

## Competing English, Spanish, and French alabaster trade in Europe over five centuries as evidenced by isotope fingerprinting

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Edited by Thure E. Cerling, University of Utah, Salt Lake City, UT, and approved September 20, 2017 (received for review May 5, 2017)

A lack of written sources is a serious obstacle in the reconstruction of the medieval trade of art and art materials, and in the identification of artists, workshop locations, and trade routes. We use the isotopes of sulfur, oxygen, and strontium (S, O, Sr) present in gypsum alabaster to unambiguously link ancient European source quarries and areas to alabaster artworks produced over five centuries (12th– 17th) held by the Louvre museum in Paris and other European and American collections. Three principal alabaster production areas are identified, in central England, northern Spain, and a major, long-lived but little-documented alabaster trade radiating from the French Alps. The related trade routes are mostly fluvial, although terrestrial transport crossing the major river basin borders is also confirmed by historical sources. Our study also identifies recent artwork restoration using Italian alabaster and provides a robust geochemical framework for provenancing, including recognition of restoration and forgeries.

alabaster | provenance | isotope fingerprinting | Middle Ages | Renaissance

A labaster was one of the preferred materials of medieval and Renaissance sculptors due to its ease of carving in elaborate detail and its translucent whiteness; it is much more than a mere substitute "if marble will not serve" (1). Alabaster was indeed chosen for many royal (2) and papal (3) effigies, for innumerable religious artworks, and for monuments.

However, the homogeneous nature and similar characteristics of alabaster from different areas make it impossible to determine its provenance on the basis of mineralogy, chemistry, and texture alone (4–6). Historically, varieties of both calcite and gypsum/ anhydrite were termed "alabaster." The first, known as "Egyptian" or "oriental" alabaster (7), quarried at the Egyptian town of Alabastron, gave its name to both materials. Even if stringent chemical distinction only became possible in the 18th century (8), sulfate and calcite alabaster were already used and distinguished in antique times (e.g., in Theophrastus' treaty on stones, ref. 9). Here we focus on the "true" alabaster, the noble variety of gypsum (CaSO<sub>4</sub>·2H<sub>2</sub>O) or anhydrite (CaSO<sub>4</sub>), widely used in medieval Europe.

The most abundant historical evidence of the European alabaster trade comes from the Midlands of England, mainly in the Dove and Trent valleys to the west of Nottingham. These deposits were considered to be the most productive and widely used European source. Alabaster from the English Midlands was worked by the "alabastermen" from the 12th century (2, 10) until 1550, when an Act of Parliament triggered the iconoclastic crisis of the Reformation, stopping all production of religious sculpture. From this time on, only funeral effigies were permitted and these continued to be produced in great number (10, 11). Early English alabaster export is documented both for artwork, e.g., to Rome (1382), Seville (1390), and Nantes (1408) (10), and for sculptural raw materials, e.g., to Fécamp Abbey in Normandy in 1414 (10, 12). After the banning of religious representations, shiploads of alabaster artworks were sent to France (13). In England, only a few sculptures escaped the plaster furnaces by being hidden and were retrieved centuries later (14). In contrast, western and northern France was inundated with outlawed English artworks.

Another well-known center of the alabaster trade was northern Spain. Spanish alabaster from the medieval quarries at Beuda, Sarral, and the Ebro Valley was the material of choice for sculptors working for the Crown of Aragon from the 14th to 16th centuries, as evidenced by the particularly large altarpieces in Aragon and Catalonia. Trade from this area spread north of the Pyrenees (15), into Portugal, and as far as the Kingdom of Naples (16).

Little is known about the distribution from other historical alabaster deposits in continental Europe. Whereas the various sources of classical marble have been studied for many decades (17, 18), the scarce documentary evidence on alabaster could not be verified by any independent geochemical method, a situation that Cheetham in his landmark monograph (10) considered as "most frustrating."

Our pilot study (19) tested isotope fingerprints (Sr, S, O) as an independent method to link artworks to their source areas, and developed an associated analytical protocol: Tiny flakes (<20 mg) of alabaster are dissolved in distilled water at 50 °C, and the dissolved sulfate is precipitated as  $BaSO_4$  for S- and

## Significance

Gypsum alabaster in medieval artwork equaled or even surpassed the importance of marble for religious sculptures and effigies. Based on isotope fingerprints, a large corpus of European alabaster sculptures are linked to historical quarries and trade in the Middle Ages and Renaissance period. We demonstrate that English alabaster export was competed on the continent by the Spanish and by a particularly long-lived (>500 y) French Alpine trade. Our study reveals historical trade routes and transport, the diversity of supply of medieval artists and workshops, as well as the presence and provenance of restoration materials.

Author contributions: W.K. and L.L. designed research; W.K., L.L., P.B., P.-Y.L.P., A.H.C., N.W., C.G., A.T.M., A.M.G., and R.A. performed research; C.G. and A.T.M. contributed new reagents/analytic tools; and W.K. wrote the paper.

The authors declare no conflict of interest.

This article is a PNAS Direct Submission.

This article contains supporting information online at www.pnas.org/lookup/suppl/doi:10. 1073/pnas.1707450114/-/DCSupplemental.

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O-isotope analysis by continuous flow isotope ratio mass spectrometer (CF-IRMS). Aliquots of the solutions are purified by ion exchange for Sr-isotope analysis by thermal ionization mass spectrometer (TIMS). Full details of the methods are provided in *SI Materials and Methods*. The sparse initial database of European historical quarries has now been greatly enlarged and includes virtually all of the known pre-19th century sites in France, plus major deposits in England, Spain, Germany, and Italy (Fig. 1 and Table S1). Strong interdeposit contrast and good intradeposit homogeneity of the isotope fingerprints (Fig. 2 and Table S2) were demonstrated, particularly when strontium and sulfur are combined. Oxygen isotopes are slightly more scattered, but are essentially consistent (albeit with some overlap) with the Sr and S data. In addition, some sources are strongly depleted or enriched in <sup>18</sup>O, strengthening the method's discriminating power (Fig. S1).

A large corpus of major alabaster artworks [66 samples including 9 already characterized (19)] has been analyzed (Tables S2 and S3). The Louvre Museum in Paris provided 19 samples from 14th–17th century sculptures. The Petit-Palais Museum in Avignon and the collection of the Chartreuse in Villeneuvelez-Avignon added 13 samples from 12 effigies, along with altarpieces from the funeral monuments of popes and cardinals of French papacy of the second half of the 14th century and during the Great Schism (1378–1417). Other individual pieces were made available from American (Cleveland Museum of Art, Chicago Art Institute), Swedish, English, and French collections, galleries, and churches.

## **Results and Discussion**

Fifteen works of art or parts of sculptural ensembles, ranging from the 14th to 17th centuries, are undoubtedly identified as Triassic alabaster from the English Midlands (Fig. 3). Our sampling strategy mainly included artwork for which English craftsmanship was not stylistically evident; consequently, English material is statistically underrepresented. The only typical "Nottingham School" carving among these samples is a 14th century altar panel that is a fragment of an Arrest of Christ from the Cluny Museum in Paris (20). In contrast, the 15th century "St. Michael striking the Evil" from the Louvre in Paris is attributed to a workshop in the Touraine province of France. These examples illustrate the medieval trade of both carvings and raw alabaster from England. More than half of the samples date from the 16th century, showing the massive export of English alabaster raw materials to France and northern Europe post-1550 in the aftermath of the Protestant iconoclastic crisis. This is illustrated by the isotope analyses (19) of the funeral monument of King Gustaf Vasa of Sweden, sculpted around 1570 by the Flemish artist Willem Boy, and by the monumental high altar of Calais produced by Adam Lottman from Valenciennes in 1624-1628. These findings support Firman's analysis (21) of an "unprecedented boom in the alabaster trade" lasting 70-80 y from 1580 before going into decline until around 1700.

Northern Spain exported heavily to southern Europe (22), but only to the Mediterranean coastal zone in France, notably the Perpignan and Narbonne areas, where alabaster arrived from Beuda by ship (19). Further evidence of local to regional supply is found in Burgundy, where small-scale medieval quarries furnished the decoration of the Jacques d'Amboise Palace in Cluny. Local supply was also evident in Provence and the French Alpine Maurienne region.

The most intriguing result of our study is the great number of non-English carvings with a very homogeneous isotope composition indicative of a French Alpine origin. Virtually all late 14th to early 15th century artworks that are linked to the popes of Avignon (11 artworks), with only one notable exception, fall in an extremely

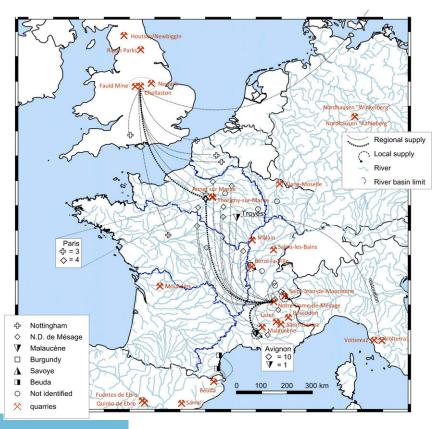
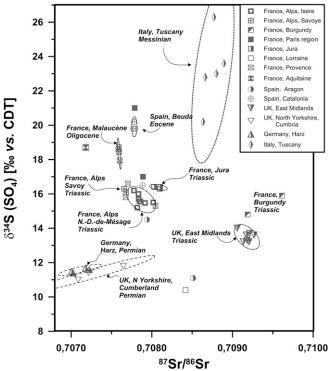


Fig. 1. Original location of analyzed artworks, attributed provenance, and schematic links to historical quarries.

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**Fig. 2.**  $\delta^{34}$ S versus <sup>87</sup>Sr/<sup>86</sup>Sr ratios of historical alabaster quarries in France, England, Spain, Italy, and Germany, including data from ref. 19. The data fields are delimited by  $2\sigma$  confidence ellipses (95% confidence level).

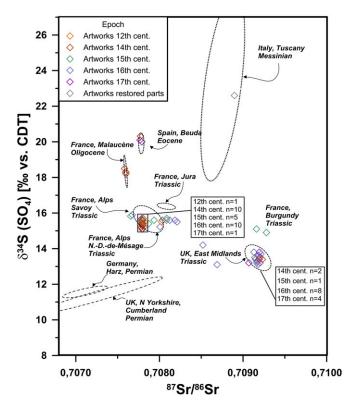
small range of  ${}^{87}\text{Sr}/{}^{86}\text{Sr}$  (0.707796  $\pm$  0.000005 n = 11) and  $\delta^{34}\text{S}$  $(15.5 \pm 0.1\%)$  vs. Canyon Diablo Troilite, CDT). This group is complemented with nine artworks from the Louvre Museum (Fig. 4), and six from other collections covering five centuries from the 12th to the 17th century (Fig. 3). The only deposit with a compatible isotope signature is Notre-Dame-de-Mésage (NDM), situated in the western French Alps, southwest of Grenoble. Here, the gypsum quarries are still accessible and alabaster is strongly represented in the local religious architecture. Two artworks in this group are noteworthy. One was collected from a fragment of the funeral chapel of the Counts of Savoy in Hautecombe Abbey. For this sample, a rare and explicit historical source exists detailing the transport of medieval alabaster. The accounting records of the castellany of Vizille (23) for 1336 make specific reference of the transfer of 45 blocks of alabaster on behalf of the Dauphin Humbert II for use by the Counts of Savoy. This exceptional journey began at "Mesatico" (NDM) with a 16-km ride using carts drawn by 110 oxen down to the navigable section of the Isere River. The cargo then proceeded by boat and finished using ox carts again for the remaining 25 km to Hautecombe Abbey, 100 km north of NDM on the shores of Bourget Lake. This source was cited as early as 1722 by Moret de Bourchenu (24), who stated that an alabaster quarry was still active in Mésage. This further proof of quarrying here agrees with our findings characterizing the second noteworthy artwork belonging to this group, the funeral monument of the Constable de Lesdiguières. Indeed, in the 17th century he owned the land where the NDM quarries are situated (25). Four other carvings, mainly 16th-17th century, show similar  $\delta^{34}$ S values but higher  $^{87}$ Sr/ $^{86}$ Sr ratios and may be attributed to other quarries in the NDM deposits.

Our study brings to light the extensive geographical distribution and longevity of the French Alpine alabaster industry, rivaling that of the English Midlands. The zone of influence of this French trade encompassed all of the eastern part of France (Fig.

Seine, and Loire. As in the English Midlands (14), river transport was possibly a key to this success with the NDM quarries situated on the banks of the historically (at least temporarily) navigable Romanche River (23), giving access, via the nearby Drac and Isère Rivers, to the Rhone and Avignon. Transloading from ship to ox carts and terrestrial transport, as shown by the Hautecombe example, gave access to the upstream parts of the Rhone basin and other major river basins. We conclude that papal Avignon was almost exclusively supplied with alabaster from NDM and not by sea from Spain via Aigues-Mortes (26). The only noteworthy Avignon exception is the funeral mon-

1), crossing the divides of three major river basins: the Rhone,

ument of Pope Urban V (1310-1370) for which we identify the Provencal alabaster of Malaucène as the source, through its highly distinctive isotope signature, strongly enriched in  ${}^{18}O$  [ $\delta^{18}O$ of  $\pm 18.8 \pm 0.4\%$  vs. standard mean ocean water (SMOW) n = 4, Figs. S1 and S2]. These quarries produced gypsum in the 13th century (27) and the sampled alabaster quarry is mentioned as early as 1458 (27). Malaucène also had a strong historical link with Avignon as it was the summer resort of Pope Clement V, and was situated much closer to the papal city than NDM (43 km compared with 240 km). However, despite the proximity and the availability of good quality of alabaster, NDM was the preferred source. One of the possible reasons is the ease of fluvial transport that was absent at Malaucène. Indeed, the role of the Rhone as principal vector for transporting bulk goods to the Papal Palace has been studied in detail (28). Land transport costs were estimated seven to nine times higher than river transport in the late Roman Empire and five times higher in the 18th century (29), so in medieval times, transport from NDM may have been cost-effective despite the five times longer distance compared with Malaucène.



**Fig. 3.**  $\delta^{34}$ S versus  ${}^{87}$ Sr/ ${}^{86}$ Sr ratios of alabaster artworks from French, American, Swedish, and English collections from 12th to 17th century, completed with nine samples from our pilot study (19). The data fields are delimited by 2 $\sigma$  confidence ellipses (95% confidence level).

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Fig. 4. Carrying of the Cross, alabaster, second quarter 14th century, Paris, Louvre Museum, Sculptures Department (France) - Inventory no. RF 3630. The material of this group was identified as from Notre-Dame-de-Mésage, Isère department, France.

Two other sculptures from our corpus are also related to Malaucène, a 14th century annunciation group initially situated in a rural church near Troyes in eastern France. This group has since been separated and the statue of Virgin Mary is conserved by the Louvre in Paris, and the Angel Gabriel by the Cleveland Museum of Arts in the United States. Our multiisotope method proves beyond reasonable doubt their common origin and raw material source. It also identifies a recent restoration, overlooked until now, where part of the base of the statue of the Virgin Mary was replaced by a visually identical but isotopically strongly contrasting material. The restoration alabaster came from the Volterra region in Tuscany, Italy, which was very active both in Etruscan and Roman antiquity and from the 18th to the early 20th centuries. However, this source has not been proved in any of the medieval and Renaissance carvings in western Europe so far analyzed, illustrating the supposed decline of the Volterra quarries until small-scale local use started again in the second half of the 16th century (30).

Our study demonstrates that isotope fingerprinting using S, O, and Sr can uniquely characterize historical sources of alabaster artwork, providing insight into previously unknown patterns of medieval stone trade and the affiliation of artworks to regional workshops or individual artists.

Even if the evidence on transport modes is indirect, our study confirms that fluvial (or marine) transport was generally privileged over terrestrial roads, possibly shorter. Nevertheless, the extension of the diffusion area of some sources (i.e., NDM) has required also the use of terrestrial transport to cross major river basin divides. This is also Cheetham's conclusion on the alabaster transport in England (14), who states, based on sparse historical records dating back to 1367, that "heavy materials were transported more commonly by road than is often supposed."

Our method benefits from the strong variations of isotope ratios of S, O, Sr in seawater and the associated evaporites through the Mesozoic (31, 32) and further age-independent contrasts related to varying continental influx and redox conditions in the evaporite basins. The large range of  $\delta^{34}$ S values of the alabaster deposits from +10.4‰ in the British Permian evaporites to +26.3‰ in the Italian Messinian deposits of Tuscany reflects mainly the evolution of seawater sulfate. Partial isolation of (sub)basins can lead to nonnegligible continental contribution to evaporite formation as shown for the East Midlands (33, 34), where it results in depletion of <sup>34</sup>S through sulfide

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oxidation and recycling of carboniferous evaporites and in more radiogenic <sup>87</sup>Sr/<sup>86</sup>Sr values due to the erosion of the surrounding massifs. Erosion of emerging zones of the crystalline basement would also explain the high <sup>87</sup>Sr/<sup>86</sup>Sr ratios (maximum 0.7096) of the Triassic Burundian deposits, which are well above the highest Mesozoic seawater values of 0.7092 (32).

The observed contrasts in signatures provide a means of forensically investigating restored artworks and forgeries. Extension of the technique to alabaster deposits from eastern Europe and the Mediterranean (35) and to artwork dating back to Antiquity seems promising, given the highly distinctive isotope signatures of Zechstein and Messinian evaporites and the major importance of gypsum alabaster in the art of ancient Mediterranean and Mesopotamian cultures.

ACKNOWLEDGMENTS. We thank the following curators, museums, collections, and galleries (in France unless stated otherwise) for providing samples and their representatives for their valuable contribution to discussions: Geneviève Bresc and Sophie Jugie from the Louvre Museum in Paris, Dominique Vingtain from the Petit Palais Museum in Avignon, Colleen Snyder from Cleveland Museum of Art (Ohio, USA), Martha Wolff and Cybele Tom from the Chicago Art Institute (Illinois, USA), Damien Berné from the Cluny National Museum of the Moyen-Age in Paris, Thierry Crépin-Leblond from the National Museum of

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the Renaissance in Ecouen. The Musée Rolin in Autun, the Musée départemental des Hautes-Alpes in Gap, the Musée des Beaux-arts in Chambéry, the Chartreuse of Villeneuve-lez-Avignon, and the Upplandsmuseet in Uppsala (Sweden) kindly provided access to artwork in their collections. Laurine Moreuil and Estelle Saint-Omer contributed to the discussions. The Townships of Cluny and Calais, and the Daniel Katz Gallery in London (UK) and the Chicago Art Institute (USA) provided both samples and financial support. British Gypsum Ltd/Saint Gobain kindly gave access and logistical support for sampling in their gypsum mine at Fauld in Staffordshire (UK). Agela Nestler and colleagues from the Geological Survey of Thüringen (Germany) collected samples of Permian alabaster from south of the Harz Mountains. Olivier Peyre provided precious historical material on the medieval gypsum production in Malaucène and Jacques Galas introduced us to the context of the Malaucène site. Philippe Bertone and Céline Laforest guided us to several other ancient extraction areas in Provence. We also thank Fabio Fratini (geologist, CNR Florence), Felice Tirabasso (quarry manager in Castellina Marittima for the Knauf company), and Gabriele Marasco in Volterra for their help collecting samples at historical alabaster extractions in Tuscany (Italy). Anthony Cooper publishes with permission of the Director of the British Geological Survey. This article was greatly improved by reviews by Jane Evans and Stephen Parry of the British Geological Survey, Rowena Stead of the French Geological Survey BRGM, and two anonymous reviewers. This study was cofunded by the Louvre Museum/Fondation des Science du Patrimoine, by the BRGM (Bureau de Recherches Géologiques et Minières), the Centre Interdisciplinaire de Conservation et Restauration du Patrimoine (CICRP), the Laboratoire de Recherche des Monuments Historiques (CRC-LRMH), and Idex Sorbonne Universités Pour l'Enseignement et la Recherche.

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