

Roman glass across the Empire: an elemental and isotopic characterization

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Abstract

This study focuses on natron glass, excavated in different areas of the Roman Empire, dated from the 1st to 5th century AD. The far-east side of the Roman Empire is accounted for by three archaeological sites, Petra and Barsinia, both in Jordan, and Gonio, in Georgia. The Italian peninsula is represented by the Iulia Felix and Embiez shipwrecks, and by samples from Augusta Praetoria. Samples from Sant Boi de Llobregat in Spain are examples of the western side of the Empire, and the sites of Tienen and Oudenburg represent the northern provinces.

No clear distinctions in glass composition and origin between the different origins in the Roman Empire can be made based on the major element composition. However, the western, central and eastern Roman Empire samples can be divided into two groups, coloured and colourless, reflected in the K₂O contents. Sr-Nd isotopes, used for provenancing geological raw materials in glass manufacturing, indicated an eastern Mediterranean origin (ϵ Nd between -2.5 and -6.0) for most of the samples. Conversely, samples with more negative ϵ Nd, between -7.02 and -10.8, indicate a western Mediterranean origin. By applying this technique on samples from well known archaeological contexts, this study demonstrates that several primary glass factories were located throughout the Roman Empire. This is contrary to what results from the late Roman to early Byzantine world indicate.

Key words: Roman Empire, natron glass, provenance, elemental analysis, Sr isotopes, Nd isotopes

Introduction

Ancient glass production makes use of three main ingredients: 1) silica in the form of sand or pure quartz, the former of the glass network, 2) soda, derived from plant ashes or from mineral natron, added as a flux to lower the melting temperature of the silica, and 3) lime, present in the silica source or added separately as limestone or shell, used as a stabilizer. Next to these main ingredients, colourants, decolourisers or opacifiers could be added to a glass batch.

Natron glass, using silica sand and mineral natron, was made in the Mediterranean and Europe from the middle of the 1st millennium BC through to the 9th century AD¹⁻³. This type of glass became the everyday, widespread and useful material

during Roman times, through the invention of glass blowing. It is, therefore, an excellent material to study manufacturing processes, production cycles, and goods distribution⁴. Where were mass producing 'primary' factories situated, and how was transport arranged? Was glass production in early Roman days primarily situated in the Near East, and was the molten material transported as chunks to 'secondary' factories to be remelted and shaped⁵⁻⁷? The homogenous composition of most Roman glass seems to indicate this. Or was this stability in chemistry due to a high number of smaller primary workshops using similar recipes? Or did intense recycling and the trade of broken glass and cullet play a role⁸?

Elemental and isotopic analysis of ancient glass will reflect the compositional signature of the ingredients with which the glass was made, thus providing potential indicators for the production location of the glass. For natron glass in the Roman era, this often means that the focus will be on the silica source, since natron is believed to come from saline lakes known today as the Wadi el Natrun (Egypt), a relatively pure soda source. Main elements used in ancient glass studies usually reflect the lime source and non-quartz minerals in the sand. Freestone et al.^{9,10} managed to distinguish five glass groups that occur commonly between the 4th and 9th centuries AD and seem to have their origin in the Syro-Palestine-Egypt region. Foy et al.^{11,12} distinguished ten compositional groups in 1st millennium AD raw and vessel glass from the Mediterranean region. Recent advances in isotopic analyses have shown that strontium and neodymium isotopic ratios are useful tools to provenance glass raw materials. Strontium can make a first distinction in the sand raw material since it is mainly incorporated in the sand via the lime-bearing material. If the Sr ratio is similar to that of modern seawater it is likely that the lime in the sand is derived from sea-shell. On the other hand the ratio could be similar to that of limestone, or possibly other minerals¹³. Neodymium isotopes are indicative of the geological age and origin of the non-quartz fraction of the sand source, an indicator that varies throughout the Roman world, making it ideal for sand provenancing.

Aims

In this study an elemental and isotopic comparison will investigate the technology used for and the occurrence of raw glass factories outside Syro-Palestine and Egypt for the early Roman period. The use of combined Sr and Nd isotopic analysis of glass allows different sand raw materials used for primary glass production to be distinguished and characterized^{1,14}. The isotope ratio data obtained for the glass samples are compared to a sand database¹⁵ which also includes relevant sands from the regions described by Pliny the Elder and primary glass from known production centres in the eastern Mediterranean^{1,14,16}.

Archaeological context

114 natron glass samples (1st to 5th century AD) excavated in different areas of the Roman Empire, were selected for compositional (n=114) and isotopic (n=87) analysis. The far east

side of the Roman Empire is accounted for by three archaeological sites, Petra and Barsinia, both in Jordan and Gonio, located in Georgia. The Italian peninsula is represented by the Iulia Felix and Embiez shipwrecks¹⁷⁻¹⁹, and by samples from Augusta Praetoria²⁰. Samples from Sant Boi de Llobregat in Spain are examples of the western side of the Empire, and the Belgian sites of Tienen¹ and Oudenburg^{21,22} represent the northern provinces. The chronology of the samples was determined by stratigraphical association.

10 Petra

The ancient city of Petra, located in south-west Jordan, played an important role in Arabian trade during Nabataean, Roman and Byzantine times. Several excavation campaigns conducted over several years yielded a multitude of glass vessels, objects and window glass which have been studied both typologically and chemically²³⁻²⁵.

In this paper 2 colourless glass fragments from the 3rd-4th century AD are considered for elemental and isotopic analysis.

Barsinia

20 The site of Barsinia is situated in north-west Jordan, 15 km west of Irbid. This site was occupied continuously from the Hellenistic until the Abbasid Period (332 BC-950 AD). It contains architectural remains (domestic nature), 4 tombs and 3 ovens (ceramic). Glass samples Ba1 and Ba10 were found in section D1, Ba2, Ba3, Ba4, Ba5 and Ba-6 were found in section B9, Ba7 was found in Tomb 1. These three sections of the site are dated to Late Roman times (3rd-4th century AD)²⁶⁻²⁸. All are coloured vessel fragments. 8 samples are analysed for their elemental and four samples for their isotopic composition.

30 Gonio

Gonio, ancient Apsaros, is located in south-western Georgia (15 km south of Batumi, near the mouth of the Chorokhi river) and comprises of a fortified city that had Roman occupation from the 1st to 4th century AD. The fortress is strategically placed upon a crossroads where the roads to the east (Turkey) and to the south (Armenia) join together; this was not only an important military location but it also played a role in the trade of goods²⁸.

In this paper we considered 8 1st-4th century AD vessel glass fragments, of which only Ge12 is colourless, for elemental analysis, and 3 samples for isotopic analysis.

Iulia Felix

The Iulia Felix shipwreck was discovered off the coast of Grado, in the north-eastern part of Italy, very close to Adria and Aquileia, two important glass working sites. It was a small cargo ship of the so-called 'corbita' type²⁹. The cargo was mainly composed of amphorae of various types. Ceramic seriation dated the cargo to the first half of the 3rd century AD^{17,18,29}. Part of the cargo was a wooden barrel filled with more than 11000 fragments of glass, totalling 140 kg. The original shapes of the different glass artefacts, mainly cups, plates and bottles, can be deduced from a typological study of the fragments. The most common colours are blue-green, pale blue and pale green, together with a good amount of colourless glass. They all represent examples of accidentally broken glass, most probably collected for recycling²⁹. 14 fragments, all colourless, were selected for elemental and

isotopic analyses.

Embiez

The Embiez was wrecked off the island of Embiez in southern France. It is dated to the end of the 2nd - beginning of the 3rd century AD³⁰. The ship was about 12 m long, and is at the moment the only known Roman ship totally dedicated to the trade of glass. The majority of the cargo was composed of raw glass, and is estimated to be c. 18 tons. In addition, the cargo was composed of about 1800 cup fragments, and window glass panels. 18 fragments, all colourless, were selected for compositional and isotopic analyses.

Augusta Praetoria

Augusta Praetoria (modern Aosta, north-west Italy) was founded in 25 BC following the Roman conquest of a territory previously inhabited by an indigenous population, the Salassi. The town commanded the access to the passes which allowed communication between the south and the north of the Alps. It was situated on the main route, of the Gallic Consular Road. It connected the eastern roman towns of Italy (Aquileia and Adria) with the western Italian towns of Augusta Praetoria and Augusta Taurinorum, and the northern provinces on the other side of the Alps. The glass fragments are dated to between the 1st and the 4th century AD²⁰. samples, all coloured ranging from blue-green to green-yellow, were selected for elemental and isotopic analyses.

80 Sant Boi de Llobregat

Sant Boi de Llobregat is a site situated in the province of Barcelona. Architectural remains were found which indicate continuous occupation from the 6th century BC until the 12th century AD. It is located 17 km from the sea on the bank of the river Llobregat, close to the important port of Tarraco. Romans occupied the city from the 1st-5th century AD, and recently there have been some indications near this site for the secondary production of Roman natron glass³¹.

19 coloured and colourless Roman glass samples, dated 1st to 5th century AD, were analyzed for their elemental composition, 11 samples were analyzed isotopically.

Oudenburg

Oudenburg was a Roman castellum situated on the Belgian coast with direct access to the North Sea and a connection to the Roman road network. The castellum started out in the 1st century AD as a small trade settlement with a small port but expanded in the 2nd century AD. In the 3rd century AD the civil settlement was abandoned and the focus became strictly military^{21,22}.

The glass samples from Oudenburg were collected during excavations of the south-western corner of the castellum. Five samples are dated to the 3rd century AD, but most of them (11) belong to the 4th-5th century AD. 16 samples were analyzed for their elemental composition, 6 samples were analyzed isotopically.

105 Tienen

The small Roman town of Tienen is situated in the Belgium province of Vlaams-Brabant and was founded in the 1st century AD close to the road from Cologne to Boulogne, and it was part of the *civitas Tungrorum*. Archaeological excavations point to the

production of several goods such as pottery, iron, and bronze as well as glass vessels. The samples that will be discussed here are 11 pale blue vessel glass fragments, associated with a 2nd century glass furnace^{1,14,32,33}. 11 samples were selected for elemental and isotopic analyses.

Analytical methods

After the removal of any external layers to avoid possible contamination from the burial environment and corrosion, 150–200 mg were taken from each sample using a diamond-coated wheel. The fragment was then washed in an ultrasonic bath with Milli-Q water, rinsed and dried at 100°C. The dry piece was then ground to a fine powder using an agate mortar.

Bulk composition

The bulk composition of the Iulia Felix samples was studied at the University of Padova with X-ray Fluorescence (XRF) spectroscopy, on a Philips PW 2400 instrument^{17,18}. Precision was better than 0.6% for major and minor elements, and about 3% for trace elements, while accuracy was within 0.5 wt% for Si, lower than 3% for other major and minor elements, and better than 5% for traces³⁴.

Elemental analyses of the Embiez samples were carried out at the University of Leuven with inductively coupled plasma optical emission spectroscopy (ICP-OES) on a Varian 720-ES (simultaneous ICP-OES instrument with axially viewed plasma)³⁵. The instrumental precision, calculated as the average relative standard deviation of the readings performed on each glass reference material, was better than 3% for major and minor elements, and accuracy was better than 10%.

Augusta Praetoria samples were analyzed at the University of Torino with ICP-OES on a Philips PU 7450 spectrophotometer²⁰. Measurement precision was better than 3% for major and minor elements.

Major and minor elements on the Tienen samples were determined by atomic emission spectrometry (AES) on a Spectrojet III spectrometer. Sodium and potassium contents were obtained from the same solutions by atomic absorption spectrometry (AAS) on a Varian Techtron AA6 spectrometer. Analytical precision was better than 0.5%, and accuracy was better than 2%¹.

Sant Boi de Llobregat, Oudenburg, Petra, Barsinia and Gonio samples were analyzed at the University of Salzburg with microprobe on a JEOL Superprobe JXA-8600 instrument. Accuracy and precision were better than 10%^{36,37}.

further passed through a column containing 2 ml Eichrom Ln.Spec resin. For this, the column was washed with 5.5 ml 0.25M HCl after adding the sample. Nd was stripped off using 4 ml 0.25 M HCl. Sr isotopic ratios were normalized to $^{86}\text{Sr}/^{88}\text{Sr} = 0.1194$, Nd isotopic ratios were normalized to $^{146}\text{Nd}/^{144}\text{Nd} = 0.7219$. Repeated static measurements of the NBS 987 standard over the duration of the study yielded an average $^{87}\text{Sr}/^{86}\text{Sr}$ ratio of 0.71025 ± 0.00002 . Repeated measurements of the La Jolla Nd standard yielded $^{143}\text{Nd}/^{144}\text{Nd} = 0.511848 \pm 0.000009$ ^{1,14}.

Sr and Nd ratios for Augusta Praetoria, Iulia Felix, Embiez,

Bulk composition analyses have been performed with different techniques and instruments on a twenty-year time-scale. Different approaches to test the comparability of data obtained with different techniques and instruments have been proposed in the literature. For instance, Paynter³⁸ suggests re-measuring a representative selection of samples with one common technique. Baxter³⁹ suggests the re-measuring of a representative set of samples as well, and describes statistical methods to subsequently correct data sets obtained in different analytical runs in order to compare and combine them. Unfortunately, re-measurement of samples was not possible for this study, as only a limited amount of sample from ancient artefacts were available.

Nevertheless, a thorough examination of the compositional data in this study allows a comparison between the different analytical runs on the basis of the good precision and accuracy of the different techniques for major element analysis. For instance, considering the worst case scenario with 10% precision and accuracy, a 0.5% K₂O concentration will result in a range of 0.45–0.55%. For all elements discussed here, the between-group variation, meaning the difference between group averages, is a (several) order(s) of magnitude greater than the analytical variability³⁹.

Isotopic analysis

For isotopic analysis, 100 mg of powdered sample were weighed into Savillex screw-top beakers and dissolved in a 3:1 mixture of 22 M HF and 14 M HNO₃ on a hot plate. Solutions were dried and the residues redissolved in aqua regia. After digestion was completed, the sample was evaporated to nearly dry and the residue was taken up into 7 M HNO₃.

The concentrations of Sr and Nd were checked using a quadrupole-based Perkin-Elmer SCIEX Elan 5000 ICP-MS instrument. An internal standard (In) was used to correct for matrix effects, signal drift, and instrument instability, and calibration was performed against an external standard containing known amounts of the elements to be determined.

Sr and Nd ratios for all the samples from Tienen¹ and Petra, and part of the Sant Boi de Llobregat samples (PD2, PD4, PD6, PD9, PD10, PD11, PD14, PD15, PD16) and the Barsinia samples (BA1, BA10) were analyzed on a six-collector Finnigan Mat 262 thermal ionization mass spectrometer (TIMS) running in multicollection mode. For the separation of Sr and Nd sequential extraction methods developed by Pin et al.⁴⁰ were utilized and slightly modified. Sr and REE were separated using 2 M HNO₃ using coupled miniaturized Teflon columns containing 50 µl of Eichrom Sr.Spec and TRU.Spec resin, respectively, and eluted with de-ionized H₂O. For the separation of Nd, the REE cut was

Oudenburg, Gonio samples, and the other Sant Boi de Llobregat and Barsinia samples, were obtained with a Thermo Scientific Neptune multi-collector inductively coupled plasma mass spectrometer (MC-ICP-MS), equipped with a micro-flow PFA-50 Teflon nebuliser, and running in static multi-collection mode.

For the isolation of Sr⁴¹, 0.1 ml of solution in 7 M HNO₃ was loaded onto a BioSpin column (BioRad) packed with 400 µl Sr specTM resin (Eichrom). After rinsing with 4 ml of 7 M HNO₃ to eliminate the matrix, the Sr fraction was eluted with 5.5 ml of 0.05 M HNO₃. A concentration-matched solution of NIST SRM

987 SrCO₃ isotopic reference material was used as an external standard (⁸⁶Sr/⁸⁸Sr = 0.1194) to correct for instrumental mass discrimination (sample-standard bracketing). To correct for the Kr interferences obtained at m/z 86 the intensity obtained for ⁸³Kr was used. On average, ⁸⁷Sr/⁸⁶Sr isotope ratios were measured with an internal precision (2s) of 0.0000439. The isolation of Nd involved a 2-step chromatographic separation⁴². The sample, taken up in 1 ml of 2 M HNO₃, was loaded onto a Micro-BioSpin column (BioRad) filled with 600 µl TRU.Spec resin (Eichrom), and was washed with 4 ml of 2 M HNO₃. The Micro-BioSpin column was then connected to an Eichrom column (0.8 cm inner diameter) packed with 1.5 ml Ln.Spec resin (Eichrom), and was rinsed with 7 ml of 0.05 M HNO₃ in order to elute the LREE (light rare earth elements) fraction from the TRU.Spec resin into the Ln.Spec resin. The Ln.Spec resin was then washed with 5 ml of 0.25 M HCl, and the Nd fraction was stripped off using 9 ml of content ranging between 56.67 and 74.85 wt%, and CaO and Na₂O contents of 4.48-11.21% and 11.79-20.53%, respectively. The concentration of MgO varies between 0.21 and 1.10 wt%, and that of K₂O between 0.29 and 1.45 wt%, suggesting the use of natron as a flux for all the samples⁴³⁻⁴⁵. The concentration of Al₂O₃ is relatively constant and varies between 1.53 and 3.61 wt%, the typical content found in ancient natron glass.

Sr and Nd isotopic compositions of the selected samples are given in Table 2, and are expressed as ratios. The ratio ¹⁴³Nd/¹⁴⁴Nd is also expressed as εNd

$$\epsilon Nd = \left(\frac{(^{143}\text{Nd}/^{144}\text{Nd})_{\text{sample}}}{(^{143}\text{Nd}/^{144}\text{Nd})_{\text{CHUR}}} - 1 \right) \times 10^4$$

where ¹⁴³Nd/¹⁴⁴Nd_{CHUR} = 0.512638, according to DePaolo and Wasserburg⁴⁶.

The ⁸⁷Sr/⁸⁶Sr ratios for the 87 selected glass samples vary between 0.70759 and 0.70947, with an average value of 0.70900 ± 0.00020. All the samples, with the exception of Tie49 (⁸⁷Sr/⁸⁶Sr = 0.70759) correspond closely to the present-day seawater signature (0.7092).

The Nd isotopic data show a wide range in composition, varying between 0.512083 and 0.512513 for ¹⁴³Nd/¹⁴⁴Nd, which is between -10.8 and -2.4 for εNd.

Discussion

Low-magnesia glass is essentially a mixture of sand and soda, where the evaporites of the Wadi el Natrun represent a relatively pure soda source. The question of provenancing glass, in terms of understanding the place where the glass was made, is hence strictly related to the sand raw material^{3,9}. Lime and alumina levels are to a degree diagnostic of the sand source⁹ as they reflect the impurities (calcite, feldspar and clay-minerals) present in the sand.

In fig. 1, the CaO versus Al₂O₃ content for all the investigated glass samples is plotted. An examination of the biplot reveals a

0.25 M HCl. For the measurements of ¹⁴³Nd/¹⁴⁴Nd, JNdi-1 reference material (Geological Survey of Japan) was used to correct for instrumental mass discrimination (¹⁴³Nd/¹⁴⁴Nd = 0.51515, ¹⁴⁶Nd/¹⁴⁴Nd = 0.7219). The intensity obtained for ¹⁴⁷Sm was used to correct the intensities obtained at m/z 144 for Sm interferences. On average, ¹⁴³Nd/¹⁴⁴Nd isotope ratios were measured with an internal precision (2s) of 0.0000219.

Results

The analytical data of the 114 glass samples under consideration are given in Table 1. Major element analyses are expressed as weight percent of element oxides.

All samples are soda-lime-silica glasses, characterized by SiO₂

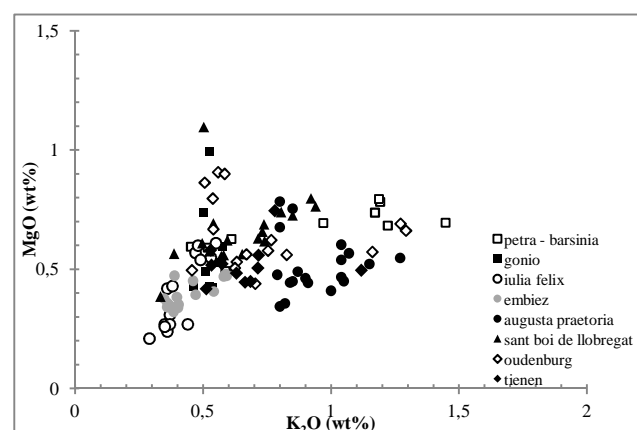


Fig. 2. MgO versus K₂O biplot of the analytical data for glass obtained in this study.

good correlation of the two elements, with a sample distribution that only limitedly respects the excavation site or the colour. Most of the colourless glass from the Iulia Felix (3rd century AD) and the Embiez (end of the 2nd – beginning of the 3rd century AD) shipwrecks show the lower CaO and Al₂O₃ values, respectively <6% and <2%. Those samples can be compared to the “antimony-only BCL” samples from Bincester, Lincoln and Colchester discussed in Paynter et al.³⁸, all 1st-4th century colourless glass. On the other hand, the highest CaO and Al₂O₃ values, respectively >8% and >2.5%, are shown in the Jordanian samples (3rd-4th century AD). Compositionally, these samples show a very good correspondence to the Levantine I glass group⁹, suggesting that this region was indeed active before the 4th century AD. The samples from Gonio, Georgia, sit in an intermediate position, with CaO content between 7.64 and 8.91 wt% and Al₂O₃ content between 1.88 and 2.74 wt%. Samples from Augusta Praetoria show a wide range in compositions. Samples 9a, 12a, 23g, 8b, 9b, 15, 20f, 20e and 22d are comparable to the “antimony-only BCL”³⁸ group, with CaO content between 4.48 and 7.17 wt% and Al₂O₃ content between 1.58 and 2.31 wt%. The other samples from Augusta Praetoria (1, 5, 7, 12b, 21b, 22e, 23d, 23e, 23i), as well as the samples from Sant Boi de Llobregat, plot in an intermediate position, that does not match any of the known compositional groups consistently, with CaO content between 6.11 and 8.71 wt% and Al₂O₃ content

between 1.98 and 3.15 wt%. Oudenburg samples, based on the elements analyzed, mainly fall in the “antimony-only BCL” area³⁸, with the exception of samples 71310, 8962, 23993 and 2960, that have a slightly higher CaO and Al₂O₃ content. Special attention has to be drawn to Tienen samples. These samples cover a range of concentrations that vary between 1.88 and 3.41 wt% for Al₂O₃ and between 5.95 and 10.25 wt% for CaO. Samples Tie12, Tie37, Tie48 and Tie49 lie in the lower corner of Levantine I group⁹. All other Tienen samples are different in composition to the Levantine I group, showing (much) lower CaO and Al₂O₃ contents.

A similar situation, with a large range in concentrations, can also be seen in the MgO versus K₂O content of the glass, typical for the non-quartz mineralogy of the sand source used (fig. 2). An

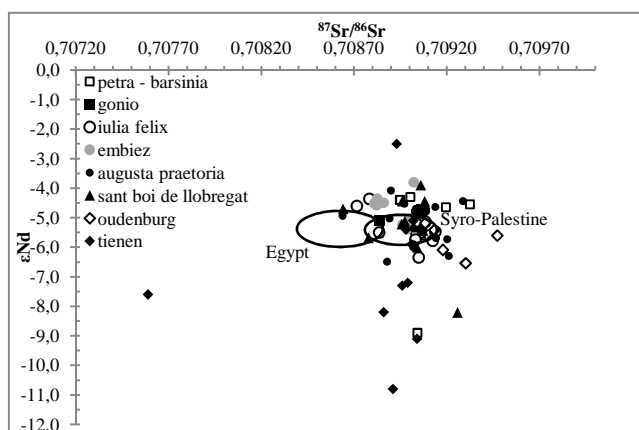


Fig. 3. ϵ Nd versus $^{87}\text{Sr}/^{86}\text{Sr}$ biplot of the analytical data obtained in this study. The typical composition of primary glass from the Syro-Palestine and Egypt is indicated¹⁶.

interesting divide in the data set can be observed, as the Italian, Spanish, Jordanian and Georgian samples show a similar trend in MgO-K₂O content, while the Belgian samples show many different compositional trends or subgroups in terms of MgO-K₂O content.

Focusing on the Italian, Spanish, Jordan and Georgian samples, a split into two sub-groups is given by the K₂O content. This separation also appears to distinguish the glass based on colour. A low K₂O group characterized by K₂O < 0.65 wt% includes all the colourless samples, together with the pale shades, for a total of 56 glass samples. A high K₂O group, with K₂O > 0.79 wt%, includes all the coloured glass (29 samples) with colour ranging from green-yellow to blue-green. The low potassium group is characterized by an average 0.47 ± 0.10 wt% K₂O content, together with 0.48 ± 0.17 wt% MgO, 2.18 ± 0.51 wt% Al₂O₃ and 6.74 ± 1.58 wt% CaO. The high potassium group is instead characterized by average 0.99 ± 0.17 wt% K₂O content, together with 0.59 ± 0.15 wt% MgO, 2.49 ± 0.45 wt% Al₂O₃ and 7.61 ± 1.66 wt% CaO. Four samples from Sant Boi de Llobregat (PD2, PD4, PD7, PD16) sit in an intermediate position, with K₂O content between 0.72 and 0.74 wt%, and MgO content between 0.62 and 0.69 wt%. The distinctive low- and high-potassium groups suggest that there were at least two separate sand sources used in the making of these glasses. A relatively mature (pure quartz) sand was used for the production of colourless glass^{2,47}, and a less pure, richer in lime alumina and potassium sand was

used for the production of the coloured glass.

Raw glass from the 4th-8th century AD primary production centers in Egypt and the Syro-Palestine has already been analyzed for its Sr and Nd isotopic composition¹⁶. Strontium is considered a proxy for the lime-rich components in the glass raw materials⁴⁸. In the Syro-Palestine samples, $^{87}\text{Sr}/^{86}\text{Sr}$ signatures close to the present-day seawater signature (0.7092) indicated the use of shell as a source of lime in the glass^{48,49}. This shell was a natural inclusion in the beach sand of the Syro-Palestine coast, which was used to manufacture the glasses. The lower $^{87}\text{Sr}/^{86}\text{Sr}$ of the Egyptian samples pointed to the use of either limestone⁴⁸ or the influence of other minerals in the sand^{50,51} relatively low in radiogenic strontium. The low variation in $^{143}\text{Nd}/^{144}\text{Nd}$ for Syro-Palestine and Egyptian primary glass, with values between -6.0 and -5.1, was consistent with the values given by the Nile-dominated sediments in the eastern Mediterranean^{52,53}.

This range of Sr and Nd isotopic values is repeated in most of the 1st-5th century AD glass samples of this study (fig. 3), suggesting that primary production was indeed taking place in the eastern Mediterranean before the 4th century AD, although, not necessarily in the same geographical areas as the aforementioned excavated primary glass units. Considering both their elemental and isotopic composition, the samples from Gonio, Georgia, (1st-4th century AD) are of particular interest. Their main element signature suggests an origin different from the Levantine I group, with lower contents of CaO, Al₂O₃ and K₂O. Their $^{87}\text{Sr}/^{86}\text{Sr}$ (0.70884 to 0.70907) and ϵ Nd (-5.08 to -4.72) signature, however, indicate a Syro-Palestinian origin, making these samples the most northerly Levantine glass found. Conversely, the Barsinia samples, Jordan, (3rd-4th century AD) could be identified as being Levantine I⁹ based on the main element composition. The less negative ϵ Nd values (-4.65 to -4.30) indicate that these glass samples were indeed produced in the eastern Mediterranean, but in a somewhat different location than the known 4th-8th century AD primary glass units.

A similar provenance can be assumed for some of the samples from Sant Boi de Llobregat (PD10, PD14, PD15, PD16, PD28), Embiez (160, 252, 264, 385, 386, 390), Iulia Felix (IB104, B82), and Augusta Praetoria (22e, 23d, 23i, 5, 12b, 15, 7, 23e, 21b), with ϵ Nd varying between -3.8 and -4.94. The $^{87}\text{Sr}/^{86}\text{Sr}$ is comparable to the present-day seawater signature, with values ranging between 0.70872 and 0.70929, and identifies the source of lime as shell. The major element compositions of these samples do not match any of the known compositional groups consistently. Sample Tie11 from Tienen, with ϵ Nd = -2.5 and $^{87}\text{Sr}/^{86}\text{Sr} = 0.70893$, shows an exceptionally high Nd signature, that at the moment does not have any similarities with published data.

On the other hand, a number of samples from Oudenburg, Augusta Praetoria, Iulia Felix, Sant Boi de Llobregat and Petra clearly have an exotic Sr-Nd isotopic composition, not corresponding to sediment signatures from the eastern Mediterranean basin. Samples 29993, 2960, 8b, 9a, B10, PD6, PE2 have ϵ Nd -6.09 to -8.90. This Nd isotopic composition is inconsistent with any sediment or raw glass in the eastern Mediterranean, but the lower ϵ Nd values correspond well to the range of isotopic values of beach and deep-sea sediments from

the western Mediterranean, from the Italian peninsula to the French and Spanish coasts and from north-western Europe^{14,54}. The primary production locations of this glass therefore most likely lie in the western Roman Empire. However, for the intermediate ϵNd values of some of this glass (-6.09 to -6.54), it cannot be discarded that the recycling of glass or the mixing of primary sources from the eastern and western Mediterranean area has given the material its isotopic composition. The $^{87}\text{Sr}/^{86}\text{Sr}$ ratios, between 0.70886 and 0.70918, are close to the present-day seawater signature, probably identifying shell as the source of lime. Three samples, 8b, PD6 and 2960, show a different $^{87}\text{Sr}/^{86}\text{Sr}$, 0.70921, 0.70926 and 0.70930 respectively, possibly indicating a different, and more radiogenic, source of lime than shell, or clay contamination.

Special attention has to be drawn to the 2nd century AD glass samples from Tienen. Though thoroughly Roman in age, the major elemental composition of samples Tie12, Tie35, Tie37, Tie48 and Tie49 corresponds well to the younger 4th-8th century AD Levantine I group⁹. Conversely, the Nd signature of these samples (ϵNd between -7.2 and -10.8) does not allow an eastern Mediterranean origin, but places their primary production locations in the western Mediterranean or north-western Europe¹⁴. Samples Tie17, Tie24, Tie41 and Tie50, though, are characterized by a typical Syro-Palestinian isotopic signature, with ϵNd between -5.1 and -5.9 and $^{87}\text{Sr}/^{86}\text{Sr}$ between 0.70898 and 0.70902, while their major elemental composition does not match any of the known compositional groups consistently. The Sr signature of most of the Tienen glass is very homogeneous, close to the present-day seawater signature and likely to be indicative of the use of shell as a lime source in the glass. Sample Tie49, however, shows a different Sr signature, with $\epsilon\text{Nd} = -7.6$ and $^{87}\text{Sr}/^{86}\text{Sr} = 0.70759$, consistent with the Sr-Nd isotopic composition of Egyptian sands¹⁴, possibly suggesting that the glass originated there. The large spread in the major element and isotopic compositions of these samples, covering the eastern and western Mediterranean and even north-western Europe, is almost certainly an indicator of the extensive recycling of this glass from different sources around the Roman Empire.

Conclusion

Neodymium and strontium are clearly useful for tracing the origin of primary glass. Nd is characteristic of the mineral fraction other than quartz in the silica raw material, while Sr is in most cases characteristic of the lime component, either attributed to the sand raw material or as a separate constituent in the form of shell. These isotopes do not replace main element analyses and both techniques should be regarded as complementary.

Based on the major element analysis, the Italian, Spanish, Jordanian and Georgian samples can be divided into two groups, coloured and colourless. The Sr and Nd isotopic composition suggests an eastern Mediterranean origin for most of these samples (ϵNd higher than -6.0), though the exact location of the primary production centres can differ from the known Egyptian and Syro-Palestinian sites. Particularly interesting are the samples from Gonio, Georgia, which based on the Nd isotopic ratio can be considered the most northerly Levantine samples. Conversely, a western Mediterranean origin is suggested for those glass samples

with ϵNd lower than -6.0.

The Northern provinces samples, from Oudenburg and Tienen, show a wide range in major elemental composition. Though not contemporary, the Tienen samples are similar in major element composition to the 4th-8th century AD Levantine I glass group⁹. The Nd signature of the glass (between -7.2 and -10.8), however, does not indicate an eastern Mediterranean origin at all, but places their primary production location in the western Mediterranean or north-western Europe¹⁴. The Sr-Nd isotopic composition of the Oudenburg samples, on the other hand, suggests a clear eastern Mediterranean origin (ϵNd between -5.1 and -6.54) for the glass. The large spread in major elemental and isotopic composition of the Tienen glass, indicating a mixed origin, is almost certainly an indicator of extensive recycling of this material from different sources around the Roman Empire⁵⁴.

By applying isotope geochemistry to samples with well dated archaeological contexts it is possible to recreate part of the ancient Roman economy system. This study not only shows that several primary factories were functioning during the 1st-4th centuries AD, but also demonstrates that they are located throughout the Roman empire and not solely in the Near East, as might be the case for late Roman/Byzantine times.

Acknowledgements

The authors are grateful to Prof. Piero Mirti and Dr. Lorenzo Appolonia for the Augusta Praetoria samples, Prof. Gianmario Molin and Dr. Alberta Silvestri for the Iulia Felix samples, Prof. Danielle Foy for the Embiez samples, Prof. Ziad al-Saad and Dr. Roland Lamprichs for the Jordan samples. A special thanks to Kris Latruwe, for his fundamental help during the MC-ICP-MS measurements. The research is financially supported by the ERC Starting Grant ARCHGLASS Grant agreement no. 240750 and FWO project no. 6.0864.09. Monica Ganio is currently working as Aspirant of the Research Foundation – Flanders (FWO).

Notes and references

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Sample	Date	Colour	SiO ₂	Na ₂ O	CaO	Al ₂ O ₃	K ₂ O	MgO	Fe ₂ O ₃	TiO ₂	MnO	total
Petra												
PE1	III-IV	colourless	67.84	17.45	6.79	2.21	0.61	0.63	0.53	0.08	0.01	96.15
PE2	III-IV	colourless	69.32	14.67	8.26	2.70	0.52	0.59	0.35	0.07	0.49	96.97
Barsinia												
BA1	III-IV	pale blue-green	65.92	15.52	9.75	3.23	1.19	0.78	0.54	0.08	0.06	97.08
BA2	III-IV	pale green	65.99	15.79	9.50	2.68	1.45	0.70	0.61	0.09	0.64	97.44
BA3	III-IV	pale blue	72.46	11.79	7.27	3.61	0.45	0.60	0.50	0.12	0.02	96.83
BA4	III-IV	pale blue	70.50	12.26	9.39	3.03	0.59	0.49	0.34	0.07	0.01	96.68
BA5	III-IV	green-yellow	66.74	14.51	10.06	3.21	1.22	0.68	0.48	0.08	0.08	97.06
BA6	III-IV	green-yellow	68.77	14.07	9.81	2.93	0.97	0.69	0.33	0.09	0.08	97.73
BA7	III-IV	green-yellow	65.18	15.30	10.83	3.25	1.19	0.80	0.52	0.10	0.14	97.29
BA10	III-IV	green-yellow	64.79	15.19	11.21	3.30	1.17	0.74	0.55	0.07	0.10	97.12
Gonio												
Ge2	I-IV	pale blue	71.37	15.58	8.28	2.46	0.46	0.43	0.27	0.04	0.36	99.26
Ge3	I-IV	green-yellow	65.48	19.24	8.36	2.04	0.52	1.00	0.66	0.14	1.55	98.99
Ge4	I-IV	green-yellow	66.89	19.27	7.66	1.88	0.50	0.74	0.61	0.13	1.44	99.11
Ge5	I-IV	pale blue	69.31	16.23	8.82	2.34	0.56	0.53	0.42	0.08	0.54	98.82
Ge7	I-IV	pale blue	70.38	15.60	8.91	2.39	0.58	0.60	0.46	0.07	0.56	99.54
Ge8	I-IV	pale blue	70.08	17.22	7.64	2.49	0.54	0.43	0.41	0.07	0.58	99.45
Ge9	I-IV	pale blue	71.88	15.58	8.05	2.49	0.53	0.43	0.32	0.06	0.40	99.73
Ge12	I-IV	colourless	70.33	14.77	8.44	2.74	0.51	0.49	0.26	0.04	1.43	99.00
Iulia Felix												
IF B05	III	colorless	70.65	19.34	4.65	1.93	0.36	0.42	0.39	0.06	0.02	97.82
IF B07	III	colorless	70.85	19.46	4.95	1.87	0.37	0.31	0.3	0.05	0.01	98.17
IF B117	III	colorless	70.04	19.22	5.54	1.89	0.38	0.43	0.41	0.08	0.02	98.01
IF C03	III	colorless	70.07	19.42	4.86	1.93	0.36	0.24	0.32	0.06	0.01	97.27
IF C11	III	colorless	70.18	19.57	4.79	1.9	0.35	0.27	0.32	0.06	0.01	97.45
IF C17	III	colorless	70.22	19.95	4.61	1.83	0.29	0.21	0.33	0.06	0.01	97.51
IF P3	III	colorless	70.62	19.69	4.75	1.96	0.44	0.27	0.34	0.06	0.01	98.14
IF P20	III	colorless	69.75	20.43	4.54	1.88	0.37	0.27	0.39	0.07	0.01	97.71
IF P23	III	colorless	69.88	20.53	4.61	1.86	0.35	0.26	0.3	0.06	0.01	97.86
IF B10	III	colorless	69.13	18.46	6.26	2.23	0.55	0.61	0.51	0.1	0.27	98.12
IF B112	III	colorless	69.56	17.92	6.1	2.16	0.53	0.55	0.53	0.1	0.25	97.70
IF B68	III	colorless	72.22	14.12	7.44	2.71	0.47	0.57	0.13	0.07	1.11	98.84
IF B82	III	colorless	70.26	14.74	7.91	2.65	0.48	0.6	0.11	0.08	1.38	98.21
IF B104	III	colorless	69.27	15.52	8.29	2.42	0.49	0.54	0.03	0.07	1.47	98.10
Embiez												
103	II-III	colorless	70.29	18.80	5.34	1.56	0.37	0.35	0.28	0.06	0.01	97.04
152	II-III	colorless	69.07	18.52	5.25	1.54	0.36	0.34	0.27	0.05	0.01	95.42
160	II-III	colorless	70.04	18.83	5.35	1.55	0.37	0.35	0.28	0.06	0.01	96.84
258	II-III	colorless	69.76	19.72	5.45	1.58	0.40	0.38	0.28	0.06	0.02	97.65
259	II-III	colorless	69.86	20.11	5.47	1.56	0.40	0.35	0.28	0.06	0.02	98.12
264	II-III	colorless	69.26	18.44	5.25	1.53	0.36	0.34	0.26	0.05	0.01	95.51
265	II-III	colorless	69.51	18.52	5.28	1.53	0.37	0.34	0.27	0.06	0.01	95.90
272	II-III	colorless	71.25	18.71	5.40	1.57	0.39	0.47	0.28	0.06	0.01	98.15
622	II-III	colorless	69.17	18.23	5.27	1.54	0.36	0.36	0.28	0.06	0.01	95.28
252	II-III	colorless	56.67	13.40	7.35	1.98	0.47	0.39	0.24	0.04	1.14	81.69
385	II-III	colorless	66.77	15.84	8.71	2.31	0.58	0.47	0.29	0.05	1.35	96.38
386	II-III	colorless	67.51	16.07	8.80	2.37	0.59	0.48	0.29	0.05	1.37	97.54
390	II-III	colorless	67.50	16.02	8.79	2.35	0.59	0.47	0.29	0.05	1.31	97.38

397	II-III	colorless	67.89	16.44	8.55	2.31	0.46	0.45	0.28	0.05	1.58	98.02
gobA	II-III	colorless	71.22	18.39	5.36	1.78	0.54	0.41	0.31	0.06	0.02	98.08
gobB	II-III	colorless	69.99	19.75	4.96	1.57	0.38	0.32	0.25	0.05	0.02	97.29
gobC	II-III	colorless	69.78	18.36	5.40	1.59	0.35	0.38	0.28	0.06	0.02	96.23
gobD	II-III	colorless	69.81	19.45	5.17	1.56	0.40	0.34	0.27	0.05	0.02	97.07
Augusta Praetoria												
1	I-III	blue-green	70.51	15.76	7.98	2.65	1.07	0.57	0.39	0.07	0.83	99.84
09A	I-II	green	67.06	16.55	7.17	2.31	1.00	0.41	0.24	0.06	0.89	95.69
12A	II-III	green-yellow	71.47	17.74	5.88	1.93	0.84	0.45	0.40	0.10	0.26	99.06
12B	I-II	blue-green	72.19	16.98	6.36	2.56	1.27	0.55	0.65	0.12	0.49	101.17
22E	I-II	blue-green	70.26	16.39	8.09	2.67	1.04	0.54	0.40	0.07	0.53	99.99
23G	I-II	blue-green	71.63	16.39	6.96	2.12	1.15	0.52	0.52	0.12	0.38	99.80
08B	II-III	green-yellow	70.21	17.70	6.75	2.21	0.80	0.79	0.88	0.22	0.03	99.59
09B	II-III	green-yellow	68.75	18.64	6.00	2.10	0.85	0.76	0.61	0.14	0.02	97.87
15	IV	blue-yellow	70.80	18.08	6.36	2.06	0.80	0.68	0.56	0.13	0.92	100.39
20F	III-IV	green-yellow	69.29	18.66	6.67	1.91	1.05	0.45	0.40	0.09	0.03	98.55
5	I-II	blue-green	70.31	15.32	8.38	2.13	0.87	0.49	0.31	0.07	0.22	98.11
7	I-II	blue-green	73.36	15.34	8.56	2.15	0.91	0.44	0.33	0.08	0.13	101.30
21B	I-III	blue-yellow	70.47	16.04	7.66	2.39	1.04	0.60	0.45	0.10	2.25	100.99
23E	I-II	blue-green	74.75	13.93	7.38	2.44	1.04	0.47	0.32	0.07	0.18	100.57
23I	I-II	blue-green	72.26	14.74	7.60	2.47	0.79	0.48	0.30	0.07	0.31	99.02
20E	III-IV	green-yellow	74.85	17.48	4.48	1.58	0.80	0.35	0.27	0.06	0.01	99.88
22D	I-II	green-yellow	74.67	16.72	4.68	1.84	0.82	0.36	0.30	0.07	0.01	99.47
23D	II	green	71.16	14.61	8.09	2.40	0.85	0.45	0.28	0.07	0.15	98.06
Sant Boi de Llobregat												
PD1	I	pale blue	68.06	16.63	6.86	2.74	0.92	0.80	0.61	0.14	0.66	97.42
PD2	I	blue	66.64	17.08	8.51	2.90	0.72	0.63	0.35	0.05	0.54	97.42
PD3	I	pale blue	65.69	16.75	7.05	2.78	0.94	0.76	0.78	0.16	0.45	95.38
PD4	I	colourless	68.98	17.46	5.84	2.52	0.74	0.62	0.48	0.11	0.39	97.14
PD5	I	pale blue	68.69	16.93	6.28	2.86	0.85	0.73	0.74	0.13	0.57	97.77
PD6	I	green-yellow	66.69	18.40	6.11	3.05	0.50	1.10	0.89	0.26	0.06	97.07
PD7	I	colourless	67.71	17.45	7.11	2.56	0.74	0.69	0.57	0.09	0.29	97.21
PD8	I	pale blue	66.51	18.08	8.12	2.92	0.59	0.62	0.31	0.04	0.46	97.65
PD9	I	pale blue	68.04	16.73	7.07	2.81	0.80	0.74	0.58	0.10	0.54	97.42
PD10	I	pale blue	67.54	17.48	8.23	2.61	0.58	0.56	0.29	0.05	0.34	97.68
PD11	I	pale blue	67.22	17.08	8.42	2.54	0.57	0.56	0.32	0.08	0.36	97.15
PD12	I	pale blue	66.68	17.32	8.71	2.63	0.65	0.56	0.25	0.06	0.44	97.30
PD14	I	colourless	66.46	17.41	9.06	2.79	0.53	0.58	0.30	0.05	0.34	97.52
PD15	I	colourless	68.91	15.54	8.32	2.88	0.62	0.52	0.27	0.06	0.32	97.45
PD16	I	purple	65.11	17.21	8.32	2.77	0.73	0.66	0.37	0.06	1.66	96.90
PD26	IV-V	colourless	68.91	19.17	5.18	2.00	0.33	0.39	0.32	0.07	0.00	96.37
PD27	IV-V	pale blue-green	67.71	15.86	7.32	2.51	0.50	0.61	0.43	0.08	1.34	96.35
PD28	IV-V	pale blue	66.89	13.84	7.94	3.15	0.54	0.69	0.70	0.10	1.43	95.28
PD29	IV-V	colourless	68.08	19.58	6.18	1.98	0.39	0.57	0.39	0.08	0.09	97.33
Oudenburg												
80993	III	blue-green	68.51	17.30	6.36	2.29	0.63	0.53	0.71	0.08	0.46	96.88
8933	IV-V	blue-green	68.04	16.70	6.86	2.46	0.67	0.56	0.51	0.07	0.35	96.21
8926 C	III	pale blue	69.63	16.44	6.55	2.39	0.70	0.44	0.43	0.09	0.45	97.12
71310	III	pale blue	68.21	16.87	6.41	2.72	1.16	0.57	0.58	0.11	0.40	97.04
8962	III	blue-green	68.57	16.64	6.75	2.59	0.77	0.62	0.82	0.10	0.30	97.16
7929	III	colourless	68.27	18.54	6.01	1.86	0.46	0.50	0.42	0.06	0.13	96.23
23993	IV-V	blue-green	68.60	16.84	7.04	2.52	0.75	0.58	0.67	0.08	0.44	97.53

22925	IV-V	colourless	69.49	18.56	6.18	2.00	0.62	0.51	0.47	0.07	0.20	98.10
2920	IV-V	colourless	67.85	18.02	6.37	2.29	0.54	0.80	0.65	0.11	1.04	97.67
23986	IV-V	pale green	63.57	19.26	5.99	2.54	0.56	0.91	1.32	0.37	1.71	96.22
23985	IV-V	pale green	63.93	18.84	6.07	2.52	0.58	0.90	1.43	0.34	1.72	96.33
2960	IV-V	blue-green	66.39	16.63	6.98	2.54	1.29	0.66	0.82	0.09	0.27	95.68
2965	IV-V	blue-green	66.88	17.00	6.87	2.37	0.83	0.56	0.63	0.12	0.43	95.68
2562	IV-V	pale green	63.77	18.98	5.96	2.35	0.51	0.86	0.82	0.19	1.25	94.69
2928	IV-V	pale blue	65.47	16.78	6.95	2.54	1.27	0.69	0.73	0.13	0.28	94.84
2927	IV-V	pale green	66.95	16.38	6.82	2.57	0.54	0.67	0.54	0.12	1.09	95.67
Tienen												
Tie 11	II	pale blue	69.17	18.93	8.41	2.60	0.53	0.52	0.28	0.02	0.42	100.87
Tie 12	II	pale blue	68.72	17.28	9.35	2.85	0.53	0.58	0.17	0.04	0.41	99.92
Tie 17	II	pale blue	70.17	17.55	7.29	2.48	0.66	0.45	0.06	0.06	0.42	99.12
Tie 24	II	pale blue	71.16	19.45	5.95	1.88	0.51	0.42	0.56	0.05	0.01	99.99
Tie 35	II	pale blue	69.65	17.24	8.30	2.68	0.72	0.56	0.48	0.04	0.98	100.64
Tie 37	II	pale blue	70.06	12.33	8.83	2.97	0.78	0.75	0.28	0.06	0.24	96.28
Tie 41	II	pale blue	69.95	16.56	8.12	2.90	0.71	0.51	0.28	0.04	0.40	99.46
Tie 45	II	pale blue	69.36	19.42	6.79	2.68	0.57	0.52	0.56	0.08	0.25	100.23
Tie 48	II	pale blue	69.23	16.45	8.80	2.92	0.63	0.49	0.06	0.04	0.36	98.96
Tie 49	II	pale blue	66.91	15.32	10.25	3.41	1.12	0.50	0.06	0.06	0.37	97.98
Tie 50	II	pale blue	70.14	17.13	8.40	2.69	0.69	0.45	0.59	0.04	0.36	100.47

Table 1. Main element composition of the considered samples. Major and minor elements are expressed in weight percent (wt%) of oxides.

Sample	Date	Colour	$^{87}\text{Sr}/^{86}\text{Sr}$	2σ	$^{143}\text{Nd}/^{144}\text{Nd}$	2σ	ϵNd	technique
Petra								
PE1	III-IV	colourless	0.70908	0.00002	0.512346	0.000010	-5.60	TIMS
PE2	III-IV	colourless	0.70904	0.00002	0.512172	0.000010	-8.90	TIMS
Barsinia								
BA1	III-IV	pale blue-green	0.70895	0.00002	0.512406	0.000010	-4.40	TIMS
BA3	III-IV	pale blue	0.70932	0.00033	0.512405	0.000070	-4.54	MC-ICP-MS
BA7	III-IV	green-yellow	0.70920	0.00042	0.512400	0.000065	-4.65	MC-ICP-MS
BA10	III-IV	green-yellow	0.70900	0.00002	0.512409	0.000010	-4.30	TIMS
Gonio								
Ge3	I-IV	green-yellow	0.70884	0.00036	0.512377	0.000065	-5.08	MC-ICP-MS
Ge5	I-IV	pale blue	0.70904	0.00033	0.512387	0.000059	-4.89	MC-ICP-MS
Ge8	I-IV	pale blue	0.70907	0.00036	0.512396	0.000061	-4.72	MC-ICP-MS
Iulia Felix								
IF B05	III	colorless	0.70904	0.00024	0.512354	0.000092	-5.54	MC-ICP-MS
IF B07	III	colorless	0.70911	0.00022	0.512351	0.000107	-5.60	MC-ICP-MS
IF B117	III	colorless	0.70904	0.00022	0.512370	0.000075	-5.23	MC-ICP-MS
IF C03	III	colorless	0.70914	0.00022	0.512357	0.000089	-5.47	MC-ICP-MS
IF C11	III	colorless	0.70906	0.00029	0.512356	0.000102	-5.50	MC-ICP-MS
IF C17	III	colorless	0.70906	0.00027	0.512361	0.000117	-5.41	MC-ICP-MS
IF P3	III	colorless	0.70908	0.00022	0.512374	0.000265	-5.15	MC-ICP-MS
IF P20	III	colorless	0.70912	0.00026	0.512342	0.000118	-5.78	MC-ICP-MS
IF P23	III	colorless	0.70908	0.00030	0.512375	0.000127	-5.14	MC-ICP-MS
IF B10	III	colorless	0.70905	0.00023	0.512313	0.000383	-6.34	MC-ICP-MS
IF B112	III	colorless	0.70903	0.00023	0.512344	0.000062	-5.74	MC-ICP-MS
IF B68	III	colorless	0.70884	0.00023	0.512356	0.000086	-5.50	MC-ICP-MS
IF B82	III	colorless	0.70878	0.00025	0.512414	0.000130	-4.36	MC-ICP-MS
IF B104	III	colorless	0.70872	0.00026	0.512402	0.000074	-4.60	MC-ICP-MS
Embiez								
103	II-III	colorless	0.70903	0.00008	0.512367	0.000158	-5.30	MC-ICP-MS
152	II-III	colorless	0.70902	0.00004	0.512372	0.000128	-5.19	MC-ICP-MS
160	II-III	colorless	0.70900	0.00005	0.512380	0.000074	-5.03	MC-ICP-MS
258	II-III	colorless	0.70904	0.00005	0.512367	0.000103	-5.30	MC-ICP-MS
259	II-III	colorless	0.70907	0.00004	0.512376	0.000151	-5.10	MC-ICP-MS
264	II-III	colorless	0.70902	0.00007	0.512443	0.000670	-3.80	MC-ICP-MS
265	II-III	colorless	0.70902	0.00007	0.512366	0.000083	-5.31	MC-ICP-MS
272	II-III	colorless	0.70902	0.00007	0.512367	0.000086	-5.28	MC-ICP-MS
622	II-III	colorless	0.70904	0.00007	0.512375	0.000151	-5.13	MC-ICP-MS
252	II-III	colorless	0.70883	0.00004	0.512415	0.000092	-4.36	MC-ICP-MS
385	II-III	colorless	0.70882	0.00004	0.512403	0.000085	-4.58	MC-ICP-MS
386	II-III	colorless	0.70882	0.00005	0.512407	0.000079	-4.51	MC-ICP-MS
390	II-III	colorless	0.70886	0.00007	0.512408	0.000100	-4.49	MC-ICP-MS
397	II-III	colorless	0.70881	0.00008	0.512406	0.000070	-4.52	MC-ICP-MS
gobA	II-III	colorless	0.70896	0.00006	0.512370	0.000089	-5.23	MC-ICP-MS
gobB	II-III	colorless	0.70903	0.00010	0.512364	0.000096	-5.35	MC-ICP-MS
gobC	II-III	colorless	0.70899	0.00007	0.512362	0.000089	-5.39	MC-ICP-MS
gobD	II-III	colorless	0.70903	0.00006	0.512369	0.000079	-5.24	MC-ICP-MS
Augusta Praetoria								
1	I-III	blue-green	0.70889	0.00003	0.512380	0.000034	-5.03	MC-ICP-MS
09A	I-II	green	0.70888	0.00002	0.512305	0.000031	-6.49	MC-ICP-MS

12A	II-III	green-yellow	0.70902	0.00003	0.512335	0.000036	-5.91	MC-ICP-MS
12B	I-II	blue-green	0.70929	0.00000	0.512411	0.000310	-4.44	MC-ICP-MS
22E	I-II	blue-green	0.70897	0.00001	0.512406	0.000030	-4.53	MC-ICP-MS
23G	I-II	blue-green	0.70902	0.00005	0.512330	0.000030	-6.00	MC-ICP-MS
08B	II-III	green-yellow	0.70921	0.00008	0.512315	0.000039	-6.29	MC-ICP-MS
09B	II-III	green-yellow	0.70902	0.00007	0.512363	0.000038	-5.36	MC-ICP-MS
15	IV	blue-yellow	0.70890	0.00000	0.512429	0.000090	-4.1	MC-ICP-MS
20F	III-IV	green-yellow	0.70920	0.00000	0.512344	0.000040	-5.73	MC-ICP-MS
5	I-II	blue-green	0.70909	0.00003	0.512397	0.000031	-4.71	MC-ICP-MS
7	I-II	blue-green	0.70909	0.00002	0.512392	0.000037	-4.79	MC-ICP-MS
21B	I-III	blue-yellow	0.70864	0.00003	0.512385	0.000031	-4.94	MC-ICP-MS
23E	I-II	blue-green	0.70908	0.00005	0.512389	0.000030	-4.86	MC-ICP-MS
23I	I-II	blue-green	0.70903	0.00002	0.512397	0.000048	-4.70	MC-ICP-MS
20E	III-IV	green-yellow	0.70914	0.00008	0.512346	0.000038	-5.69	MC-ICP-MS
22D	I-II	green-yellow	0.70905	0.00005	0.512362	0.000053	-5.38	MC-ICP-MS
23D	II	green	0.70914	0.00002	0.512400	0.000038	-4.64	MC-ICP-MS
Sant Boi de Llobregat								
PD2	I	blue	0.70897	0.00002	0.512375	0.000010	-5.12	TIMS
PD4	I	colourless	0.70904	0.00002	0.512330	0.000010	-6.00	TIMS
PD6	I	green-yellow	0.70926	0.00002	0.512217	0.000010	-8.20	TIMS
PD9	I	pale blue	0.70896	0.00002	0.512371	0.000010	-5.22	TIMS
PD10	I	pale blue	0.70896	0.00002	0.512414	0.000010	-4.44	TIMS
PD11	I	pale blue	0.70907	0.00002	0.512360	0.000010	-5.43	TIMS
PD14	I	colourless	0.70908	0.00002	0.512410	0.000010	-4.44	TIMS
PD15	I	colourless	0.70906	0.00002	0.512439	0.000010	-3.90	TIMS
PD16	I	purple	0.70864	0.00002	0.512391	0.000010	-4.70	TIMS
PD28	IV-V	pale blue	0.70878	0.00040	0.512385	0.000036	-4.94	MC-ICP-MS
PD29	IV-V	colourless	0.70912	0.00040	0.512347	0.000118	-5.68	MC-ICP-MS
Oudenburg								
71310	III	pale blue	0.70947	0.00040	0.512351	0.000067	-5.61	MC-ICP-MS
7929	III	colourless	0.70904	0.00042	0.512374	0.000055	-5.16	MC-ICP-MS
23993	IV-V	blue-green	0.70918	0.00036	0.512326	0.000075	-6.09	MC-ICP-MS
22925	IV-V	colourless	0.70913	0.00038	0.512360	0.000081	-5.42	MC-ICP-MS
2920	IV-V	colourless	0.70908	0.00037	0.512372	0.000054	-5.19	MC-ICP-MS
2960	IV-V	blue-green	0.70930	0.00041	0.512303	0.000067	-6.54	MC-ICP-MS
Tienen								
Tie 11	II	pale blue	0.70893	0.00001	0.512511	0.000009	-2.5	TIMS
Tie 12	II	pale blue	0.70899	0.00002	0.512267	0.000009	-7.2	TIMS
Tie 17	II	pale blue	0.70902	0.00001	0.512378	0.000010	-5.1	TIMS
Tie 24	II	pale blue	0.70902	0.00002	0.512376	0.000013	-5.1	TIMS
Tie 35	II	pale blue	0.70886	0.00001	0.512219	0.000009	-8.2	TIMS
Tie 37	II	pale blue	0.70891	0.00002	0.512083	0.000006	-10.8	TIMS
Tie 41	II	pale blue	0.70901	0.00001	0.512337	0.000009	-5.9	TIMS
Tie 45	II	pale blue	0.70904	0.00001	0.512174	0.000008	-9.1	TIMS
Tie 48	II	pale blue	0.70896	0.00001	0.512262	0.000005	-7.3	TIMS
Tie 49	II	pale blue	0.70759	0.00001	0.512249	0.000010	-7.6	TIMS
Tie 50	II	pale blue	0.70898	0.00001	0.512362	0.000008	-5.4	TIMS

Table 2. Isotopic composition for the selected samples.