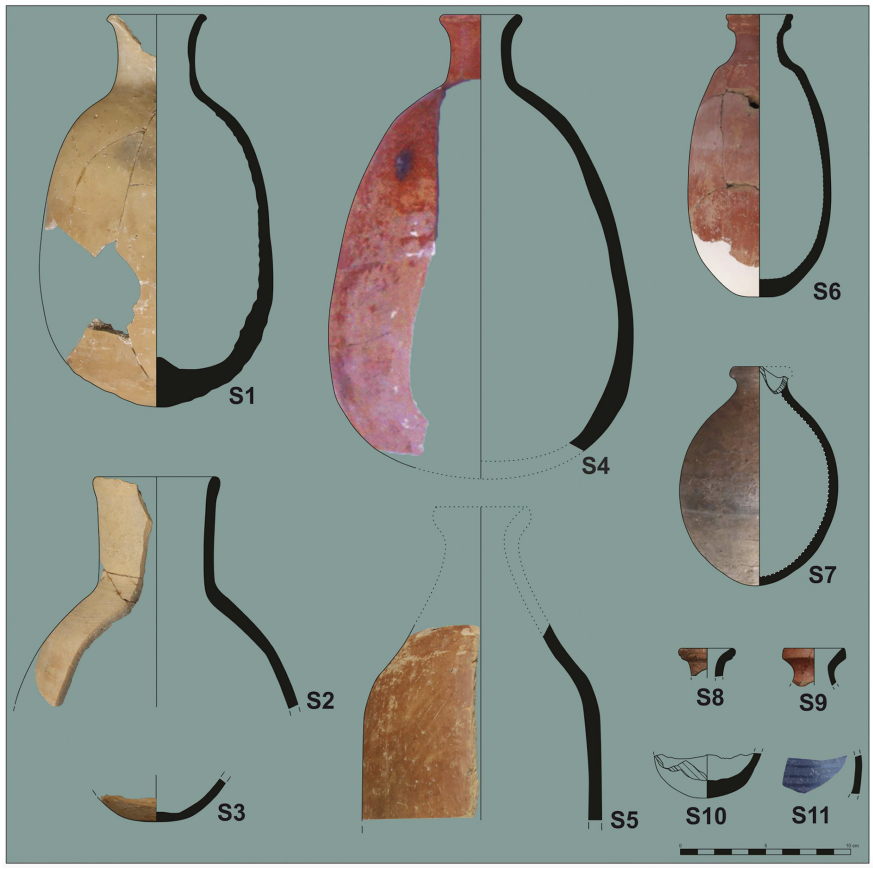

FIGURE 2 Bottle fragments from the site of Küllüoba (S1–S5 are analysed with GC-MS in this study).

To our knowledge, this study represents the first attempt to characterize the content of these bottles from an archaeometric perspective. The analysis by Ay et al. (2014), which showed the content of a very small container (6 × 3 cm) as a Mn oxide-based cosmetic paste, was purportedly on a 'Syrian bottle', but both size and shape (cf. Figure 2, d) do not fit in Sconzo's narrow typology of 'Syrian bottles' (Sconzo, 2014), nor more broadly in the category of perfume bottles.

To follow is a description of the samples under analysis, their typology and their retrieval context in order to shed light on their dating and possible function. We then present our methodological approach, followed by the results of the biochemical analysis. Lastly, we discuss these results in light of contemporary archaeological and textual evidence for products with similar characteristics across the Bronze Age Near East. Based on their chrono-typology, the biochemical results and the historical parallels, we suggest that the specimens from Küllüoba may be considered containers for scented oils and they currently represent the oldest archaeological evidence in the Near East for the exchange of this commodity.

THE ARCHAEOLOGICAL CONTEXT

The site of Küllüoba (39°55′83.65″N/30°74′32.54″E) is an ongoing excavation with a well-documented stratigraphic sequence spanning over 1300 years (*c*.3300–1950 cal BCE) (Efe & Ay-Efe, 2007; Türkteki, 2021). The settlement is about 4 ha in size and around 2650–2550 cal BCE (levels IVD–

IVC) is characterized by a compound of public buildings surrounded by domestic structures (Efe & Fidan, 2008). Several finds indicate the participation of Küllüoba in supra-regional trade networks, particularly between 2500 and 2200 BCE (Efe, 2007, 2020).

All vessels analysed here belong to level IIIA, datable to *c*.2450–2200 cal BCE (cf. the additional supporting information for radiocarbon dates). This phase is only preserved in what had been the central courtyard within the public buildings' compound (levels IVD–IVC) and is characterized by a palaeosurface dotted with large numbers of pits. These structures stand out from the hundreds of other domestic refuse pits at the site because they contain animal bones and fragmented drinking and eating vessels in quantities larger than average, as well as a variety of intact artefacts that seem intentionally deposited including pots, tools, ornaments, figurines and prestige goods. In several cases whole animal carcasses were also deposited in ways that may indicate a ritual significance, and pits were often lined with plaster or clay and closed after the deposition (Türkteki & Başkurt, 2016). These features are paralleled in several other contemporary sites in the region and seem to represent the last stage of a feasting event with some ritual significance (Türkteki & Başkurt, 2016). All ceramic bottles under analysis were retrieved from such pits and were thus probably related to the feasting events.

THE CERAMIC BOTTLES

The bottles found at Küllüoba (Figure 2) are all produced in ceramic fabrics that are otherwise not documented at the site (cf. the additional supporting information for ware description), and in shapes that are not otherwise found at Küllüoba or other central Anatolian sites. They were also manufactured with techniques alien to the local ceramic production, including the use of the fast potter's wheel and the employment of complex pyrotechnology (Türkteki, 2010).

The former excavation director originally described all these ceramic containers as 'Syrian bottles' (Efe, 2007). While this term was originally employed only for ceramic products originated along the Middle Euphrates (cf. Figure 1) (Kühne, 1976; Orthmann & Rova, 1991; Schachner & Schachner, 1995), it has subsequently and misleadingly been applied also to vessels with similar function but different shapes found in Anatolia, the Aegean and South Eastern Balkans (Efe, 2007; Leshtakov, 2002; Sconzo, 2014). In the most detailed chrono-typological treatment to date, Sconzo (2014) defined a narrow set of characteristics that describe true 'Syrian bottles', that is, products that can be confidently attributed to workshops in the Middle Euphrates region:

- They are generally made in luxury wares (i.e., very fine and thin-walled fabric without visible inclusions, well-fired in controlled firing environments likely by professional ceramicists).
- Are characterized by small sizes (generally between 0.3 and 1.5 L in volume).
- They are handleless, footless, with a rounded or pointed base, very thin-walled with a short neck and an everted rim.
- Are produced in four main types: two with globular bodies (types 1 and 2) and two with more elongated bodies (types 3 and 4).

An analysis of shapes, fabrics and surface treatments (cf. the additional supporting information for a detailed description) suggests that S6–S9 may belong the so-called 'Syrian bottles'(cf. Sconzo, 2014, types 2, 3). The same origin can be suggested for S11: despite being very fragmentary, the peculiar ring-burnished surface treatment confidently makes it part of the so-called Black Euphrates Banded Ware (Sconzo, 2015). On the other hand, several other bottles from Küllüoba seem to pertain to other ceramic productions, perhaps spatially closer to central Anatolia. For instance, S4 finds its best parallel at Tarsus-Gözlükule in Cilicia (Goldman, 1956), and the same region might be considered as the origin of S1 and S2. For S3 and S5 there is not enough preserved profile to understand the typology, but both are not local to the Küllüoba repertoire. Examples that we are confident are not 'Syrian bottles' (S1, S2, S4 and S5) are in general also much bigger than those that are likely originated along

the Middle Euphrates (Figure 2, S6–S9, S11), containing between 1.2–2.7 and 0.3–0.6 L, respectively (cf. the additional supporting information for data on volume). As argued below in the discussion, both the typological and the size differences in the Küllüoba dataset might be reflected in their different contents.

MATERIALS AND METHODS

Sample preparation

The choice of the samples to submit to ORA was partly dependent on the sherd's size and possibility to break portions of it, and partly on their accessibility for analysis. S8–S11 were too small for destructive analysis, and some of the intact/reconstructable vessels are stored at Eskişehir Museum, from which we could not secure the permit for analysis (S6 and S7). This left five samples of the 11 available bottles from Küllüoba (Figure 2) to be selected for ORA, and unfortunately we were not able to select any of the vessels that we securely identified as 'Syrian bottles', though we suggest that S3 might in fact have been one. The additional supporting information provides documentation regarding their archaeological context and their manufacturing details.

After removing about 0.5 mm of each fragment's surface to avoid contamination from soil and fingerprints, a sample weighing about 4 g was taken from the rim and body parts of the artefacts, which were more likely to contain ORs (Copley et al., 2003; Eerkens, 2005; Evershed et al., 2002, 2008; Leitch et al., 2016; Olsson & Isaksson, 2008; Regert, 2011; Vykukal et al., 2021). The sample size (4 g for two extraction) and the injection volume (5 µl) were selected in order to increase the detection of the molecules with low concentrations in the samples. The samples were then ground using an agate mortar and prepared for total lipid extraction. All solvents and chemicals used (obtained from Sigma-Aldrich, Darmstadt, Germany) were of analytical and chromatographic grades and employed without further purification. Glassware was washed three times in a solvent, and nitrile gloves were worn at all times.

Acid-catalysed extraction

A one-step, acid-catalysed, direct extraction–methylation method, which has been successfully applied by many researchers analysing the lipid residues in ancient ceramics, especially from South East Europe and the Middle East (Leclerc et al., 2018; Mileto et al., 2017; Oras et al., 2017; Papakosta et al., 2015, 2019), was employed for the samples under study. Lipid residues were extracted and methylated, following the protocol described by Papakosta et al. (2019) and Correa-Ascencio et al. (2014), with some modifications. According to the method, 2 g of the sample containing 50 µl of *n*-tetratriacontane (1000 mg/l) added as internal standard were heated with 6 ml of a mixture of MeOH and 98% H_2SO_4 (5:1, v:v) at 70°C for 4 h, and then cooled. Lipids were extracted with *n*-hexane (3 × 2 ml) and separated off after centrifugation (2500 rpm, 3 × 5 min).

Base-catalysed extraction

Since the method used for the extraction of lipid residues is not suitable for relatively more polar molecules such as wine biomolecules and minor compounds, the method proposed by Pecci et al. (2013) was used with minor modifications. According to the method, 2 g of sample were extracted with 1 M KOH (2 × 4 ml) in water in a bath at 70°C for 120 min. After cooling and centrifugation, the supernatant was acidified with 1 ml of 37% HCl and the polar molecules were extracted with ethyl acetate (2 × 4 ml), then separated off after centrifugation (2500 rpm, 2 × 5 min).

GC-MS analysis

n-Hexane and ethyl acetate phases collected for ORA were dried down under a gentle N_2 flow and further silylated with 100 µl of *bis*(trimethylsilyl)trifluoroacetamide (BSTFA) with 1% trimethylchlorosilane (TMCS) at 70°C for 1 h. The aim of the silylation process is to decrease the polarity of the analyte, to increase its stability and improve the chromatographic separation (Moldoveanu & David, 2019). The extract was dissolved in 100 µl of *n*-hexane and filtered through a polytetrafluoroethylene (PTFE) filter and analysed in the GC-MS system. The Agilent (CA, USA) 7890 N-5975C GC system, consisting of an MS detector and thermostated column oven, was employed for analysis. The operating conditions of the GC-MS system equipped with Agilent HP-5MS capillary column (30 m × 0.25 mm × 0.25 µm, [5%-phenyl]-methylpolysiloxane) were as follows: the temperature programme of the column oven was held at 50°C for 1 min, then increased at 5°C/min from 50 to 300°C, and held at 300°C for 9 min. The temperature of the injection block was 300°C; the carrier gas was He; the flow rate was 0.7 ml/min. The sample injection volume was 5.0 µl on splitless mode; the electron ionization mode of MS was 70 eV. The temperatures of source and quadrupole in MS block were 230 and 150°C, respectively. The MS was set to the scan mode between *m/z* 40 and 650, with a scan rate of 1.24/s. The first 10 min each of analysis were defined as the solvent delay time. A blank was performed for each sample using the same extraction protocols, taking great care in order to check for contamination introduced in the laboratory and to avoid misinterpretations. Chromatographic peaks belonging to the sample and similarities to the blank were carefully eliminated. Analyses were performed in triplicate and the results were given with relative standard deviations (RSDs). The *m/z* ratios of the chromatographically separated compounds were automatically compared by the MS library data, and the relevant compounds were identified. Compounds identified with a 90% or greater match with the MS library were considered. While the compounds determined with 80–89% match were carefully evaluated, matches below 80% were not considered. The percentage by weight of the compounds detected in the ORA obtained as a result of the relevant extraction was calculated automatically by the device software. Tables 1 and 2 report the percentages of each detected compound in the ORs obtained through the relevant extraction.

RESULTS

Acid-catalysed extraction

Lipid concentrations well above 5 µg/g were detected in all samples, sufficient to make reliable interpretations (Evershed et al., 2008; Reber et al., 2019), and the one-step, acid-catalysed, direct extraction–methylation method used was found to be a useful approach for the recovery of ORs from archaeological ceramics. The total amount of lipid extracted and the ratios of the detected fatty acids were determined by the peak area of the internal standard with known concentration. The additional supporting information presents the lipid concentration of the five samples obtained by the GC-MS analysis and indicates that an average of 244.30 µg/g lipid ratio was detected. This result shows a relatively good preservation.

The results in Table 1 show that palmitic (C16:0) and stearic (C18:0) acids were predominant in the samples, as expected. They varied from 12.27% (S3) to 47.68% (S2) and 9.91% (S5) to 71.40% (S3) for palmitic (C16:0) and stearic (C18:0) acids, respectively. The total ratio of saturated fatty acids (SFAs) and the total ratio of monounsaturated fatty acids (MUFAs) were found to be highest in S2 (91.08%) and S5 (15.17%). In terms of MUFA structure, oleic acid (C18:1) was detected in all samples, while palmitoleic (C16:1) acid was observed only in S4 and S5 samples. Linoleic (C18:2) acid was detected in S2, S4 and S5. Especially S5 stands out with its relatively high oleic (C18:1) and linoleic (C18:2) acids content at 13.01% and 9.39%, respectively. The chromatogram of the fatty acid composition of S3 is given in Figure 3(a), while the chromatograms of other samples are given in

TABLE 1 Organic residue compositions of ceramics obtained by acid-catalysed extraction.

			S1		S2		S3		S4		S5	
No.	**Compound**	**CX**	**Mean**	**RSD**	**Mean**	**RSD**	**Mean**	**RSD**	**Mean**	**RSD**	**Mean**	**RSD**
1	Caprylic (octanoic) acid	C8:0	0.60	0.03	0.64	0.03						
2	Pelargonic (nonanoic) acid	C9:0	1.60	0.03	0.61	0.03						
3	Capric (decanoic) acid	C10:0	0.80	0.03	0.49	0.04	0.11	0.27	0.47	0.02		
4	Tetradecane	C14			0.29	0.03						
5	Suberic (octanedioic) acid	C8:0							1.27	0.03		
6	Lauric (dodecanoic) acid	C12:0	1.19	0.02	3.02	0.03	0.16	0.19	3.36	0.01	0.62	0.03
7	Azelaic (nonanedioic) acid	C9:0					0.26	0.12	2.11	0.01	0.88	0.03
8	Myristic (tetradecanoic) acid	C14:0	3.66	0.02	1.88	0.06	0.88	0.03	9.07	0.01	4.82	0.02
9	Pentadecanoic acid	C15:0	1.09	0.09	0.50	0.02			1.53	0.01	1.35	0.03
10	Palmitoleic (9-hexadecenoic) acid	C16:1							1.20	0.09	2.16	0.04
11	Palmitic (hexadecanoic) acid	C16:0	43.85	0.02	47.68	0.02	12.27	0.04	38.30	0.01	34.75	0.00
12	Eicosane	C20					0.56	0.04				
13	Margaric (heptadecanoic) acid	C17:0							0.59	0.02		
14	Linoleic (9,12-octadecadienoic acid) acid	C18:2			0.40	0.38			0.55	0.02	9.39	0.01
15	Oleic (9-octadecenoic) acid	C18:1	1.04	0.51	0.82	0.26	3.12	0.05	7.16	0.01	13.01	0.01
16	Stearic (octadecanoic) acid	C18:0	34.73	0.02	36.26	0.02	71.40	0.04	14.06	0.01	9.91	0.02
17	Docosane	C22					1.02	0.25				
18	Tricosane	C23					0.78	0.26				
19	Arachidic (eicosanoic) acid	C20:0					0.78	0.13	0.35	0.00		
20	Dehydroabietic acid	C20					2.82	0.07				
21	Tetracosane	C24					0.46	0.02				
22	Pentacosane	C25					0.46	0.46				
23	Hexacosane	C26					0.59	0.02				
24	Heptacosane	C27					0.36	0.08				
	Other		11.44		7.40		1.09		19.98		23.11	
	Hydrocarbon saturated		0.00		0.29		4.22		0.00		0.00	
	Hydrocarbon unsaturated		0.00		0.00		0.00		0.00		0.00	
	SFA		87.52		91.08		85.86		71.10		52.33	
	MUFA		1.04		0.82		3.12		8.36		15.17	
	PUFA		0.00		0.40		0.00		0.55		9.39	

Note: S, sample; RSD, relative standard deviation (obtained from three replicate analyses).

the additional supporting information. No labels were added to the peaks of compounds with an MS match of less than 80%. Peaks that were found to be impurities by blank tests were labelled.

In order to assess the potential origin of these fatty acids, their ratios can be useful, but they need to be compared with and integrated with the presence/abundance of other compounds. For instance,

TABLE 2 Organic residue compositions of ceramics obtained by base-catalysed extraction.

		S1		S2		S3		S4		S5	
No.	**Compound**	**Mean**	**RSD**	**Mean**	**RSD**	**Mean**	**RSD**	**Mean**	**RSD**	**Mean**	**RSD**
1	Butyric (butanoic) acid			2.05	0.36						
2	Lactic acid					7.75	0.09	1.48	0.01	1.43	0.02
3	Caproic (hexanoic) acid			4.48	0.54	0.69	0.20	1.43	0.05	0.13	0.00
4	Acetic acid	0.16	0.06			2.23	0.15	0.48	0.15	0.12	0.25
5	Malonic (propanedioic) acid			1.29	0.02	0.31	0.10	0.16	0.06	0.21	0.05
6	Oxalic (ethanedioic) acid	1.26	0.04	3.58	0.00			3.69	0.05	3.10	0.03
7	2-Ethylhexanol	1.06	0.03					0.48	0.06	0.12	0.25
8	Enanthic (heptanoic) acid			0.62	0.47	0.84	0.10	0.58	0.03		
9	Benzoic acid	0.40	0.05	0.60	0.47	1.03	0.07	0.68	0.04	0.38	0.05
10	Caprylic (octanoic) acid	0.13	0.08	1.20	0.46	2.09	0.08	1.79	0.04	0.41	0.02
11	Succinic (butanedioic) acid	0.40	0.03	0.56	0.46	1.42	0.01	1.04	0.03	0.63	0.02
12	Securinine							0.21	0.14		
13	Pelargonic (nonanoic) acid	0.45	0.07	1.09	0.47	6.39	0.04	2.90	0.02	1.19	0.03
14	2-Hydroxyheptanoic acid					0.53	0.36	0.25	0.04	0.19	0.00
15	Glutaric (pentanedioic) acid	0.20	0.10					0.35	0.03	0.18	0.00
16	Capric (decanoic) acid	0.21	0.10	0.40	0.45	1.40	0.19	1.20	0.06	0.38	0.16
17	Vanillin					0.41	0.07				
18	Adipic (hexanedioic) acid	0.53	0.32			0.63	0.10	1.12	0.02	1.10	0.03
19	Pimelic (heptanedioic) acid							0.79	0.01		
20	Lauric (dodecanoic) acid	0.35	0.03	1.93	0.47	0.44	0.09				
21	Phthalic acid	19.36	0.00	3.93	0.01	6.75	0.01	52.62	0.00	47.65	0.00
22	*Cis*-aconitic acid					6.67	0.07				
23	Azelaic acid	1.08	0.03					3.20	0.02	2.37	0.04
24	Myristic (tetradecanoic) acid			0.39	0.03			0.79	0.01	0.59	0.02
25	Sebacic acid							0.29	0.03		
26	Palmitic (hexadecanoic) acid									0.51	0.02
27	Stearic (octadecanoic) acid									0.31	0.00
28	Dehydroabietic acid					1.70	0.10				
29	2-Monopalmitin	2.50	0.11	2.07	0.47	5.50	0.11	0.38	0.03	1.13	0.03
30	Palmitinic acid-glycerin-(1)-monoester	5.90	0.01	7.29	0.46	22.78	0.04	3.76	0.01	8.41	0.67
31	Stearinic acid-glycerin-(2)-monoester	18.30	0.01	32.09	0.47	1.76	0.24			0.33	0.09
32	1-Monostearin	40.56	0.03	41.03	0.51	7.91	0.13	0.50	0.13	2.99	0.12
33	Arachidic (eicosanoic acid)			0.74	0.04						

Note: S, sample; RSD, relative standard deviation (obtained from three replicate analyses).

if stearic acid (C18:0) is lower than palmitic acid (C16:0), and pentadecanoic acid (C15:0), margaric acid (C17:0) and oleic acid (C18:1) are also present, then the investigated residue may be a fatty compound obtained from ruminant animals, which may be fat or dairy products (Regert, 2011). In the analysed samples, S4 fits this fatty acid profile (Table 1). Also, ruminant carcass fats are characterized by a higher amount of C18:0 compared with C16:0 (Regert, 2011), and only S3 conforms to this pattern. The presence of animal fats is often reported in ointments, perfumes and medicinal

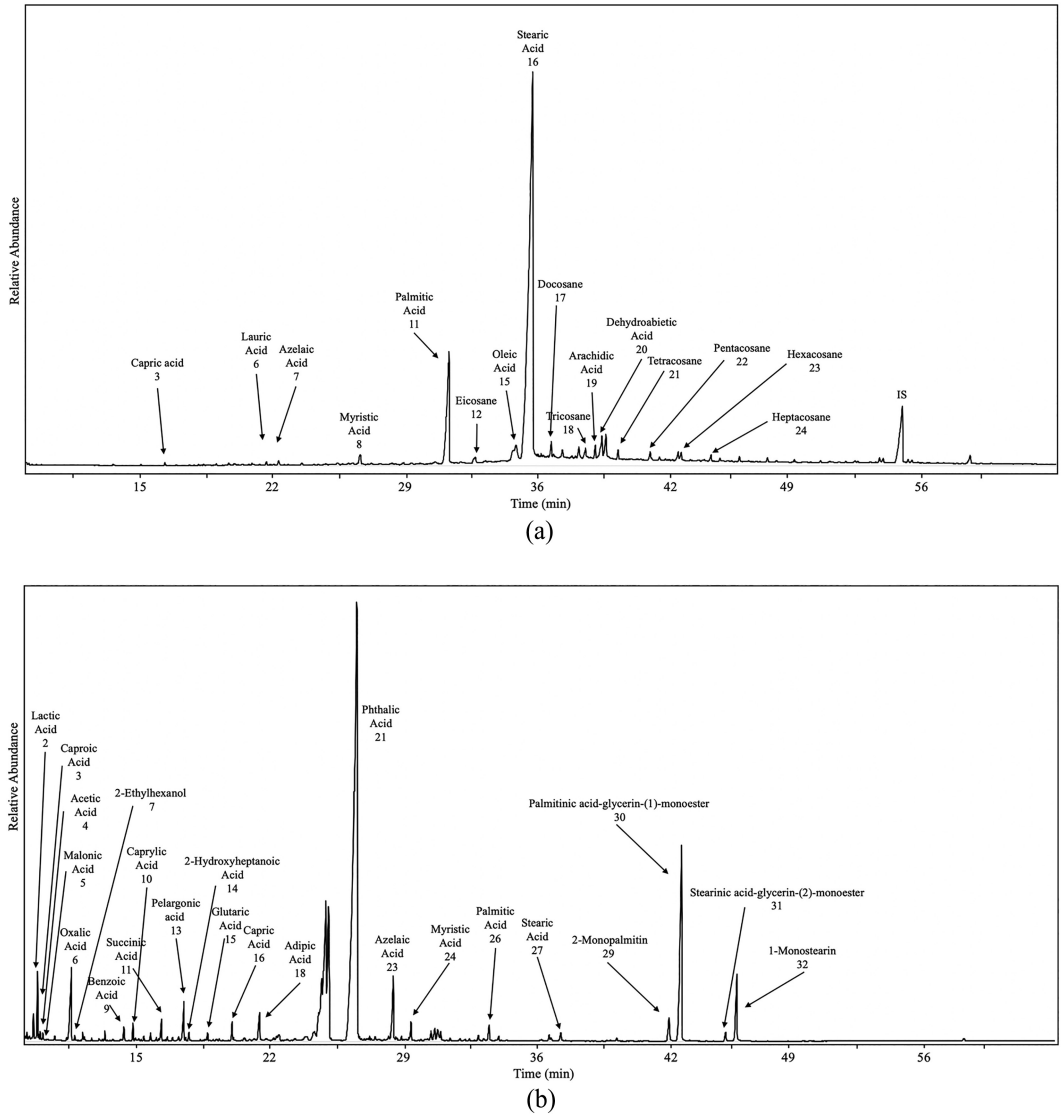

FIGURE 3 Chromatograms of samples (a) S3 from acid-catalysed extraction and (b) S5 from base-catalysed extraction (cf. Tables 2 and 3 for the output sequences of each biochemical compound; IS: internal standard).

balms, as is the case of Etruria and Cretan perfume vessels from the eighth to the sixth centuries BCE (Bodiou et al., 2008; Giachi et al., 2013). Furthermore, fatty acids with odd carbon numbers, which generally indicate an animal fat (Evershed et al., 1997), are determined in all samples (Table 1). In co-presence with their branched homologues, these are generally considered a good marker for animal fats (Regert, 2011). In this case, their absence gives an indication that the OR detected may not be of animal origin.

The most common and most stable oxidation products of unsaturated fatty acids are dicarboxylic acids, which survive particularly well in geographies with dry climates (Regert et al., 1998) such as the Near East. Dicarboxylic acids ranging from C5 to C12, with azelaic acid as the main compound (Eerkens, 2005; Hansel et al., 2011; Regert, 2011), have, for instance, been determined in ceramic samples from a 4000-year-old Nubian burial (Gülaçar et al., 1989). Considering both extraction results, azelaic acid was determined in S1 and S3–S5, while adipic acid was observed in S1 and

TABLE 3 Biochemical compounds of the analysed bottles and their suggested origin.

Sample(s)	Main molecular compounds	Interpretation
S4	More stearic acid than palmitic acid with the presence of pentadecanoic acid, margaric acid and oleic acid	Fat or dairy products
S3	More stearic acid than palmitic acid	Ruminant carcass fats
S1, S3–S5	More palmitic acid than stearic acid with the presence of dicarboxylic acids and oleic acid	Plant-originated oil
S3	Dehydroabietic acid and vanillin	Tree resin
S3	*Cis*-aconitic acid	Fat or plant-based oil

S3–S5, and pimelic and sebacic acids were detected in S4 (Tables 1 and 2). Especially azelaic acid is a hydrolysis product of unsaturated fatty acids with a double bond at the ninth carbon position in the original fatty acids (Hudlicky, 1990), such as oleic acid (C18:1). Roffet-Salque et al. suggest that dicarboxylic acid compounds are more likely to be found in plant oils (Roffet-Salque et al., 2017). The presence of dicarboxylic acids and oleic acid (C18:1) strengthens the possibility that an oleic acid-rich plant oil or derivative mixtures may have been a major component of samples S1 and S3–S5. Under anaerobic conditions, oleic (C18:1) acid tends to transform into azelaic (C9:0) and palmitic (C16:0) acids through oxidative decay (Copley et al., 2005; Regert et al., 1998). S1, S4 and S5 contain oleic (C18:1) acid along with azelaic acid (C9:0) and a large amount of palmitic acid (C16:0) (Table 1), suggesting that the organic residue (OR) in these samples may be of plant-originated oil.

On the other hand, the identification of *n*-alkanes provided key indications for the presence of products derived from leafy vegetables (Eerkens, 2005; Kimpe et al., 2004). S3 has *n*-alkanes such as eicosane, docosane, tricosane, tetracosane, pentacosane, hexacosane and heptacosane (4.2% in total), while tetradecane was detected in small proportions in sample S2 (Table 1). However, since the odd-numbered ones do not dominate the even-numbered alkane species, it should be considered that the current data might potentially be the result of plastic contamination. Another common biomarker in archaeological ceramics is diterpenoids, which is of great importance in interpretative terms (Colombini, Modugno et al., 2005; Manzano et al., 2016; Ribechini et al., 2008). Of these, dehydroabietic acid and its oxidation by-product, 7-oxo-dehydroabietic acid are characteristic compounds of a resin from the Pinaceae family (Blanco-Zubiaguirre et al., 2019). The presence (2.8%) of such compound in S3 is also supported by the presence of vanillin (see below), which may have originated from degraded wood (cf. Table 2). Previous studies have documented the use of tree resin as a waterproofing material (sealant), as an additive to wine, and also as a component of medicinal and cosmetic ointments in ancient pharmacopeia (Colombini, Giachi et al., 2005). Given the size of the ceramic bottle from which S3 derives, and the co-occurrence of animal and vegetable fats in the OR, the latter possibility seems more plausible.

Base-catalysed extraction

The organic acid residue composition of the five samples obtained by GC-MS analysis is given in Table 2. Benzoic, caprylic (C8:0), succinic, pelargonic and capric acids were present in all samples, varied from 0.38% (S5) to 1.03% (S3), from 0.13% (S1) to 2.09% (S3), from 0.40% (S1) to 1.42% (S3), from 0.45% (S1) to 6.39% (S3), and from 0.21% (S1) to 1.40% (S3), respectively. For the fatty acids, the highest proportions were found in sample S3. Lactic, caproic, acetic, malonic, oxalic, 2-heptanoic, glutaric, adipic, lauric, azelaic and myristic acids were residues seen in more than 50% of all samples (Table 2). The acetic acid detected may have instead originated from the ethyl acetate used for sample treatment. The chromatogram of the composition of S5 obtained as a result of base-catalysed extraction is given in Figure 3(b), while the chromatograms of other samples are given in the additional supporting information. It is observed that the selected high sample amount and high

injection volume give peaks of plastic contaminants at high rates thanks to the basic environment of the current extraction method (Figure 3b). The detected plastic contamination is thought to come from the samples rather than from the laboratory. For this purpose, the blank analysis chromatograms performed for the S5 sample are given in the additional supporting information. As can be seen from the relevant chromatograms, a contamination from the laboratory environment could not be detected within the scope of the current detection limits.

Benzoic acid is detected in most archaeological ceramic samples. Biomolecules that may indicate fermented products in analysed samples were succinic (Blanco-Zubiaguirre et al., 2019; Fujii et al., 2019; Garnier & Valamoti, 2016; Pecci et al., 2017; Zhang et al., 2018), glutaric (Fujii et al., 2019), and malonic (Pecci et al., 2017) acids. Table 2 shows that all samples contain most of the detected fermentation-related molecules. These are, however, on their own not sufficient to indicate the presence of fermented beverages or products, since they are products of natural degradation or fermented products that may have formed during the degradation process.

In addition, a variety of low-molecular-weight aliphatic carboxylic acids such as butyric (C4:0), caproic (C6:0), enanthic (C7:0), caprylic (C8:0), pelargonic (C9:0), capric (C10:0) and lauric (C12:0), as well as medium molecular weight aliphatic carboxylic acids such as myristic (C14:0), palmitic (C16:0), stearic (C18:0) and arachidic (C20:0) acids were also present in different proportions across the sampled dataset (Table 2). Lactic acid, a chemical by-product of anaerobic respiration by bacteria and soil microorganisms (Zhang et al., 2018), was observed as a structure that is a non-original component of ORs in samples S3–S5. The presence of 2-monopalmitin, palmitinic acid-glycerin-(1)-monoester, stearinic acid-glycerine-(2)-monoester and 1-monostearin also considered as a product of triglyceride hydrolysis. *Cis*-aconitic acid was found in S3 and is naturally found in the highest concentration in cow's milk and has also been detected in several different foods such as Japanese pumpkin, purple mangosteen, shallots (all of which are not present in the Near East during this period), as well as opium poppy and root vegetables (Foodb, 2021).

Vanillin, a phenolic compound and one of the main biochemical components found in clove essential oil (Uddin et al., 2017) and rosemary (McGovern et al., 2009), was found in sample S3. However, the inability to detect components such as eugenol, eugenol acetate, fenchone, camphor, borneol and cuminaldehyde makes it unlikely that the OR may have originated from these plants. It is thought that the vanillin detected in sample S3 may originate from a tree (pine?) resin (Romanus et al., 2009) or pine wood/dust, which is also documented in the same sample by the presence of dehydroabietic acid.

Sample S4 also contained securinine, a bioactive compound that is the main alkaloid present in the roots of plants belonging to the genera *Phyllanthus*, *Securinega* and *Flueggea*, present also in South West Asia (Klochkov & Neganova, 2021). Securinine has a wide variety of biological properties including the activities of acetylcholinesterase inhibitory, antimalarial, antimicrobial and antifungal, and is also known as a potent biochemical that stimulates the central nervous system (Klochkov & Neganova, 2021). However, in order to increase the confidence in the identification of the molecule, it is obvious that comparative analyses should be performed with chemical standards rather than the GC-MS library result.

DISCUSSION

The shape of the analysed ceramic bottles indicates that they contained liquids, and their relatively small volume also suggests that they were functionally different from transport jars and pithoi, which could reach up to 400–700 L (Türkteki, 2020). They instead probably contained high-value/low-bulk commodities whose perceived value may have allowed their exchange across relatively large distances. Furthermore, even considering the possibility that slightly different soil conditions at the site may have affected the preservation of different organic compounds and that some bottles may have been reused, the presence of different OR profiles within each sample (Table 3) suggests that they probably did not contain the same liquid.

Several compounds including caproic, caprylic, pentadecanoic and margaric acids suggest the co-presence of animal fat in all samples, albeit in different proportions. In addition, both the abundance of oleic acid in co-presence with dicarboxylic acids and the presence of large amounts of palmitic acid compared with stearic acid indicates that the main component of the contents of bottles S1, S4 and S5 is likely to be a plant-based oil. Conversely, in sample S2 there is no good evidence for the presence of plant-based oils. In particular, S3 stands out for the presence in significant proportions of three compounds otherwise not detected even in trace in the other samples. These include *cis*-aconitic acid (6.7%), which may have originated either from cow's milk, opium poppy or root vegetables. Furthermore, dehydroabietic acid (1.7%) shows the inclusion of a conifer resin, a hypothesis also supported by traces of vanillin. Finally, it is worth mentioning that S2 and S3 also stand out from the remaining samples because they come from the largest and smallest bottles, respectively (cf. Figure 2), suggesting that shape might have been associated with a particular product.

Even if we cannot understand in detail the recipes of the liquids contained in Küllüoba's bottles, they have clear parallels with both textual and archaeological evidence for scented oils and medicinal ointments. For instance, GC-MS analysis on mid-second millennium BCE Red Lustrous Wheelmade Ware vessels, later than but similar in size to the vessels analysed here, has instead identified different mixtures of plant and animal oils, resin, beeswax and/or bitumen in their OR profiles (Steele & Stern, 2017). Similarly, GC-MS analysis on early first millennium BCE Phoenician flasks identified their content as a scented oil (Namdar et al., 2013).

Furthermore, a recent analysis of palaeobotanical remains from a workshop in Ebla's Palace G (datable to the late 24th century BCE, thus roughly contemporary with Küllüoba's bottles in level IIIA) revealed the collection of wild plants to be used in medicinal concoctions. They include Euphorbiaceae and opium poppy (two important psychoactives), as well as terebinth (an important source of resin) and other substances known to have curative properties in traditional medicine (Peyronel et al., 2014). For the Mycenean period (*c*.1400–1200 BCE), there is archaeological evidence for scented oil workshops at Zakros (Crete) and Pylos (mainland Greece) (Shelmerdine, 1985). From the same historical context, several Linear B texts mention the use of wine, honey, resins, tree barks, herbs and spices for the production of scented oils (Shelmerdine, 1985). Furthermore, a substantial group of cuneiform and Linear B texts spanning the late third to the late second millennia BCE document both the use of oil-based medicinal ointments (Fairbairn et al., 2019; Fales, 2012; Fronzaroli, 1998; Geller, 2010) and the trade of aromatic oils in the Eastern Mediterranean and Near East (Fappas, 2012; Palmer, 2003; Shelmerdine, 1985).

In light of this, we can confidently suggest that the Küllüoba's bottles did contain various types of scented/medicinal ointments, whose different recipes might have been related to differences in their morphological shape. Given their association with well-documented, secure stratigraphic contexts radiocarbon dated to *c*.2450–2200 cal BCE, they currently represent the earliest direct archaeological evidence for the existence of such commodities in Anatolia.

Their shape, fabric, surface treatment and production techniques indicate that the analysed bottles were not locally produced at Küllüoba, but likely several hundred kilometres to the south-east (Türkteki, 2010). Some of the examples from Küllüoba (S6–S9, S11, not analysed by GC-MS) can confidently be attributed to the 'Syrian bottles' group and likely originated on the Middle Euphrates. Others (S1–S2, S4) were probably produced elsewhere, and we suggest Cilicia as a possible candidate for their origin. This hypothesis relies not only on typological parallels with similar vessels found at Tarsus-Gözlükule but also on the spatial distribution of wild and domesticated *Olea europea*, whose product (olive oil) was possibly the main component of ORs from S1 and S3–S5 (described by the analytical results as plant-based oil). Palaebotanical and palinological archives from the Holocene record the presence of olive trees along the Aegean and Mediterranean coasts as far inland as the Middle Euphrates valley, but notably not on the central Anatolian plateau (Figure 1) (Carrion et al., 2013; Oflaz et al., 2019). This is confirmed by the analysis of Küllüoba's palaeobotanical assemblages, where olive tree remains and olive pits are notably absent (Çizer, 2015). While it remains possible that olive oil might have been shipped to inland Anatolia and used locally to make scented

oils, several observations make it unlikely: (1) there is no evidence for the existence of oil-transport amphorae in this period, (2) there is no archaeological evidence for scented oils' workshops anywhere in the region, and (3) the bottles' typology is alien to central Anatolia. The most plausible solution is that these goods had been manufactured in an area where olive oil production is attested during the Early Bronze Age, packaged with locally made vessels, and then inserted into the supra-regional exchange networks. If we accept this hypothesis, then the analysed bottles from Küllüoba represent the earliest direct evidence for the exchange of scented/medicinal oils between Anatolia and the Near East.

Regarding the possible use of these liquids, Hittite, Mycenaean and Mesopotamian texts document the use of scented oils in a large range of ritual activities including ritual offerings, anointment of rulers, preparation of the body for burial and building foundation rituals (Felli, 2012; Shelmerdine, 1985). At Küllüoba, all the bottle examples were found in the so-called 'feasting pits' clustered at the centre of the settlement's public area during level IIIA (Türkteki & Başkurt, 2016). These deposits show evidence for large communal gatherings that involved food consumption and (alcoholic?) drinking, the discard of tableware employed in the celebration, and the deposition of intact objects/animal carcasses before the sealing of the pits with clay.

The Küllüoba 'pit horizon' is part of a broader phenomenon observed at several sites in the region, where apparent remains of feasting for dozens or hundreds of individuals were buried together with animal carcasses and whole objects—in some cases with a perceived high value such as gold and silver vessels (Bachhuber, 2009; Kouka, 2011; Türkteki & Başkurt, 2016). These 'feasting pits' are in all cases associated with public/elite buildings and it can be argued they likely pertained to some form of ritual officiated by (or in presence of) elite individuals in front of their community. Intriguingly, the archaeological evidence is matched by later Hittite texts describing purification rituals where whole animal carcasses were buried (often in conjunction with grinding stones) to appease divinities of the underground (Türkteki & Başkurt, 2016; Türkteki & Türkteki, 2021). In this context, the ceramic bottles under study might have been deposited whole as offerings (thus as 'sacrificed' luxury commodities), or alternatively their content might have been employed in some format during the ceremony itself.

CONCLUSIONS

This study represents the first archaeometric effort to shed light on the content of small bottles/flasks produced in the Near East during the third millennium BCE and thought to contain scented/medicinal oils. Our results confirm once again the success of GC-MS as a method to analyse ORs derived from archaeological contexts, and the high concentration of ORs in all the samples analysed indicates that the post-depositional environmental conditions at Küllüoba were particularly conducive to good preservation. Our results also confirm the idea that the content of most of the analysed samples was a base of plant oil with additional ingredients that varied from sample to sample, but included animal fat, plant resin and other plant-based products whose exact nature is beyond the capabilities of detection of GC-MS. These recipes are reminiscent of Bronze Age archaeological and textual evidence for similar products that were produced and traded across the Aegean and the Near East.

ACKNOWLEDGEMENTS

The authors thank Ezgi Çakır and Şeyma Nur Akış, undergraduate students in Selçuk University Biochemistry Department, for their efforts during the study.

DATA AVAILABILITY STATEMENT

The data that support the findings of this study are available in the supplementary material of this article.

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PEER REVIEW

The peer review history for this article is available at https://publons.com/publon/10.1111/arcm.12852.

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Additional supporting information can be found online in the Supporting Information section at the end of this article.

How to cite this article: Tarhan, İ., Massa, M., Türkteki, M., & Türkteki, S. (2023). Toward an understanding of the exchange in ancient scented oils through organic residue analysis of Bronze Age Near Eastern ceramic bottles by GC-MS. *Archaeometry*, *65*(4), 833–849. https://doi.org/10.1111/arcm.12852