

Tracing Medieval and Renaissance alabaster artwork back to quarries: A multi-isotope (Sr, S, O) approach.

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ABSTRACT

Multi-isotope fingerprinting (sulphur, oxygen and strontium isotopes) has been tested to study the provenances of medieval and Renaissance French and Swedish alabaster artwork. Isotope signatures of historical English, French and Spanish alabaster source quarries or areas reveal highly specific, with a strong intra-group homogeneity and strong inter-group contrasts, especially for Sr and S isotopes. The chosen combination of isotope tracers is a good basis for forensic work on alabaster provenance allowing verification of hypotheses about historical trade routes as well as identification of fakes and their origin. The applied analytical techniques of continuous flow isotope ratio mass spectrometry (CF-IRMS) and thermal ionisation mass spectrometry (TIMS) only require micro-samples in the low mg range thus minimising the impact on artwork.

Keywords: alabaster, provenance, sculpture, multi-isotope tracing, forensics, sulphur, oxygen, strontium, isotopes.

Introduction

Gypsum alabaster ($\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$) and to a much lesser extent anhydrite alabaster (CaSO_4) have been used throughout history for artworks and as ornamental stone due to their fine-grained texture and colour. Alabaster is white in its pure form, often with Fe-rich brown veins, suggesting marble, but its softness makes it easier to work. When considering stone monuments, like other types of artefacts, it is crucial for art historians or museum conservators to correctly assign the provenance of the raw materials. This allows them to validate or invalidate hypotheses about historical trade routes based on iconographic or stylistic comparisons. The fact that it is easy to work makes alabaster a prime material for forgery and spectacular alabaster fakes are known, like the supposed Egyptian “Armana princess” (Hardwick 2009) and the “Assyrian” reliefs both forged by Shaun Greenhalgh of Bolton early this century and sold to reputable museums as genuine for considerable sums of money (http://en.wikipedia.org/wiki/Shawn_Greenhalgh). A thorough characterisation of the raw material of different provenances would help with forensic studies to recognise and backtrack such forgeries. Yet, in contrast to marble artwork, only a few studies have so far addressed the mineralogical, geochemical and isotopic fingerprints of the raw source alabaster compared to those of alabaster sculptures (e.g. Gale *et al.* 1988; Costagliola *et al.* 2001; Ligeza *et al.* 2001; Castro *et al.* 2008; Playà *et al.* 2012). It has been shown that sulphur and oxygen isotopes of gypsum plaster can be used to trace its origin (Usdowski 2001; Kloppmann *et al.* 2011), and this combination of isotopes together with $^{87}\text{Sr}/^{86}\text{Sr}$ ratios were used in the present pilot study of isotope fingerprinting undertaken on French and Swedish alabaster artworks. Significant contrasts between marine gypsum deposits of different ages and locations can be expected due to the systematic shifts of $\delta^{34}\text{S}$, $\delta^{18}\text{O}_{\text{SO}_4}$ and $^{87}\text{Sr}/^{86}\text{Sr}$ in seawater over the geological timescale (Burke *et al.* 1982; Claypool *et al.* 1980; Denison *et al.* 1998). These changes are preserved in the isotope signatures of evaporite layers formed from the seawater. Gale *et al.* (1988) were the first to combine strontium isotopes with sulphur isotopes to assess the provenance of Minoan Bronze Age alabaster artwork found in Crete, Thera and on the Greek mainland (Mycenae, Tiryns). The strong isotopic contrast of marine Mio-Pliocene gypsum deposits in central Crete with Triassic or Permo-Triassic evaporites from western Greece allows confirming the local origin of Cretan artwork and Cretan provenance for part of the mainland artworks (Mycenae).

Additional variations in S and O isotopes for a given formation can also be due to input from non-marine sulphur sources such as the oxidation of sulphides or presence of organic sulphur, which are typically depleted in ^{34}S with respect to marine sulphate. Recycling of older sulphate deposits by dissolution and redeposition causes further variation (Taylor 1983). Strontium can also be provided from silicate and carbonate weathering so that continental inputs can potentially mix with marine Sr in coastal evaporite basins. Strontium derived from Rb-containing silicate weathering will have higher $^{87}\text{Sr}/^{86}\text{Sr}$ ratios, as ^{87}Rb decay produces radiogenic ^{87}Sr .

Sulphur and oxygen are major components in calcium sulphates and Sr easily substitutes for Ca in the gypsum crystal lattice, or is found in the form of tiny celestite crystals (SrSO_4), so that microsamples (in the low mg range) are sufficient to perform the chosen isotope analyses (S and O by continuous flow isotope ratio mass spectrometry (CF-IRMS) and Sr by thermal ionization mass spectrometry (TIMS)). The small samples required considerably limit the impact on artwork. The present study investigates the possibility of linking alabaster artwork to source quarries or areas by using multi-isotope fingerprinting. The artwork studied came from French and Swedish museums and historical monuments. The choice was guided by whether or not conservators or art historians had already established relatively strong hypotheses on the provenance of the raw materials. The possible source samples were collected as close as possible to the historical quarries, in different regions of France, Northern Spain and England.

Materials and Methods

Sampling:

Nine samples of medieval and Renaissance sculpture fragments of different provenances in France (Burgundy, Lorraine, Roussillon, Pyrenees) and Sweden were analysed together with twenty one samples taken from or near ancient gypsum quarries (Fig. 1, Table 1, Table 2) from France (Jura, Alps, Provence, Burgundy, Lorraine), Spain (Aragon and Catalonia), England (Derbyshire and Staffordshire).

Ebro Valley (Aragon, Spain): The abundance of continental Miocene evaporites in the middle Ebro Valley (Perrier 2002; Utrilla *et al.* 1992) allowed the extraction of large

quantities of plaster-grade gypsum and alabaster for building stones and ornate carvings. These range from antiquity, including the Roman Caesaraugusta city (Lapuente *et al.* 2009) to the present. The development of alabaster artwork mainly from the 14th to 17th century was closely related to the extension of the Kingdom of Aragon and had its climax in the 16th century with monumental altarpieces in the cathedrals of Zaragoza, Huesca, Logroño, Velilla de Ebro, and Tarragona (Barral I Altet and Manote 1987; Perrier 2002).

Beuda (Girone, Catalonia, Spain): The approximately 1000 m thick marine evaporitic Beuda Formation made up of alternating shales and anhydrite was deposited in a restricted basin in the South Pyrenean foreland in the Eocene (upper Lutetian, Vergès *et al.*, 1992). The Beuda alabasters are among the most highly reputed materials for medieval sculpture, not only within the zone of influence of the crown of Aragon, but also north of the Pyrenees and as far as the kingdom of Naples (Español 2003). The first quarries were opened around 1327-28 and the material was used in Catalonia, Aragon, Valencia, French Roussillon and Bézier region as well as in Southern Italy. Alabaster from the Girona province was highly prized for its purity up to the middle of the 15th century (Español 2003).

Sarral (Tarragona, Catalonia, Spain): The lacustrine Eocene (Priabonian) gypsum of the southern margin of the Ebro basin (Ortí *et al.* 2007) was formed in shallow hypersaline lakes, fed by groundwater that had dissolved Mesozoic (Triassic and lower Jurassic) evaporites (Utrilla *et al.* 1992; Ortí *et al.* 2011). The Sarral deposits were used from the Middle Ages to present time (Ortí Iglesias 2005). During the 14th and 15th centuries, alabaster workers for the Aragonese crown seem to have preferred Beuda alabaster. However, the first sculpture workshops opened in the 16th century and from then on the Sarral alabaster was widely used (Ortí Iglesias 2005).

East Midlands (England): The calcareous Mercia Mudstone Group (formerly called the Keuper Marls^{*}) of the East Midlands contains several evaporite horizons that have been exploited for alabaster since medieval times (Cannan 2010; Young 1990). These include

* The lithostratigraphic term “Keuper” is strictly speaking only valid for the Germanic Trias north of the Alps and corresponds roughly to the chronostratigraphic term of the “Upper Triassic” (Carnian, Norian and Rhaetian stage), starting in fact in the Landinian (Middle Triassic). Given the scarcity of fossil remains in the Keuper sediments, it is difficult to link them precisely to the chronostratigraphic stages and we will, in the following, employ “Keuper” as lithostratigraphic term.

workings in the Tutbury Gypsum at Chellaston in Derbyshire and Fauld in Staffordshire, plus the Newark Gypsum from near Newark in Nottinghamshire; all three sites were sampled for our study. In this area gypsum beds were deposited in hypersaline lakes on a large peneplain with periodic marine influx (Taylor 1983). The first documented alabaster carvings from Tutbury go back as far as AD 1160-70 (Firman 1984). By the end of the 15th century, Nottingham had become an important centre of the alabaster industry and of the international trade in altarpieces, exported to Denmark, Holland, France, Spain, Italy (Firman 1964). A major factor in the decline of Nottingham alabaster workshops was the fact that in 1550 the English Parliament banished all religious representations, with the exception of tombs and effigies. Consequently, large quantities of alabaster artwork in England were destroyed or exported, mainly to France (Firman 1964; Edwards 1966). However, extraction continued and English alabaster was exported and used in workshops on the Continent including Mechelen in Belgium which was the veritable hub of the alabaster trade in Europe in the 16th to 17th centuries (Wustrack 1982; Boye Petersen and Boye Petersen 1999).

Lorraine (France): Upper Triassic gypsum and anhydrite deposits outcrop frequently in the Eastern Paris Basin, originating from a lagoonal/marine transgression, during the opening of the Germanic basin (Pomerol 1974). Even without historical and textual references the possibility of alabaster extraction in the Lorraine region exists. In the northern part of Lorraine, near Metz and Thionville, the Upper Keuper formation consists of red marls including gypsum and anhydrite layers or lenticular masses. The sampled Klang quarry was exploited for plaster during the 19th and 20th centuries, but could also be an alabaster source as it is located not far (15 km) from the Mosel River, which was an important transport route during Roman and medieval times, being a tributary of the Rhine. Other quarries, nearer to Nancy, exploit Upper Triassic formations, including a massive anhydrite unit associated with marls and halite.

Burgundy (France): The Upper Triassic gypsum outcrops at a few places in Burgundy, close to the western limits of the evaporitic Keuper basin. It was exploited both for plaster and as alabaster for sculpture. Two historical quarries have to be cited. The first one is located at Berzé-la-Ville (Saône-et-Loire), where gypsum alternates with clay deposits, attributed to lower Keuper. This quarry, located near Cluny, was under control of Cluny abbey and used for the Jacques d'Amboise palace at the beginning of the 16th century. The alabaster of Cluny was widely known, according the correspondence of 1510 between Marguerite d'Autriche

and Jean Lemaire (Poiret 2003). The second one is the Mâlain quarry, sampled for our study, located at 20 km from the west of Dijon (Côte-d'Or), in a Carnian (and/or Norian) clay and dolomite formation. It was possibly mainly exploited for plaster, but may have been an alabaster source as early as Roman times (1st century), when the city was called Mediolanum (Roussel 1979)

Jura (France): In the French Jura, marine Upper Keuper gypsum and anhydrite layers alternate with marls, dolomites and sandstones. Most of the gypsum quarries which provided alabaster are located in an allochthonous series that was deformed, but remained unmetamorphosed. Gypsum and anhydrite have been extracted for sculpture mainly at Salins-les Bains, which was sampled for our study at the Saint-Lothain and Poligny quarries. Saint-Lothain is attested as a source by textual references (Poiret 2003) for the construction of the rood screen, altarpiece, tombs (delivered in 1512) and architectural ornamentation of the Royal Monastery of Brou (Bourg-en-Bresse, France). Daville (1936) also suspects Roman activity in this quarry.

French Alps: Massive Triassic gypsum and anhydrite layers have been and are still exploited on the French side of the Alps. These marine evaporites were tectonically overprinted during the Alpine orogeny (Miocene), and more or less metamorphosed. Two alabaster workings are notable and have been sampled for this study. The Notre-Dame-de-Mésage quarry, near Grenoble, is well known and has been exploited in modern times, probably mainly during the 19th century (Debelmas 1990). However earlier uses, even for export, are suspected. Alabaster from this quarry is supposed to have been used in 1519-1520 for three tombs produced by Martin Claustre, sculptor in Grenoble, for the chapel of castle of Thouars (in western France) at the behest of Louis II de la Trémoille (Marchegay 1876; Reymond 1907). There are also quarries in the Maurienne valley, analysed in our study, which are nowadays exploited for gypsum plaster, where it is possible to obtain the more or less coarse grained variety of alabaster. In the past, this alabaster was used locally in architecture and for the ornamentation and sculpture in the cathedral of Saint-Jean-de-Maurienne.

Provence (France): Has alabasters of similar age and overall geological context to those from Provence and the southern French Alps. In Provence, Triassic gypsum horizons also formed diapirs during Jurassic and Eocene/Oligocene extensional phases along main regional

faults. The sampled Beumes-de-Venise alabaster belongs to a Triassic gypsum diapir that rose during the Rupelian (Upper Oligocene) along the Nîmes fault. Gypsum has been extracted from tens of outcrops for plaster production. Alabaster is present in small amounts and it was used locally for vernacular sculptures until the 19th Century (Guiomar 2005). It could have played a role in the manufacture of decorative sculptures when the papacy was settled in Avignon and later when King René reigned over Provence in the 14th and 15th centuries.

location of Figure 1

The provenance and other background information for the investigated artworks are provided in Table 2 and Figure 1.

Uppsala Cathedral, Sweden Gustav Vasa monument: This alabaster monument, carved in the late 16th century by the Flemish sculptor Willem Boy (around 1520-1592) carries four small obelisks, one of which was damaged by a fire in the cathedral in 1702. The remnants, sampled for our study, have been preserved, after the obelisk was replaced by a plaster version in the 1890s. In a letter dated June 11th 1572 it is stated that Willem Boy was leaving Antwerp to go to England to get alabaster for the rest of the Vasa monument after having finished the statues of the king and his two first wives in 1571. The complete monument was delivered to Uppsala in the 1570s, but because of a fire in the cathedral it remained unfinished until 1583 when the inscriptions were carved.

Oudart de Bournonville Tomb (Museum of Fine Arts, Arras, France): Fragments of this funeral monument from northern France were discovered in 1840 in the church St-Martin of Hénin-Beaumont in the Pas-de-Calais and are now conserved in Arras Fine Art Museum. It was sculpted in 1585 from alabaster that was probably reused from a previous monument. A Nottingham provenance was supposed by conservators because of painting traces on the sheltered side, indicating a first use that has not been determined. The design of these paint remains are typically of the "Nottingham fashion" (Sautereau 2008).

Sculpted facade of Jacques d'Amboise Palace (Cluny, Saône et Loire, France): The facades of the two towers of this palace, constructed at the end of the 15th century for Jacques d'Amboise, abbot of Cluny from 1485 to 1510, are entirely ornamented with alabaster sculptures. The raw material is likely to come from the close-by Berzé-la-ville quarries on the Abbey territory.

“Retable de la Déploration” altarpiece (Nancy, France): This altarpiece, conserved in the Musée Lorrain in Nancy (eastern France), was sculpted in the 16th century. According to the museum inventory, the sculptural origin of this piece is unknown, but it is stylistically attributed to a south Netherlands workshop.

Statue of St Antoine - Jube of Royal Monastery of Brou (Bourg-en-Bresse, France): The church of the Royal Monastery of Brou was built between 1513 and 1532 for Margaret of Austria. It hosts seven statues, formerly part of the rood screen, including the statue of St Antoine which like five others is made from alabaster that is attributed to the Saint Lothain quarries (Briat-Philippe 2006; Sarda 2006). This location is situated 100 km north of Bourg-en-Bresse and 30 km from the sampled quarries in Salins-les-Bains.

Statue of Notre-Dame de Bethléem (Narbonne, France): This statue, 180 cm in height, is part of the sculptural decoration of a chapel of Narbonne Cathedral to which it has conferred its name. Both in style and material it is different from the rest of the sculptural content of the chapel, even though it is considered as contemporary (3rd quarter of the 14th century; Pradalier-Schlumberger 1998).

High altar of Perpignan Cathedral (France): The high altar of the Gothic cathedral was carved over a long period (1573-1631) by two successive workshops (Claudio Parret and later Jordi Lleonart) using a white alabaster very likely from Beuda, Catalonia (Hernandez 2002).

Analyses:

The isotopic composition of sulphur is expressed in the usual delta notation as a per mil (‰) deviation of the heavy-to-light isotope abundance ratio ($^{34}\text{S}/^{32}\text{S}$, $^{18}\text{O}/^{16}\text{O}$) in the sample ($\delta^{34}\text{S}$), with respect to the Canyon Diablo troilite (CDT) standard. Oxygen isotopes are reported as $\delta^{18}\text{O}$ with respect to the standard mean ocean water (SMOW) standard. Strontium isotopes are reported as $^{87}\text{Sr}/^{86}\text{Sr}$ ratios.

The samples for sulphur and oxygen isotopes were crushed and shaken for 72 h in contact with 100 ml of Millipore® distilled water adapting the Italian standard method for determining soluble salts in stone samples (NORMAL 1983). Sulphates were precipitated as BaSO_4 from the filtered solution by adding BaCl_2 solution. The precipitate was then filtered

off and left to dry and a fraction ($\approx 350 \mu\text{g}$) of BaSO_4 was mixed with vanadium pentoxide in a tin capsule (Giesemann *et al.* 1994), injected in a flash combustion elemental analyzer (Flash EA) where BaSO_4 is reduced to SO_2 at $1700\text{-}1800^\circ\text{C}$. The purified SO_2 is analysed for S isotopes by a continuous flow isotope ratio mass spectrometer (CF-IRMS: Thermo Delta Plus XP). An aliquot of the BaSO_4 ($\approx 200\mu\text{g}$) is placed in a silver capsule, injected in a high temperature conversion elemental analyzer (TC/EA) reactor with a graphite insert at 1450°C . The resulting CO is analysed by CF-IRMS for oxygen isotopes. Sulphur and oxygen isotopes are always measured twice. We also tested direct injection of the $\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$ into the Flash EA and TC/EA units as an alternative to BaSO_4 precipitation. Whereas sulphur isotope ratios are very coherent for both methods ($\delta^{34}\text{S}_{\text{direct}} = 1.07$, $\delta^{34}\text{S}_{\text{BaSO}_4} = 1.27$, $R^2 = 0.954$ for $n=22$ $\delta^{34}\text{S}$ values determined on the same samples), oxygen isotopes obtained by direct introduction of gypsum show a systematic shift towards lower $\delta^{18}\text{O}$ values. This is probably due to the isotope exchange of crystal water oxygen with SO_2 ; we consider that this method is unreliable for oxygen isotopes except probably for anhydrite samples. Analytical uncertainty is estimated at 0.4‰ for both $\delta^{34}\text{S}$ and $\delta^{18}\text{O}$. A theoretical minimum requirement of 5 mg of BaSO_4 for quadruple analysis corresponds to 3.7 mg of gypsum sample. In practice, to take into account precipitate losses on filters and to be able to repeat the analysis in case of problems, around 7 mg of gypsum should be available as a minimum.

Chemical purification of Sr was performed using an ion-exchange column (Sr-Spec) before mass analysis according to a method adapted from Pin and Bassin (1992), with total blank < 1 ng for the entire chemical procedure. After chemical separation, around 150 ng of Sr was loaded onto a tungsten filament with a tantalum activator and analysed with a Finnigan MAT262 multi-collector thermal ionisation mass spectrometer (TIMS). The measured $^{87}\text{Sr}/^{86}\text{Sr}$ ratios were normalised to an $^{88}\text{Sr}/^{86}\text{Sr}$ of 0.1194 and then adjusted to the NBS987 standard value of 0.710240. An average internal precision of $\pm 10 \times 10^{-6}$ ($2\sigma_m$) was currently obtained during this study and the reproducibility of the $^{87}\text{Sr}/^{86}\text{Sr}$ ratio measurements was tested through repeated analyses of the NBS 987 standard for which we obtained a mean value of $0.710230 \pm 9 \times 10^{-6}$ (2σ ; $n = 12$) during the period of analysis. Depending on the Sr content of the samples (from 3500 to 290 mg/kg), between 1 and 5 mg of alabaster would be sufficient for Sr isotope analysis, but since the results are not known in advance a 5 mg sample should be considered as a minimum.

Results and Discussion

Quarry samples

The $\delta^{34}\text{S}$ values of all the French samples from the Alps and the alpine foreland (Provence, Jura) fall in a narrow range of +15.9 to +17.0 ‰ vs. CDT, whereas $\delta^{18}\text{O}$ values are much more variable for this group (+11.2 to +17.5 ‰ vs. SMOW) falling in the typical range for Keuper evaporites (Fig. 1). Samples from Côte d'Or (F, Burgundy) and Sarral (E, Catalonia) plot in the same characteristic range as samples of Upper Triassic age (Pearson *et al.* 1991). Catalan samples from Beuda are significantly enriched in ^{34}S with a $\delta^{34}\text{S}$ around +20 ‰ with the lowest $\delta^{18}\text{O}$ values of the data set (+11.3 to +11.8 ‰) similar to values reported for the same site by Inglés *et al.* (2009) and Utrilla *et al.* (1992); in comparison, Spanish samples from Aragon and French samples from Lorraine are depleted in ^{34}S ($\delta^{34}\text{S}$ of +10.4 to +14.5 ‰). The English samples of Triassic Chellaston and Tutbury gypsum fall within the previously published $\delta^{34}\text{S}$ range of +12 to +14 ‰ (Taylor 1983).

$^{87}\text{Sr}/^{86}\text{Sr}$ ratios for raw alabaster vary significantly from 0.7076 to 0.7096 and allow the discrimination of French Alps alabaster *sensu lato* from the Upper Triassic East Midlands alabaster which has a much more radiogenic and very well defined signature (0.709151 ± 0.000058 , $n=5$). This is again very similar to the previous work of Taylor (1982) on the Tutbury and Fauld gypsum deposits which had a mean value of 0.709156 ± 0.000060 ($n=8$) and no significant difference between white parts and brown veins. The Spanish Catalan marine Eocene alabaster from Beuda has a value of 0.70781 and a very low variation (± 0.000013 , $n=3$) and falls in the range of Triassic gypsum from French Alps and foreland, but can be discriminated by higher $\delta^{34}\text{S}$ values. This is contrary to the continental Eocene gypsum of Sarral, which is indistinguishable from the French Keuper samples, a sign of the recycling of Triassic evaporites into the Eocene. The Ebro basin Miocene gypsum shows a relatively wide range both for $\delta^{34}\text{S}$ and $^{87}\text{Sr}/^{86}\text{Sr}$ (Fig. 3). The depletion in ^{34}S of the continental Miocene gypsum of the central Ebro basin has been explained by S reduction in organic-rich lake sediments and subsequent oxidation (Utrilla *et al.* 1992).

The East Midlands samples and the Mâlain Burgundy samples contain much more radiogenic ^{87}Sr than upper Triassic (Keuper) marine deposits (Burke *et al.* 1982). Here, continental influence has to be suspected with significant input from silicate weathering. The

highest $^{87}\text{Sr}/^{86}\text{Sr}$ ratio of 0.709614 was observed for the Upper Triassic (Upper Keuper) Mâlain sample from Burgundy.

Combining $\delta^{34}\text{S}$ and $^{87}\text{Sr}/^{86}\text{Sr}$ leads to a strong discrimination (Fig. 3) with well-defined and clearly distinguished fields, partly due to the variations over time of seawater signals, partly due to the contribution of non-marine sources, especially for strontium. Strontium concentrations are much less discriminating as local variations can be quite important (e.g. by a factor of 3 for the Beuda quarry and by a factor of 6 for East Midlands (Nottingham) alabasters) whereas $^{87}\text{Sr}/^{86}\text{Sr}$ ratios are very homogeneous for each site.

Artwork

The studied artwork from Roussillon region in southern France (Narbonne (NDB) and Perpignan (PER-1 and PER-B) cathedrals), shows a clear Beuda (Catalonia)-type signature for all three isotope systems; this attribution was also suspected on the basis of stylistic and historical analyses (Hernandez 2002). No alabaster was available to quarry in the Roussillon area and the raw material had to be imported from the other side of the Pyrenees, first by ox-cart for over 50 km to the Roses (Girona) harbour, then onwards by ship (Hernandez 2002).

An English origin was confirmed for the tombstone sculpture of Oudart de Bournonville (TOB) and for the funeral monument of Gustav Vasa of Uppsala (UPPS) in Sweden. Both funeral monuments (1570 and 1562-83 respectively) fall in the period after 1550 when the manufacture of images was forbidden by the English Parliament. Even if memorial tombs were still allowed to be made, it was mainly the raw material rather than carved alabaster that was now exported. This continued a long tradition of commerce in uncarved alabaster with France that is noted as early as 1414 (Edwards 1966) and which also took place with other countries. At this time, Mechelen (Belgium, Figure 1) had become an important centre for alabaster sculpture under the patronage of the Duchess of Burgundy, Margaret of Austria (Boye-Petersen and Boye-Petersen 1999). The local workshops here used raw materials from different origins including Alsace-Lorraine (France) the Harz mountains (Germany) and England (Wustrack 1982). The sculptor of the Gustav Vasa funerary monument, Willem Boy, worked in Mechelen from 1562-65 through 1566-68 and between 1570-75 (Bengtsson 2010), it is therefore likely that the sculpture transited via the Mechelen workshop.

A local origin is also possible for the analysed artwork from Burgundy and Jura. The statue of St Antoine from Bourg-en-Bresse (STA) shows isotopic similarities to the Jura gypsum, as expected given the textual sources indicating Saint-Lothain quarry as its origin. However, alabaster from the Palais Jacques d'Amboise (PJA) at Cluny in the Burgundy region is quite enriched in ^{87}Sr ; this is also a feature that characterises the sample from Mâlain (Burgundy), even if the original quarry is most probably that of Berzé-la-Ville, located at less than 10km.

No clear hypothesis can be formulated on the provenance of the alabaster used for the Retable de la Déploration (RDD) altarpiece conserved in Nancy (France). $\delta^{18}\text{O}$ and $\delta^{34}\text{S}$ values indicate a Keuper deposit, which could be affiliated to the East Midlands quarries, but $^{87}\text{Sr}/^{86}\text{Sr}$ ratios seem too low. In the present study, the only analysed Lorraine alabaster deposit is from Klang (Moselle), which shows a rather different isotope signature, much more depleted in ^{34}S and with a lower $^{87}\text{Sr}/^{86}\text{Sr}$ ratio (Fig. 3). Here, clearly, more data on regional and supra-regional historical alabaster quarry sources are needed to establish the multi-isotope method as an operational tool for alabaster provenance studies

Conclusions

The chosen combination of isotope tracers (S, O, Sr) is proving promising for forensic work on alabaster provenance in Europe for several reasons:

- The elements S and O are major constituents of calcium sulphate and are associated with Sr as a ubiquitous minor compound. This fact together with the chosen analytical techniques (continuous flow IRMS, TIMS) permits the use of microsamples in the low mg range ensuring that damage to sculptures is minimal.
- Both S and Sr show high intra-group homogeneity and strong inter-group contrasts, *conditio sine qua non* for forensic work on artwork provenance. The oxygen isotopes are more variable for most of the groups and show some overlap for different deposits.
- The low analytical errors on all the parameters might allow for further refinement, in particular within the groups that have large geographic extents such as the French alpine and peri-alpine Keuper evaporite province.

Our study confirms that, even if local to regional supply was used whenever possible, as demonstrated by the examples from French Jura and Burgundy, renowned sources of Renaissance alabaster had international dissemination (Fig. 1). The Beuda quarries are

proved to have delivered raw material to the French regions north of the Pyrenees; trade with the Kingdom of Naples is testified for the middle of the 15th century (Espanol 2003); English quarries in the larger vicinity of the important Nottingham workshop exported alabaster, directly or indirectly, as far as Sweden, Iceland (Reykjavik) and Spain (Santiago de Compostela) (Edwards 1966).

Despite this work, the database on samples from historical quarries needs to be considerably enlarged to take into account the variability of raw materials from Europe and the Mediterranean basin (Woods 2010). It also needs to be extended to extra-European sources especially when aiming to backtrack forgery. It is likely that some overlap of isotope signatures cannot be avoided, especially within the widespread Messinian evaporites that are present from Tuscany to Sicily (Costagliola *et al.* 2001). Other sources of medieval and Renaissance alabaster artwork that need considering in future research include the Permian (Zechstein) evaporites from the periphery of the German Harz mountains and from Lower Silesia (Poland); Polish Miocene gypsum from Podolia (present Ukraine) quarried from 1560 onward; Belgian alabaster and English deposits in Somerset and Cumbria.

Given the need to track down and exclude modern alabaster forgeries, such as the faked "Assyrian" carvings that were probably made with imported Italian alabaster, a worldwide isotope database of ancient and modern alabaster sources would be invaluable.

location of Tables 1 and 2

location of Figure 2

location of Figure 7

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Table 1

	Sample	Provenance	Quarry/Situation	Geological age	Legend	$^{87}\text{Sr}/^{86}\text{Sr}$	$2\sigma(m)$ $^{87}\text{Sr}/^{86}\text{Sr}$	$\delta^{34}\text{S}$ (‰ vs. CDT)	$\delta^{18}\text{O}_{\text{SO}_4}$ (‰ vs. SMOW)	Sr mg/kg)
France	39-02 Zone blanche	Salins-les-Bains, Jura	quarry in the town centre (46°57'00" N - 05°52'45"E)	Trias, upper Keuper	F-Jura1	0,708099	0,000010	16,3	13,1	696
	39-02 Zone veinée	Salins-les-Bains, Jura	quarry in the town centre (46°57'00" N - 05°52'45"E)	Trias, upper Keuper	F-Jura2	0,708104	0,000008	16,4	14,0	848
	39-14a	Salins-les-Bains, Jura	quarry « Le Boisset » (46°54'35"N - 05°53'48"E)	Trias, upper Keuper	F-Jura3	0,708030	0,000008	16,4	14,2	1170
	73-06a	Saint-Jean-de-Maurienne, Savoie, Alps	quarry « Les Rossières » (45°15'35" N - 06°19'15"E)	Trias	F-Maur1	0,707682	0,000008	16,1	11,7	1414
	73-06c	Saint-Jean-de-Maurienne, Savoie, Alps	quarry « Les Rossières » (45°15'35" N - 06°19'15"E)	Trias	F-Maur2	0,707655	0,000010	16,3	11,2	1700
	Mâlain	Mâlain, Côte d'Or, Burgundy	quarry (47°20'00"N - 04°47'25"E)	Trias, Carnian or Norian	F-Mala1	0,709614	0,000008	15,9	13,7	794
	84-70a	Beaumes-de-Venise, Vaucluse, Provence	outcrop domaine d'Urban (44°07'48"N - 05°01'35"E)	Trias	F-Beau1	0,707680	0,000008	16,7	17,5	458
	84-70b	Beaumes-de-Venise, Vaucluse, Provence	outcrop domaine d'Urban (44°07'48"N - 05°01'35"E)	Trias	F-Beau2	0,707959	0,000008	17,0	14,0	3522
	38-32	Notre-Dame-de-Mésage, Isère, Alps	quarry (45°04'20"N - 05°44'35"E)	Trias, Keuper	F-Mésa1	0,707835	0,000009	16,0	12,7	1840
	57-17	Klang-Moselle, Lorraine	quarry (49°19'15"N - 06°21'40"E)	Trias, Upper Keuper	F-Mose1	0,708418	0,000008	10.4	15,7	958

Spain	Quinto de Ebro	Quinto de Ebro, Aragon		Miocene	E-QEbr1	0,707939	0,000008	14.5	14,5	926
	Fuentes de Ebro	Fuentes de Ebro, Aragon	quarry (41°30'14.91" N-0°37'49.71"W)	Miocene	E-FEbr1	0,708510	0,000008	11.1	16,0	494
	Sarral	Sarral, Catalogne	quarry (41°26'31.64"N-1°13'15.56"E)	Eocene	E-Sarra1	0,707886	0,000008	16.5	14,7	2782
	Beuda 1	Beuda, Catalogne	quarry (42°14'12.01"N - 2°44'02.35"E)	Eocene	E-Beud1	0,707794	0,000008	19.8	11,3	2792
	Beuda 2	Beuda, Catalogne	quarry (42°14'12.01"N - 2°44'02.35"E)	Eocene	E-Beud2	0,707781	0,000008	20.2	11,8	982
	Beuda 3	Beuda, Catalogne	42°14'12.01"N - 2°44'02.35"E	Eocene	E-Beud3	0,707769	0,000009	19.8	11,6	836
England	Chellaston	Chellaston, Derbyshire	outcrop close to the Medieval quarry (52°52'21.39"N-1°25'36.03"O)	Trias, Keuper	GB-Nott1	0,709209	0,000009	13.7	14,2	290
	Triassic 3	Newark Nottinghamshire	Kilvington quarry, 52°58'51.65"N 0°48'44.12"O	Trias, Keuper	GB-Nott2	0,709058	0,000007	14.0	14,1	944
	Tutbury 6	Fauld Mine, Staffordshire	Fauld Mine Staffs. White parts 52°50'37.85"N 1°44'16.94"O	Trias, Keuper	GB-Nott3a	0,709182	0,000007	13.3	11,7	1507
	Tutbury 6	Fauld Mine, Staffordshire	Fauld Mine Staffs . Brown veins 52°50'37.85"N 1°44'16.94"O	Trias, Keuper	GB-Nott3b	0,709134	0,000007	13.2	11,1	1655
	Tutbury 7	Fauld Mine, Staffordshire	Fauld Mine Staffs .52°50'37.85"N 1°44'16.94"O	Trias, Keuper	GB-Nott4	0,709170	0,000006	13,6	11,8	961

Table 2

Country	Sample	Artwork / current location	Original monument/ Situation	Legend	$^{87}\text{Sr}/^{86}\text{Sr}$	$2\sigma(m)$ $^{87}\text{Sr}/^{86}\text{Sr}$	$\delta^{34}\text{S}$ (‰) vs. CDT)	$\delta^{18}\text{O}_{\text{SO}_4}$ (‰) vs. SMOW)	Sr mg/kg)
France	TOB Lion 4	Oudart de Bournonville Tomb - Museum of Fine Arts, Arras, France	Church St-Martin de Hénin-Beaumont, Pas-de-Calais, France (50°25' 15" N, 02° 56' 54" E)	TOB-4	0.709204	0.000008	13,2	13,7	992
	TOB Fragment C	Oudart de Bournonville Tomb -Museum of Fine Arts, Arras, Pas-de Calais,France	Church St-Martin de Hénin-Beaumont, Pas-de-Calais, France (50°25' 15" N, 02° 56' 54" E)	TOB-C	0.709179	0.000009	13,12*	nd	921
	71-09	Sculpted facade of Jacques d'Amboise Palace-Cluny ,Saône et Loire; France	Jacques d'Amboise Palace, Cluny, Saône et Loire, France 46° 26' 02" N, 04 39' 33" E)	PJA	0.709279	0.000008	14,9	13,2	1064
	Inv. 61-9-13	Retable de la Déploration alarpiece	Musée historique Lorrain, Nancy, France (48°41' 49" N, 06° 10' 47" E)	RDD	0.708690	0.000008	13,1	14,2	1050
	Bg. B Jub. Ant.	Statue of St Antoine - Jube of Royal Monastery of Brou, Bourg-en-Bresse, Ain, France	Jube of Royal Monastery of Brou, Bourg-en-Bresse, Ain, France (46° 11' 53" N, 05° 14' 11" E)	STA	0.708008	0.000008	15,2*	nd	1116
	11-49a	Statue of ND de Bethléem - Chapel ND de Bethléem, Cathedral of Narbonne, Aude, France	Chapel ND de Bethléem, Cathedral of Narbonne, Aude, France (43° 11' 05" N, 03° 00' 05" E)	NDB	0.707772	0.000007	20,3*	13,4	964

	PER-1	Altarpiece, of Perpignan Cathedral - Pyrénées-Orientales, France	Choir of cathedral, Perpignan, Pyrénées-Orientales, France (42° 42' 2.51" N et 2° 53' 50.43" E)	PER-1	0.707768	0.000007	20,1	11,1	834
	PER-B	Altarpiece of Perpignan Cathedral - Pyrénées-Orientales, France	Choir of cathedral, Perpignan, Pyrénées-Orientales, France (42° 42' 2.51" N et 2° 53' 50.43" E)	PER-B	0.707789	0.000009	20,0	10,4	915
Sweden	UPPS1	Gustav Vasa monument, fragments of broken obelisk - Uppsala Cathedral, Sweden	59°51'29.49"N 17°38'3.54"E Uppsala Cathedral, Sweden	UPPS	0.709205	0.000008	13,5	12,7	1153

* $\delta^{34}\text{S}$ obtained by direct reduction of gypsum

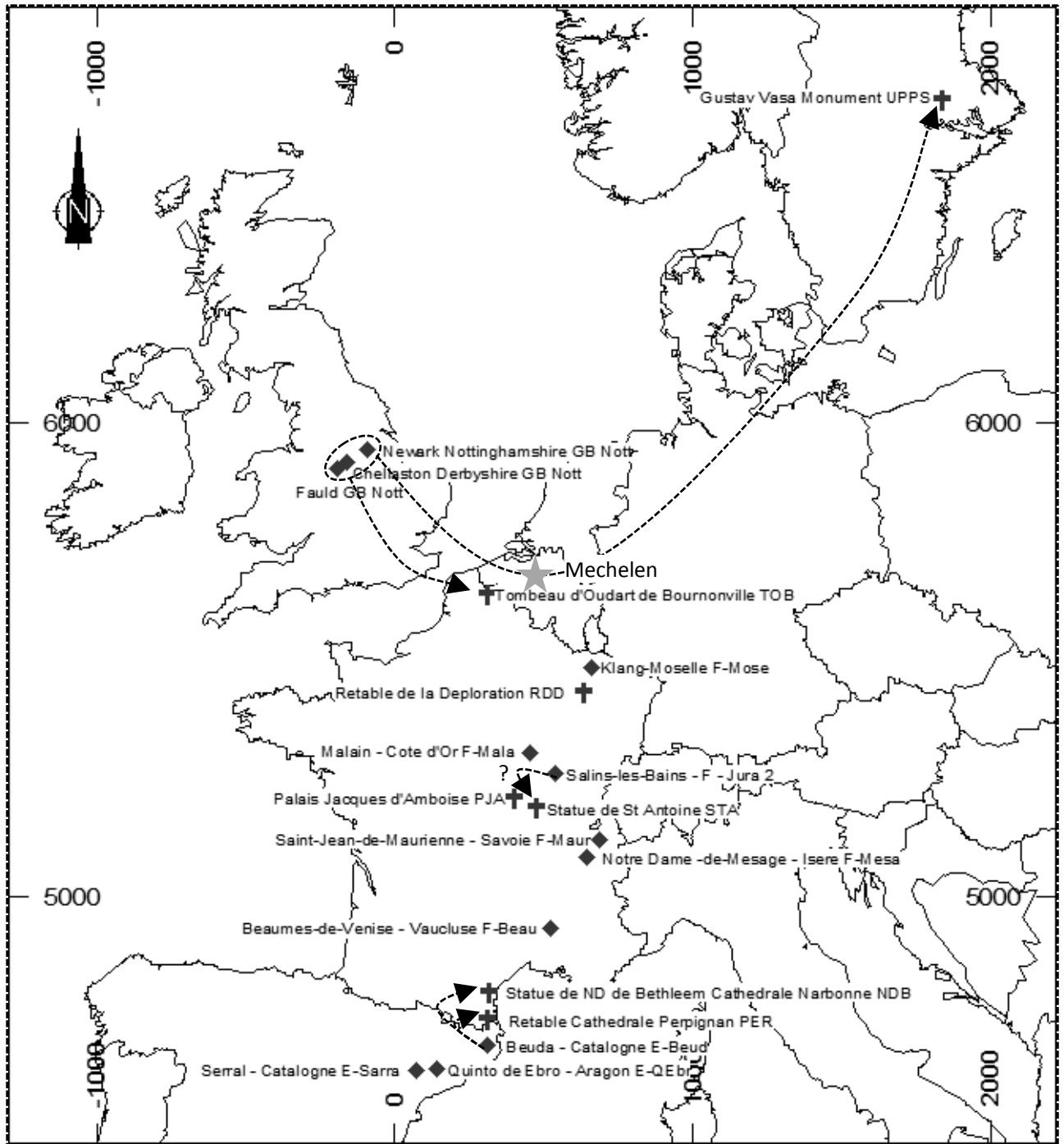


Figure 1 Locations of the investigated historical alabaster exploitations (◆) and alabaster artwork (⚡), arrows indicate the possible links between investigated historical alabaster exploitations and artwork as concluded from this study.

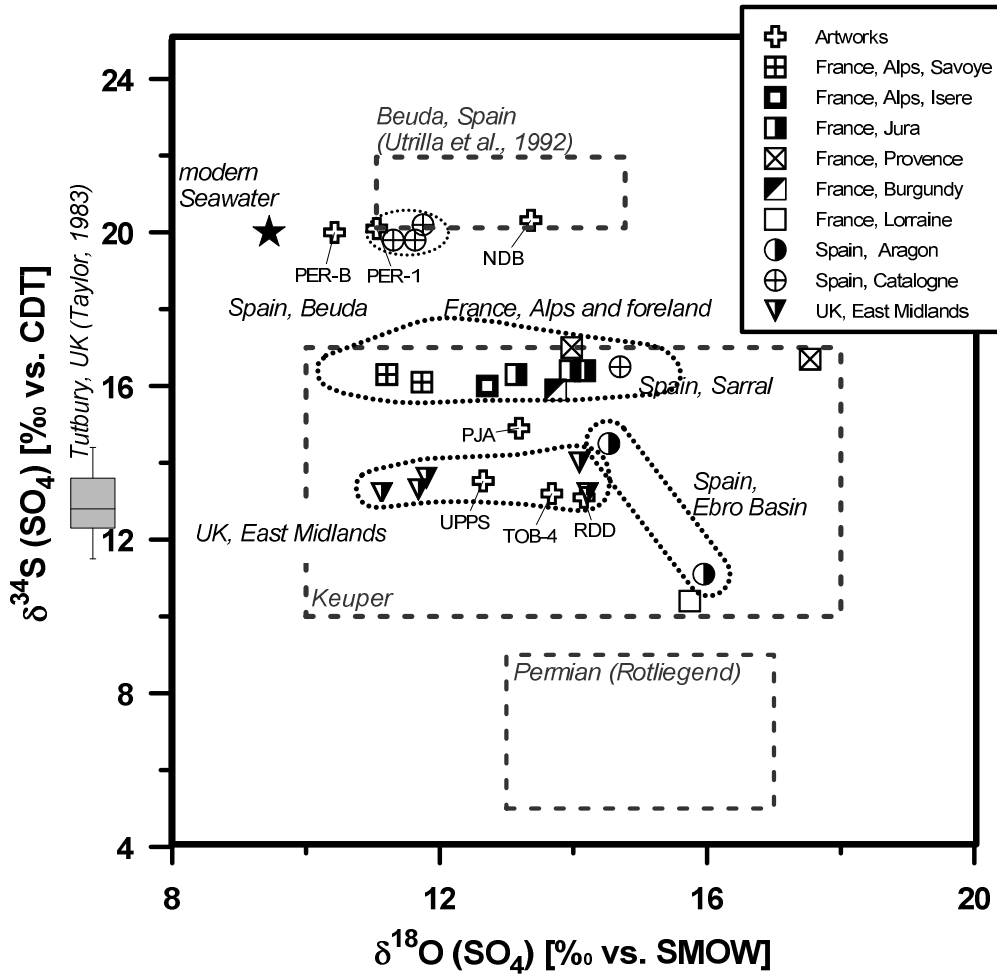


Figure 2 : Sulphur and oxygen isotope ratios of alabaster quarries and artwork. For comparison: Keuper and continental Permian signatures (Pearson *et al.* 1991), Beuda quarries (Utrilla *et al.* 1992).

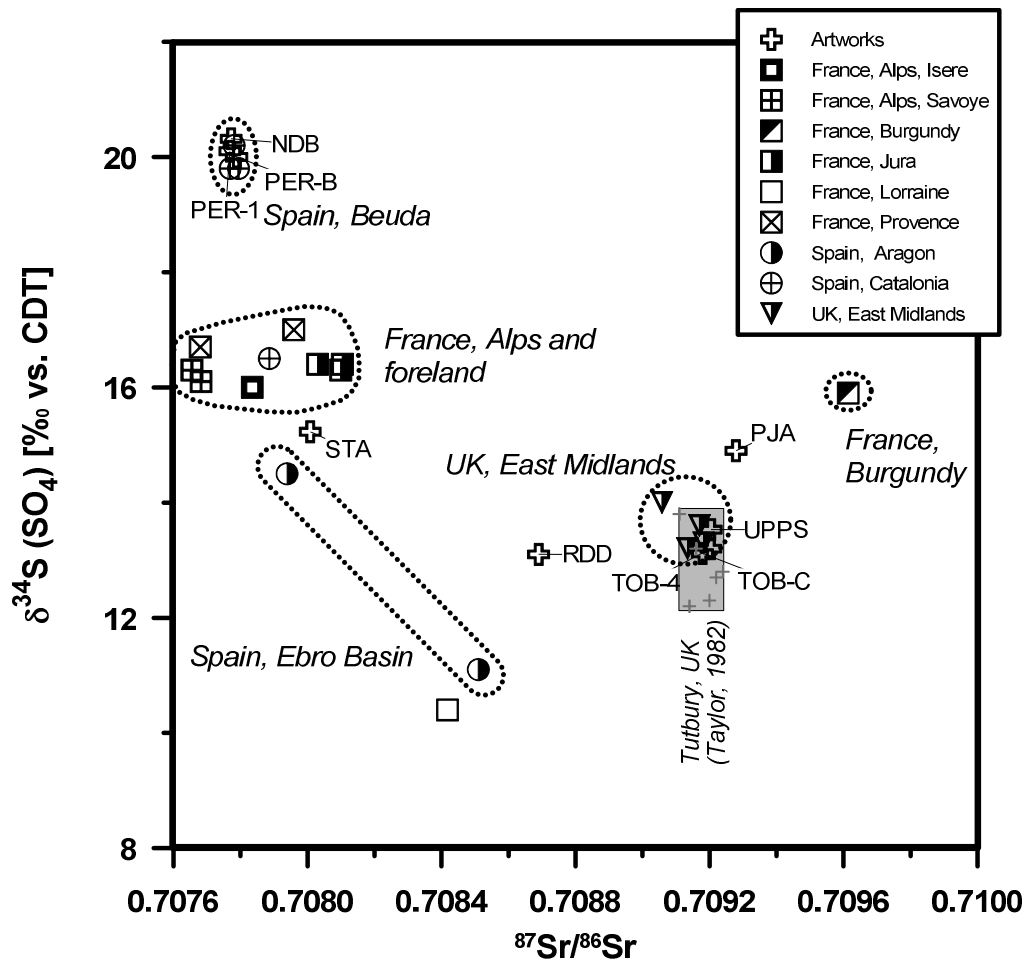


Figure 3 : Sulphur and strontium isotope ratios of alabaster quarries and artwork. For comparison: isotope signatures of Tutbury gypsum (Taylor 1982)

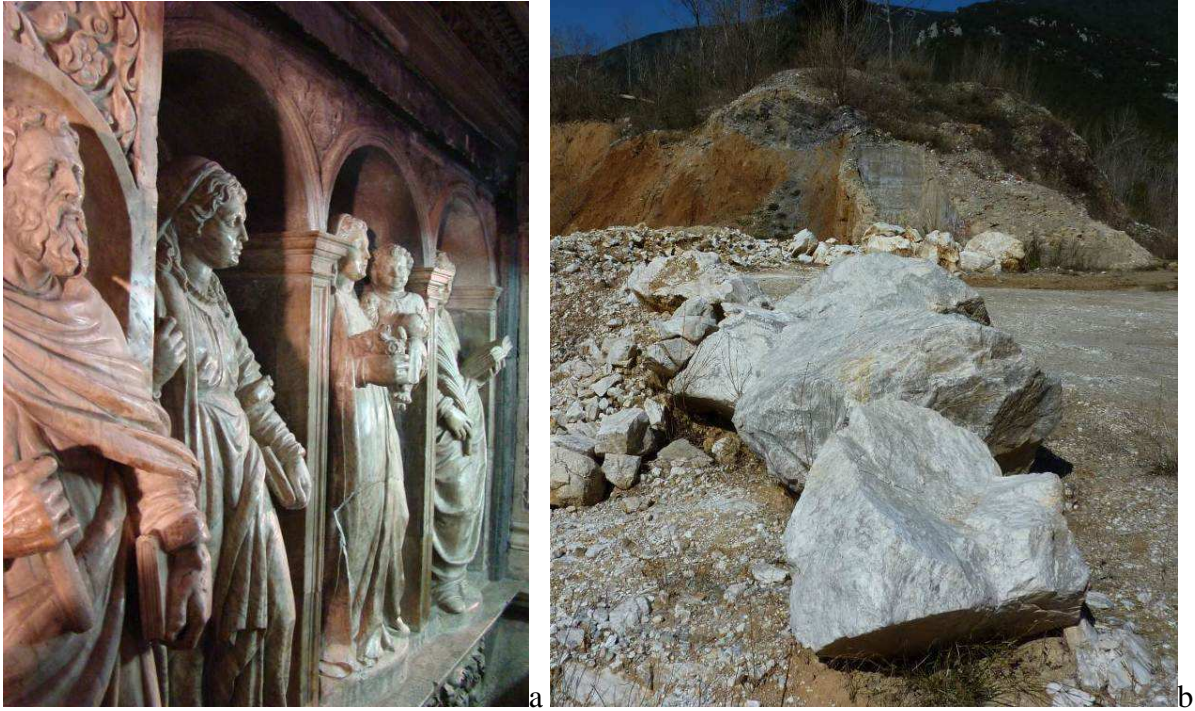


Figure 4 (a) High Altar of the Perpignan Cathedral, 1573-1690. 4 (b) Alabaster blocks in the Beuda quarry, Catalonia, Spain.

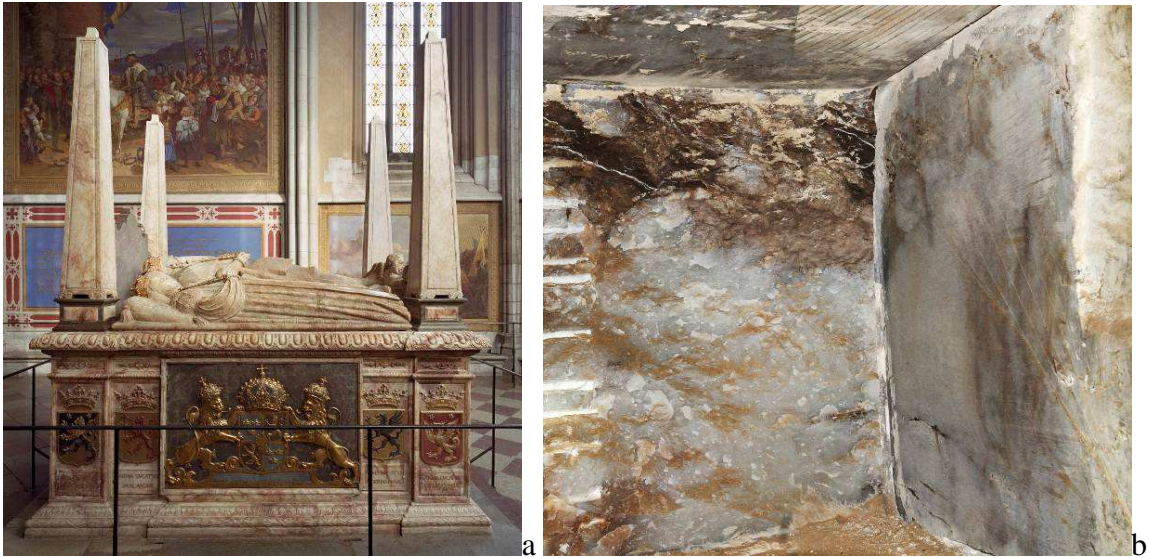


Figure 5 (a) Funeral Monument of King Gustav Vasa of Sweden, Uppsala Cathedral 1562-1583. The discussed replaced obelisk is the second from the right; 5 (b) Alabaster extraction underground at Fauld Mine from the Tutbury Seam, East Midlands (UK).