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CINNAMALDEHYDE IN EARLY IRON AGE PHOENICIAN FLASKS RAISES THE POSSIBILITY OF LEVANTINE TRADE WITH SOUTH EAST ASIA

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ABSTRACT

Small ceramic flasks with thick walls and narrow openings were produced in Phoenicia. These flasks were common in Phoenicia, the southern Levant and Cyprus in the early Iron Age, namely in the 11th–mid-9th centuries BCE. Their shape, size, decoration and find-contexts suggest that they contained some precious materials and were part of a commercial network operating in these regions. We analyzed the lipid contents of 27 such containers from 5 archaeological sites in Israel using gas chromatography coupled with mass spectrometry (GC-MS). The organic extractions of 10 of these flasks contained cinnamaldehyde (C9H8O), a major component of cinnamon. In antiquity the cinnamon tree grew only in South and South East Asia. As cinnamaldehyde is found in small quantities in some modern potential contaminants, possible contamination of the small flasks with this compound was carefully assessed. Significantly, two recently excavated small flasks that were not handled directly contained relatively high concentrations of cinnamaldehyde. Other vessel types from the same archaeological sites and in some cases the same contexts did not contain cinnamaldehyde. Thus it is unlikely that the presence of cinnamaldehyde in the flasks results from contamination. This finding raises the intriguing possibility of long distance trade in the early Iron Age, assuming that the extracted cinnamaldehyde is indeed derived from the bark of the cinnamon tree. This is consistent with other suggestions that trade from South/South East Asia to the West took place at such an early date.

KEYWORDS: Residue analysis, GC-MS, Iron Age, cinnamaldehyde, flasks, South East Asian trade, Cinnamon

1. INTRODUCTION

The collapse of most Late Bronze Age polities and economic systems in the east and central Mediterranean ca. 1200 BCE, was considered to have been accompanied by a cessation of long-range trade activities, ushering in a 'dark Age' (Ward and Joukowsky, 1992). Archaeological evidence indicates that this is not so. During the entire early Iron Age (ca. late 12th–mid-9th centuries BCE) exchanges between various Mediterranean regions, as far apart as the Levant and the Atlantic coast of Iberia, are well attested, involving mainly metal and luxury items, such as prestigious metal objects and various items of personal adornment (Celestino et al., 2008, Coldstream, 2000, Crielaard, 1997, Gilboa et al., 2008, Kourou, 2008, Mederos Martín and Ruiz Cabrero, 2006, Niemeyer, 1999, Nijboer, 2008b, Nijboer, 2008a, Pare, 2006, Sherratt, 2012, Sommer, 2007, Sommer, 2010). As opposed to the Late Bronze Age, however, in the early Iron Age there is very little evidence for sustained inter-regional exchange in commodities packed in ceramic containers, with one notable exception; small decorated ceramic containers that were shipped from Phoenicia to Cyprus (Gilboa et al., 2008). These containers can generally be divided into two groups. The best known and extensively discussed is the so-called "Phoenician Bichrome" group that consists mostly of rounded jugs of varying sizes, which feature a typical decorative syntax and decorative motifs. They are usually considered the best indication of early Phoenician trade (Aubet, 2000, Bikai, 1987, Gilboa, 1999). The second group comprises small flasks, which received far less scholarly attention, though they circulated in the Levant more extensively than Phoenician Bichrome containers, and were shipped much more frequently to Cyprus (Gilboa et al., 2008, Karageorghis and Iacovou, 1990, Mazar, 1985). In Phoenician chronological terms this export occurred from Ir1a to Ir2a, and in Cypriot terms from Late Cypriot IIIB to Cypro-Geometric III, roughly the late 12th–mid-9th centuries BCE (Gilboa and Sharon, 2003). This group, henceforward "the small flasks", is the focus of this study.

The small flasks are typically 18–20 cm high (their size is not standard), with narrow necks and apertures (1 cm or less in diameter), and with a volume of circa 50 ml (Fig. 1). They are made of a rather coarse fabric and are usually decorated with simple concentric circles in one or two colors. However, even though they are quite humble in appearance, their decoration stands in contrast to the very dull and unadorned vista of Phoenician pottery of the early Iron Age. In fact, the small flasks and the Bichrome containers are the only ceramic vessels in Phoenicia that were systematically decorated (Gilboa, 2005, Gilboa and Sharon, 2003). This suggests that their decoration had a specific function, possibly because they were meant to be 'marketed'. Furthermore, most of the small flasks have walls around 1cm thick (Fig. 1) that are much thicker than those of all other contemporary Phoenician ceramic vessels. This presumably represents a special effort to avoid breakage. Indeed, many of them survived intact, even in the habitation contexts of stratified tells, making them good candidates for residue analysis.

Figure 1. Example of a small flask (no. 2267 from Tell Qasile)

All these characteristics, in conjunction with the fact that in the southern Levant the small flasks are frequently found in 'elite' contexts such as treasuries and temple store-rooms (e.g., the Tell Qasile examples in Table 1), suggest that they may have been used for the distribution of some precious material. Here we focus on one particular compound extracted from 10 out of the 27 small flasks analyzed, namely cinnamaldehyde. Cinnamaldehyde is the major organic compound in cinnamon oil (65-95% of the bark oil volatiles, species dependent), giving it its flavor and odor (Senanayake and Wijeskera, 2004). This pale yellow, viscous liquid occurs naturally in the bark of cinnamon trees and other species

of the genus Cinnamomum (Senanayake and Wijeskera, 2004, Neish, 1968) that in ancient times grew only in South and South East Asia. Here we raise the possibility that cinnamon mixed with a liquid was stored in the small flasks.

Table 1. Details of the small flasks sampled. All vessels were sampled by drilling, other than those marked *, where the samples were broken off with pliers, and those marked ▪, where the samples were scraped from the inner side of the item using dental tools. Locus numbers are indicated only in instances where vessels from the same depositional units were sampled as controls (Tables 3, 4).

Lab no.	Site	Description	Decoration	Context	Condition and treatment
2019▪	Tell Qasile L227	Small lentoid flask	Dark red circles	Area C Iron 1a, Temple 200 store-room	Body intact, handles and upper neck glued; touched and washed externally; long-term storage; inner lower part scraped
2192	Dor	Very thick rounded flask	Red circles and additional decorations	Area G, Iron 1a/b domestic cult room in elite setting	Half a flask; washed inside and outside; long term storage.
2269	Tell Qasile L188	Small lentoid flask, asymmetric	No painted decoration	Area C Iron 1b Temple 131 store-room	Intact; touched and washed externally; long term storage.
2271	Tell Qasile L188	Small rounded/asymmetric flask	Black and red circles	Area C Iron 1b Temple 131 store-room	Mostly intact, upper part of neck glued; washed externally; long term storage.
2272▪	Tell Qasile L188	Small lentoid, slightly asymmetric flask	No painted decoration	Area C Iron 1b Temple 131 store-room	Body intact, handles and neck glued; touched and washed externally; long-term storage; inner lower part scraped
2024▪	Yoqneʻam	Small lentoid flask	Faint black circles	Area A Iron 1b, cottage industry complex	Intact; touched and washed externally; long-term storage; inner lower part scraped
2267	Tell Qasile L188	Small lentoid flask	Dark red circles	Area C Iron 1b Temple 131 store-room	Body intact, handles and neck glued; touched and washed externally; long-term storage.
2276	Dor	Small lentoid flask	Faint gray circles	Area G Iron 1a industrial context	Fragmentary, upper part only; handled and washed; long-term storage.
2380	Dor	Small lentoid to rounded flask	Orange circles	Area D2 Iron 1b storage complex	Near complete; washed and glued from a few fragments.
2194▪	Kinneret	Small lentoid flask	Red circles	Area U Iron 1b domestic context	Upper part only; handled and washed.
2275	Dor	Small lentoid flask	Red circles	Area G Iron 1|2 domestic context	Fragmentary, upper part of vessel only; handled and washed.
2278*	Dor L09D2-386	Small flask (not yet restored and exact shape unknown)	Red circles	Area D2, Iron 1a store-room in elite dwelling	Complete, found in pieces; fresh sample from excavation; untouched,

					unwashed.
2279	Dor	Small lentoid flask	Red circles	Area B Iron 1a|b public structure	Nearly intact flask; washed and handled; long-term storage.
2281*	Dor	Small lentoid flask (not yet restored and exact shape unknown)	Red circles	Area D2 Iron 1a room in elite dwelling	Near complete; fresh sample from excavation; untouched, unwashed.
2190	Kinneret	Small lentoid flask	No apparent painted decoration	Area U Iron 1b, domestic context	Large fragment only; handled and washed.
2191	Kinneret	Small lentoid flask	Faint red circles	Area U Iron 1b domestic context	Near complete, washed and glued from several fragments.
2268	Tell Qasile L188	Small lentoid flask	White slip, no painted decoration	Area C Iron 1b Temple 131 store-room	Intact; touched and washed externally; long-term storage.
2195▪	Kinneret	Flask	Unclear	Area U Iron 1b domestic context	Fragmentary; handled and washed.
2242*	Dor	Medium-sized lentoid flask	Red circles	Area D5 Ir1a storage context	Intact, lower part only; unclear if washed or touched inside.
2244	Dor	Medium-sized lentoid flask	Red circles	Area D2 Iron 1b storage complex	Intact, lower part only; handled and washed.
2265	Tell Qasile L134	Small lentoid flask	No painted decoration	Area C Iron 1b cultic platform in Temple 131 hall	Intact; touched and washed externally; possibly soaked in water; long-term storage.
2266	Tell Qasile L188	Small lentoid flask	No painted decoration	Area C Iron 1b Temple 131 store-room	Intact; touched and washed externally; long-term storage.
2270	Tell Qasile L188	Small lentoid flask, slightly asymmetric	Dark red circles	Area C Iron 1b Temple 131 store-room	Body intact; touched and washed externally; handles and neck glued; long-term storage.
2273	Tell Qasile L188	Small lentoid flask	Dark red circles	Area C Iron 1b Temple 131 store-room	Intact; touched and washed externally; long-term storage.
2277*	Dor	Small lentoid flask	No painted decoration	Area D5 Iron 1 unclear context	Nearly intact, handles and neck missing; fresh from excavation untouched, unwashed.
2280*	Dor	Small lentoid flask	Red circles	Area D2 Iron 1|2 cottage industry complex	Near complete, found in fragments; fresh sample from excavation; untouched, unwashed.
5177	Nahal Patish	Small lentoid flask	Dark red circles	Area B Iron 1b temple	Intact, neck missing; fresh from excavation; unwashed.

2. MATERIALS AND METHODS

2.1. Vessels Analysed

Only small flasks found in modern Israel were analysed, since the small flasks in Cyprus are routinely acid-washed and those from Phoenician sites in Lebanon are inaccessible to us. We analyzed 27 small flasks from 5 archaeological sites in modern Israel (Fig. 2), comprising sites in southern Phoenicia, its immediate environs

and neighboring Philistia: Dor (11 flasks), Kinneret (4), Yoqne'am (1), Tell Qasile (10) and Nahal Patish (1) (Table 1). They range in age from Ir1a to the Ir1|2 transition, ca. the 11th–late-10th centuries BCE. The 27 small flasks are from different sites, depositional environments and different environments of preservation. They were also handled and stored in different ways. Several small flasks were intact and stored for about three decades, after being washed externally. Other small flasks were fragmentary and hence they were handled, washed and some were also glued. Four flasks were unearthed while this project was being conducted and were not touched with bare hands prior to analysis. Table 1 lists the preservation states of the flasks and the ways they were handled during and after excavation.

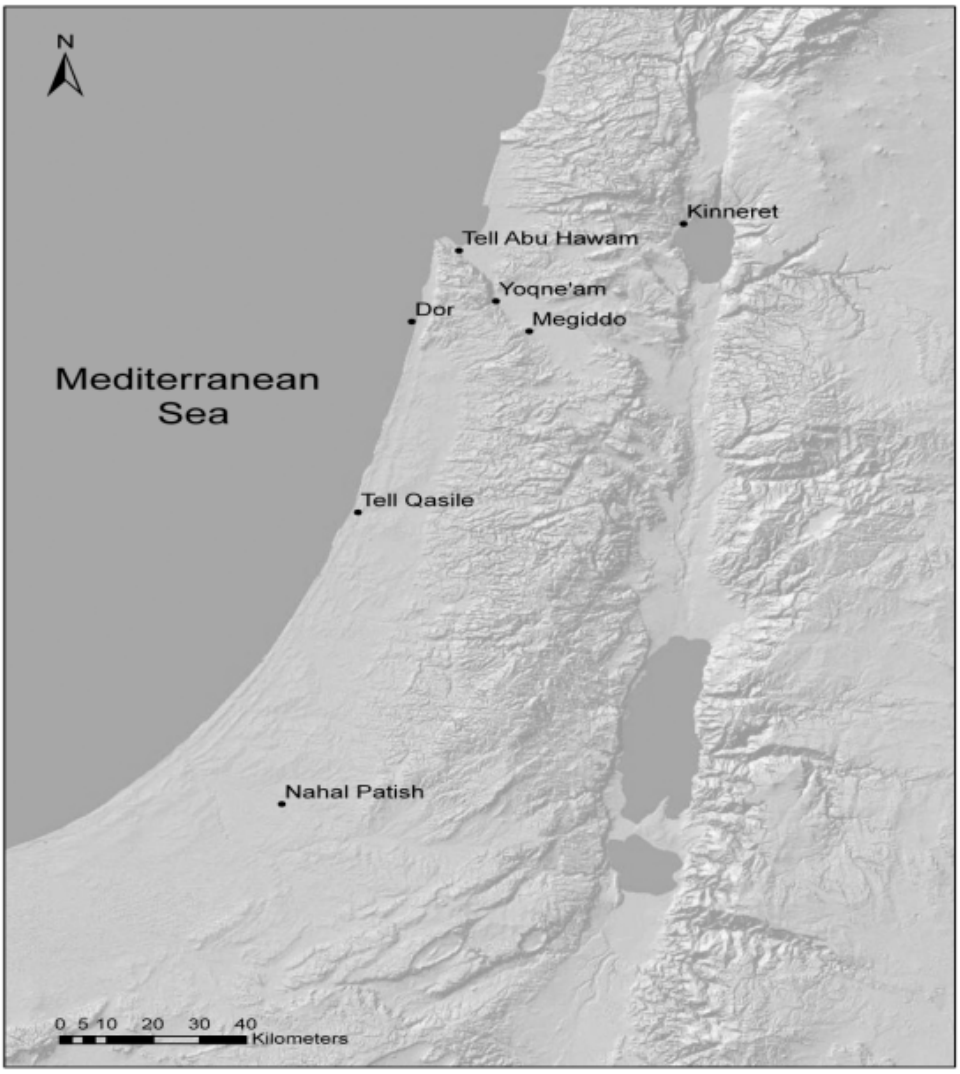

Figure 2. Map of the southern Levant showing main sites mentioned in the text

2.2. Sampling

The intact flasks, nearly intact flasks and flasks that were restored prior to analysis were sampled by the slow drilling of a hole as close as possible to the bottom of the vessel. In five cases the inner faces of intact flasks were scraped with metal dental tools. With fragmentary vessels, small pieces of ceramic were broken off with pliers, fragmented and ground manually to a powder in an agate mortar and pestle. Most samples

weighed around 1 g, but due to curatorial restrictions some of the samples extracted were smaller, but not less than half a gram. Table 1 also lists the sampling details for each vessel.

2.3. Lipid Extraction

Glassware was soaked overnight in fuming nitric acid, washed carefully with distilled water, and then washed with acetone, followed by dichloromethane and dried in a fume hood. Powdered samples were extracted twice with 10 mL of solvent (dichloromethane: methanol, 2:1 volume ratio) followed by sonication for 10 minutes. The samples were centrifuged for 5 minutes at 3500 rpm. The supernatant was transferred to a clean glass vial. The solvents were removed by evaporation under a gentle stream of nitrogen. Prior to analysis 50-100 μL of N,O-bis (trimethylsilyl) trifluoroacetamide containing 1% trimethylchlorosilane was added to the dry extracts followed by heating at 65 °C for 20 minutes. One μL of each sample was injected into the gas chromatograph (GC) coupled with a mass selective detector (MSD).

2.4. Identification of Lipids with Gas Chromatography/Mass Spectrometry (GC/MS)

GC/MS analyses were carried out using a HP7890 gas chromatograph coupled with a HP5973 mass spectrometer (electron multiplier potential 2 KV, filament current 0.35 mA, electron energy 70 eV; the spectra were recorded every 1s over the range m/z 50 to 800) using a splitless injection mode. An Agilent 7683 autosampler was used for the sample introduction. A 30 m, 0.32 mm ID 5% cross-linked phenylmethyl siloxane capillary column (HP-5MS) with a 0.25 μm film thickness was used for separation, forwarded by 1 m of fused silica deactivated high temperature pre-column (0.25 mm ID). Helium was used as a carrier gas at a constant flow of 1.1 mL s–1. An isothermal hold at 50 °C was kept for 2 minutes, followed by a heating gradient of 10 °C min^{-1} to 345 °C, with the final temperature held for 10 minutes. The injection port temperature was 220 °C. The MS interface temperature was 300 °C. Peak assignments were carried out with the aid of library spectra (NIST 1.6) and compared with published data.

2.5. Quantification of Lipids Using Gas Chromatography (GC)

GC analyses were carried out using a HP6890 gas chromatograph equipped with a flame ionization detector (FID), a splitless injection mode and a 30 m, 0.32 mm ID 5% cross-linked phenylmethyl siloxane capillary column (HP-5MS) with a 0.25 μm film thickness. Helium was used as a carrier gas at a constant flow of 1.1 mL s^{-1}. An isothermal hold at 50 °C was kept for 2 minutes, followed by a heating gradient of 10 °C min^{-1} to 345 °C, with the final temperature held for 10 minutes. The injection port temperature was 220 °C.

Quantification of the amounts of lipids per unit weight ceramic using gas chromatography was conducted on 5 samples that were initially found to contain cinnamaldehyde, and that we were allowed to re-sample. Two of these samples (2278, 2281) were freshly excavated, not touched with bare hands, but placed in plastic bags. Cinnamaldehyde and 2 other molecules (myristic acid and tartaric acid) were quantified separately based on calibration curves using standards (from Sigma Aldrich). Myristic acid and tartaric acid were first silylated. The quantification of the total lipid extracts (TLEs) of each sample was based on the areas under all the peaks eluted from the GC column against the calibration curve for myristic acid, after silylation.

2.6. Adsorption Experiments

Ten milliliters of 10-2 M mixture of cinnamaldehyde, tartaric acid and myristic acid were dissolved in DCM: MeOH (1:1, v:v) and were then added to custom-made ceramic cups (60% clay, 25% chalk, 15% quartz sand, and fired at 550, 770 and 940 ºC). They were left for 48 hours to adsorb and were then extracted and analysed as described above. The procedure was repeated separately for cinnamic acid. The estimated error is based on duplicate injections into the GC.

3. RESULTS

Table 2 shows the inventory of all the lipids identified in the 27 flasks analyzed. The diversity of compounds extracted from the flasks presumably reflects variations in their contents, different states of preservation, and because most

of these vessels were handled during excavations and obtained from store-rooms, contaminants due to handling and storage. It is therefore a daunting task to try to understand the origins of all these molecules. We however were impressed by the fact that 10 of the 27 flasks analyzed contained cinnamaldehyde (Table 2, Fig. 3) and none of the 17 other vessels analysed as controls contained cinnamaldehyde (Tables 3 and 4). Furthermore, two of the 4 flasks that were recently excavated and therefore untouched by human hands, contained relatively high concentrations of cinnamaldehyde (Table 2, nos. 2278, 2281, last column). We therefore decided to focus on this compound, both because its presence is unexpected as it is a relatively unstable compound, and intriguingly a likely "natural" source is the bark of the cinnamon tree. In antiquity the cinnamon tree grew only in South/South East Asia, thus raising the possibility of long distance trade between this region and the Levant.

Table 2 lists all the compounds identified in the extracts from the 27 small flasks. Some extracts contained diacids, borneol, isoborneol and coumarins. In addition, monoacylglycerides, with C_{12} to C_{18} fatty acid chains were detected in most of the flasks and in other vessel types (Tables 2, 4). Further decomposition products of the triacylglycerides found in the samples, such as monoacylglycerides and fatty acids were present in the small flasks as well (Table 2).

Table 2. Molecular components in the total lipid extracts of the small flasks

Lab no.	Major components	Fatty acids (TMS)	AGs	Alcohols	Others and contaminants	Quantification (μg/g);Cin(TLE)
2019	cinnamaldehyde	$C_{9:0}$, $C_{12:0}$, $C_{14:0}$, $C_{16:0}$, $C_{18:1}$, $C_{18:0}$	MAG_{16}, MAG_{18}		K_{11}, TA, *i*-$PrC_{14:0}$, diacid, WE_{12}, WE_{16}, WE_{18} phthalate	
2192	benzoic acid, cinnamaldehyde	$C_{8:0}$, $C_{9:0}$, $C_{10:0}$, $C_{12:0}$, $C_{13:0}$, $C_{14:0}$, $C_{16:0}$, $C_{18:0}$			K_{11}, phenol, TA, *i*-$PrC_{12:0}$, *i*-$PrC_{14:0}$ phthalate	65 (150)*
2269	benzoic acid, cinnamaldehyde	$C_{11:0}$, $C_{12:0}$, $C_{13:0}$, $C_{14:0}$, $C_{16:0}$, $C_{18:0}$, $C_{20:0}$, $C_{22:0}$	MAG_{16}, MAG_{18}	$C_{12}ol$ $C_{13}ol$	K_{11}, TA, *i*-$PrC_{12:0}$, isoborneol, *i*-$PrC_{14:0}$ phthalate	
2271	benzoic acid, cinnamaldehyde	$C_{9:0}$, $C_{10:0}$, $C_{12:0}$, $C_{13:0}$, $C_{14:0}$, $C_{16:0}$, $C_{18:0}$, $C_{20:0}$	MAG_{14}, MAG_{16}, MAG_{18}		monosaccharide, TA, *i*-$PrC_{12:0}$, *i*-$PrC_{14:0}$ squalene, $C_{15:0}$, cholesterol, phthalate	90 (480)*
2272	benzoic acid, cinnamaldehyde	$C_{12:0}$, $C_{14:0}$, $C_{16:0}$, $C_{18:0}$	MAG_{16}, MAG_{18}		TA, *i*-$PrC_{12:0}$, *i*-$PrC_{14:0}$ cholestenol, squalene, cholesterol, phthalate	
2194	cinnamaldehyde	$C_{9:0}$, $C_{12:0}$, $C_{14:0}$, $C_{16:0}$, $C_{18:1}$, $C_{18:0}$, $C_{20:0}$			coumarin, *i*-Pr $C_{12:0}$, *i*-$PrC_{14:0}$ phthalate	
2275	benzoic acid, cinnamaldehyde	$C_{10:0}$, $C_{12:0}$, $C_{13:0}$, $C_{14:0}$, $C_{16:1}$, $C_{16:0}$, $C_{18:0}$	MAG_{16}, MAG_{18}	$C_{13}ol$ $C_{22}ol$ $C_{24}ol$	*i*-$PrC_{12:0}$, *i*-$PrC_{14:0}$ $C_{15:0}$, $C_{17:0}$, cholestenol, squalene, cholesterol	460 (1750)*

2278	cinnamaldehyde	$C_{14:0}$, $C_{16:0}$, $C_{18:0}$	MAG_{12}		*i*-Pr$C_{14:0}$ phthalate	1280 (3065)*
2279	benzoic acid, cinnamaldehyde	$C_{12:0}$, $C_{14:0}$, $C_{16:0}$, $C_{18:0}$	MAG_{16}, MAG_{18}		borneol, *i*-Pr$C_{14:0}$ phthalate	
2281	benzoic acid, cinnamaldehyde	$C_{9:0}$, $C_{12:0}$, $C_{14:0}$, $C_{16:0}$, $C_{18:0}$, $C_{20:0}$, $C_{22:0}$	MAG_{14}, MAG_{16}, MAG_{18}	C_{18}Ol	*i*-Pr$C_{12:0}$, *i*-Pr$C_{14:0}$, $C_{16:0}$ME, $C_{18:0}$ME, K31, K33, K35 cholestenone	270 (1245)*
2024		$C_{9:0}$, $C_{12:0}$, $C_{13:0}$, $C_{14:0}$, $C_{16:1}$, $C_{16:0}$, $C_{18:0}$			ketone, TA, *i*-Pr$C_{12:0}$, *i*-Pr$C_{14:0}$ $C_{15:0}$	
2267		$C_{9:0}$, $C_{10:0}$, $C_{14:0}$, $C_{16:0}$, $C_{18:0}$	MAG_{16}, MAG_{18}	C_{23}ol	diacid, TA, *i*-Pr$C_{14:0}$ phthalate	
2276		$C_{8:0}$, $C_{9:0}$, $C_{10:0}$, $C_{12:0}$, $C_{13:0}$, $C_{14:0}$, $C_{16:1}$, $C_{16:0}$, $C_{18:0}$	MAG_{14}, MAG_{16}, MAG_{18}	C_{22}ol	TA, *i*-Pr$C_{14:0}$ glycerol, $C_{15:0}$, squalene, oleamide tricaprylate, cholesterol, phthalate	
2380		$C_{14:0}$, $C_{16:0}$, $C_{18:0}$, $C_{20:0}$	TAG_{14}, TAG_{16}, TAG_{18}		TA, *i*-Pr$C_{14:0}$, *i*-Pr$C_{16:0}$, *i*-Pr$C_{18:0}$, WE_{14}, WE_{16} phthalate	
2190		$C_{9:0}$, $C_{12:0}$, $C_{14:0}$, $C_{16:0}$, $C_{18:1}$, $C_{18:0}$		C_{18}ol	*i*-Pr$C_{12:0}$, *i*-Pr$C_{14:0}$	
2191		$C_{10:0}$, $C_{12:0}$, $C_{14:0}$, $C_{16:0}$, $C_{18:1}$, $C_{18:0}$			*i*-Pr$C_{14:0}$, $C_{15:0}$, amide, amine, cholestenol, phthalate	
2268		$C_{12:0}$, $C_{14:0}$, $C_{16:0}$, $C_{18:0}$	MAG_{14}, MAG_{16}, MAG_{18}	C_{15}ol	*i*-Pr$C_{14:0}$ phthalate	
2195		$C_{18:0}$			 amine, phthalate, alkanes	
2242					 phthalate	
2244		$C_{16:0}$, $C_{18:0}$			 phthalate	
2265		$C_{12:0}$, $C_{16:0}$, $C_{18:0}$			Benzoic acid, MAG_{16}, MAG_{18} squalene, phthalate, cholessterol	
2266		$C_{6:0}$, $C_{9:0}$, $C_{10:0}$, $C_{16:0}$, $C_{18:0}$			MAG_{16}, MAG_{18} phthalate	
2270		$C_{8:0}$			K_{11}, C_{18}ME alkanes	

2273					K_{11}, C_{18}ME alkanes	
2277					 phthalate, alkanes	
2280		$C_{16:0}$, $C_{18:0}$			K_{11}, *i*-Pr$C_{18:0}$ phthalate	
5177		$C_{16:0}$, $C_{18:0}$			 phthalate, squalene	

TA– tartaric acid; $C_{x:y}$ – fatty acid with *x* carbons in a chain and *y* degrees of unsaturation; C_xME – methyl ester of fatty acid with *x* carbons in a chain; $C_x$ol – alcohol with *x* carbons in a chain; K_x – ketone with *x* carbons in a chain; MAG/TAG$_x$ – mono/tri acylyglycerides with *x* carbons in a chain attached to the glyceride-backbone; *i*-Pr$C_{x:y}$ – isopropyl conjugation of the given fatty acid; $C_{15:0}$, $C_{17:0}$ – biomarkers of microbial activity (Dudd et al, 1998); squalene – known contaminant of skin lipids, result of bare-handed handling, mostly found along with cholesterol and its by-compounds; glycerol tricaprylate - dilution agent for bone glue; phthalate – plasticizer.

cin (TLE) - Quantification is given for the amounts of cinnamaldehyde out of the total lipid extracts. *Value in parenthesis is the amount of total lipid extract in μg/g ceramic. We assume that the uncertainties are similar to those measured in Table 5.

Isopropyl esters of fatty acids were also observed. Some alcohols and monosaccharides were also detected, as well as small amounts of tartaric acid. One flask contained high molecular weight ketones. The presence of the above compounds is consistent with liquids being stored in the small flasks. Note too that many of the detected compounds are also present in animal fats and plant oils (Behrman and Gopalan, 2005, Christie, 2003, Espenshade and Hughes, 2007). Tartaric acid is thought to be rare in nature, except for grapes. Thus the presence of tartaric acid in ancient residues has long been considered a marker for wine (Guasche-Jané et al., 2006, Guasche-Jané et al., 2004, McGovern, 2009, Michel et al., 1993) though this has recently been debated (Isaksson et al., 2010, Barnard et al., 2010, DeBolt et al., 2006, Stern et al., 2008).

Since all the samples were stored in plastic bags prior to analysis, almost all contain phthalate esters (Table 2) (Shantha and Ackman, 1991, Tomita et al., 1977). Penta- and heptadecanoic acids ($C_{15:0}$ and $C_{17:0}$) were detected in 5 of the lipid extracts (Table 2). These are often attributed to bacterial activity (Dudd et al., 1998), although they are present in animal fats as well (Řezanka and Sigler, 2009). Other possibly extraneous compounds such as squalene, cholesterol, cholestenol and various amides were detected in some of the extracts (Table 2). However, these compounds, excluding squalene, have also been demonstrated to be present in animal fats and plant oils (Behrman and Gopalan, 2005, Christie, 2003, Espenshade and Hughes, 2007). Therefore, a major concern is that the cinnamaldehyde present in 10 of the small flasks is also a modern contaminant.

3.1. The Possibility that Cinnamaldehyde is a Modern Contaminant

Cinnamaldehyde is common in many modern materials that could have come in contact with the archaeological items during their handling or storage, such as insect repellants, sunscreens, hand creams and insecticides (Weller-Fahy et al., 1980, Huang and Ho, 1998, Sharma, 2011). Since contamination is a fundamental issue in cases where the items analyzed were handled with bare hands during excavation and/or are from storage, the best option is to find small flasks during excavation, and then remove them from the field without direct contact with human hands and store them appropriately. We succeeded in obtaining 4 such small flasks (Table 1, nos. 2277, 2278, 2280, 2281). For items originating from storage the best option is to compare them to relevant controls, namely other contemporary vessels from the same storerooms, sites/field projects, and preferably even the same depositional units. We used both these options, as follows:

a) Intact flasks that were freshly excavated or sampled only on the inside. Four flasks from Tel Dor were unearthed while this project was in progress. They were removed from the field with metal tools (rather than with bare hands), were transferred directly to the laboratory and not handled in any way prior to analysis. Two of these flasks (2277, 2280) did not contain cinnamaldehyde, proving that external contamination of cinnamaldehyde from the sediment in which they were buried did not occur. Two small flasks contained cinnamaldehyde in high concentrations (2278 and 2281) (Table 2, last column). This observation strongly supports the possibility that cinnamaldehyde in these two small flasks is derived from their original contents. We do note that even these samples contained some phthalate esters, most probably from the plastic bags that were in contact with the flasks.

b) Other Early Iron Age Containers. Table 4 shows the lipids extracted from 9 other early Iron Age containers, most of them of the Phoenician Bichrome group (Table 3, section A). None of these vessels contained cinnamaldehyde, even though they were clearly contaminated with other molecules. Six of these containers are from the same sites and same excavation projects from which the flasks that contained cinnamaldehyde were obtained (Dor and Qasile). These control containers were treated in the same manner as the small flasks during and after the excavations: all of them were extensively handled, some of them glued for restoration, they were stored in the same store-rooms under identical conditions and underwent the same extraction and analysis procedures in the laboratory. The other three containers are from another old excavation at a Phoenician site (Tell Abu-Hawam). These vessels too were extensively handled, and stored in various store-rooms for nearly a century. They also did not contain cinnamaldehyde. All these observations are consistent with the small flasks not being contaminated by cinnamaldehyde.

c) Vessels from the Same Depositional Units as the Small Flasks. Six ceramic vessels of different types were analyzed from the same loci at Tell Qasile that produced small flasks nos. 2019, 2269, 2271 and 2272, with cinnamaldehyde (Table 3, section B). None of these other vessels contained cinnamaldehyde.

Two kraters were sampled from the same context as Dor flask 2278, and they too contained no cinnamaldehyde (Table 3, section B). All these vessels underwent identical treatment as the small flasks during and after excavation, by the same individuals, some were glued for restoration and they were stored in the same store-rooms.

Table 3. Details of other vessels sampled as controls. Locus numbers are indicated only for vessels originating from the same depositional units as small flasks with cynnamaldehyde.

Lab. No.	Site	Description	Decoration	Preservation
A. Other early Iron Age containers (see also nos. 2021, 2022 below)				
2608	Dor	Small lentoid flask	Bichrome	Upper part only; handled and washed; long-term storage
2609	Dor	Jug	Bichrome	Large fragment; handled and washed; long-term storage
2610	Dor	Small lentoid very thick flask	Bichrome	Nearly half a vessel; handled, washed, glued; long-term storage
2611	Dor	Rounded jug	Bichrome	Large fragment; handled and washed; long-term storage
2025	Tell Abu Hawam	Rounded flask	Red slipped	Intact; handled and otherwise treatment unknown; long-term storage
2026	Tell Abu Hawam	Rounded jug	Bichrome	Intact, rim and part of neck missing; handled and otherwise treatment unknown; long-term storage

2027	Tell Abu Hawam	Rounded jug	Bichrome	Intact, half the neck missing and hole in body; handled and otherwise treatment unknown; long-term storage
2020	Tell Qasile	Rounded Bichrome jug	Bichrome	Intact other than neck/handle; handled, washed, glued; long-term storage
2023	Tell Qasile	Small lentoid flask	Bichrome	Intact; handled, unknown if washed; long-term storage
B. Vessels form the same depositional units as the small flasks with the cinnamaldehyde				
3205	Dor L09D2-386	Krater	--	1/4 vessel; handled, washed; stored since 2009
3206	Dor L09D2-386	Krater	--	1/2 vessel; handled, washed, glued; stored since 2009
2021	Tell Qasile L188	Large two handled flask	Bichrome	Complete; handled, washed, glued; long-tem storage
3202	Tell Qasile L188	Store-jar	--	Fragmentary; handled, washed, glued; long-term storage
3201	Tell Qasile L188	Lamp	--	Intact; handled, washed; long-term storage
2022	Tell Qasile L227	Large spoon flask	Red slip and black decoration	Intact; handled and washedexternally; long-tem storage
3200	Tell Qasile L227	Dipper juglet	--	Nearly intact other than neck; handled, washed, glued; long-term storage
3203	Tell Qasile L227	Bowl	--	Intact; handled, washed; long-term storage

Table 4. Molecular components in the total lipid extracts of the control vessels listed in Table 3. Abbreviations as in Table 2

Lab no.	Cinnamaldehyde	Fatty acids	AG	Alcohols	Contaminants and Others
A.					
2608		$C_{16:0}$, $C_{18:0}$			alkanes, phthalate
2609		$C_{16:0}$, $C_{18:0}$			alkanes, phthalate
2610		$C_{16:0}$, $C_{18:0}$			alkanes
2611					phenol, K11, alkanes, phthalate
2025		$C_{16:0}$, $C_{17:0}$, $C_{18:0}$			alkanes, phthalate, nicotin acid
2026		$C_{10:0}$, $C_{12:0}$, $C_{14:0}$, $C_{15:0}$, $C_{16:1}$, $C_{16:0}$, $C_{18:1}$, $C_{18:0}$			alkanes, phthalate, cholestenol,
2027					alkanes, phtalate
2020					alkanes, phtalate
2023		$C_{12:0}$, $C_{14:0}$, $C_{15:0}$, $C_{16:0}$, $C_{18:1}$, $C_{18:0}$			alkanes, phthalate, cholestane
B.					
3205		$C_{14:0}$, $C_{16:0}$, $C_{18:0}$			phthalate
3206		$C_{9:0}$, $C_{14:0}$, $C_{16:0}$, $C_{18:0}$			benzoic acid
2021		$C_{9:0}$, $C_{10:0}$, $C_{12:0}$, $C_{14:0}$, $C_{16:1}$, $C_{16:0}$, $C_{18:1}$, $C_{18:0}$	MAG_{16}	$C_{18}Ol$, $C_{19}Ol$	alkanes, phthalate, cholestenol, cholesterol

3202		$C_{9:0}$, $C_{16:0}$, $C_{18:0}$			alkanes, phthalate
3201		$C_{16:0}$, $C_{18:0}$			benzoic acid, alkanes, phthalate
2022		$C_{14:0}$, $C_{16:0}$, $C_{18:1}$, $C_{18:0}$			alkanes, phthalate
3200		$C_{9:0}$, $C_{14:0}$, $C_{16:0}$, $C_{18:1}$, $C_{18:0}$			phthalate
3203		$C_{9:0}$, $C_{16:0}$, $C_{18:0}$			phthalate

We therefore conclude from all the above observations that the cinnamaldehyde found in the 10 small flasks is most likely derived from the original contents of the vessels and not from contaminants.

3.2. The Preservation of Cinnamaldehyde and the Absence of its Breakdown Products

Cinnamaldehyde is sensitive to aerobic oxidation and breaks down into cinnamic acid, benzaldehyde and benzoic acid (Friedman et al., 2000). It has been shown that selective oxidation of cinnamaldehyde to benzaldehyde occurs under mild conditions in the presence of a polymeric catalyst (Yang et al., 2013).

It is therefore surprising that cinnamaldehyde is preserved at all, let alone in relatively large concentrations. We do not know why the cinnamaldehyde is preserved, but suspect that it is due to the adsorbed cinnamaldehyde molecules being more stable than the non-adsorbed molecules. We note for example that in bone the relatively unstable non-collagenous proteins are relatively well preserved compared to the more stable collagen.

It is known that the former are closely associated with the mineral phase and are hence assumed to be stabilized (Hare et al., 1980, DeNiro and Weiner, 1988, Tuross et al., 1989). Note too that Friedman et al. (2000) showed that cinnamaldehyde in cinnamon essential oil is highly resistant to oxidation, as opposed to the pure cinnamaldehyde.

Among the breakdown products of cinnamaldehyde, benzoic acid is present in nearly all the flasks that produced cinnamaldehyde, though in small amounts (Table 2; Fig. 3). Benzaldehyde has too low a mass to be detected using the standard injection procedure applied here.

However, it is still surprising that the direct breakdown product, namely cinnamic acid, is absent in the extracts. In order to better understand this we carried out a series of experiments in which cinnamaldehyde and cinnamic acid were adsorbed separately into three different modern ceramic substrates.

We found that under the conditions used to extract the lipids from the archaeological ceramics, the cinnamaldehyde was readily extracted (Table 5). Cinnamic acid however was not extractable, nor was it present in the extracts containing cinnamaldehyde.

Note that the same phenomenon was detected for another acidic molecule, tartaric acid, namely that it was extracted in negligible amounts after being absorbed into different modern ceramic substrates.

Table 5. Lipid extracts (μg/g) after placing 10 mL solution of standard mixtures (10^{-2} M) onto ceramic substrates fired to 550, 770 and 940 ºC. The estimated error is based on duplicate injections into the GC. Numbers in ( ) show the yields obtained. --- not analysed

	550 ºC	**770 ºC**	**940 ºC**
Cinnamaldehyde	4360±75 (33%)	4440±50 (33%)	2830±50 (21%)
Myristic acid	2700±65 (12%)	3330±75 (14%)	1996±0 (9%)
Tartaric acid	58±3	61±1	65±10
Cinnamic acid	0	0	---

As cinnamic acid and tartaric acid are charged, they probably interact strongly with the mineral surfaces exposed in the pores of the ceramic. This inability to extract cinnamic acid and tartaric acid using the commonly used organic solvent-based extract protocol applied here, may account for the absence of cinnamic acid in the extracts from the flasks. We cannot however exclude the possibility that cinnamic acid is not present in the ceramics.

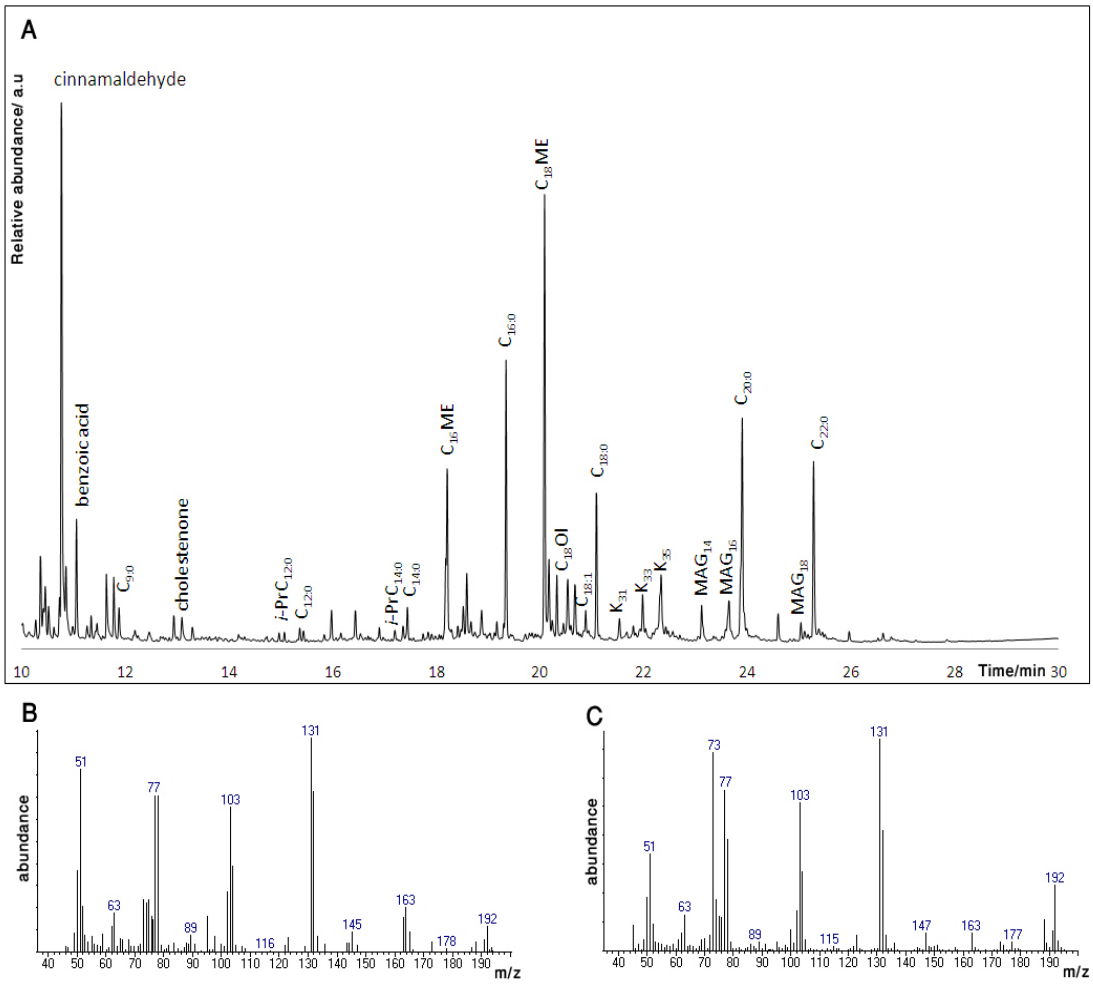

Figure 3. A. Gas chromatogram of flask 2281, demonstrating the presence of cinnamaldehyde in its organic extract; B. MS of the cinnamaldehyde in the archaeological extract. C. MS of cinnamaldehyde standard, purchased from SAFC (cat. no. W22,861-3-K)

4. DISCUSSION

Cinnamaldehyde was extracted from 10 of the 27 small flasks, including two that were untouched. Cinnamaldehyde was absent in all the controls. We therefore conclude that this compound was derived from the substances stored in the small flasks in antiquity and was not likely to have been introduced during deposition or after the vessels were excavated.

4.1. Possible Sources of Cinnamaldehyde

Cinnamaldehyde should be found in almost every plant since it is generated as part of the Shikinic acid pathway that ends up with the formation of lignin (Whetten and Sederoff, 1995, Neish, 1968), one of the major structural materials of terrestrial plant cells (Clark, 1991, Kim et al., 2000). However, in most of the plant kingdom this lignin biosynthesis pathway involves very low concentrations of the intermediate reactants, including cinnamaldehyde. In fact, cinnamaldehyde has been found in trace quantities in only a few plant species (Clark, 1991). The only plant that is endemic to the Levant, including Israel, which has been reported to contain cinnamaldehyde in its essential oil is Laurus nobilis (Caredda et al., 2002), but the amounts are negligible. Moreover, no other major compounds of this plant's essential oil, such as monoterpenes, linalool or 1,8-cineole, were detected in any of the small flask extracts. We conclude that it is unlikely that Laurus nobilis is the origin of the cinnamaldehyde in the small flasks.

The only plant group that accumulates large quantities of cinnamaldehyde is Cinnamomum, also of the Lauraceae family. The large amounts of cinnamaldehyde found exclusively in the

Cinnamomum species are attributed to a kinetic bottleneck in the lignin formation mechanism that results in its accumulation. Cinnamaldehyde is one of the three major compounds of cinnamon (Tomaino et al., 2005). The Cinnamonum family has more than 100 species, all native to South and Southeast Asia (Clark, 1991). The stem bark oil of Cinnamomum verum (Cinnamomum zeylanicum) contains 65–97% cinnamaldehyde out of its total volatiles (Senanayake and Wijesekera, 1989). The volatile oil from its leaves contains no cinnamaldehyde (Singh et al., 2007). Cinnamomum cassia contains almost 95% cinnamaldehyde in both its bark and leaf oil (Senanayake and Wijeskera, 2004). On the other hand, Cinnamomum camphora contains no cinnamaldehyde in its essential oil (Ravindran et al., 2004), and thus it can be excluded from the list of possible plant sources.

Cinnamomum verum occurs naturally mainly in Sri Lanka and southern India, and also in the Tenasserim hills of Myanmar (Ravindran et al., 2004). Cinnamomum cassia (Chinese/Vietnam cassia) is native mainly to Southeast China and Vietnam, also to Laos and Myanmar (Kim Dao, 2004, Ravindran and Babu, 2004, Ravindran et al., 2004, Senanayake and Wijeskera, 2004).

If cinnamon bark is the source of the cinnamaldehyde in the small flasks, we do not know how in ancient times this compound was extracted from the bark and entered into the ceramic. As the apertures of these flasks are small, only powdered bark or a liquid extract could have been introduced into them.

4.2. Preliminary Historical Observations and Implications

In the 11th–10th centuries BCE, cinnamon could only have originated in South/South East Asia since these plants did not grow naturally, nor were they cultivated anywhere else before early modern times (Pearson, 2003). To date the earliest literary attestations of South/South East Asian spices reaching the 'West' relate to the 6th-5th centuries BCE. Most notable is Herodotus' description of cassia and cinnamon (Herod. Histories 2.86, 3.107, 111). By Roman times the European spice trade with Asia was extensive and is well documented (Miller, 1969, Sidebotham, 2011, Tomber, 2008). Our findings imply that such trade may have begun at least half a millennium earlier than hitherto assumed.

It is interesting to note that occasionally remains of Far Eastern spices have been reported from other 'Western' contexts preceding the second half of the 1st millennium BCE, and even preceding our finds. Cloves, native to the Moluccas in eastern Indonesia, were reported from Terqa on the middle Euphrates, and dated to ca. 1600 BCE (Buccellati and Buccellati, 1983). Nutmeg remains, from the same region, were identified at Deir el-Bahari in Egypt, dating to the 18th Egyptian Dynasty (16th–14th centuries BCE) (Naville et al., 1913). These two finds, however, were not fully described and cannot be well evaluated. In contrast, black pepper grains of Indian origin were positively identified in the mummy of Pharaoh Ramesses II (13th century BCE). These were well documented by Plu (Plu, 1985) and they attest unequivocally that indeed, spices from South East Asia reached the 'West' already during the Late Bronze Age. Also well documented is a cinnamon flower which is later than our flasks, but still earlier than the conventional date for the beginning of long-range Asian spice trade. It was identified in a late Iron Age (7th century BCE) context at Samos in the east Aegean (Kučan, 1995). These findings have largely been overlooked by scholars dealing with trade in the second and first millennia BCE.

There are additional indications for such early contacts between South/South East Asia and the West. Most relevant to this study is the multidisciplinary SEALINKS research project, investigating contacts between Asia, Arabia and Africa from a longue durée perspective (Boivin et al., 2009, Boivin and Fuller, 2009, Fuller et al., 2011). Inter alia these scholars suggest that faunal and botanical translocations, especially crop transfers between India and China on the one hand and east Africa on the other, attest to rather systematic trade between these regions starting about the beginning of the second millennium BCE and increasing in scope during the Middle and Late Bronze Ages (their Phase III). According to this study, maritime activities subsequently intensify in the Iron Age (Phase IV). The second millennium contacts are seen as the precursors of the South and South East Asian

spice trade. Our findings are consistent with these observations.

Much more needs to be done in order to contextualize these finds and identify additional evidence for early Iron Age long-range trade between the Far East and the Levant. Only then can we begin to understand the commercial networks over this vast expanse in this period. This entails, for example, addressing the allusion to cinnamon and cassia in texts in the Old World (such as Middle and Late-Kingdom Egyptian texts and the Hebrew Bible) as well as independent information provided by other media (Caubet and Yon, 2006). Above all, it calls for a consideration of the societies that participated in these networks, consumption contexts, the modes of exchange and the routes / interaction spheres through which all this could have been achieved.

5. CONCLUSIONS

We identified cinnamaldehyde in small lentoid-shaped Phoenician flasks of the early Iron Age. The possibility that this compound is a depositional or post-depositional contaminant is unlikely.

The most probable source of cinnamaldehyde is the plant Cinnamomum, which until early modern times grew only in South East Asia. This observation raises the intriguing possibility that long-range spice trade from the Far East westward may have taken place some 3000 years ago.

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