[Supplementary material]

Birch-bark tar in the Roman world: the persistence of an ancient craft tradition? Martine Regert^{1,*}, Isabelle Rodet-Belarbi^{1,2}, Arnaud Mazuy¹, Gaëlle Le Dantec¹, Rosa Maria Dessì¹, Stéphanie Le Briz¹, Auréade Henry¹ & Maxime Rageot^{1,3}

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S1. Description and context of discovery of the Roman hinges investigated

Roman hinges are composite objects comprising an empty, cylindrical bone piece frequently made of a sawed-off section from a long and tubular bone (a cattle - Bos Taurus metatarsus). Two models can be distinguished: a small one with one regular hole (type A XI, 2; Béal 1983: 102) and a long one with two regular holes, one above the other, pierced on one face (type A XI, 1; Béal 1983: 102). The other pieces that compose hinges are a wooden cylinder conforming to the internal cavity of the bone and wooden pegs (Fremersdorf 1940). The centre of the bone is plugged with the wooden cylinder. Each piece is held to the next by a system of tenons and mortise located at its ends. The cylinders may be adjusted together to produce hinges which are able to twist on the same axis but in different directions. The wooden pegs are fitted inside the holes of the cylindrical bone and alternately, inside the holes bored into the edge of the box or cupboard and into the lid (or door). Some hinges come from ancient excavations and their contexts of discovery are unknown (Lyon, Besançon). Others were found in urban contexts, sometimes in handcraft areas (Autun, rue des Pierres; Fréjus, les Poiriers). Several examples were found in sanctuary areas (Vendeuil-Caply), rooms of domus (Amiens, Palais des Sports), in dumps (Amiens, îlot de la Boucherie and Garage Citroën), gutters (Nice, Cimiez), in pit fillings (Reims, rue Maucroix) or in various embankments (Amiens, îlot de la Boucherie; Narbonne, clos de la Lombarde; Fréjus, îlot Camelin).

S2. Details of the methodology employed in the chemical analysis

DI-MS analyses

A micro-grain, typically the size of a pinhead or even smaller, was placed in a Pyrex vial on the probe that was introduced into the ionisation chamber of the mass spectrometer. The probe was heated from 50°C to 325°C at a rate of 40°C min⁻¹ which allows the thermal desorption of the organic molecules present in the sample. Mass spectra were recorded in the electron ionisation mode at 70 eV on a Shimadzu QP210 ultra equipped with a quadrupole. The mass range was scanned from m/z 50 to 950. The source temperature was fixed at 200°C. To avoid any memory effect, a blank was acquired between the analyses of successive samples with an empty Pyrex glass.

GC-MS analyses

The samples were extracted with dichloromethane (HPLC grade) and dried under a gentle stream of nitrogen at 40°C. Trimethylsilation was performed with 50 μ L of BSTFA with 1% TMCS, 1 μ L of pyridin and 1 μ L of CH₂Cl₂ dichloromethane for 30 min at 70°C. After evaporation of the derivatising agent to dryness, the sample was re-dissolved in 100 μ L of dichloromethane.

One µL of the solution obtained was then injected into the split/splitless injector (splitless mode) on a GC-MS apparatus Shimadzu GC2010 plus QP2010 ultra. The temperature of the injector was fixed at 300°C. The gas chromatograph was equipped with a DB-5HT column (15m length, 0.32mm i.d., 0.1µm film thickness). Helium was used as carrier gas. Gas flow at the head of the column was of 3mL min⁻¹. The temperature of the gas chromatograph oven was programmed as follows: 50°C for 1 min, 20°C min⁻¹ to 150°C, 4°C min⁻¹ to 350°C, 10 min at 350°C.

Mass spectra were recorded in the electron ionisation mode at 70eV. The source was held at 200°C and the transfer line at 280°C. The mass range was scanned from m/z 50 to 800 in 0.25 s.

S3. Textual analysis and textual databases exploited

The research focused on the following Latin terms occurring in texts from Classical Antiquity to the Middle Ages: the lemmas *betulla*, *bitumen*, *cera*, *gluten*, *pix* and *resina* (Table S1). *Betulla* is the Latin word used by Pliny the Elder to refer to the birch tree. The other terms (*bitumen*, *cera*, *gluten*, *pix* and *resina*) belong to the same lexical field. They are often used as synonyms for a viscous, sticky and combustible substance, whether or not of plant origin

lemmas	Betulla	Bitumen	Cera	Gluten	Pix	Resina
	(birch)					
LLT (Library of Latin Texts)	3	410	1685	117	720	474
PL (Patrologia Latina)	1	319	1412	155	450	207
MGH (Monumenta Germaniae	0	73	389	49	140	21
Historica)	0	15	507	ч <i>у</i>	140	21
CBMA (Corpus Burgundiae Medii	0	0	0	0	1	0
Aevi - Corpus of medieval Burgondy)	0	0	U	0	1	0
Chartae Galliae	0	0	0	0	0	0
Original charters prior to 1121	0	0	0	0	0	0
Charters from 1121 to 1220	0	0	0	0	0	0

Table S1. Summary of the Latin text databases investigated and occurrences of searchwords. Numbers in parenthesis correspond to the occurrence related to birch.

In Old French texts, research was focused on the terms *boul* for birch, and *brai* and *glu* for viscous and sticky substances of vegetal origin, sometimes obtained from birch, sometimes produced by other plants. In Modern French, brai de bouleau means "birch-bark tar". Coming from Scandinavia (braeda - cf. FEW. One should note that the word brai first appeared in areas where the substance thus mentioned could help caulking ships), the word brai was used in Normandy, and from there went to England (after the Norman Conquest of England) and then to France. As for the word glu (glue), it is quite frequent in Old French texts where it refers to sticky substances of vegetal origin, mostly holly, and it often appears as a means to capture birds (cf. DMF, GD, FEW). Although it appeared in Old French texts at the end of the twelfth century (cf. TLFi), we chose not to include the word goudron (from the Arabic qatran), since it is generally used for fossil substances. For the same reason, we did not consider the word *bitume*. In the *DMF*, the only occurrences quoted appear in translations from ancient technical texts (one of a Hippocrates' medical treatise, the other of a Vegetius' military treatise). Moreover, another occurrence which is not in the DMF (it can be read in Regnaud le Queux' Baratre infernal, c. 1480: see La Vision de Tondale. Les versions françaises de Jean de Vignay, David Aubert, Regnaud le Queux, éd. Mattia Cavagna, Paris, Champion, 2008 (Classiques français du Moyen Âge, 159): 296–97), which is meant to explain the viscosity of Hell's "ground" through a comparison with bitumen and which is not to be read in the Latin text which usually inspires Regnaud (the anonymous Speculum

morale), is however glossed with a reference to Ovide's *Metamorphoses* (liber IX), where *bitumen* means the well-known mineral product of earth. So, all medieval occurrences of *bitume* refer to antic uses, and none relates to birch tar. As for the words *poix* and *resine*, they relate to resinous plants which are not birch, while *cire* mainly means "beeswax". The main online dictionaries and reference lexicons, based on a corpus of Old French texts, were consulted. For each word, these resources propose information related to definition and etymology and provide occurrences of the word with mention of the written sources and the portion of the text containing the term of interest (Table S2).

 Table S2. Summary of the French medieval text databases queried and occurrences of search words.

	Boul	Brai	Ritume	Cire	Glu	Goudron	Poir	Resine	
	(birch)	Drut	Duume	ene	0111	Gouaron	1 000		
AND	1	1	2	30	7	_	_	_	
CREALSCIENCE	0	0	_	_	_	_	_	_	
DMF	7	3	2	84	31	6	12	4	
GDC	10	0	5	25	5	10	8	7	

The characteristics of each database queried are provided below. It is noteworthy that some of the terms of interest had other meanings completely unrelated to this research. The context in which each word was detected helped isolating the meanings relevant to this investigation. Finally, dictionaries of Gaulish language were consulted to assess the origin of the terms related to birch (Delamarre 2001; Lambert 2003; Walter & Avenas 2017).

Latin text databases

Library of Latin Texts, Brepolis, Brepols Publishers Online (LTT-O)

(http://www.brepols.net/Pages/BrowseBySeries.aspx?TreeSeries=LLT-O)

This database is composed of two series. The *LLT-A* series contains 78 million Latin words collected in more than 3800 texts from 1200 authors, from Classical antiquity to the twentieth century. LLT Series B is a supplement to Series A, operational since 2009. "In total, the present version of the *LLT-B* includes 901 works in addition to 5804 diplomatic charters: it is now possible to search more than 36.4 million forms, drawn from more than 900 works that

are attributed to approximately 450 authors" (<u>https://about.brepolis.net/library-of-latin-texts-series-b/</u>).

Patrologia Latina (PL) is the electronic version of the collection of the same title published by Jacques-Paul Migne in the nineteenth century. The PL includes Latin works spanning a millennium, from Tertullian (d. AD 230) to Pope Innocent III (d. AD 1216), but also includes works from AD 1216 onwards

(http://www.mlat.uzh.ch/MLS/xanfang.php?corpus=2&lang=0).

Monumenta Germaniae Historica (MGH)

MGH concern the medieval history of Germany and Europe, including Church history. The database eMGH includes the series falls into five main divisions, *Antiquitates*, *Diplomata*, *Epistolae*, *Leges* and *Scriptores*, with an additional smaller division of *Necrologia*. Many subsidiary series have also been established, including a series of more compact (*Scriptores in usum scholarum*) and special studies (MGH Schriften)

(https://about.brepolis.net/monumenta-germania-historicae-emgh/).

CBMA « Corpus de la Bourgogne du Moyen Âge » (Corpus of medieval Burgundy) (http://www.cbma-project.eu/)

This textual database of the Burgundian region is composed of nearly 29 000 charters and more than 30 manuscripts.

TELMA platform [Electronic processing of manuscripts and archives] from the IRHT (Institut de Recherche et d'Histoire des Textes) (http://www.cn-telma.fr/) Telma is a service and distribution platform for electronic publishing of corpus of ancient sources. It offers approximately ten text databases, including:

- Chartae Galliae (http://www.cn-telma.fr/publication/chartae-galliae)
- Original charters prior to AD 1121 (http://www.cn-telma.fr//originaux/index/)
- Charters from AD 1121 to 1220 (http://www.cn-telma.fr//originaux2/index/)

Chartae Galliae is a database devoted to writings related to France until the end of the thirteenth century. It was set up within the framework of the ANR (French National Research Agency) Espachar project (*Les espaces de la charte : les caractéristiques régionales et des réseaux d'influence dans l'établissement des actes*). Still active, it is regularly complemented with the integration of the twelfth century Cistercian charters (within the framework of the ANR Charcis programme). The database currently contains nearly 40 000 medieval charters of France.

The database *Original charters prior to AD 1121* provides the original texts, and soon photographic reproductions, of all the original charters prior to 1121 preserved in France. It consists of more than 5000 charters.

The database *Charters from AD 1121 to 1220* is a complement to the previous one (original charters prior to 1121) that has been constituted by adding the corpus of charters of the following century, currently preserved in Lorraine, *i.e.* more than 700 charters to date.

French medieval text databases

CREALSCIENCE

This database (https://crealscience.github.io/Dictionnaire/) results from an ANR (French National Research Agency) project started in 2010 (http://www.agence-nationale-recherche.fr/Projet-ANR-10-CREA-0007). The Crealscience project (Genesis of a scientific language: the case of medieval French) is focused on the genesis of French as a scientific language in Medieval times (Ducos & Salvador 2011). Within the framework of this project, a dictionary of Medieval Scientific French (DFMS) is being produced (oral communication I. Vedrenne). The letters A, B, C, should be published soon.

Dictionary of Middle French (Dictionnaire du Moyen Français (DMF)) (http://www.atilf.fr/dmf/)

This is an electronic dictionary on the French language of the Middle Ages (AD 1300–1500), initiated in 1981, developed within the ATILF laboratory in Nancy, and still evolving. This dictionary currently proposes 65 720 entries, 470 125 citations. The text corpus of the DMF consists partly of the FRANTEXT database (220 full texts) and about 500 other 'partial' texts.

Dictionary of Old French language and all its dialects from the 9th to the 15th century by Frédéric Godefroy (GD)

It is completed by the GDC (complement au Godefroy).

(https://classiques-garnier.com/godefroy-dictionnaire-de-l-ancienne-langue-francaise.html). This dictionary is the essential element of the lexicography of Old French. It provides an invaluable source of information with more than 160 000 entries, and approximately 370 000 extracts from authors from Middle Ages. The dictionary relies on several sources including printed sources, manuscripts, charters and documents of archives, of the ninth to fifteenth centuries AD.

Anglo-Norman Dictionary Online (AND)

(http://www.anglo-norman.net/gate/index.shtml?session=SNWK28160T1532684621) The Anglo-Norman language corresponds to the language in use in England following the Norman Conquest from AD 1066 until the middle of the fifteenth century. This online dictionary is a continuation of the second edition of the dictionary edited by Louise W. Stone and Wiliam Rothwell. Entries A to F are based on the text from the paper version of this second edition. For letters G to L, the paper version was abandoned in favour of the online version. The M–Z entries are based on the text from the first edition of the dictionary and will be gradually replaced as the new electronic version progresses.

To date, for the letters A to E, there are 10 600 nouns, and more than 26 000 entries, as well as 155 000 citations.

Französisches Etymologisches Wörterbuch (FEW), by Walther von Wartburg (continued under the direction of Jean-Pierre Chambon and Jean-Paul Chauveau), Bonn/Leipzig/Berlin/Paris/Bâle /Tübingen, 1928-... (https://apps.atilf.fr/lecteurFEW/) This is an etymological and historical dictionary of the Galloroman language (French and dialects of oïl, francoprovençal, occitan, gascon). The *FEW* aims to provide the most complete possible picture of the Galloroman lexicon from a genetic perspective, with analysis of phonetic, morphological and semantic changes, and precise references to sources. This work, begun by Walther von Wartburg between 1910 and 1940, is today continued by the ATILF group [*Analyse et Traitement Informatique de la Langue Française*] in Nancy (France). The general index of the dictionary consists of 25 fascicles and more than 16 700 pages. A selective index was published in 2003 under the direction of Eva Buchi. Accessible online, it contains 2370 pages and 275 295 forms deemed representative of the 25 volumes of the *FEW*, or about one-twentieth of the total.

Trésor de la Langue Française informatisé (TLFi) (http://atilf.atilf.fr/tlf.htm)

This is the computerised version of the *Trésor de la Langue Française*, a dictionary of the nineteenth and twentieth centuries in 16 volumes and one supplement, listing 100 000 words with their history, their definition (270 000 definitions) and examples presenting the words in their context (430 000 examples of occurrences).

Base textuelle FRANTEXT de l'Atilf (FRANTEXT) (http://www.frantext.fr)

This is a database including 5350 references and 251 000 000 words. Initiated and developed within the ATILF laboratory in Nancy, *Frantext* is has been online since 1998 and is still evolving. The texts studied date from ninth to twenty-first century, 325 are in medieval French. Each word is given in context.

S4. Translations

Pliny the Elder, Natural History, Book XVI, translated from the Latin by H. Rackham (1938).

XXX.

The service-tree delights in cold places, but even more the birch. The latter is a Gallic tree, of a remarkable white colour and slenderness, a cause of terror as supplying the magistrates' rods of office; it is also easily bent to make hoops and likewise the ribs of small baskets, and the Gauls extract from it bitumen by boiling. These trees are accompanied into the same regions by the may also, the most auspicious tree for supplying wedding torches, because according to the account of Masurius it was used for that purpose by the shepherds who carried off the Sabine women; but at the present time the hornbeam and the hazel are most usually employed for torches.

LXIX.

Cato attributes to the willow the third place in the estimation of the countryside, and puts it before the cultivation of the olive and before corn or meadowlandand this is not because other kinds of withes are lacking, inasmuch as the broom, the poplar, the elm, the blood-red cornel, the birch, the reed when split and the leaves of the reed, as in Liguria, and the vine itself and brambles after the thorns have been cut off serve as ties, and also the hazel when twisted and it is surprising that any wood should make stronger ties after being bruised by twisting; nevertheless it is the willow that has the properties specially required for this purpose.

LXXVII.

The trees that have the coldest wood of all are all that grow in water; but the most flexible, and consequently the most suitable for making shields, are those in which an incision draws together at once and closes up its own wound, and

which consequently is more obstinate in allowing steel to penetrate; this class contains the vine, *agnus castus*, willow, lime, birch, elder, and both kinds of poplar. Of these woods the lightest and consequently the most useful are the *agnus castus* and the willow; but they are all suited for making baskets and things consisting of flexible wicker-work. Also they are shiny and hard, and easy to use in carvings. Plane has flexibility, but of a moist kind, like alder; a drier flexibility belongs to elm, ash, mulberry, and cherry, but it is heavier. Elm retains its toughness most stoutly, and is in consequence the most useful wood for the hinges and frames of doors, because it is not liable to warp, only it should be put the other way up, so that the top of the tree is towards the lower hinge and the root above.

Figures

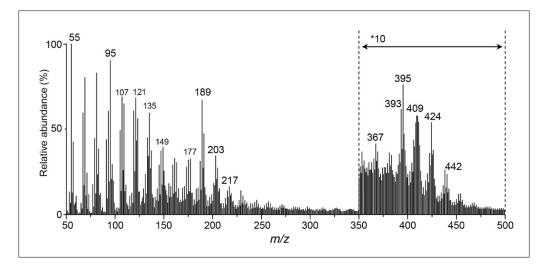


Figure S1. Mass spectrum of sample MR6253, obtained by DI-MS, typical of birchbark tar. The spectrum corresponds to the sum of the spectra between 4 and 7 min on the desorption curve after substraction of the background between 1 and 1.5 min. The region of the spectrum from m/z 350 to 500 is magnified (×10).

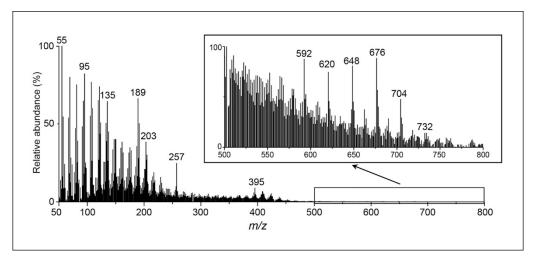


Figure S2. Mass spectrum of sample MR6261, obtained by DI-MS, characteristic of a mixture of birch-bark tar and beeswax. The spectrum corresponds to the sum of the spectra between 5 and 6 min on the desorption curve after substraction of the background between 1 and 1.5min. The detail of the spectrum (topright) shows the peaks characteristic of the molecular ions of beeswax palmitate esters.

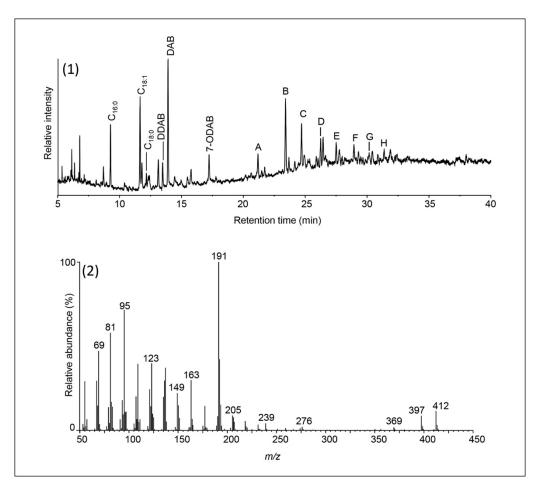


Figure S3. Chromatogram (1) obtained by GC-MS analysis of sample MR6248 showing the presence of fatty acids, diterpenoid markers and a series of hopanes

components (AtoH). DDAB: dehydro-dehydroabietic acid. DAB: dehydroabietic acid. 7-ODAB: 7-oxo-dehydroabietic acid. The components labeled A to Hal lpresent a base peak at m/z191 in mass spectrometry, with molecular weight of 370 (A), 398 (B), 412 (C), 426 (D), 440 (E), 454 (F), 468 (G) and 482 (H) corresponding to hopane scomponents with respectively 27, 29, 30, 31, 32, 33, 34 and 35 carbon atoms. The mass spectrum of compound C is compatible with that of 17a-hopane published in 1974 by Kimble et al., as shown on the second part of this figure (2). Table S3. List of samples investigated in this study and indication of the materials identified (bbt: birch-bark tar. FA: fatty acids. PR:pine resin. BW: beeswax). ND: not documented. AE: ancient excavations.

Town	Archaeological site	Complementary	Chronology	Lab	Type of	Analyses	Materials
		archaeological		number	object	carried	identified
		information				out	
Amiens	AIB	36	AD 60–70	MR6288	Hinge part	DI-MS	bbt
		38	<i>c</i> . AD 70	MR6289	Hinge part	DI-MS	bbt
			10/190-10/250			DI-MS &	
		139	AD	MR6290	Hinge part	GC-MS	bbt
			AD 140–			DI-MS &	
	Garage Citroën	600	180/200	MR6286	Hinge part	GC-MS	bbt
						DI-MS &	
		667	AD 40–100	MR6287	Token	GC-MS	bbt
						DI-MS &	
	PDS	303	AD 125–130	MR6283	Hinge part	GC-MS	bbt
			AD 100/110-			DI-MS &	
		650	125/130	MR6284	Hinge part	GC-MS	bbt
						DI-MS &	
		900	AD 125–130	MR6285	Hinge part	GC-MS	bbt

			Mid-first to				
		Région 1, Insula	mid-second				
Augst	Augusta Raurica	22, 1964-1603	century AD	MR6269	Hinge part	DI-MS	bbt
			Mid-first to				
		Région 1, Insula	mid-second				
		24, 1959-5193	century AD	MR6265	Hinge part	DI-MS	bbt
		Région 1, Insula					
		25,1977-10977	1st century AD	MR6263	Hinge part	DI-MS	bbt
			Mid-first to				
		Région 1, Insula	mid-second				
		29, 1961-13747	century AD	MR6266	Hinge part	DI-MS	bbt
			Mid-first to				
		Région 1, Insula	mid-second				
		30,1962-10895	century AD	MR6264	Hinge part	DI-MS	bbt
			Mid-first to				
		Région 1, Insula	mid-second				
		30,1959-12118	century AD	MR6267	Hinge part	DI-MS	bbt
		Région 1, Insula	First–Fourth				
		50, 1982-12385	centuries AD	MR6270	Hinge part	DI-MS	bbt
		Région 17, D,	First century				
		1973-14151	AD	MR6268	Hinge part	DI-MS	bbt

		Région 2, E, 1985-	First century				
		83552	AD	MR6262	Hinge part	DI-MS	bbt
			Gallo-Roman				
Autun	Rue des Pierres	EVI - 62	times	MR6256	Hinge part	DI-MS	bbt
Besançon	Museum collection	41501 -	Gallo-Roman	MR6274	Hinge part	DI-MS	Hydrocarbons
		2013.0.1628	times				
			Gallo-Roman				
		40831- 852.15.52	times	MR6275	Hinge part	GC-MS	FA + bbt
		44424 -	Gallo-Roman				
		A.2005.6.135	times	MR6276	Hinge part	GC-MS	bbt
		41502 -					
		2013.0.1629/South	Gallo-Roman				
		Gaul	times	MR6277	Hinge part	DI-MS	bbt
			Gallo-Roman			DI-MS &	
Chartres	Cœur de ville	C191-5024-1	times	MR6271	Hinge part	GC-MS	bbt
			Gallo-Roman			DI-MS &	
		C191-5047-2	times	MR6272	Hinge part	GC-MS	bbt
			Gallo-Roman		Cylinder		
		C 190-5282-3	times	MR6273	part	GC-MS	bbt
			First century			DI-MS &	
Fréjus	Îlot Camelin	1041	AD	MR6252	Hinge part	GC-MS	bbt

						DI-MS &	
		3633	ND	MR6254	Hinge part	GC-MS	bbt
		3630	ND	MR6255	Hinge part	DI-MS	191 - bitumen
			Augustus:			DI-MS &	
	Les Aiguières	n°527	60/70 AD	MR6250	Hinge part	GC-MS	bbt
						DI-MS &	
	Les Poiriers	300482	400–550 AD	MR6251	Hinge part	GC-MS	bbt
			Gallo-Roman				
Lyon	ND, AE	136-608	times	136-608	Hinge part	DI-MS	bbt
			Gallo-Roman				
		143-615	times	143-615	Hinge part	DI-MS	bbt
			Gallo-Roman				
		144-616	times	144-616	Hinge part	DI-MS	bbt
	Verbe incarné	170-771-6-56	Gallo-Roman	MR6245	Hinge part	DI-MS	Insufficient
			times				material
		Carré L6 dernier					
		sol 2e état, 81-1-8-	Gallo-Roman				
		-8	times	MR6244	Hinge part	DI-MS	bbt
		ZA5 carré 9	Gallo-Roman				
		citerbe, 80-1-6-40	times	MR6246	Hinge part	DI-MS	bbt

Narbonne	Clos de la Lombarde	10007	Gallo-Roman	MR6253	Decorative	DI-MS &	bbt
			times		plaque	GC-MS	
_		Cimiez sud					
		caniveau	Gallo-Roman				
Nice	Cimiez	décumanus I	times	MR6257	Hinge part	GC-MS	bbt
			Gallo-Roman			DI-MS &	FA+PR +
		Transfert Masséna	times	MR6258	Hinge part	GC-MS	bitumen
Poitiers	Jardins du	5045	First half of the	MR6259	Hinge part	Not	Insufficient
	Pyugarreau		second century			analysed	material
			AD				
			Third century			DI-MS &	
Reims	Bd Henrot	OI 742, US 518	AD	MR6281	Hinge part	GC-MS	bbt
			Third century				
		OI 1035, US 1334	AD	MR6282	Hinge part	DI-MS	bbt
_			Third century				
	Rue Maucroix	OI 789, US 1070	AD	MR6278	Hinge part	DI-MS	bbt
_			Third century				
		OI 1043, US 398	AD	MR6279	Hinge part	DI-MS	bbt
		OI 153, US 158	No context	MR6280	Hinge part	DI-MS	bbt
			Gallo-Roman				
Rom	Le Parc 2011	125/10	times	MR6261	Hinge part	DI-MS	bbt + BW

			Gallo-Roman			DI-MS &	
	ND, AE	83S8 DEP	times	MR6293	Hinge part	GC-MS	bbt
			Gallo-Roman				
		R3 S1 1006	times	MR6294	Hinge part	DI-MS	bbt
			Gallo-Roman			Not	Insufficient
		R4?? 2003	times	MR6295	Hinge part	analysed	material
			First-Second			DI-MS &	
Saintes	Rue Daubonneau	359/3001	century AD	MR6260	Hinge part	GC-MS	bbt
Vendeuil			AD 180–			DI-MS &	
Caply	2013	2002 - 11	220/230	MR6291	Hinge part	GC-MS	bbt
			AD 180–			DI-MS &	
		2050 - 60	220/230	MR6292	Hinge part	GC-MS	bbt
Saint-	Rue Colbert	ND	Medieval:	MR6296	Die	GC-MS	FA
Maximin-			twelfth-	MR6297	Die	GC-MS	BW
la-Sainte-			fifteenth				
Baume			centuries AD				

Number	Site name	Reference	Archaeological	Site location	Type of object	Date	Period	Number of	Materials
on map			context		/ material			samples	identified
								investigated	
12	Cuciurpula	Rageot et	Early Iron Age	Site located at an	Free lumps and	Ninth-	Early Iron	Two free lumps and	Birch-bark tar
		al. (2016)	village	altitude of 1000m,	residues on	sixth	Age	seven visible	and other
				in the south-	ceramic vessels	centuries		organic residues	materials (pine
				central part of		BC		sampled on ceramic	resin, beeswax)
				Corsica, on the		(phases 0		vessels	
				Punta di		to 2)			
				Cuciurpula					
11	Vetulonia,	Morandi et	Graveyard	Northern Tuscany,	Tin-band	c. ninth–	Early Iron	Four micro-samples	Birch-bark tar
	graveyard of	al. 2018		Italy (42°51034'N	decorated	eighth	Age	of a black organic	
	Colle			10°58016'E, 335m	funerary urn	centuries	(Villanova	substance	
	Baronico			asl)		BC	culture)	decorating the urn	
2	Argancy	Regert &	Chariot burial	Argancy, Moselle,	Organic		Early Iron	One sample	Birch-bark tar
		Rolando		France	residue in a		Age		
		(2002)			chape of a				
					sword				

Table S4. List of sites from Early Iron Age to Roman period in which birch-bark tar was chemically identified. NI: not indicated.

1	Le Clos des	Langlois et	Hallstatt	Eterville, France	Organic	End of	Early Iron	One sample	Birch-bark tar
	Lilas	al. (2005)	necropolis		residue on a	sixth	Age		
					fibula	century-			
						beginning			
						of fifth			
						century			
						BC			
3	Eckwersheim	Courel et	Hallstatt	Eckwersheim, NE	"Adhesive	625–550	Early Iron	One sample	Birch-bark tar
		al. (2018:	necropolis,	France	used to stick	BC	Age		
		72)	tomb 6008		two bronze		(Hallstatt		
					half-spheres to		D1)		
					form a				
					pendant"				
4	Heuneburg	Rotländer	Hallstatt	Herbertingen-	Residues on	600–450	Early Iron	One sample	Birch-bark tar
		(1986)	settlement	Hundersingen, SO	ceramic vessels	BC	Age		
				Germany			(Hallstatt		
							D)		
5	Hochberg	Hayek et		Lower Austria	"Ancient glued		Early Iron	One sample	Birch-bark tar
		al. (1990:			joint on a		Age (R. H.		
		2042,			pottery (grave		C/D)		
		1991: 155)			gift)"				

7	Stillfried	Hayek et		Lower Austria	"Contents of		Early Iron	One sample	Birch-bark tar
		al. (1990:			pottery bowl"		Age (R. H.		
		2042,					C/D)		
		1991: 155)							
8	Odenburg	Sauter		Hungary	Material used		Early Iron	Five samples	Possibly birch-
	Burgstall	(1967)			to repair		Age		bark tar (result
					ceramic vessels		(Hallstatt)		based on IR
									analysis)
9	Kaptol-	Doracic et		Croatia	Reparation of		Early Iron		Birch-bark tar
	Gradci	al. (2012)			ceramic vessels		Age		
10	Belogradets	Ribechini		Belogradets,	Samples from	Eighth-	Thraco-	Five samples	Birch-bark tar
		et al.		Bulgaria	the golden	seventh	cimmerian		
		(2011)			sheath of a	centuries	period		
					Thracian	BC			
					dagger				
12	Cuciurpula	Rageot et	Early Iron Age	Site located at an	Free lumps and	Beginning	Early Iron	Three free lumps	Birch-bark tar
		al. (2016)	village	altitude of 1000m,	residues on	of the sixth	Age	and 24 visible	and other
				in the south-	ceramic vessels	century		organic residues	materials (pine
				central part of		BC (phase		sampled on ceramic	resin, beeswax)
				Corsica, on the		3)		vessels	

				Punta di					
				Cuciurpula					
6	Langenlebarn	Sauter et	Tomb in a	District of Tulln,	Adhesive used	<i>c</i> . 600 BC	Early Iron	One sample	Birch-bark tar
		al. (2002)	tumulus	Lower Austria	to fix clay		Age		
					figurines		(Hallstatt)		
13	Wetwang	Stacey	Chariot burial	East Yorkshire,	Adhesive in the	400–100	Late Iron	Two samples	Birch-bark tar,
		(2004)		England	harness fittings	BC	Age		possibly mixed
									with conifer
									product
15	Les Vairies	Sicard	Gallic rural	Saint-Sauveur-des-	Five tar loaves	170–10			Birch-bark tar
		(2017)	settlement	Landes, Ile-et-		BC			(unpublished
				Vilaine, France					analytical data)
14	Orval	Langlois &	Chariot burial	Coutances,	Adhesive used	Third–first	Late Iron	Five samples	Birch-bark tar
		Regert		Manche, France	to fix a coral	centuries	Age (La	analysed, four with	
		(2007)			decor on parts	BC	Tène)	birch-bark tar	
					of the harness				
					fittings				
17	Vincelles	Poplin		Vincelles, Yonne,	Adhesive used		Late Iron		Birch-bark tar
		(1971)		France	to repair a		Age (La		(unpublished
					pottery		Tène)		analytical data)

18	Lavansaari	Reunanen	Funerary	Island of	Samples from	1900–2200	Pre-Roman	Two samples	Birch-bark tar
		et al.	context	Lavansaari (russ.	wooden	BP	Iron Age		
		(1993)		Mostnyi ostrov),	containers				
				eastern part of	holding burnt				
				Gulf of Finland	bones of the				
					deceased				
16	Grand Aunay	Regert et	Enclosures and	Yvré-l'Evêque, 5	Free lumps and	Third-first	Late Iron	Eight lumps and	Birch-bark tar
		al. (2003)	ditches	km from Le Mans,	residues on	centuries	Age (La	organic residues on	
				Sarthe, France	ceramic vessels	BC	Tène)	ceramic vessels	
19	West Cotton	Charters et		West Cotton,	Roman Ecton		Roman	One sample	Birch-bark tar
		al. (1993)		Northamptonshire,	jar was		period of		
				GB	recovered		Great		
					repaired by		Britain		
					birch-bark tar				
20	Catterick	Dudd &		Yorkshire, Great	"Contents of a	Roman	Roman		Birch-bark tar
		Evershed		Britain	small,	sediments	period of		
		(1999)			enamelled		Great		
					vessel"		Britain		
21	Naintré	Ribechini		A few km from	Wooden	Second	Gallo-	One adhesive, a	Birch-bark tar
		et al.		Poitiers (France)	spindle	half of the	roman	brownish-black	
		(2011)				third	period	material, used to fix	

						century		together two pieces	
						AD		of an archaeological	
								spindle	
22	Albersto	Bergström		Province of	One tar loaf,	First	Roman Iron	One sample	Mixture of
		(2004)		Södermanland,	weight: 204g.	century	Age		birch-bark tar
				eastern Sweden		AD			and animal fat
23	Vellensby	Karg et al.	A woman's	Island of	Two chewing		Late Roman	One of the chewing	Birch-bark tar
		(2014)	grave	Bornholm,	gum-like		Iron Age	gums	(low amount)
				Denmark	objects with				and plant oil
					dental				
					impressions				
24	NI (not	Nordby	Grave contexts	Norway	Brownish-	<i>c</i> . 500 BC–	Norwegian	Thirty-seven	Birch-bark tar
	shown on	(2009)			black ring of	AD 570	Early Iron	caulking remains	
	Figure 6)				varying size in		Age		
					the sediment				
					(linked with				
					lath-walled				
					wooden or bark				
					containers)				

Number	Site name	Reference	Archaeological	Site location	Type of object	Date	Period	Number of	Materials
on map			context		/ material			samples	identified
								investigated	
А	Put Blanc	Connan <i>et al</i> .	Sublacustrin	Sanguinet Pond,	Vase 99-14	750–400	Early Iron	One sample	Conifer pitch
		(2002)	site	Landes, France	with carbonised	BC	Age		
					residue				
Н	La Castellina	Garnier (2003)	Etruscan site	La	Etruscan	700–300	Etruscan		Mixture of
				Castellina,Lazio,	amphorae	BC			pine tar/resin
				Italy	coating				and beeswax
В	Archaic	Connan (2002)	Waterlogged	Marseille, France	Ship caulking	Sixth	Archaic		Mixture of
	Greek		site			century	Greek		conifer tar
	shipwreck-					BC			and beeswax
	Jules Verne								
	VII								
С	Cassidaigne	Garnier (2003)	Waterlogged	Provence coast,	Coating	Sixth	NI		Pine resin
	shipwreck		site	France		century			
						BC			
D	La Palud II	Garnier (2003)	Waterlogged	Provence coast,	Greek	Sixth	Archaic		Pine tar
	shipwreck		site	France	amphorae	century	Greek		
					coating	BC			

Table S5. List of sites from Early Iron Age to Roman period in which conifer products were chemically identified. NI: not indicated.

Е	Pointe	Garnier (2003)	Waterlogged	Provence coast,	Greek	Sixth	Archaic		Pine tar
	Lequin 1A		site	France	amphorae	century	Greek		
	shipwreck				coating	BC			
F	Pisa San	Garnier (2003)	Ancient	Pisa, Tuscany,	Amphorae	400–200	Etruscan		Pine tar
	Rossore		harbour	Italy	coating	BC			
J	Roman	Jerković et al.	Waterlogged	Adriatic sea, near	Greco-Italian	Middle	Roman	One sample	Pine resin/tar
	amphora	(2012)	site	Vis Island, Croatia	amphora type	of the	Republic		
	from Vis				Benoit	second			
					Republicane-	century			
					II/Lamboglia	BC			
					coating				
Ι	Chiusi	Colombini et al.	Etruscan	Chiusi, Tuscany,	Unguentarium	Second	Etruscan	One sample	Mixture of
		(2009: 1490)	necropolis	Italy	content	half of		("some contents	plant oil,
						Second		of the	pine resin
						century		unguentarium,	and pistaccia
						BC		approximately	resin
								10mg")	
G	Bouvier	Garnier (2003)	Waterlogged	Porto Vecchio,	Dressel 21-22	75–60	Roman		Pine tar
	shipwreck		site	Corsica, France	amphorae	BC	Republic		
					coating				

F	Pisa San	Colombini et al.	Ancient	Pisa, Tuscany,	Waterproofing,	First	Etruscan	10 samples	Pine resin
	Rossore	(2003)	harbour	Italy	caulking and	century	and		and tar
					painting	BC-	Roman		
					materials of	second	ages		
					Roman ships	century			
						AD			
Κ	Monte Poro	Izzo et al. (2013)	Subaqueous	Monte Poro,	Dressel 1	First	Roman	NI	Pinaceae
			contexts	Calabria, Italy	amphorae	century	Republic		resin and tar
					coating	BC			
0	London,	Stern <i>et al.</i> (2008)	NI	London, United	Amphorae	AD 0-	Romano-	Seven samples	Pine resin
	Regis House			Kingdom	coating	200	British		
U	Chalon/Saône	Connan &	Waterlogged	Chalon/Saône,	Ship caulking	AD 0-	Gallo-	NI	Conifer tar
	shipwreck	Nissenbaum	site	Bourgogne-		200	Roman		
		(2003)		Franche-Comté,					
				France					
М	Carlisle	Stern <i>et al.</i> (2008)	Various sites	Cumbria, United	Camulodunum	NI	Romano-	Five residues of	Pine resin
				Kingdom	186 and		British	different	
					Gauloise flat-			amphorae	
					based amphorae				

Т	Melun-	Garnier (2003)	NI	Melun, Île-de-	Gallo-Roman	AD 50-	Gallo-		Pine tar
	Vernin			France, France	amphorae	100	Roman		
					coating				
W	Sud Perduto	Garnier (2003)	Waterlogged	Bonifacio,	Dressel 28	AD 75-	Roman		Pine tar
	II shipwreck		site	Corsica, France	amphorae	100	period		
					coating				
Х	Villa B	Ribechini et al.	Roman villa	Oplontis, Naples,	Glass	First	Roman	Seven samples	Pinaceae
		(2008)		Italy	unguentaria	century	period		resin +
						BC-first			beeswax and
						century			another wax
						AD			
S	Rouen	Garnier (2003)	NI	Rouen, Normandy,	Gallo-Roman	AD 100-	Gallo-		Pine tar
				France	amphorae	300	Roman		
					coating				
L	Sommaränge,	Hjulström <i>et al</i> .	Dark coloured	Uppland, Sweden	Free lamp in	AD 240-	Late	Thirteen from	Pine tar
	Fullerö,	(2006)	circles		small funnel-	540	Roman	funnel-shaped	
	Highway E4				shaped pits		Iron Age	pits	
R	Guernsey	Connan et al.	Gallo-roman	Island of	Piece of pitch	c. AD	Roman	One sample	Pine tar
		(2002)	shipwreck	Guernsey, United		280	period		
				Kingdom					

Y	Eastern	Papageorgopoulou	Funeral	Thessaloniki,	Embalming	c. AD	Roman	One sample from	Conifer and
	cemetry of	et al. (2009)		Greece		300	period	the compact-	pistacia
	Thessaloniki							particle material	resins
N	Northview	Brettell et al.	Burials	Purton, Wiltshire,	Debris	Late	Late	Four samples	Pinaceae
	Hospital	(2015)		UK	associated with	third-	Roman		derivative +
					skeletal	fourth	period		Pistacia spp.
					elements	century			
						AD			
Q	Poundbury	Brettell et al.	Burials	Dorchester,	Residues	Fourth	Late	13 samples	Pinaceae
	Camp	(2015)		Dorset, UK	associated with	century	Roman		derivative
					plaster or	AD	period		
					textiles				
Р	280	Brettell et al.	Burials	Around London,	Samples within	Mid-	Late	10 samples	Pinaceae
	Bishopsgate,	(2015)		UK	lead coffin	Fourth c.	Roman		derivative
	site K					AD	period		
V	Sanguinet	Connan <i>et al</i> .	Sublacustrin	Sanguinet, Landes,	Production	AD 400-	Gallo-	One sample	Conifer tar
		(2002)	site	France	residue	600	Roman		

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