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Sulphur, fats and beeswax in the Iberian rites of the sanctuary of the *oppidum* of Puente Tablas (Jaén, Spain)

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ABSTRACT

This paper discusses the results of chemical analysis with regard to a major ceramic set retrieved from the sanctuary of Puerta del Sol. The site dates back to the 5th century BC and lies in the Iberian *oppidum* of Puente Tablas in Jaén, Spain. This is the first research paper on the Iberian culture to identify and describe accurately a sanctuary rite according to data based on chemical analysis. The chemical markers identified here confirm the occurrence of native sulphur, vegetable fat, ruminant animal fat and beeswax in the vessels under study. Methodologically, lipid analysis took place in two stages: GC–MS analysis of all the vessels, and HPLC–APCI-MS of the vessels with a lipidic profile capable of specifying the occurrence and the structure of triacylglycerols.

1. Introduction

The complexity of Iberian rites becomes apparent in cult spaces such as the sanctuary Puerta del Sol of the *oppidum* of Puente Tablas, in Jaén. Unlike the basic, standard pattern, it is an exceptional Iberian sanctuary that comprehends a large number of specific properties (spatial, symbolic, material, functional, etc.). These properties result in a sanctuary that follows a complex mythology of the female deity. The female deity supports a number of rites that define the aristocratic cult of an Iberian lineage in the late 5th century BC and in the first half of the 4th century BC (Ruiz et al., 2015).

As a complex set of rites and ceremonies, the Iberian cult may manifest as a wide range of expressions. In turn, these are related with a variety of properties and deities with specific variants (Rueda, 2011). The Iberian cult expresses a number of rules and norms on religious ceremonies (of fertility, healing, death, heroification, marriage, youth, etc.), and these can be observed in a range of spatial and chronological contexts (Rueda, 2013). In this general frame, it is important to remark that ritual behaviour's manifestations and materializations can be delimited (Bell, 1992), because cultural practice reflects specific actions and, naturally, sets in a specific liturgical structure (Insoll, 2011).

Rites materialize as 'artefacts' that act as active instruments for the achievement of the rite, and as images that narrate the moment chosen for a wider recognition. From a methodological point of view, this approach to the study of rites requires the analysis of behaviour models that find a reflection in the archaeological record. These models allow approaching symbolic patterns as a conditioned expression that is connected with the liturgical structure. The latter often uses a sanctioned language that is also socially understandable. In the case of the Iberians, these processes can be tracked down via a range of materials and which demand an articulated, interdisciplinary research method (Rueda, 2013). Food offerings and/or sanctuary commensality are a clear example: they can be fully understood only by use of articulated, complementary diagnostic techniques such as chemical analysis, archaeozoological and carpological analysis, ceramic typology, iconographic studies and studies of written sources.

This paper deals with the chemical analysis of contents in a large ceramic set of the ritual area of the sanctuary Puerta del Sol. The aim is to gain a deeper knowledge of the rites performed in the sanctuary. The chemical analysis is complemented and fed back by the information provided by archaeological analysis, by the study of written sources and by the analysis of the wildlife and seeds (Montes, 2015; Rodríguez and Pradas, 2015; Ruiz et al., 2015). The method used for the chemical analysis is based on the joint use of gas chromatography–mass spectrometry (GC–MS) and high-performance liquid chromatography–atmospheric

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pressure chemical ionization-mass spectrometry (HPLC–APCI-MS) (Craig et al., 2009).

Chemical analysis of contents in ceramic vessels, specifically of lipids, has made substantial progress in the past 30 years. Use of GC–MS for research on trimethylsilylderivates (TMS) in the 90 s allowed identifying particularly relevant markers in archaeology, such as mono-, di-, triacylglycerols (TAGs) and wax esters (Evershed et al., 1990, 1992). The method prevailing until then relied on the formation of fatty acid methyl esters (FAMEs) for subsequent GC–MS analysis. Use of the latter method meant the loss of the information contained in acylglycerols and wax esters. The improvement in the methods available has thus resulted in more accurate identification of highly relevant archaeological contents such as animal and vegetable fats, beeswax and vegetal wax, drinks and resins (Colombini et al., 2005; Evershed et al., 1991, 2008; Garnier et al., 2003; Parras et al., 2011; Regert et al., 2001, 2003; Sánchez et al., 1998).

A range of variants have followed the development of the above method over the past few years. In some specific cases of analysis of ceramic materials, pyrolysis has been added as the extraction system for GC–MS (Py-GC–MS) (Garnier et al., 2003; Legnaioli et al., 2013). Still, Py-GC–MS has been used more often for other materials. Recent progress in MS and hyphenated techniques allows the identification of carbon stable isotope compositions. Using a highly sensitive device such as a gas chromatograph–combustion–isotope ratio mass spectrometer (GC–C–IRMS) and plotting the $\delta^{13}C$ values of methyl palmitate against those of methyl stearate, several animal and marine fats can be identified, and the occurrence of milk can be attested (Craig et al., 2011; Cramp et al., 2014; Evershed et al., 2008; Heron et al., 2013; Regert, 2011). The third variant, used for this paper along with GC–MS, is HPLC–APCI-MS.

Unlike former chromatographic techniques, HPLC–APCI-MS has been used recently for the analysis of lipidic contents in Archaeometry. In fact, this technique can isolate and identify intact TAGs more accurately than other techniques. Thus, it is possible to associate TAG profiles with a range of fat types by HPLC–APCI-MS (Saliu et al., 2011). First, TAGs with a high content of unsaturated fatty acids (oleic, linoleic, linolenic) are associated with vegetal fats (Fasciotti and Pereira, 2010; Kimpe et al., 2001). Second, TAGs of saturated fatty acids are associated with animals; some of them are so specific that they can be considered biomarkers of fat of specific animal species (Kimpe et al., 2002; Romanus et al., 2007; Saliu et al., 2011). Finally, the difference between ruminant animal fat and non-ruminant animal fat is based on the proportion of palmitic and stearic acids in the *sn*-2 position of TAG (P:S ratio) (Evershed et al., 2002; Regert, 2011; Saliu et al., 2011).

HPLC has a lower potential in archaeological research, even if it has led to identification of a range of contents in ceramic vessels. Thus, a substance that could be identified as olive oil was identified in lamps (Kimpe et al., 2001), and ruminant fat was identified in cooking vessels too, both in the late Roman materials of the site of Sagalassos (Turkey) (Kimpe et al., 2002). In the latter case, the TAG profiles found in the residue were compared with the TAG profiles of present-day animal fat samples used as a reference. Using the same basis of comparison, Craig et al. (2009) presented a dataset obtained from research on food residue absorbed in Early Iron Age ceramics from the site of Gordion (Turkey). HPLC–APCI-MS has also proved efficient for the identification of ruminant fat, as attested by the comparison with GC–MS and GC–C–IRMS analysis (Romanus et al., 2007). HPLC–APCI-MS analysis has also been recently considered useful for TAGs analysis in a piece of research on 18th century majolica ceramic apothecary jars and conserved in the ancient Apothecary of the Aboca Museum (Sansepolcro, Arezzo, Italy) (Saliu et al., 2011).

2. The archaeological context

The ceramic material analysed here comes from the sanctuary Puerta del Sol, in the Iberian *oppidum* of Puente Tablas (Jaén, Spain). The Iberians are known to have been the most advanced culture in the Iberian Peninsula during the Iron Age. The Iberian world developed from the 6th century BC and reached its zenith during the 5th to the 3rd century BC. Geographically, the Iberian culture spread over the south, east and north of the Iberian Peninsula, as well as over the southeast of present-day France, and represented a clearly Mediterranean culture (Ruiz and Molinos, 1998).

The *oppidum* of Puente Tablas lies 4 km north of Jaén (Andalusia, Spain) (Fig. 1). It is a triangular plateau of approximately 6 ha of surface with a pointed shape towards the northwest. It is heavily fortified with walls and towers except at the base of the triangle, which looks on River Guadalbullón and, therefore, forms a natural defence (Fig. 2). Chronologically, the settlement was occupied by the Iberians from the late 8th century BC to the late 3rd century BC (Ruiz and Molinos, 2008, 2015).

The sanctuary Puerta del Sol is next to the east gate of the *oppidum*. It is the main entrance, at the end of a 14 m corridor, and it is also where an anthropoid stone stele stands that represents the female deity (Ruiz et al., 2015). The construction has a surface of 300 m^2 and is arranged as three terraces and several rooms with a stone board and mud walls with a covering. In the more important rooms, the covering is gypsum (Figs. 3 and 4) (Parras et al., 2015; Sánchez et al., 2014).

Access is through the first terrace across a yard (Room R). This room also contains a bullskin-shaped stone altar. This is the landmark for access to the *antecella* (Room Q) and to the area dedicated to the divinity, *cella* (Room F). The latter houses the small chapel-like room (Room M) that would contain the stele for a part of the year.

The side of the yard leads to the second terrace, which is divided into several functional spaces. A quadrangular-shaped, two-storey tower lies to the east. The inside has a gypsum covering. Two *bothroi* made with remains of lamb sacrifices were at the entrance (Room J). A yard with two paved levels lies opposite (Rooms U and Z). This space has been interpreted as used for rites. Four small caves and three small altars are on the west corner. The altars are of various shapes: quadrangular, eye-shaped and ear-shaped. The caves, insofar as liminal spaces in the symbolic sense, are physically marked off from the construction space by a small stone wall. This is a *temenos* whose threshold was not allowed to be trespassed. Actually, no offerings have been recorded inside the caves. This is in sharp contrast with the record of votive offerings in Rooms U and Z, where storage vessels, small vases and a loom weight stand out. The loom weight combines a bullskin and a lotus flower in a small stamp. Attic grave offerings, namely two cups and a red-figure krater are especially remarkable. The krater depicts an initiation before the statue of a female deity, probably Aphrodite or Artemis. The fourth wall is separated by a wall. The entrance is through a trapeze-shaped room (Room V). This is a room of limited access, because this is the largest and deepest cave, and the one where the myth of the deity is born.

The third terrace is the highest one, and it is used for service. Specifically, a cistern for water supply through a channel that runs across the sanctuary has been attested here. Water is a highly symbolic element and a key functional element in this cult space.

The female deity defines the cult of this sanctuary and sanctions aristocratic practice. The stone stele depicts a lady wearing a mitre and with her arms folded on her belly and her hands holding a small circular element, perhaps intended to represent the sun. This deity is associated with equinoxes as vital landmarks of the ritual calendar. These landmarks signal their location opposite the entrance corridor and would receive the first sunbeams at dawn. Only some minutes later, the second stage of the mythical narrative of the deity takes place. The sunbeams enter the fourth cave and create a scenographic effect of a visual representation of the sacred. This religious event strengthens collective identity by way of the construction of their mythology (Hamilakis, 2011). These two stages make up the sequence of a myth based on the association between the sun and the deity. This myth describes the deity's return to her dwelling, the cave. Alternatively, it can be interpreted as her

Fig. 1. Location of the *oppidum* Puente Tablas.

reunion with a dead male god or a hero who, as a result, rises again (Ruiz and Molinos, 2015).

3. Method

The first stage of the chemical analysis was GC–MS analysis of a set of 126 ceramic vessels. HPLC–APCI-MS analysis was then used on the samples that showed any fat-related marker, in order to identify the TAG types and obtain the P:S ratio in the *sn*-2 position. Regardless of the specificities of the HPLC–APCI-MS analysis described below, the following remarks on the method should be mentioned.

The most efficient TAGs separations have been obtained using reverse-phase HPLC (Craig et al., 2009; Holčapek et al., 2003; Kimpe et al., 2001, 2002; Lísa et al., 2011; Romanus et al., 2007; Saliu et al., 2014). Reverse-phase HPLC is based on the equivalent carbon number (ECN) (Holčapek et al., 2003; Lísa et al., 2011). This parameter helps understand the elution order of these species in a chromatogram, because the TAG retention time increases as its ECN increases too. The mass

Fig. 2. Aerial photo of the *oppidum* of Puente Tablas.

Fig. 3. East gate and sanctuary Puerta del Sol.

spectrum in most TAGs typically showed protonated molecular ions $[M + H]^+$ and diacylglycerol ions $[M\text{-}RCO_2]^+$.

Use of APCI as an ionization source has been proved to contribute to relevant data collection in AAB and ABC TAGs mass spectrometry. The data are not only about which fatty acids are in each TAG, but also about the position in which the fatty acids estearify each of the three positions of the glycerol skeleton. This allows an estimate of the regioisomeric composition of TAGs based on the contrast in the relative abundance of diacylglycerol $[M\text{-}RCO_2]^+$ fragments. The formation of ion $[M\text{-}RCO_2]^+$ resulting from the loss of acyl group in the *sn*-2 position is energetically less favourable with respect to the ions formed by rupture of acyl in the *sn*-1 or *sn*-3 position (Fauconnot et al., 2004; Jakab et al., 2003; Leskinen et al., 2007; Mottram et al., 1997, 2001). This is the case of SSP/SPS regiomers (S: stearic acid and P: palmitic acid). Both are difficult to distinguish, because they present the same ions $[M\text{-}RCO2]^+$ i.e. $[SS]^+$ and $[SP]^+$ with an m/z ratio of 607 and 579, respectively. Taking this into consideration, these isomers can be distinguished according to the $[SS]^+/[SP]^+$ ratio: the SSP isomer's ratio is close to 1, whereas the SPS isomer's ratio is considerably lower.

This regioselective analysis leads to the identification of a fatty acid in the *sn*-2 position of glycerol. This is crucial for the analysis of contents in view of its direct association with the type of residue present. Vegetable fat can thus be distinguished from animal fat. In the latter case, it may be even possible to tell ruminant from non-ruminant fat based on the proportion of palmitic and stearic acid in the *sn*-2 position. Ruminant fats show a P:S ratio in the *sn*-2 position of ca. 60:40, whereas the ratio is closer to 95:5 in non-ruminants (Evershed et al., 2002; Regert, 2011; Saliu et al., 2011).

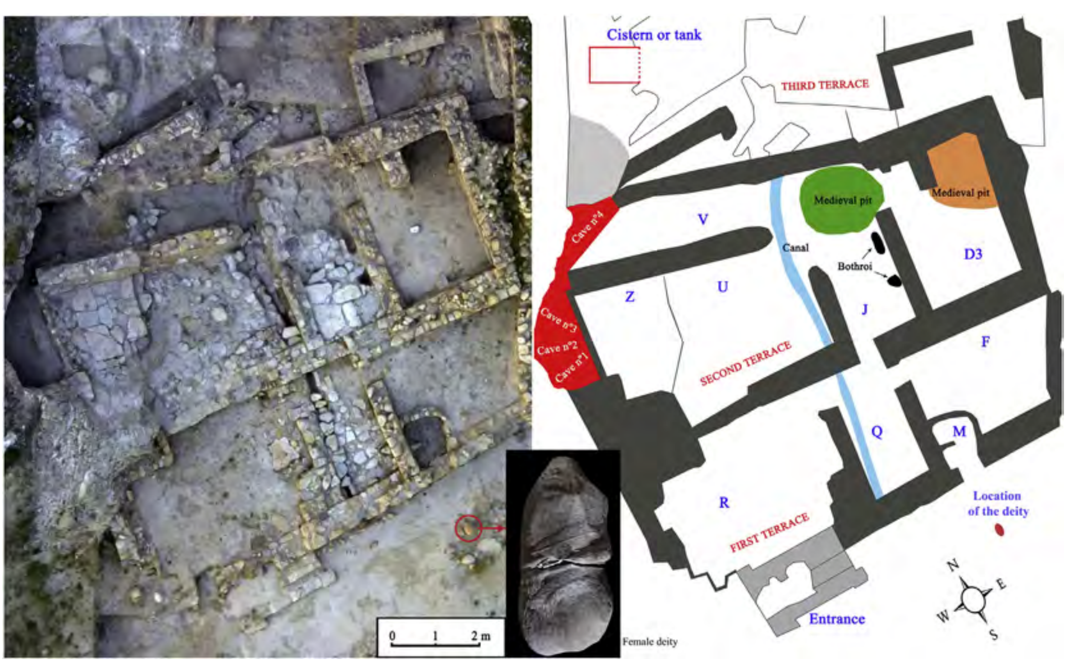

Fig. 4. Photo (left) and plan of the sanctuary Puerta del Sol (right) and distribution of rooms, caves and stele of female deity.

3.1. Sampling

The set under study consists of 126 vessels retrieved from the 8 main rooms of the sanctuary's two first terraces (Rooms R, Q, F, M, Z, U, J, D3). Only one (Room V) was discarded, because it did not contain any archaeological material. The vessels were selected according to the availability and identification of a minimum number of vessels. This second criterion took into consideration the availability of clearly different bases, rims, walls, types of ceramic pastes and sizes, in order to ensure no two parts of the same vessel were analysed. This sampling method resulted from two major causes: the delay in the detailed study of the archaeological materials (it would have undoubtedly identified accurately the types of vessels in the sanctuary), and the need to carry out the research as fast as possible in order to prevent potential degradation following the retrieval and storage of the archaeological materials.

The final sample thus included a significant range of vessels. Among these, the following stand out as the most common ones: storing vessels (mainly amphorae) and small vessels (mainly bowls and dishes) (Table 1). The sampling area was the area of the vessel that remained, so the sample was sometimes taken from the base and sometimes from the body of the vessel.

3.2. Gas Chromatography–Mass Spectrometry (GC–MS)/High Performance Liquid Chromatography–Atmospheric Pressure Chemical Ionization-Mass Spectrometry (HPLC–APCI-MS)

3.2.1. Archaeological samples

Approximately 4 g of the bottom or of the body of the vessel was sampled. The fragments of archaeological vessels selected for analysis were wrapped each in a piece of dark paper and stored in a freezer at least at −20 °C until their analysis. The fragments were then taken out of the freezer and a sample was collected. Any remains of soil were removed with an electric hand-drill. The sample was then ground to the appropriate size in an agate mortar. 2 g were taken for the analysis.

3.2.2. Lipid extraction

Extraction is in accordance with the procedure described in Evershed et al. (1990). 10 μL of tetratriacontane (internal standard) and 10 mL of the mixture chloroform/methanol ($CHCl_3$:MeOH) (2:1 *v*/*v*) were added to 2 g of the ground ceramic fragment. Lipids were extracted with ultrasound for 15 min. The solution was centrifuged (3500 rpm, 5 min) eliminating the remaining ceramics and removing the supernatant where the lipids are solved. This process was repeated twice. The extract portions were combined into one for solvent evaporation under N_2 stream. The dry extract was solved again in 500 μL of $CHCl_3$. The extracts were stored in a deep freeze (−20 °C) until required for GC–MS and HPLC–APCI-MS.

3.2.3. Derivatization

A 100 μL aliquot was removed and transferred to a smaller vial. This volume was evaporated to dryness under an N_2 stream. N,O-bis-(trimethylsilyl) trifluoroacetamide (BSTFA) with 1% trimethylchlorosilane (TMCS) was used as a derivatization agent. The derivatization reaction took place with 20 μL of this reagent at 70 °C for 30 to 40 min. When the reaction was over, the vial was cooled and the remaining derivatizing agent was evaporated under an N_2 stream. The sample was then dissolved again in 50 μL of cyclohexane. An amount of 1 μL of the sample was injected into the chromatograph.

Table 1
Sample number and distribution in the sanctuary arranged by specific rooms. US: undetermined shape.

Room	No. of samples	No. of vessels and types
R	12	9 storing vessels, 1 dish/bowl, 2 bowl
Q	8	5 storing vessel, 2 US
F	19	14 storing vessel, 2 dish, 3 US
M	1	1 storing vessel
Z	14	2 storing vessel, 2 dish/bowl, 10 US
U	39	8 storing vessel, 6 dish/bowl, 1 patera, 1 pot, 23 US
J	21	10 storing vessel, dish/bowl, 2 pot, 7 US
D3	12	4 storing vessel, 1 dish/bowl, 7 US

3.2.4. GC–MS analysis

The analyses were performed using a quadrupole type HP 5989B MS coupled to an HP 5890A series II plus GC. The samples were introduced by on-column injection into a 15 m × 0.25 mm I.D. fused silica capillary column, coated with poly(dimethylsiloxane) stationary phase with 0.1 μm film thickness. Helium was used as the carrier gas (purity 99.99%) at a flow speed of 1.2 ml/min. The GC oven temperature programme was as follows: initially at 50 °C, held for 2 min, ramp to 350 °C at 10 °C/min, held for 10 min.

The mass spectrometer conditions were an emission current of 100 μA, an electron energy of 70 eV and an ionization source temperature of 300 °C. Total ions measurements were obtained in the mass spectrometer that scanned from m/z 50–900 at a scan rate of 1,15 scan/s. The GC–MS capillary interface was maintained at a temperature of 350 °C.

3.2.5. HPLC–APCI-MS analysis

A 100 μL aliquot was removed and transferred to a smaller vial, evaporated to dryness under an N_2 stream and solved again in 200 μL of isopropanol. An amount of 20 μL of sample was injected into the chromatograph.

The analyses were performed on an Agilent 1290 Infinity HPLC system. The final HPLC method for TAG analysis used the following conditions: a chromatographic column Zorbax Eclipse Plus C18 (150 mm × 4.6 mm, 1.8 μm particle size), column temperature of 30 °C and the mobile phase gradient, at a flow rate of 0.4 mL/min, of 3 min 50% A (methanol) + 50% B (isopropanol), 10 min 10% A + 90% B, 20 min 100% B, held for 5 min. The mobile phase was prepared fresh everyday before the analyses.

The HPLC system was connected to an Agilent 6220 TOF (time-of-flight) mass spectrometer equipped with an APCI interface operated in the positive ionization mode, using the following operation parameters: capillary voltage, 3000 V; vaporizer temperature: 375 °C; nebulizer pressure, 60 psi; drying gas flow rate, 5 L/min; gas temperature, 325 °C; corona current: 4 μA. The time-of-flight analyser was used in the mass range m/z of 100–1300, with a scan time of 1 s.

4. Results

GC–MS positive results were obtained for 41 out of 126 vessels, i.e. 32.53%. Lipidic markers were found in 16 out of the above 41 vessels, thus qualifying for HPLC–APCI-MS analysis. This technique yielded more and better information of the TAGs in 8 vessels. The P:S index in the *sn*-2 position was recorded in 6 of them (Table 2).

The chemical markers recorded confirm the occurrence of native sulphur, vegetable and animal fat and beeswax in the ceramic vessels. Their combinations and mixtures vary in 16 vessels, whereas in the remaining 25 only sulphur is identified. Regardless of the interpretation of these results and their meaning in the context of the sanctuary's ritual (see below), the identification of these contents is based on the following evidence.

The most frequent chemical marker is *native sulphur*. It occurs in 34 vessels: in 25 of them it occurs alone (Fig. 5) and in the remaining 9 it occurs combined with vegetable fat, animal fat and/or beeswax. Most of the vessels where sulphur was recorded were found in Room U (25) and, to a lesser extent, in Room J (6). The occurrence of sulphur in the krater and in one of the Attic ceramic cups retrieved in Room Z is worthy of special note.

Table 2
GC–MS and HPLC–APCI-MS results. US: undetermined shape, A: amphorae, M: myristic acid, Pd: pentadecanoic acid, P: palmitic acid, Po: palmitoleic acid, Ma: margaric acid S: stearic acid, O: oleic acid, L: linoleic acid, W: wax ester, W-(OH): wax hydroxymonoester, K: ketone.

Sample-room	Chemical markers (GC–MS + HPLC–APCI-MS)	Content	Typology
5-J	S_8	Sulphur	Cup
7-J	S_8	Sulphur	Storing vessel (A?)
8-J	S_8	Sulphur	Storing vessel (A)
10-J	S_8	Sulphur	US
13-J	S_8	Sulphur	US
14-J	C16:0, C18:0, C24(OH), C26(OH), C28(OH), C30(OH), C32(OH), C34(OH), nC27, nC29, nC31, nC33, W40, W42, W44, W46, W48, W50, W52, W40(OH), W42(OH), W44(OH), W46(OH), PS, SS, PMP, PPdP, PPP, PPO, POP, PMaP, PSP, SPO, SPMa, SPS, SSO, SMaS, SSS, P:S ratio *sn*-2: 55/45	Beeswax and ruminant fat	US
16-J	S_8	Sulphur	Storing vessel (A)
23-U	C16:0, C18:1, i-C18:1, C18:0, PP, PS, W44, W46, W48, W50, PP, PS, SS, LLL, LPL, LOL, PLO, OLO, PPP, OOP, OOO, PSP, OSO, SPS, SSS, P:S ratio *sn*-2: 60/40	Ruminant and vegetable fat	US
24-U	S_8	Sulphur	US
26-U	S_8	Sulphur	Dish/bowl
27-U	S_8, C15:0, C16:1, C16:0, C17:0, C18:1, C18:0, C24(OH), C26(OH), C28(OH), C30(OH), nC27, nC29, nC31, nC33	Sulphur. Mixture vegetable oil + animal fat?	US
28-U	K31, K33, K35, K33-unsaturated, K35-unsaturated, PoPPo, PoPO, PPP, OPO, OOO, PSP, SPO, SPMa, SPS, SSS, P:S ratio *sn*-2: 55/45	Ruminant fat	US
30-U	S_8, C24(OH), C26(OH), C28(OH), C30(OH), C32(OH), C34(OH), nC27, nC29, nC31, W40, W42, W44, W46, W48, W50, W52, W40(OH), W42(OH), W44(OH), W46(OH)	Sulphur and beeswax	US
35-U	S_8, K29, K31, K33, K35, K33-unsaturated, K35-unsaturated, MMP, PMP, PPP, POP + PPO, OOO, PSP, POS, SPS, SSS, P:S ratio *sn*-2: 60/40	Sulphur and ruminant fat	Dish/bowl
36-U	S_8, C16:0, C18:0, PP, PS, SS, PPP, PPO, POP, OPO, OOP, PSP, POS, SPS, SOS, SSS, P:S ratio *sn*-2: 68/32	Sulphur, ruminant and non-ruminant fat mixture?	US
38-U	S_8, W40, W42, W44, W46, W48 (very low concentration)	Sulphur and beeswax?	Small pot
39-U	S_8	Sulphur	Dish/Bowl
40-U	S_8	Sulphur	Dish/Bowl
42-U	S_8	Sulphur	Storing vessel (A?)
43-U	S_8	Sulphur	Storing vessel (A?)
46-U	S_8	Sulphur	US
47-U	S_8	Sulphur	Closed cup
48-U	S_8	Sulphur	Closed cup
50-U	S_8	Sulphur	Storing vessel (A)
51-U	S_8	Sulphur	US
52-U	S_8	Sulphur	US
54-U	S_8, C16:1, C16:0, C17:0, C18:1, i-C18:1, C18:0, PP, PS, SS, W44, W46, W48, W50	Sulphur, ruminant fat, beeswax?	Patera
55-U	S_8, C15:0, C16:1, C16:0, C17:0, C18:1, i-C18:1, C18:0, K31, K33, K35, K33-unsaturated, K35-unsaturated	Sulphur, animal fat (ruminant?)	Dish/bowl
56-U	S_8, C16:0, C18:1, i-C18:1, C18:0	Sulphur, animal fat (ruminant?)	US
57-U	S_8	Sulphur	US
59-U	S_8, C16:0, C18:1, C18:0	Sulphur, animal fat?	Storing vessel (A?)
60-U	S_8	Sulphur	US
76-Z	S_8	Sulphur	US
77-Z	S_8	Sulphur	US
79-D3	S_8	Sulphur	US
109-R	S_8	Sulphur	Bowl
125-F	S_8	Sulphur	US
132-F	C14:0, C15:0, C16:0, C17:0, C18:1, C18:0, PP, PS, SS, PMP, PPP, PPO, PSP, SPO, SPS, SSS, P:S ratio *sn-2*: 62/38	Ruminant fat	Bowl
134-F	C16:0, C18:1, C18:0, PP, PS, PO, SS, MPP, PPP, PPO, PPS, OPO, POS, OOO, OOS, LLL, LPL, LOL, PLP, PLO, POP, OOP, OOO, PSO + POS, SSO	Vegetable and animal fat	Storing vessel (A?)
135-F	C14:0, C15:0, C16:1, C16:0 C17:0, C18:1, C18:0 LL, LPL + LOL, PLP, POL, PPP + PPO + POP + OPO + OOP + OOO, PSP, SPO, SPS, SSS	Vegetable and animal fat	US
136-F	C14:0, C16:0, C18:1 i-C18:1, C18:0	Animal fat	Storing vessel (A)

It is worth remarking that the area of the *oppidum* of Puente Tablas does not lie on any sulphur outcrop, whether volcanic or sedimentary, or on any sulphur water spring. The nearest sulphur mine is approximately 150 km away, in the provinces of Murcia, Albacete and Granada.

The second most frequent type of contents is *animal fat*. In some cases, HPLC–APCI-MS analysis allowed to specify it is ruminant animal fat. The interpretation of the results is based on the following.

The occurrence of animal fat is evidenced, first, by the high content in stearic acid (C18:0) compared with oleic acid (C18:1), by the occurrence of volatile fatty acids (lauric C12:0 and myristic C14:0), and by the prevailing occurrence of saturated acids TAGs in the *sn*-2 position (pentadecanoic C15:0, palmitic C16:0, margaric C17:0, stearic C18:0). Secondly, the identification of ruminant animal and non-ruminant animal fat is based on the following:

- Mixture of isomers of oleic acid (double bond at 9, 11, 13, 15 or 16 positions) characteristic of ruminant animals.
- Occurrence of odd number of carbon atoms fatty acids, associated with ruminant animals: pentadecanoic acid (C15:0), margaric acid (C17:0), nonadecanoic acid (C19:0) and seldom heneicosanoic acid (C21:0) (Colombini and Modugno, 2009; Evershed et al., 2002; Regert, 2011).
- Within saturated TAGs, triacylglycerol MaSS (Ma: margaric; S: stearic) is related exclusively with ruminant animal (bovine and ovine), while APS (A: arachidic; P: palmitic; S: stearic) only occurs in pig fat (Craig et al., 2009; Saliu et al., 2011).

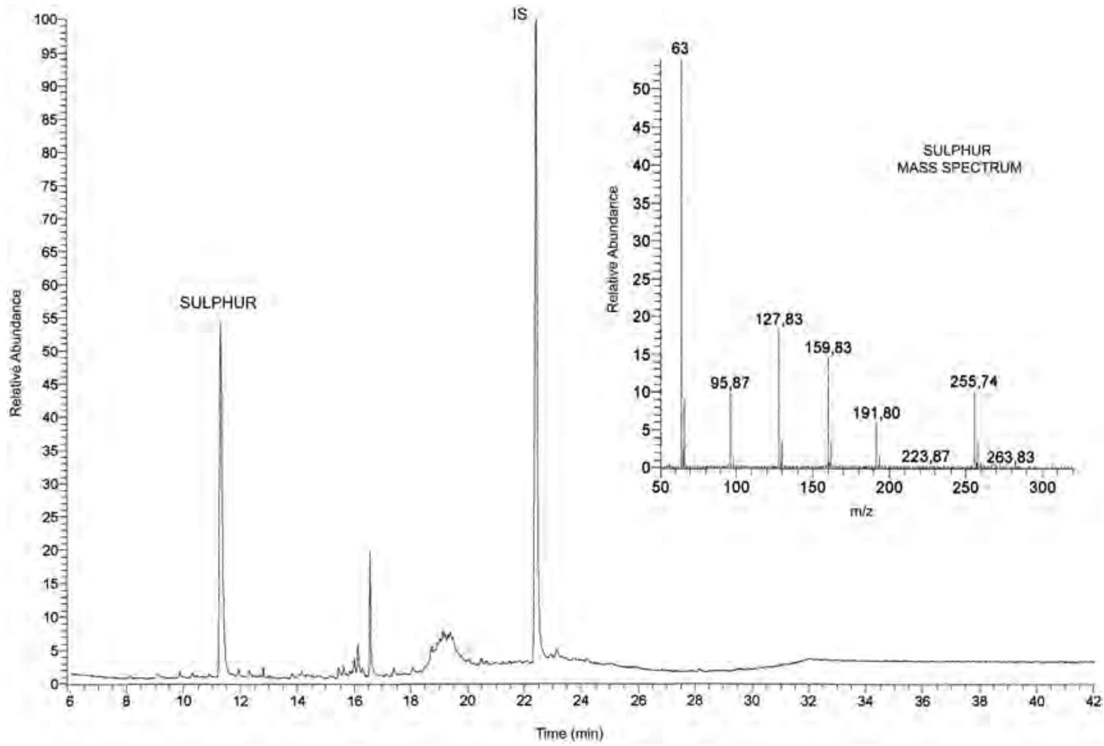

Fig. 5. GC–MS of the vessel 8-J: Sulphur.

- P:S index in the *sn*-2 position 60:40 for ruminant animal fat and 95:5 for non-ruminant animal fat (Craig et al., 2009; Evershed et al., 2002; Regert, 2011; Saliu et al., 2011).

According to the above, animal fat has been identified in 15 vessels. In three cases animal fat is the only content (28-U, 132-F, 136-F) and in the rest of vessels it is mixed with vegetable fat, sulphur and/or beeswax (Fig. 6). While some samples were highly degraded and most identification criteria could not be applied, other samples retained clear lipidic profiles. Ruminant animal fat has been clearly identified in at least 5 vessels (samples 14-J, 23-U, 28-U, 35-U, 132-F) based on the P:S index in the *sn-2* position around 60:40 (Fig. 7). The P:S index in sample 36-U is 68:32, which suggests the possibility of a mixture of ruminant animal and non-ruminant animal fat.

Ketones eluting at a retention time of 20 to 24 min have also been recorded in samples 28-U, 35-U and 55-U. This includes 29 to 35 carbon atoms ketones: hentriacontan-16-one (K31), tritiacontan-16-one (K33) and pentatriacontan-18-one (K35). Also, 33 and 35 carbon atoms monounsaturated ketones eluting immediately before their saturated counterparts were recorded (Fig. 8). These species normally appear in lipidic extracts as a result of fat degradation after intense and/or sustained heat (Evershed et al., 2002). Lack of heat signs or evidence in the vessels under study suggests that the fat was first altered during preparation of a substance or food in a different vessel, and then poured into the vessel under study.

Vegetable fat has been clearly identified, mixed with animal fat, in vessels 23-U, 134-F and 135-F, mainly based on the occurrence of triacylglycerols LLL, LPL, LOL, OLO, PLP, PLO, POL (L: linoleic; O: oleic; P: palmitic) that are clearly identified by HPLC-APCI-MS (Fig. 9). The sample of vessel 27-U does not show any of the above triacylglycerols: it shows a very high content of oleic acid and, as a result, it is difficult to classify.

The fourth main content is *beeswax*. Although there is some evidence of its occurrence in four vessels, it has been identified clearly in two of them: vessel 14-J, with ruminant animal fat, and 30-U, with sulphur (Figs. 10 and 11). The most characteristic chemical markers of beeswax identified in the samples are (Charters et al., 1995; Regert et al., 2001; Tulloch, 1970):

- Series of C23 to C33 n-alkanes displaying a unimodal distribution with a strong odd-over-even predominance;
- Series of W40–W54 carbon number palmitic acid wax esters with a main constituent that contains 46 carbon atoms.
- Long-chain alcohols, resulting from hydrolysis of wax esters, with an even carbon number usually from C24(OH) to C34(OH), maximizing at C30.

The alkanes and long-chain alcohols were missing from samples 38-U and 54-U. Only low-intensity peaks were identified, which represent palmitic acid wax esters.

5. Discussion

Individual identification of the contents of the vessels is followed by the interpretation of their role in the context of the sanctuary. This approach is based on the individual interpretation of each of the contents identified, whether alone or in combination with other contents. The latter is limited by degradation processes (mainly oxidation and hydrolysis). These processes may have affected chemical markers to various degrees. As a result, the combination of contents identified advises a conservative interpretation, because such combinations may not be entirely representative of the original vessel contents. Thus, a number of ritual activities and procedures that may be associated with the chemical markers recorded can be described.

The highest occurrence of vessels with chemical markers of contents is in three rooms: U, J and F. Room U (25 vessels), which is associated with Room Z, is also associated with caves and water rituals. Room J (7 vessels) can be interpreted with regard to two ritual deposits of lambs in small pits. Rooms U and J make 32 out of the 41 vessels that yield positive results. Room F (5 vessels) is equally relevant, because

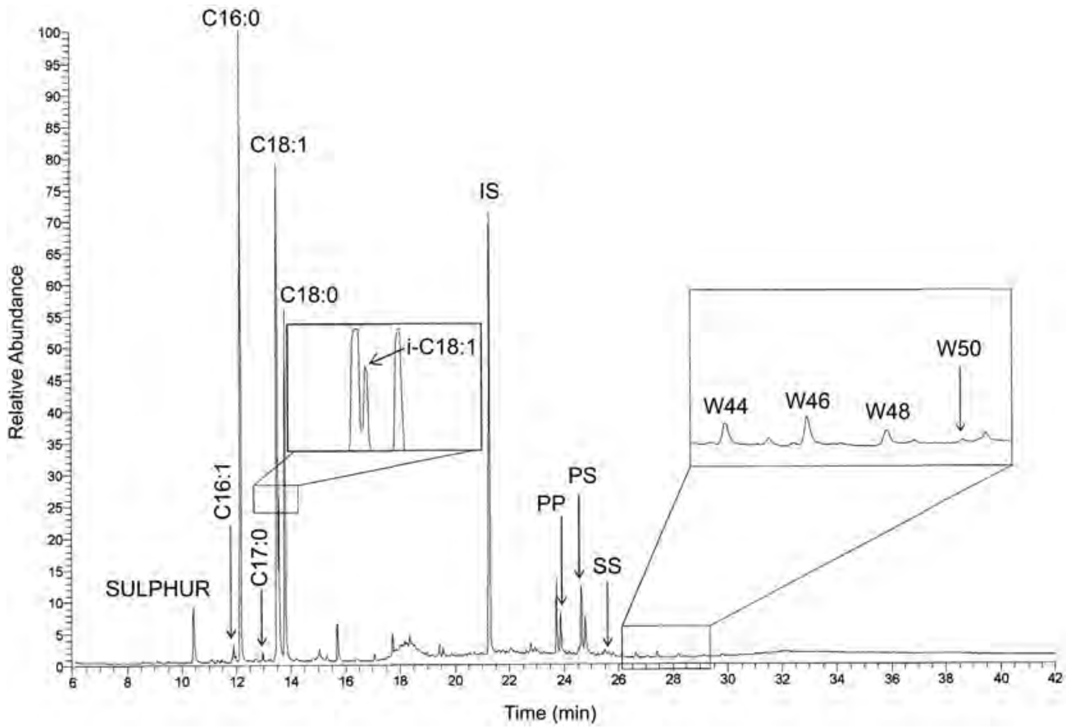

Fig. 6. GC–MS of the vessel 54-U: Sulphur, ruminant fat and beeswax?

that is the *cella* where the stele of the female deity is during the most part of the year and, therefore, must have been the setting of regular offerings. The four remaining vessels are associated with rooms as follows: 2 vessels in Room Z, 1 in Room D3, and 1 in Room R (Table 3).

5.1. Sulphur

Native sulphur appears as the most significant finding in the context of an Iberian ritual space. Use of sulphur is not foreign to the Mediterranean culture of the Iberian society. Actually, it is clearly relevant and useful in a range of domains ranging from religion and economy to human and animal care (Bassani, 2011). The preclassical and classical Greek and Roman literary sources available describe an interesting range of uses for sulphur (Rodríguez, 2012).

Homer's *The Iliad* and *The Odyssey* (Homero, 2001, 2007) show how sulphur is used for purification of objects and rooms. In *The Iliad*, Achilles purifies a cup with sulphur before using it during a rite of consumption dedicated to Zeus. In the latter, *The Odyssey*, Odysseus instructs his nurse Eurycleia to fumigate the palace with sulphur, so the pollution caused by the suitors' death disappears. A similar process of sacralization of a palace can be found in Euripides' *Helen* (Euripides, 2010), where Theonoe instructs her servants to purify the palace with burning sulphur before she enters, so that she might ‘breathe heaven's pure air’. In Euripides' *Iphigenia in Tauris* (Euripides, 2011), Iphigenia also asks Thoas whether to purify the temple of Artemis, possibly with sulphur. A final example of the properties of sulphur is that it is also apt for human use, as in Ovid's *Metamorphoses* (Ovidio, 2012), where Aeson is purified three times with fire, three times with water and three times with sulphur.

The association between water and sulphur is also attested throughout Greek-Roman Antiquity by mention of springs with sulphur and medical uses. Use of sulphur has often been associated with sanctuaries and healing deities such as Asklepios/Aesculapius or Mefitis (Edlund-Berry, 2006; Bassani, 2011). Sulphur water could also spring out of caves associated with nymphs, and could be used against skin diseases (Håland, 2009; Peréx, 1997).

With the above literary references in mind, the data supplied by the archaeological context and the information obtained from chemical analysis allows interpreting the role played in rites by the vessel contents found.

The relevance of water for sanctuaries and its traditional association with sulphur as a purifying and beneficial element bear witness to an apparently reasonable connection between themselves. In this case, water belongs in a liturgical scenography where libation practices become prominent, especially near the three caves, even if still as complements to sacrifices. Lack of chemical markers of wax and fat in 25 vessels suggests the use of water in those vessels. Both the water and the ceramic vessel would be purified by pouring sulphur inside. Purified water would remain in storing vessels and would be used during religious libation ceremonies, from smaller vases both Greek (the krater and the cup) and local (dishes and bowls).

The water running through the channel would also receive symbolic amounts of sulphur and would thereby purify the areas and the rooms of the sanctuary. The water channel that cuts across the sanctuary collects the water from the upper terrace at a point near cave no. 4, which is where the deity returns, then runs across Room U by the deity's *cella* and leads outside, where the representation of the deity would stand during the equinoxes.

Another purifying use of sulphur would be intended to prepare the space of the sanctuary for the sacred rite by setting fire to a small part of the sanctuary. The sharp smell of sulphur smoke could also mask unfavourable odours and eliminate the impurity of death. The physical and spiritual state of cleanliness would favour the connection (or a channel of communication) with the female deity (Rodríguez, 2012).

5.2. Offerings with animal fat, vegetable fat and/or beeswax with sulphur

Chemical markers of vegetable fat, animal fat and beeswax have been recorded to co-occur with sulphur. The occurrence of sulphur in the vessels where the former substances have been recorded too can be interpreted as caused by the need to purify the offering contained in the vessels. Alternative interpretations are also possible, assuming the composition of the original contents has not become degraded.

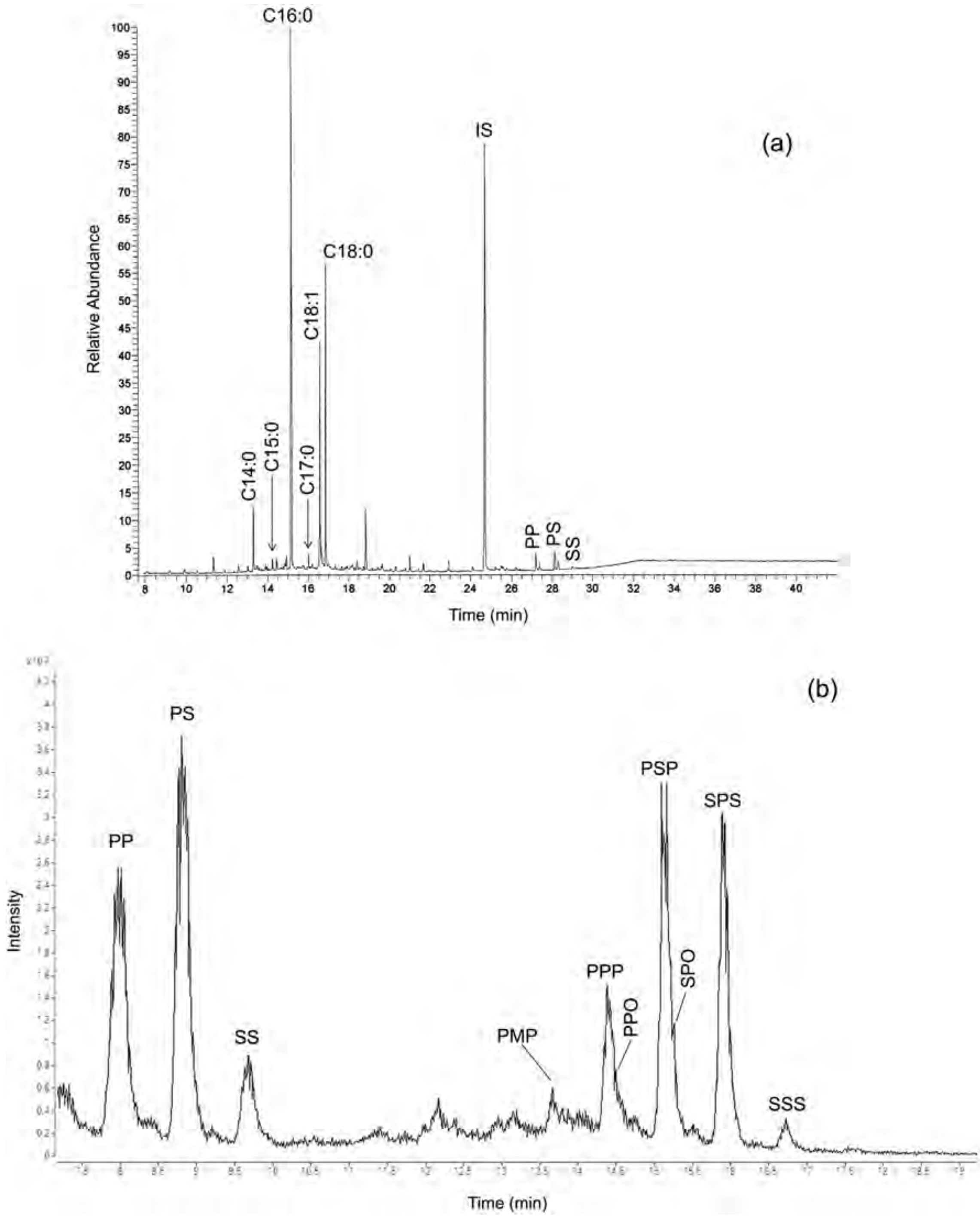

Fig. 7. GC–MS (a) and HPLC–APCI-MS (b) of the vessel 132-F: Ruminant fat.

Use of a number of criteria allowed establishing virtually exclusive occurrence of ruminant animal fat in the offerings. The P:S index in the *sn*-2 position proved particularly useful. In line with this association, the animal record of the sanctuary reveals prevailing occurrence of bovine and ovine bones compared with pig bones (Ruiz and Molinos, 2015). Lack of non-ruminant animal fat, except when mixed with ruminant animal fat, as in sample 36-U, suggests that the Iberians of Puente Tablas did not use pig parts in the vessels, for offerings and/or for consumption. In Puente Tablas, pig offerings were laid on the ground inside a ritual pit next to the east gate of this *oppidum*.

A similar model has been identified in other Iberian contexts, e.g. the Iberian sanctuary of Atalayuelas (Fuerte del Rey, Jaén) or the cemetery of La Noria (Fuente de Piedra, Málaga). Although the archaeological record also proves the occurrence of pig materials, it is virtually exclusively ruminant animal materials that are processed and stored in the vessels, and then offered in the cemeteries and the cult spaces (Sánchez et al., 2011).

Concerning the origins of the vegetable fat, the antracological and carpological analysis of the sanctuary, the palace and the dwellings of the *oppidum* must be taken into consideration too. These analyses evidence that the most frequent vegetable species that can be the source of vegetable fat are wild olives and almonds. Acorns and linseeds have been recorded to a lesser degree (Montes, 2015; Rodríguez and Pradas, 2015).

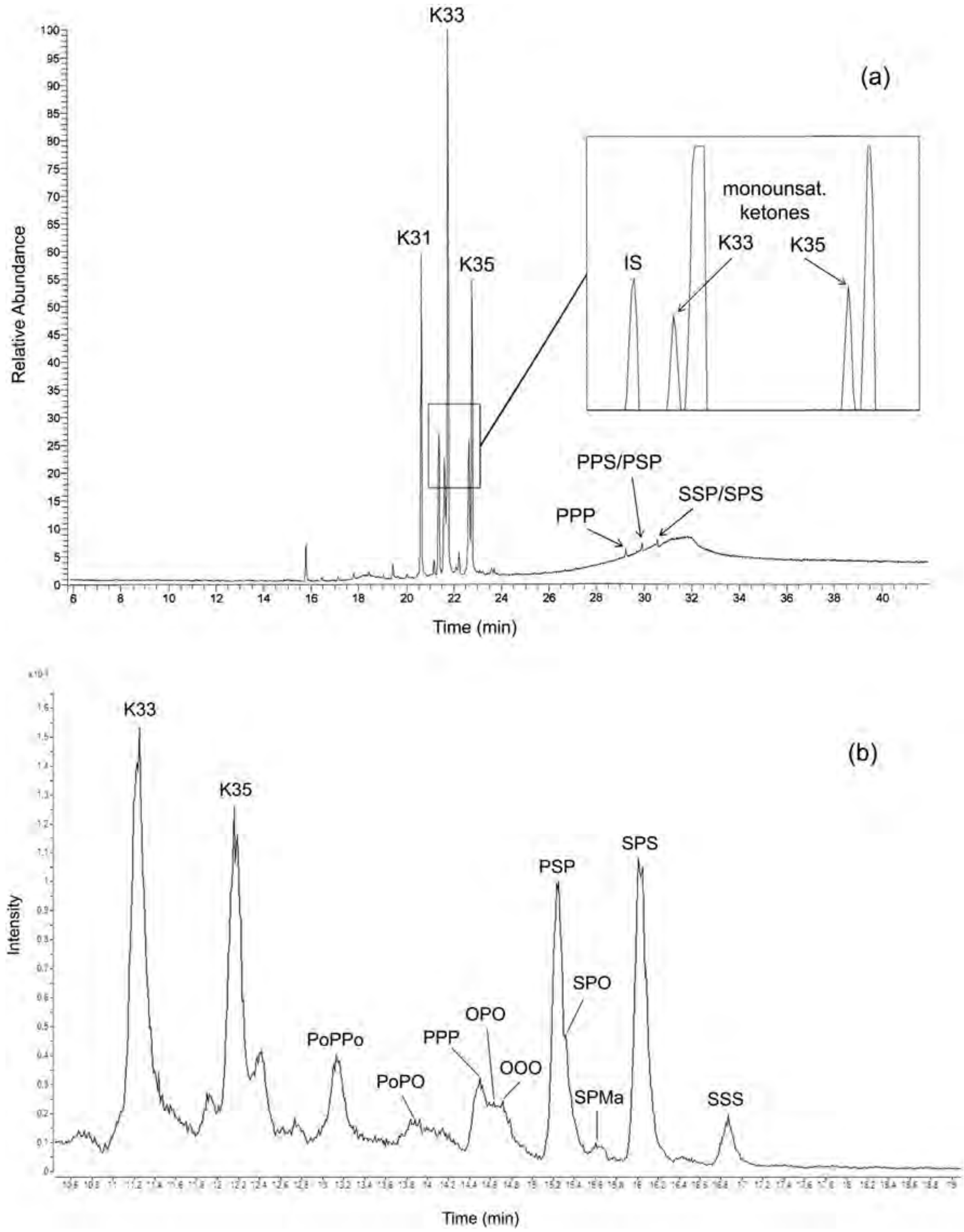

Fig. 8. GC–MS (a) and HPLC–APCI-MS (b) of the vessel 28-U: Ruminant fat.

While oil extraction from olive pulp is possible by use of unsophisticated artisan crushing methods, dried fruits demand use of a rotating grinding mill. Such a mill produces an oily pulp from which small amounts of oil can be obtained by hand pressure. Present-day artisan extraction of oil from the dried fruit known as *argan* is a clear example of this process (Charrouf and Guillaume, 2008; López and Alba, 2009). The occurrence of rotating grinding mills in Puente Tablas is scarce: only one has been recorded, specifically near the palace. This may mean that olive or almond oil production was at small-scale and intended as medicines or for rites rather than for everyday consumption. Oil extraction from seeds is more difficult and unlikely, because it requires high pressure and chemical processes that were not within the reach of protohistoric societies.

Another type of contents associated with sulphur is beeswax. Sample 30-U is the clearest case of this substance, even if it unfortunately belongs to a vessel of undefined shape. Use of beeswax in Prehistory and Protohistory has been interpreted variously: for medical use, as a cosmetic, for rites, as a protection against corrosion, for paints, for boat building, as a glue, as an insulating substance, for consumption of mead and other alcoholic drinks, and as fuel for lighting (Evershed et al., 1997; McGovern et al., 2013; Regert et al., 2001). The range of interpretations makes the spatial and temporal contexts become essential

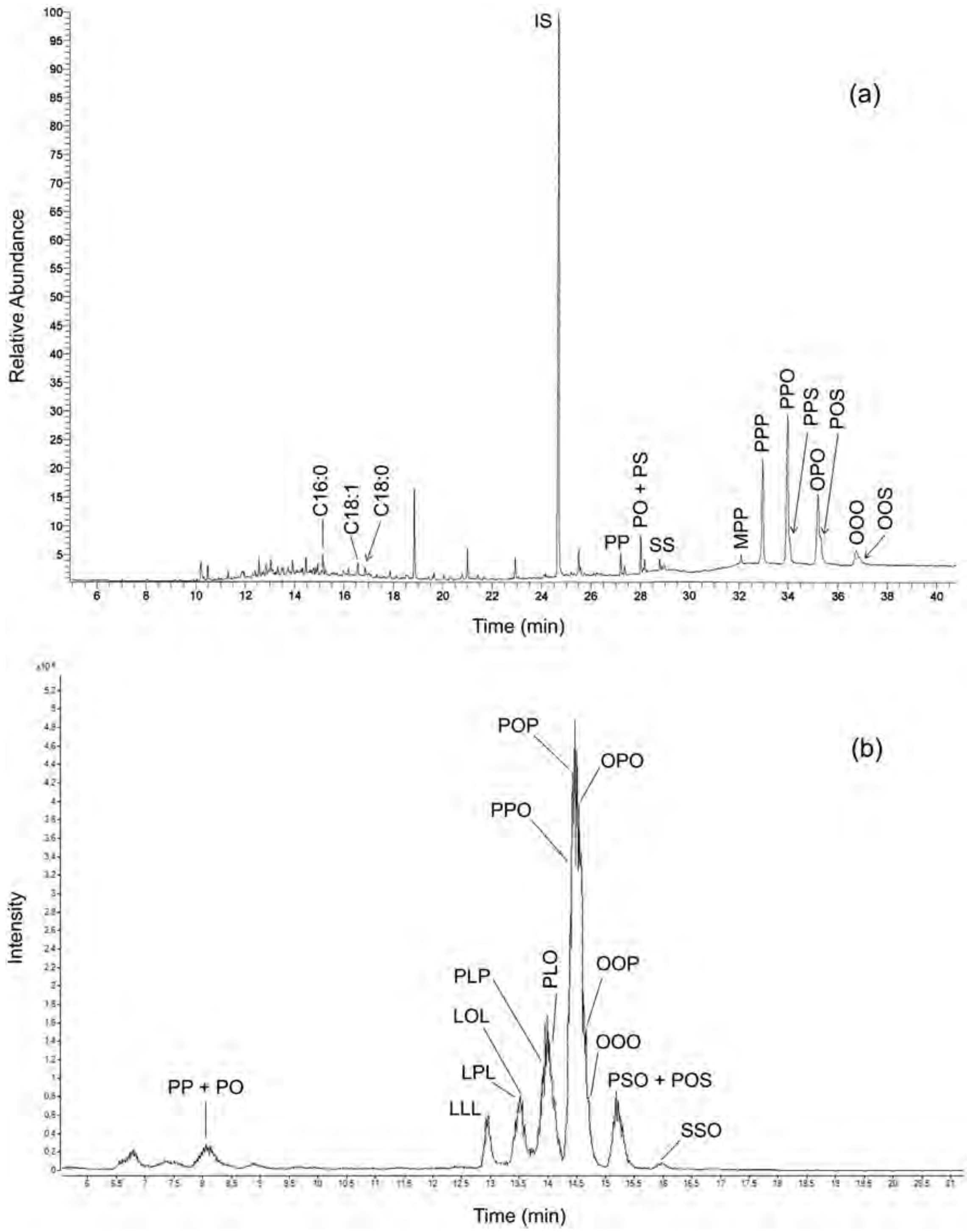

Fig. 9. GC–MS (a) and HPLC–APCI-MS (b) of the vessel 134-F: Vegetable and animal fat.

for an interpretation related with beeswax. In the case of the vessels of the sanctuary, the interpretation should also take into consideration the sacred context and the co-occurrence with sulphur and animal fat. Beeswax may have been used as a covering to seal the inside of vessels, or may be associated with the occurrence of honey and/or mead. Being offerings, these substances may also have been in contact with sulphur.

The combination of sulphur with beeswax and vegetable and/or animal fat may be related to healing mixtures intended as treatments for humans or animals. These mixtures or balms were offered to the deity for increased healing power. This may explain why they have been found in the vessels of Room U. The healing power of sulphur is attested in the writings by Pliny the Elder. Pliny laid emphasis on the increased power of sulphur when mixed with fat as plasters and balms. He also remarked it was effective against abscesses, facial herpes, asthma and kidney or lower back pain (Plinio el Viejo, 2002).

The treatment of animals with sulphur for various purposes is also mentioned frequently by ancient roman writers, e.g. Ovid and Virgil in *Fasti* and *Georgics*, respectively. Epizootic diseases were a constant

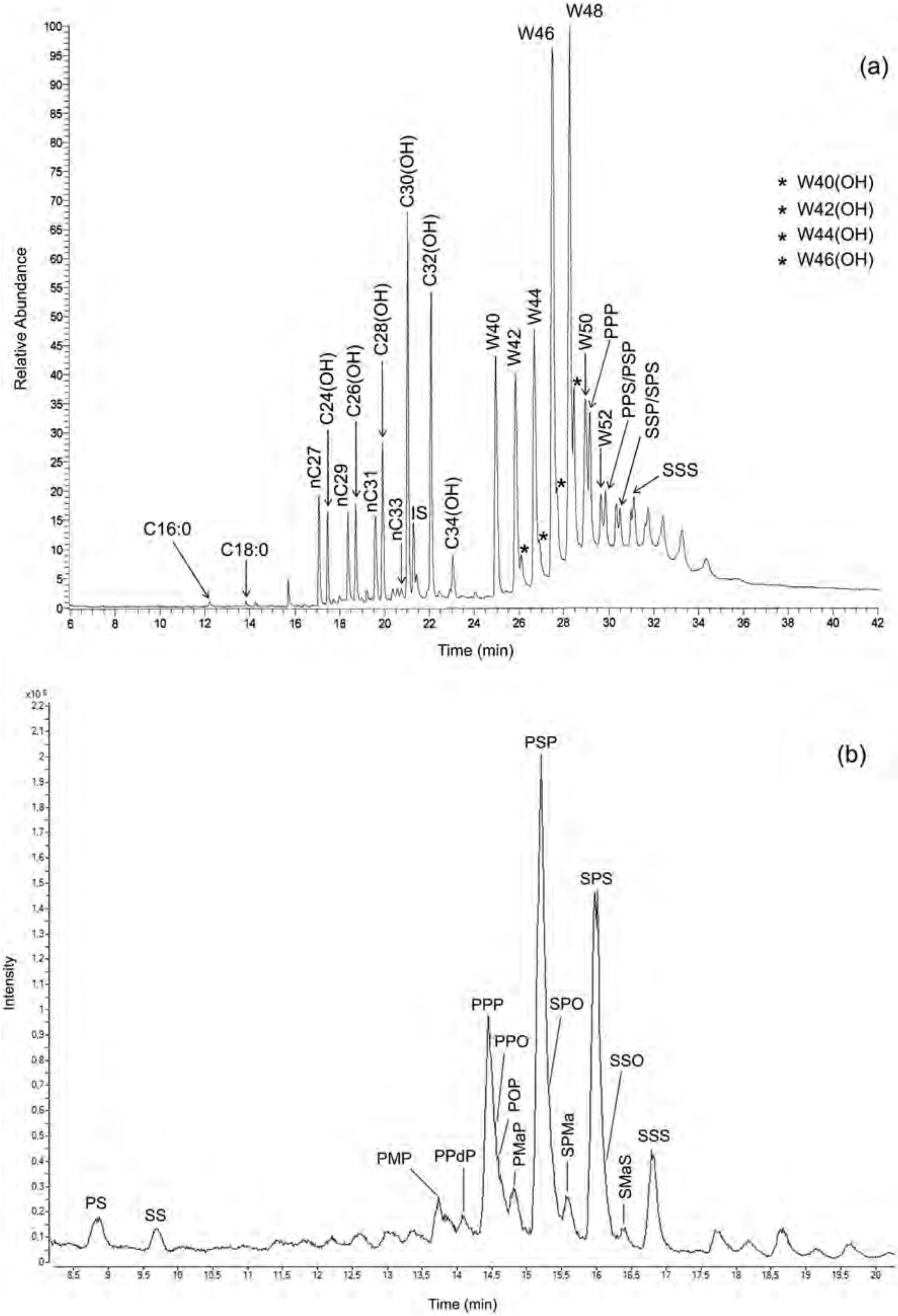

Fig. 10. GC–MS (a) and HPLC–APCI-MS (b) of the vessel 14-J: Beeswax and ruminant fat.

threat to sheep farmers. Native sulphur was used together with pitch, wax and oil against psoroptic mange, that is, 'sheep-scab', and foot rot and other skin diseases. The paste was smeared on the infected parts. Sulphur was also used to clean stables by smoking, as described in the rituals dedicated to the goddess Pales (Santillo Frizell, 2004).

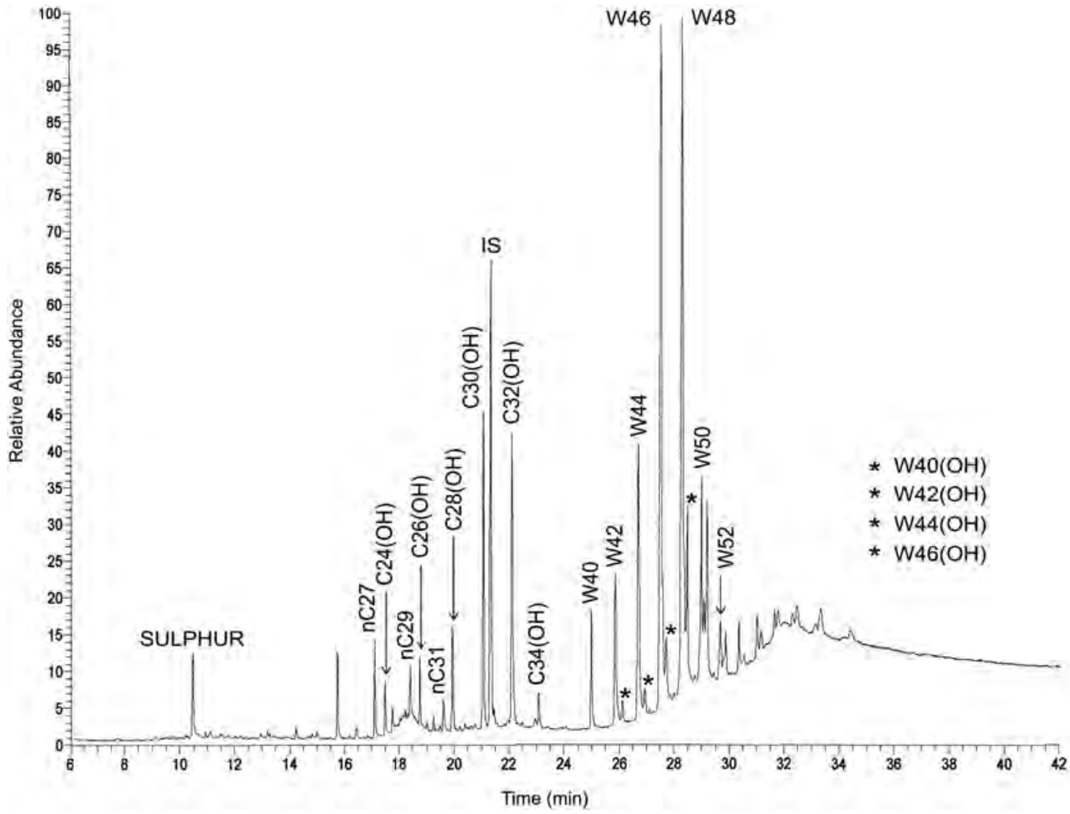

Fig. 11. GC–MS of the vessel 30-U: Sulphur and beeswax.

5.3. Offerings with fats and waxes

As to the vessels where vegetable fat, animal fat and beeswax have been identified, the following remarks must be made. Firstly, the lack of sulphur may be due to postdepositional factors. Secondly, this is a small set of vessels (7 units), most of which belong to Room F, the deity's *cella*. This room hosted 4 vessels. Animal fat was found in vessel 136-F, ruminant animal fat in vessel 132-F, and a mixture of vegetable fat and animal fat in vessels 134-F and 135-F. Only one vessel revealed remains of sulphur, but this vessel did not display any other chemical marker. Two of the three remaining vessels were laid in Room U and another in Room J. As noted for vessel 35-U, the contents found in Room U had been heated, which degraded the ruminant animal fat still present.

Finally, the combination of beeswax with ruminant animal fat may be interpreted as remains of fuel for burning, if it is to be taken as undegraded contents of other markers (undetermined shape vessel 14-J). This vessel would thus be used as a lamp (Evershed et al., 1997).

Table 3
Combinations of contents and their distribution in the sanctuary according to specific rooms. SV: storing vessel, D/B: dish/bowl, C: cup, B: bowl, PO: pot, PA: patera, US: undetermined shape.

Content	Total no.	Room: no. of vessels	Typology
Sulphur	25	U:14	3 SV, 3 D/B, 2 C, 6 US
		J: 6	3 SV, 1 C, 2 US
		Z: 2	2 US
		D3:1	1 US
		R:1	1 B
		F: 1	1 US
Animal fat	3	U: 1	1 US
		F:2	1 SV, 1 B
Sulphur + animal fat	5	U: 5	1 SV, 2 D/B, 2 US
Sulphur + beeswax	2	U: 2	1 PO, 1 US
Sulphur + animal fat + vegetable fat	1	U: 1	1 US
Sulphur + animal fat + beeswax	1	U: 2	1 PA
Animal fat + vegetable fat	3	U:1	1SV, 2 US
		F:2	
Animal fat + beeswax	1	J: 1	US

6. Conclusions

The results obtained are the first rigorous approach to a chemical marker-based analysis of Iberian rites. An accurate interpretation of chemical markers is obtained from the study of a considerable number of vessels, from their spatial distribution, from the data supplied by the written sources, and from the antracological and carpological analysis.

Methodologically, joint use of GC–MS and HPLC–APCI-MS appears as an adequate research procedure. HPLC–APCI-MS of archaeological fat residue supplies relevant information that could not be attained with current GC–MS techniques. This information is especially relevant as regards the most useful TAGs for research on archaeological contents.

The identification of sulphur along with vegetable fat, ruminant animal fat, and beeswax is a novelty that brings to light the range of chemicals used for rites and for offerings. Also, it has become possible to identify which rooms were more closely associated with the rites of the female deity.

Purifying and healing substances like sulphur, ruminant animal food offerings, probably bovine and/or ovine, vegetable fat from olives or almonds, again both as food and as medicines, and beeswax as a sealing substance, as a healing substance, as fuel or associated with food or alcoholic drinks are only some of the interpretations that bring our eyes back to a hypothesis that has been defended in the past for the Iberian society: the wide range of rites held around the year and in the form of sacrifices, offerings and libations. The rites of passage, which were so important in the Iberian religion or world of beliefs (Rueda, 2013), would explain the cult of this sanctuary in the main. Other practices would take place alongside the former, probably at different points of the year, as fertility or healing rites.

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