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Case study

Analysis of medieval organic remains: Incense in SE Spain

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ABSTRACT

The medieval organic materials recovered from Visigothic strata (8th century CE) at El Tolmo de Minateda (Hellín, Albacete, Spain) and from 11th century CE strata at La Graja (Higueruela, Albacete, Spain) were initially identified as incense remains used in religious ceremonies. Utilizing SEM, EDS, FT-IR, and GC–MS techniques, we have refined our understanding of these samples. The El Tolmo materials contain aromatic compounds from various botanical sources, especially conifer resins, indicating a local origin. In contrast, the La Graja samples, dating from the Muslim period, show a predominance of benzaldehyde and benzyl alcohol, differing significantly from the El Tolmo findings. The El Tolmo remains, which still emit an incense and smoke aroma, represent a local formulation of ritual incense using nearby botanical species. Conversely, the La Graja remains appear to have undergone intense pyrolysis. The presence of iron on the surface of these samples suggests they may have been in contact with iron, either in containers made of this material or in wooden-handled iron tools as plant-based adhesives.

1. Introduction

The discovery of a compact, yellowish or brown-blackish mass during the excavation of a Visigothic church at Tolmo de Minateda (Hellín, Albacete, Spain), which easily crumbled into powder and emitted an incense-like aroma, led us to hypothesize that these were remnants of incense used in religious rituals in the early 8th century CE. Similarly, the finding of numerous pellets, each less than 1 cm in length and not forming a compact mass, by another team during the excavation of the Muslim village of La Graja (Higueruela, Albacete, Spain), and the proximity of a small mosque, prompted us to consider connections. This persuaded us to undertake both studies in parallel.

The use of materials rich in aromatic compounds that are volatilized by the heat provided by coal embers characterizes incense. Some common notes in incense fragrances include frankincense, myrrh, balsamic and smoky, but also those of spices such as cinnamon, clove and pepper. Incense is usually made by

Abbreviations: AMU, atomic mass unit; FT-IR, Fourier-transform infrared spectroscopy; GC–MS, Gas chromatography-mass spectrometry.

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combining various natural products of different origins, so their formulations can be infinite. Although the classic botanical sources such as frankincense, myrrh, benzoin and storax are characteristic resins, leaves and stems of remarkably diverse botanical origin also enter the composition of incense.

The earliest evidence of incense use are texts and images that illustrate its use, or containers with remains of chemically identifiable resins. Incense use was well-documented in ancient Egypt (2686–2181 BCE), in ancient Mesopotamia (2050 BCE), the Indus Valley Civilization (2600–1900 BCE) and ancient China (1600–1046 BCE), being used in ancestral worship and religious rituals [1–4].

Chemical analyses of residues on ancient pottery vessels from sites such as Umm an-Nar in Oman and Tayma in Saudi Arabia have revealed traces of frankincense (*Boswellia* spp.) and myrrh (*Commiphora* spp.), suggesting their use as incense or medicine [5,6]. Residue analysis on pottery from sites in East Africa, such as Unguja Ukuu in Tanzania (7–8th cent. CE), has detected the presence of copal residues (Burseraceae), suggesting its use as incense [7,8].

GC–MS analysis of residues from incense burners found in medieval churches in Belgium, revealed the presence of aromatic compounds consistent with the use of incense in religious practices [9].

2. Research aim

Organic materials recently recovered from two medieval sites in the province of Albacete, southeastern Spain—El Tolmo (8th century CE) and La Graja (11th century CE)—were initially believed to be incense.

When crushed, El Tolmo samples give off a perfume reminiscent of “incense”. On the contrary, the sample from La Graja does not give off a particular odor, but it also appeared in an area close to a religious context (mosque) within a settlement dedicated to cattle raising. Therefore, our starting hypotheses for this last sample were incense or a veterinary material.

We intend to approach the identification of the botanical origins of these samples based on their physicochemical characteristics, considering that the time elapsed, more than a millennium in both cases, and the conditions of deposition and conservation may have modified some of them. In the case of El Tolmo, the materials were recovered from strata buried atop a hill inhabited by humans since the Bronze Age. This hill, composed of calcareous materials, rises 50 m above the surrounding floodplain, significantly reducing the impact of moisture. Additionally, the presence of some gypsum may have contributed to the preservation of the materials.

In La Graja, the studied habitat is situated within a “dehesa,” an open evergreen oak forest that occupied the remnants of the medieval settlement. This site does not appear to have suffered a fire that caused its abandonment, so the evidence of pyrolysis is due to the intentional use of fuels. The stratum depth is shallow, exposing the remains to changes in temperature and humidity throughout the annual cycles since their deposition.

3. Material and methods

3.1. Analytical techniques employed

The methodologies used are Scanning Electron Microscopy with Energy-dispersive X-ray spectroscopy, Fourier Transform infrared spectroscopy (FT-IR), and gas chromatography coupled with mass spectrometry (GC–MS) [10,11]. Each technique provides us with relevant but partial evidence and from the integration of the whole we expect a closer vision of the solution for the proposed problems. Details on sample preparation instrumental and procedures are available in Supplementary Material and Methods section. Information on comparison samples is available in Supplementary Table 1.

3.2. The sites

3.2.1. El Tolmo de Minateda

The archaeological site of El Tolmo de Minateda (Hellín, Albacete, Spain), dominates the natural route between the interior plateau and the southeastern coast of the Iberian Peninsula. It was a strategic point throughout its history. Since 1988, the University of Alicante has documented a long sequence of occupation from prehistoric to medieval times. It was an important pre-Roman and Roman settlement. It was occupied again in the second half of the 6th century CE. At this time, the revitalization of El Tolm*o* is magnified by the presence of the episcopal see of *Eio*, archaeologically documented by a church, a baptistery, an *episcopium* and a funerary area around the head and foot of the religious building [12] 17 (Fig. 1). The liturgical and administrative life of the diocese must have maintained its original function until the middle of the 8th century, when the meaning of the buildings changed being reconverted to domestic uses or abandoned.

The stratigraphic phase is framed in the second half of the 8th century CE when a functional transformation of the buildings of the old diocese occurred. At this moment, the smallest spaces are adapted to domestic use through specific modifications (orange clay pavements, elevation of the level of circulation, construction of hearths, changes of hollows, etc.). At the same time, the first symptoms of plundering of architectural material appear. Still, the main structures are standing. The sample analyzed in this work come from this stratigraphic phase and was found in the patio that connects the church with the episcopal palace (Fig. 1).

In the case of the episcope, and the patio where the sample appears, this new floor extends through all the building rooms, except for the basilica, and through some sectors of the square, also showing a new domestic functionality.

The sample studied here comes from one of these levels of clay pavement, so the “*incense*” reached El Tolmo at some time before the second half of the 8th century. Based on the current data, we cannot exclude that they are remains of incense used in religious activities associated with the episcopal see, but also it could be remains of materials used in domestic spaces in the middle of the 8th century without a religious purpose. The material is solid, compact, black and shiny, but it can very easily be crushed in a glass mortar resulting in a chestnut-colored powder strongly aromatic recalling smoke and incense (Fig. 1). Samples analyzed (SEM, FT-IR and GC–MS) include 62319M2 and 62478M3 coming from two distinct parts of the same bulk.

3.2.2. La Graja

At the site of La Graja (Higueruela, Albacete, Spain), three archaeological excavation campaigns have been conducted since 2020 as part of the research project “The Andalusian settlement in the dry lands: the south-eastern area of La Mancha” [13,14]. This is an *alquería* (a small rural settlement of farmers). It is one of the dozens this type that extended throughout eastern La Mancha in the Andalusian period, specialized in rainfed agriculture and livestock production. The *alquería* of La Graja is one of the largest, with 35 houses. Excavations have documented a large house made up of two nuclei around courtyards, with a total area of about 725 m^2, as well as a modest congregational mosque that stood in the center of the settlement and served the inhabitants of the village and other nearby settlements (Fig. 2).

The domestic unit had a silo dug out of the rock. The importance of livestock farming is attested by the presence of numerous

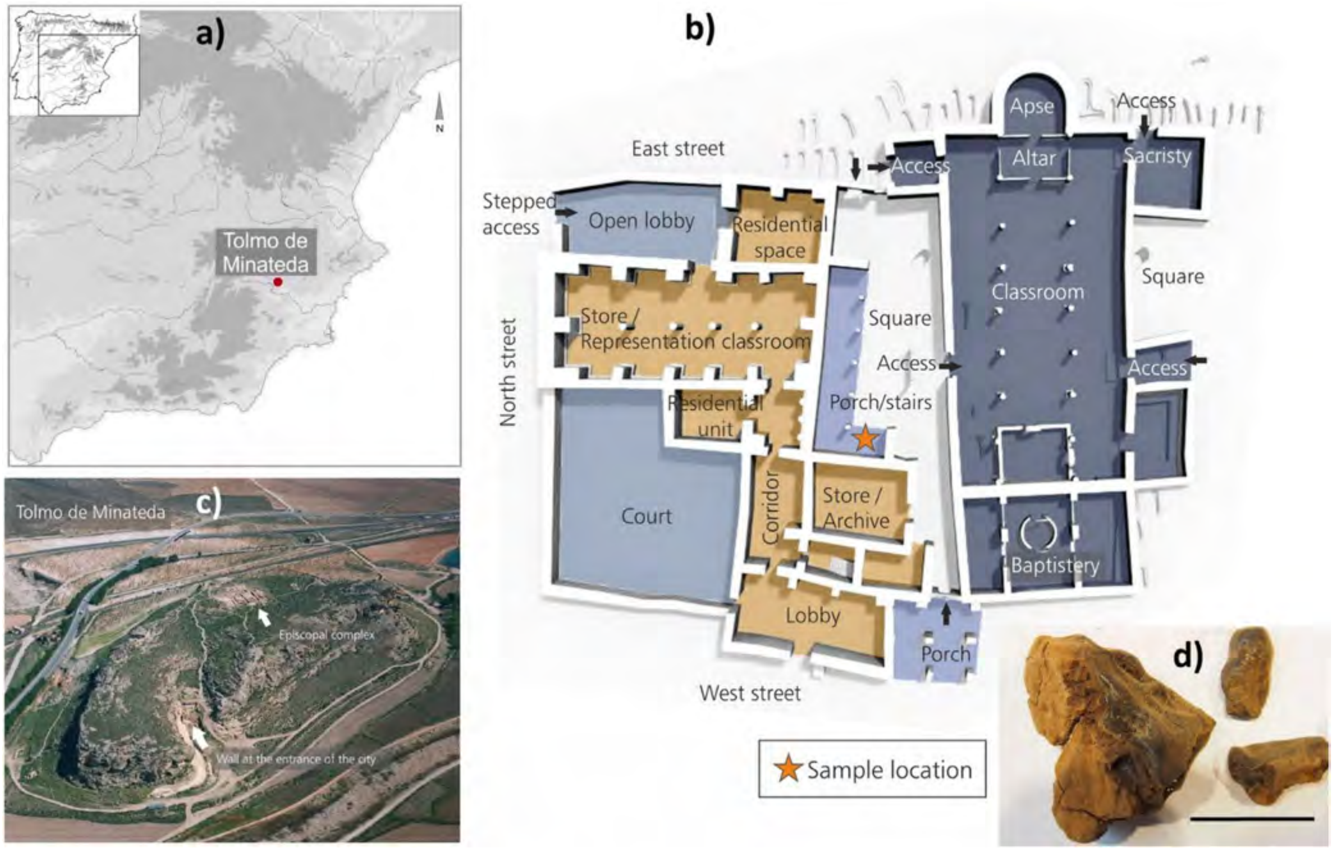

Fig. 1. Location and samples from medieval El Tolmo (Minateda, Hellín, Albacete, Spain); (a) Situation of El Tolmo within eastern Spain. (b) Plan of religious buildings and sample location. (c) Aerial view of the site. (d) Tolmo "incense," scale = 5 cm. Images: Victoria Amorós.

fenced and courtyards linked to the dwellings. In fact, the complete skeleton of a sheep has been found and can be dated back to the 11th century by C14 analysis. In addition to agriculture and livestock farming, the peasants who lived in this village had access to the commercial circuits of the time, where they marketed their surpluses, especially wool. There is evidence that they seem to have opted for a certain specialization, generating surpluses to sell or exchange and thus gain access to manufactured goods that arrived from abroad, such as glazed pottery or metal tools (brass thimbles, knives and iron horseshoes) which could have come from urban workshops such as Chinchilla or Jorquera.

There were recovered some fragile bluish-blackish tears, resin-like (Fig. 3), among wood coals in several samples of the same stratum. Samples analyzed: 08M1S41 (SEM, EDS, FT-IR and GC–MS) and 07 (GC–MS), from two different samples of coals and resins from the same zone.

4. Results

4.1. Field emission scanning electron microscopy images

The Field Emission Scanning Electron Microscopy (FE-SEM) images of El Tolmo samples (Supplementary Fig. 1) reveal a compact vitreous structure, like that of other comparison materials analyzed such as frankincense, myrrh or benzoin.

The FE-SEM images of La Graja samples (Fig. 3) reveal a more complex structure with a compact vitreous structure, often with cavities, and an irregular external surface covered with granules and needles, in which the element iron (Fe) can be distinguished by Energy-dispersive X-ray spectroscopy.

4.2. Fourier transform infrared spectra

The FT-IR spectra are plotted in Fig. 4 and show notable differences between the La Graja sample and the two samples from El Tolmo. The information provided for the materials from both deposits by these spectra will be discussed in the following section.

4.3. GC–MS analyses

The analysis of El Tolmo 62478M3, resulted in 109 peaks, starting at 9.109 min with benzaldehyde and ending at 31.849 min with methyl-abietate. The first identifiable peak of sample 62319M2 was toluene at 4.512 min, ending at 32.636 min with methyl-abietate, totalizing c. 200 peaks.

From La Graja were analyzed two samples: Sample 07, resulted in c. 60 peaks, starting at 6.840 min with p-Xylene and ending at 25.962 min with octadecane. The first identifiable peak of sample 08M1S41 was p-xylene at 6.855 min, also ending at 25.962 min with octadecane and totalizing c. 40 peaks. The latter sample had several siloxane peaks that are artifacts of the analysis process.

What is evident is the significant poverty in volatile elements in the samples from La Graja with respect to those from El Tolmo (Table 1, Fig. 5). This suggests quite different properties and uses of the recovered materials.

5. Discussion

5.1. Incense from El Tolmo

Considering the analytical results, we can venture an interpretation of the nature of the 'incense' sample from El Tolmo by com-

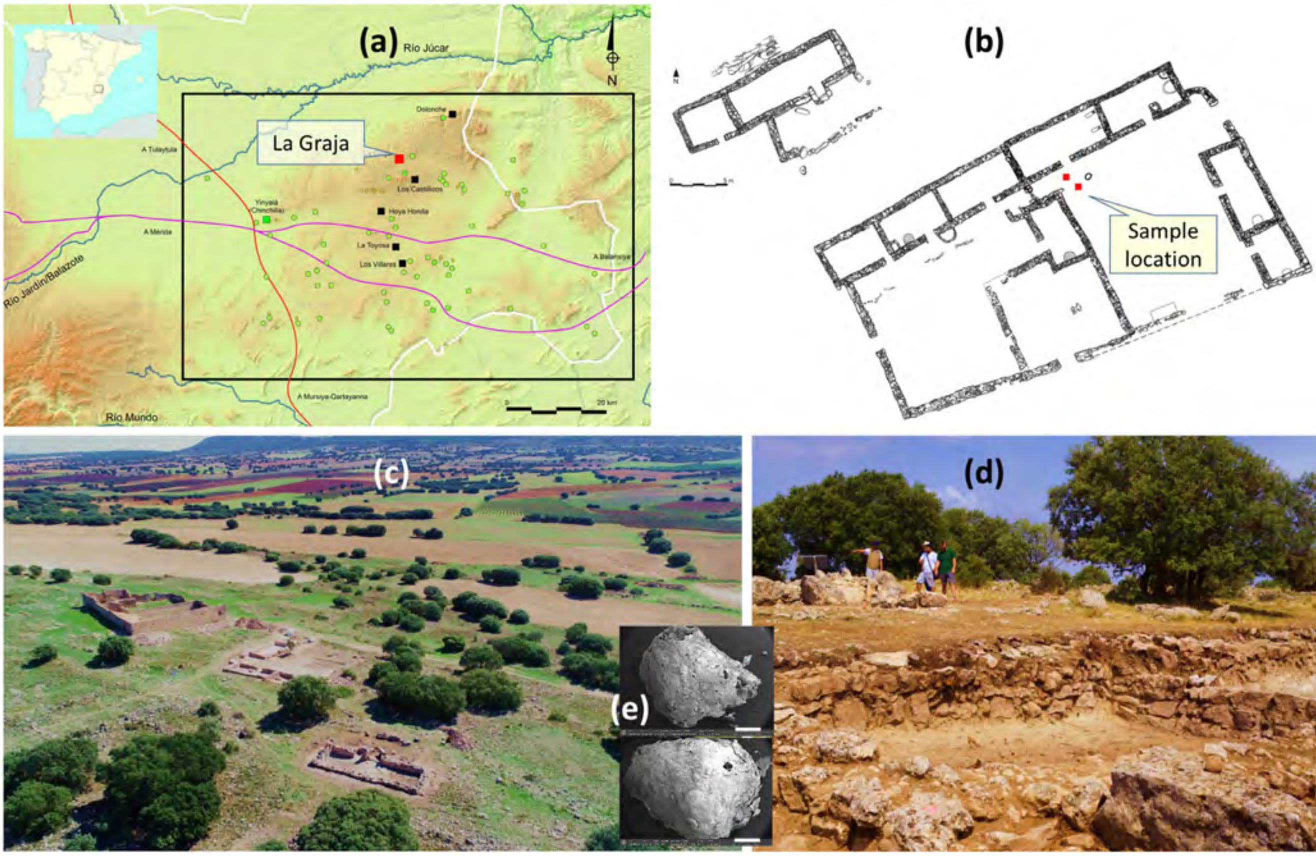

Fig. 2. Location and samples of Medieval Alquería de La Graja (Higueruela, Albacete, Spain): (a) Alquería de La Graja within eastern Spain. (b) Plan of buildings with sample location marked in red. (c) Aerial view of the site. (d) Area where the resin was recovered. (e) Pellets of resin, SEM images, scale 1 mm. Images: a-d by José Maria Moreno Narganes; e by Teresa Coronado and Javier Valera. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

paring it with relevant literature. The sample is a partially burnt mixture of resins and other plant parts.

In the FT-IR spectrum of sample 62319M2, the band at 3368 cm^{-1} corresponds to the O–H stretching mode of carboxylic acids or alcohols. The displacement of its position to lower energies and great width indicates the existence of O–H···O hydrogen bonds. The C=O stretching band at 1700 cm^{-1} agrees with the existence of hydrogen-bonded COOH groups.

In this case, the C–H stretching bands of alkyl groups at 2827, and 2868 cm^{-1} are clearly observed. Bands attributable to aromatic rings are observed at 1599 and 1563 (C–C stretching), and 727–875 cm^{-1} (C–H bending).

The band at 1239 cm^{-1} can be assigned to the C–OH stretching mode and is described in the literature as characteristic of resins [16]. The other bands (1445, 1382, 1034–1170 cm^{-1}) cannot be assigned with confidence, since both C–H or C–O–H bending, and C–O stretching bands usually appear in this region [17].

The FT-IR spectrum in sample 62478M3 is quite like that of sample 62319M2. In both cases we have characteristic bands of carboxylic acids with aliphatic chains and aromatic rings. The main differences between 62319M2 and 62478M3 occur in the zone between 690 and 905 cm^{-1}, where 62478M3 gives a greater number of bands, which could be due to the presence of aromatic rings with different substitution patterns.

Several botanical sources can explain the FT-IR bands detected, such as sandarac, pine resins and tars, mastic, and labdanum, bands which can also be recognized from the Sevillian incense mixture (Supplementary Table 2). Therefore, based on the FT-IR analysis, it is likely that the mass recovered at El Tolmo is a mixture of resins from local sources (pine or juniper resin, beeswax or propolis, mastic from lentisk) or imported from other parts of the Iberian Peninsula (labdanum) or North Africa (sandarac of *Tetraclinis articulata*) (Supplementary Table 2).

The GC–MS data, as detailed in Supplementary Table 3, corroborate the FT-IR analysis results. Based on the GC–MS analysis, the samples contain a complex mixture of sesquiterpenes and diterpenes (see Table 1 and Fig. 5). Potential natural sources for these compounds are listed in Supplementary Table 3. Diterpenic resins are typically derived from plants in the order *Pinales*, including pines, junipers, cypress, and related species [18]. Thus, the presence of diterpenes in our samples suggests that conifers might be among the sources of these materials. Compounds present in concentrations exceeding 2.5 %, such as various diterpenes derived from abietic acid, support the identification of pines and junipers as sources of some fundamental components of the El Tolmo mass. The detection of retene further indicates that resins from pines, such as *Pinus halepensis* or *P. pinaster*, are contributors. Additionally, calamenene could originate from mastic (*Pistacia lentiscus*), associated with Mediterranean incense trade [8], or from tar derived from *Juniperus oxycedrus* L.

Cadalene may derive from labdanum of *Cistus ladanifer* [19], or from resins of Spanish, Phoenician, and prickly junipers (*Juniperus thurifera, J. phoenicea*, or *J. oxycedrus*). The presence of benzaldehyde suggests potential involvement of seeds from a Rosaceae species or might result from the elevated temperatures the mass was exposed to during its deposition and preservation approximately thirteen centuries ago.

However, the detection of polycyclic aromatic hydrocarbons (PAHs) such as phenanthrene and naphthalene implies that the

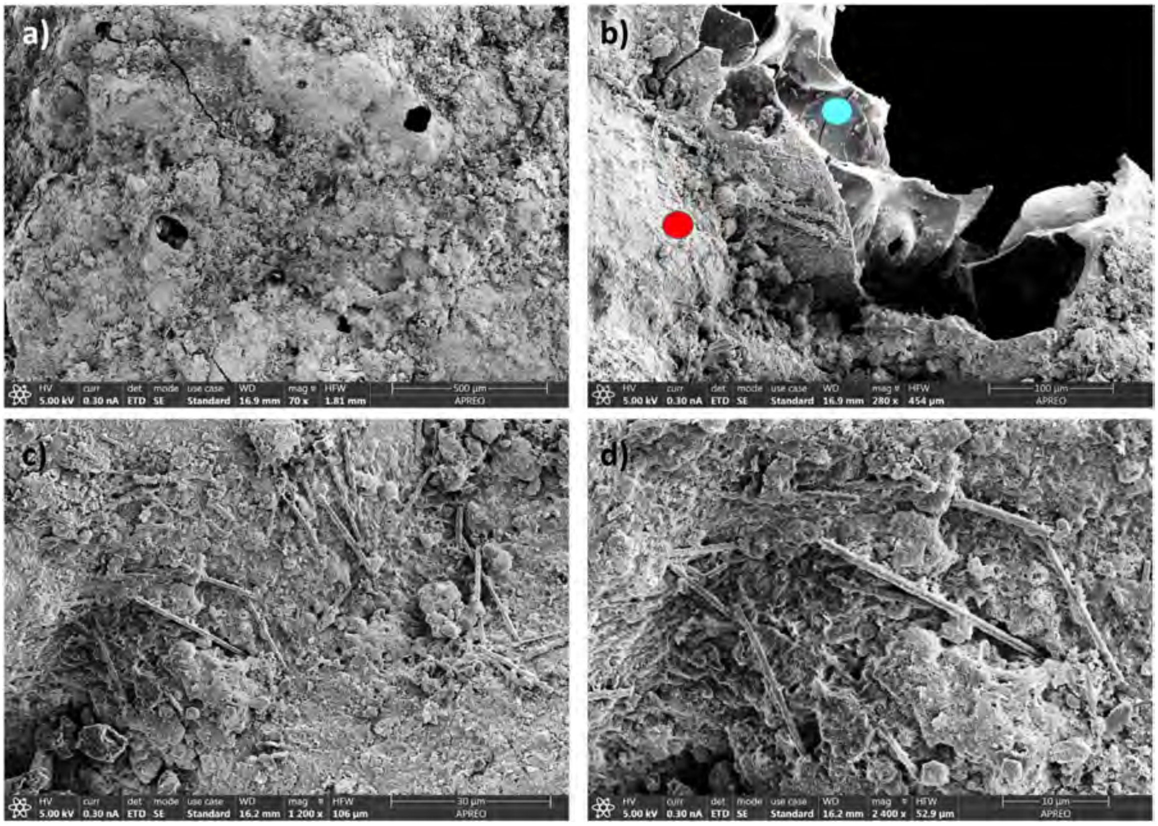

Fig. 3. SEM images of "resin" samples from medieval La Graja (Higueruela, Albacete, Spain); (a) Graja 08M1S41, scale = 500 μm; (b) Graja 08M1S41, scale = 100 μm. Red dot marks the zone where EDX reveals the presence in the surface of iron (Fe), blue dot marks the inner zone where Fe was not found; (c) Graja 08M1S41, scale = 30 μm; (d) Graja 08M1S41, scale = 10 μm. Images: Teresa Coronado Parra. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

sample may also include tar or a partially burnt resin mixture. The presence of anthracene reinforces this possibility, as such PAHs are commonly associated with tar [20]. Additionally, compounds like eugenol, fenchol, guaiacol, and guaiazulene suggest the presence of other plant materials, such as labdanum, rosemary, and sage, within the sample.

A notable characteristic of the incense analyzed from El Tolmo is the absence of markers for ingredients typical of modern Catholic ritual incenses, such as cinnamic acid, benzoic acid, and cinnamyl alcohol from storax (*Styrax officinalis* L.) and benzoin (*Styrax benzoin* Dryand.) [21], incensol or boswellic acid derivatives from frankincense (*Boswellia sacra* Flück. = *Boswellia carteri* Birdw.) [8,9,11,22], furanoeudesma 1,3-diene from myrrh (*Commiphora myrrha* (T.Nees) Engl.) [23], santalol from sandalwood (*Santalum album* L.) [24], and others, which are imported from East Africa, the Arabian Peninsula, India, or East Asia. Despite this, the incense from El Tolmo emits an aroma that falls within the range of conventional incenses.

The solid, compact, black, and shiny appearance of the material, along with its strong aromatic properties that recall smoke and incense, are consistent with the characteristics of resinous materials that were commonly used for incense burning in medieval times. The fact that the material can easily be crushed into a chestnut-colored powder also supports the possibility that it is a resinous material. Early medieval recipes for incense are known [25], but the present work opens a new line of research on the elaboration of incense based almost exclusively on the use of local resources as it was among the Etruscans [26], c. 4th century BCE, and, later, in times of Dioscorides [25], 1st century CE.

Spain is one of the countries in Europe where there is a deep-rooted tradition of creating specific mixtures such as those of the various confraternities of Seville, although at present imported ingredients prevail. Isidore of Seville (7th century CE) mentions on incense (*incensum*) that it is so called because it is 'consumed by fire' (*igne consumere*) when it is offered; and a *Tetraidos formulae* of incense made from four ingredients [26]. There is evidence of the use of incense in Christian rituals in Spain during the 5th to 8th centuries CE, particularly within monastic contexts [27] and cathedrals. This evidence is derived from the presence of censers and other related objects, as well as from the study of texts such as conciliar canons [28]. However, there has been a lack of analytical studies on these incenses. Therefore, the present study provides significant new evidence and advances our understanding of medieval incenses and their composition.

5.2. Resin pellets from La Graja

Although we initially hypothesized that the samples from La Graja constituted a ritual incense used for purifying and cleaning the small mosque—given the pellets' shape, translucent nature, size of less than 1 cm, and blue to black color—the analyses contradict this hypothesis and suggest other types of materials.

The Energy-Dispersive X-ray Spectroscopy (EDS) analysis detected the presence of iron (Fe) in most of the surface of the pellet of resin while it was missing in the inner part (sample 08M1S41).

The analysis of the FT-IR spectrum of sample 08M1S41 offers relevant information. The band around 3200 cm^{-1} is typical of the OH stretching mode of alcohols. The displacement of its position at lower energies and its great width indicates the existence of O–H···O hydrogen bonds. It could also be due to the OH groups of carboxylic acids, but the absence of significant bands over

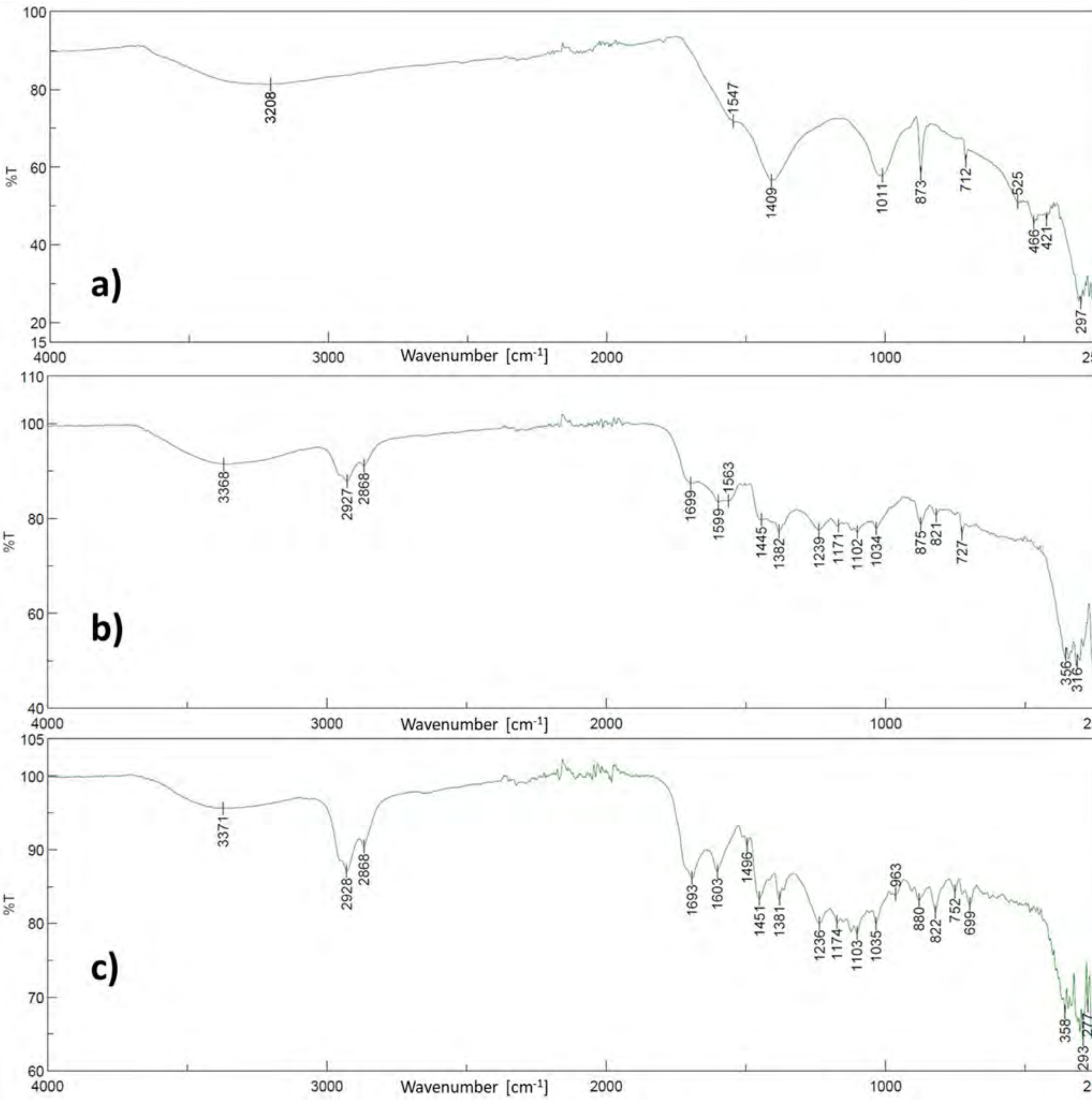

Fig. 4. FT-IR Spectra; (a) 08M1S41, Alquería de La Graja (Higueruela, Albacete, Spain); (b) 62319M2, El Tolmo (Minateda, Hellín, Albacete, Spain); (c) 62478M3, El Tolmo (Minateda, Hellín, Albacete, Spain). Images: Juan Gil.

1700 cm^{-1} allows us to rule out this possibility. Again, bands assignable to aromatic rings are observed (1547, 873 and 712 cm^{-1}), together with others attributable to C–H deformation and C–O stretching vibrations (1409 and 1011 cm^{-1}) It is worth noting the absence of C–H stretching bands, which should be observed over 2900 cm^{-1}. This could be because the sample has undergone some thermal degradation process, since the heat treatment of wood samples causes the bands above 2900 cm^{-1} to widen and decrease in intensity until they are indistinguishable [29]. The spectrum of 08M1 is compatible with the presence of alcohols, with aliphatic chains and aromatic rings. The sample has suffered intense degradation.

Several botanical sources can explain the FT-IR bands detected, such as sandarac, pine resins and tars, mastic, tree wood, bark, and labdanum, bands which can also be recognized from the Sevillian incense mixture. However, the band at 3268 cm^{-1} is only found in adhesive samples elaborated through bark pyrolysis or from Leguminosae gums (Supplementary Table 4).

During carbonization or pyrolysis organic compounds can undergo thermal degradation that can lead to the formation of volatile compounds, including benzyl alcohol and benzaldehyde among others [30]. The presence of compounds such as p-cymene, limonene, camphor, tetradecane, and cedrene suggests a potential botanical origin (Supplementary Table 5). In an archaeological context, the presence of octacosane, eicosane and hentriacontane suggests the potential use of natural waxes that contain these compounds for their protective and water-repellent functions.

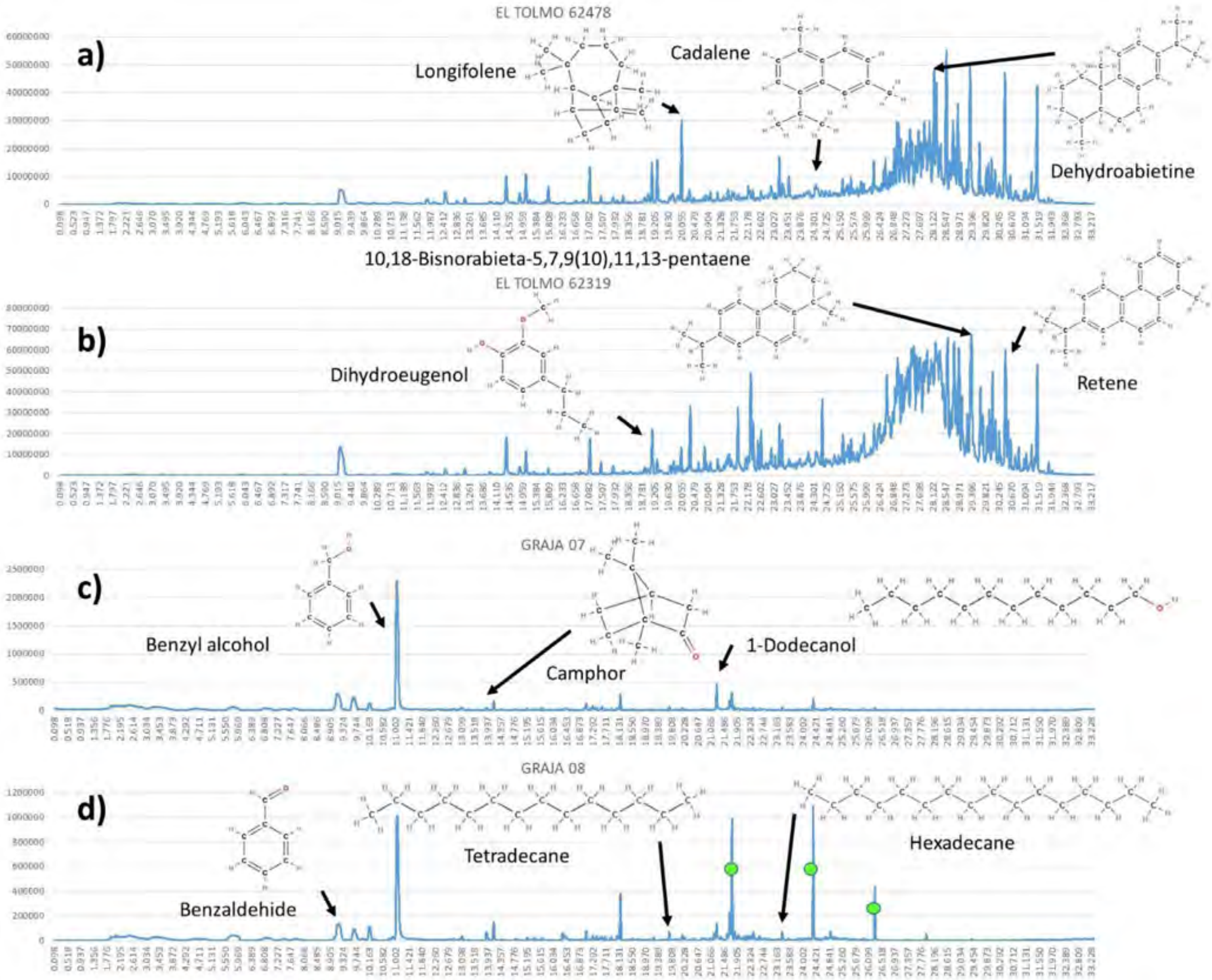

Fig. 5. GC–MS Spectra; (a) 62478M3 El Tolmo (Minateda, Hellín, Albacete, Spain). (b) 62319M2 El Tolmo (Minateda, Hellín, Albacete, Spain). (c) Sample 07, Alquería de La Graja (Higueruela, Albacete, Spain). (d) 08M1S41, Alquería de La Graja (Higueruela, Albacete, Spain). Green dots mark peaks due to artifacts: Silane and Siloxanes. Images: Diego Rivera. 2d chemical structures adapted from PubChem [15] and https://molview.org/.

Based on the identified compounds (Table 1, Supplementary Table 5), the substance appears to be a coniferous resin mixed with leguminous gum, wood tar, and beeswax. While its specific use remains unknown, it possesses preservative or adhesive properties.

Since the Neolithic tree bark tars (pure or mixed with Pinaceae resin, beeswax and fat/oil) have been used in Europe [31]. The presence of appreciable amounts of *p*-xylene and toluene in our sample suggests that our resin contains the pyrolysis products of wood [32,33].

The resin pellets were found mixed with fragments of charred wood, oak. Notably, a bronze thimble was also discovered. The presence of iron residues on the surface of the analyzed drops suggests they were in close contact with an iron object and, given their composition, may have functioned as an adhesive. However, the iron residues could also be due to accidental contact with iron materials from the building or storage in an iron container.

There are many examples of the use of adhesives in the prehistory and history. Adhesive production is one of the earliest forms of transformative technology, predating ceramics, and metallurgy by over 150,000 years [34]. Adhesives made of materials such as birch bark and pine tar, bitumen, resin, gum, lime mortar, soybeans, rice, animal hides, and blood can be found in the archaeological record around the world [35]. Depending on the type of adhesive used, different production sequences were required. Numerous plant materials were used but also beeswax and other materials from animals. A recent study [36] suggested that archaeological resin-wax mixtures were subjected to thermal pre-treatment before their use as adhesives. Heating these mixtures led to the formation of new chemical features, due to the creation of hybrid species between resin diterpenes and beeswax compounds. This could partly explain the heterogeneous composition of the La Graja pellets.

In the context of the Iberian Peninsula, ancient evidence of binding materials is sparse but significant. Notably, Upper Paleolithic adhesive residues have been identified at Cueva Morín, Northern Spain [37]. Further evidence remains scarce; however, a noteworthy analysis of residues from pottery vessels at the Roman villa of São Cucufate (Vidigueira, southern Alentejo, Portugal, 1st century CE) detected trace amounts of resin acids and diterpenoids. These findings suggest the use of coniferous resin as an adhesive during that period [38].

There are parallels that show in the area the development of technologies based on local resources of great value such as "barrilla" (from *Halogeton sativus*) to lower the melting temperature of

Table 1
Relevant compounds identified (>1 %), using GC–MS technique, in the "Incense" samples from Visigoth levels of El Tolmo de Minateda (Hellín, Albacete, Spain) and Medieval Muslim La Graja (Higueruela, Albacete, Spain).

Compounds	CAS N°	% Area	Q	% Area	Q
El Tolmo		**62478M3**		**62319M2**	
Benzaldehyde	100-52-7	1.22	1	3.31	1
1,6-dimethyl-naphthalene,	575-43-9	0.27	2	2.21	1
Eudalene	490-65-3	2.04	2	1.7	1
Cadalene	483-78-3	4.06	2	2.84	1
Calamenene	483-77-2	0.28	2	2.89	1
Endo-Borneol	464-45-9	0.69	2	1.48	2
Longifolene	475-20-7	1.75	1	0.72	1
α-Muurolene	10208-80-7	–	–	1.80	1
Methyl 3,4,5-trimethoxycinnamate	20329-96-8	0.98	3	1.02	3
4-Ethylguaiacol	2785-89-9	0.75	2	1.22	2
Dihydroeugenol	2785-87-7	0.79	2	1.54	2
2-methyl-phenanthrene,	2531-84-2	2.09	2	1.25	4
3,6-dimethyl-phenanthrene,	1576-67-6	1.25	1	–	–
Dehydroabietine	5323-56-8	9.22	1	4.35	2
10,18-Bisnorabieta-8,11,13-triene	32624-67-2	2.19	2	3.18	2
10,18-Bisnorabieta-5,7,9(10),11,13-pentaene	6566-19-4	4.17	1	5.90	1
Retene	483-65-8	3.22	1	3.99	1
Methyl dehydroabietate	1235-74-1	2.57	1	3.19	1
La Graja		**07**		**08M1S41**	
Benzyl alcohol	100-51-6	35.33	1	24.8	1
Benzaldehyde	100-52-7	8.8	2	6.25	2
Camphor	76-22-2	0.51	1	1.01	2
Octacosane	630-02-4	1.13	2	1.17	3
Cyclododecane	294-62-2	0.17	2	2.37	1
Hexadecane	544-76-3	0.35	1	1.60	1
Eicosane	112-95-8	1.87	2	–	–
Hentriacontane	630-04-6	0.17	3	1.17	3
1-Dodecanol	112-53-8	3.4	2	–	–
p-Xylene	106-42-3	1.99	1	1.74	2

Notes : Quality code (Q) : 1, $q \geq 95$ %; 2, 80 %$\leq q < 95$ %; 3, 65 %$\leq q < 80$ %; 4, $q < 65$ %. CAS N°: CAS Registry Number (number assigned by the Chemical Abstracts Service).

silica sand in the manufacture of glass [39,40], or "*esparto*" as a source of materials for cordage, basketry and for caulking boats [40–42], so it cannot be ruled out that local incenses also reached considerable levels of quality. The formulation of adhesives, on the other hand, seems to be part of a long European tradition dating back to the Neolithic period [14,31].

6. Conclusions

The study of the remains through the combination of different analytical techniques has allowed us to suggest that the sample from El Tolmo (8th century CE) was most probably used as incense, but in its elaboration, mostly local species of Cupressaceae, Pinaceae and Lamiaceae were used. The presence of imported exotic ingredients is not totally ruled out but is not necessary to explain the detected compounds and functionalities.

In the case of the samples from La Graja (11th century CE), we are dealing with a complex material low in volatile compounds. The ingredients may have included tars obtained by pyrolysis of tree bark, conifer resins, and beeswax. However, we did not find any tools in the site excavation with this resin, only isolated pellets, so we cannot conclude a definite use for it. This could prompt other research teams to verify the presence of similar pellets in their samples from other medieval excavations, which might otherwise be overlooked, thus enhancing the understanding of their significance. Furthermore, the recovery and analysis of tools combining iron and wood are essential to search for analogues of the "resin" pellets, which could confirm their use as an adhesive, following a millennia-old tradition in Europe.

For both the La Graja and El Tolmo samples, implementing solution extraction GC–MS or pyrolysis GC–MS would significantly improve the detection of non-volatile compounds, providing a more comprehensive chemical profile. These techniques would broaden the range of detectable compounds and offer structural insights into the residues, complementing our current data.

Future research should also focus on the experimental creation of incenses, based on resins and local aromatic plants, that mimic or approximate those discovered in El Tolmo. Additionally, it is crucial to continue the chemical study of medieval containers used in religious contexts to better understand the diversity of incenses used at the time.

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Declaration of generative AI and AI-assisted technologies in the writing process

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Supplementary materials

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