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# Geochemical heterogeneity of sand deposits and its implications for the provenance determination of Roman glass

Dieter Brems\* , Jente Pauwels, Annelore Blomme, Rebecca B. Scott, and Patrick Degryse

Department of Earth and Environmental Sciences, KU Leuven, Leuven, Belgium

**Abstract** During the production of natron glass most features of the raw materials are lost and only some bulk geochemical characteristics have potential as provenance indicators. To determine the primary origin of archaeological glass artefacts, suitable sand raw materials have to be accurately characterised. In this respect, information about the possible variation in geochemical properties within a silica source is also vital to account for potential (partial) overlap of different sources. In this study, it is shown that the variation in major and minor elemental composition of beach sand on a local scale is smaller than the variation in Roman natron glass. Therefore, a single sand deposit can be seen as a relatively homogeneous source of silica for glass production. Nd isotopic signatures are identical for all samples analysed. The isotopic composition of Sr however varies considerably due to local variations in the relative proportions of carbonates and silicates (mostly feldspar).

**Keywords** provenance studies; natron glass; raw materials; beach sand; Sr isotopes; Nd isotopes

Subject classification codes: include these here if the journal requires them

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## Introduction and objectives

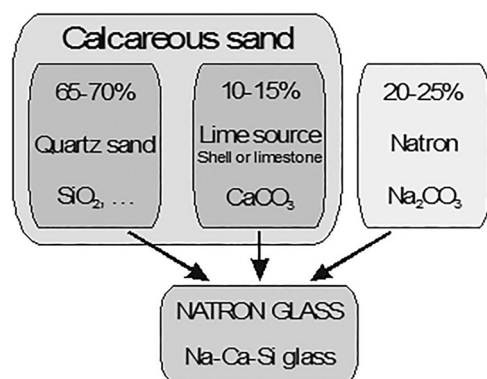
Natron glass was produced by melting quartz sand, natron and lime at temperatures of about 1100°C (Fig. 1). As a result of the use of natron as a relatively pure source of soda, this type of soda–lime–silica glass has characteristically low K<sub>2</sub>O and MgO concentrations which readily discriminates it from plant ash glass (Brill 1970). Natron glass dominates the archaeological record in the Mediterranean area and the rest of Europe from the fifth century BC until the ninth century AD (Freestone 2006; Sayre and Smith 1961; Wedepohl, Simon and Kronz 2011).

Large quantities of natron glass were produced in large tank furnaces in a process called primary production (Freestone, Gorin-Rosen and Hughes 2000; Freestone, Ponting and Hughes 2002; Freestone, Greenwood and Gorin-Rosen 2002; Gorin-Rosen 1995, 2000; Nenna, Picon and Vichy 2000; Picon and Vichy 2003; Tal, Jackson-Tal and Freestone 2004). This raw glass was then traded throughout the known world to be remelted and shaped into objects in secondary workshops (Foy, Picon and Vichy 2000; Freestone, Gorin-Rosen and Hughes 2000; Nenna, Picon and Vichy 2000). Numerous secondary production

sites are found throughout the Mediterranean and Europe (Lauwers 2007; Nenna 2007; Price 2005). Archaeological evidence for primary production centres is far less common. Only a limited number of these primary workshops were unearthed in Egypt and Syro-Palestine (Freestone and Gorin-Rosen 1999; Gorin-Rosen 1995, 2000; Picon and Vichy 2003; Tal, Jackson-Tal and Freestone 2004). During the Late Roman and Byzantine period, raw natron glass was probably exclusively produced in these regions. For the earlier Hellenistic and Roman period, it has been suggested that primary natron glass production took place in the same region (i.e. the eastern Mediterranean), but also in the western part of the Empire (Brems et al. 2012a; Degryse and Schneider 2008; Degryse et al. 2014). However, no primary furnaces dating to this period have been found and their exact location remains unknown. A number of classical writings from the first century AD, such as those of Pliny the Elder, mention the production of raw glass using sand raw materials from Italy, Gaul and Spain, but this was never confirmed by archaeological finds.

The provenance determination of ancient glass artefacts is complicated by the non-straightforward relationship between the raw materials used and the

\*Corresponding author, email: dieter.brems@kuleuven.be

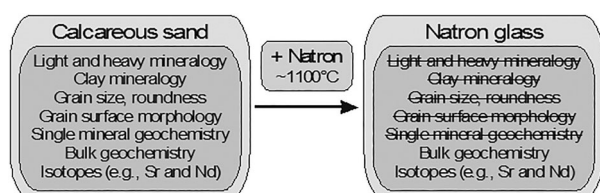


**Figure 1** Schematic illustration of the different components making up a glass batch.

resulting glass. During the primary glass production process the raw materials are completely melted, mixed and homogenised. As a result, their original crystallographic and mineralogical features are lost and only some geochemical characteristics are preserved, which can be useful to provenance ancient glass (Fig. 2). Since natron glass is essentially a blend of three major ingredients, i.e. quartz sand, natron and lime, the composition of the glass will be a mixture of components coming in with these raw materials. Furthermore, ancient glass was often coloured or decoloured by adding small amounts of specific minerals. These colourants would introduce a wide variety of trace elements, which do not normally occur in high concentrations in the other raw materials (Brems and Degryse 2014a, c).

As an extra complication, it is possible that during the actual manufacturing of glass objects in secondary workshops raw glass from more than one primary factory was mixed (Freestone, Ponting and Hughes 2002; Freestone, Price and Cartwright 2006). Also recycling of glass cullet appears to have been common practice (Degryse et al. 2006a; Freestone, Ponting and Hughes 2002; Jackson 1997; Mirti, Lepora and Saguì, 2000; Mirti et al. 2001). As a result, the geochemical characteristics of the final glass artefact will combine features of all these different sources, blurring the original (primary) signatures.

So how can it be proven that primary Roman natron glass was produced outside of the known production places in Israel and Egypt? Without any actual



**Figure 2** Schematic representation of the transformation of sand raw materials into natron glass with an indication of different characteristics that can be useful for provenance studies.

archaeological evidence from excavations or any detailed information from ancient texts, it is very difficult to pinpoint the exact location of any primary production centres in other parts of the Empire.

Recently, researchers have taken a different approach to the problem by studying the geochemical composition of potential glassmaking raw materials and mapping the distribution of suitable sand sources. A first substantial database with major and minor elemental data for potential sand raw materials from the western Mediterranean was presented by Brems et al. (2012b). Supplementary isotopic and trace element data were published by Brems et al. (2013a, b) and Brems and Degryse (2014a). These databases provide a first frame of reference for glass studies. It was shown that suitable sand raw materials for natron glass production are relatively rare. Only a limited number of possible sources could be identified in the western Mediterranean (Brems and Degryse 2014b; Brems et al. 2012b, c).

After the identification of potentially suitable sand raw materials, an accurate characterisation of their geochemical properties is essential in order to correlate them with the different glass groups that have been identified. In this respect, information about the possible variation in these geochemical properties within a silica source is also vital to account for potential (partial) overlap of different sources. Variations in the geochemical characteristics within a sand deposit can occur due to a heterogeneous distribution of different mineral phases caused by for example different local hydraulic conditions. For instance, the local concentration of heavy minerals would cause higher concentrations of  $\text{Fe}_2\text{O}_3$  and  $\text{TiO}_2$ . Variations in the ratio of shell fragments to feldspar could result in changes in the Sr isotopic signature of the sand.

The aim of this research paper is to investigate the possible variation in geochemical properties within a single sand deposit. The focus will mainly be on the major and minor elemental compositions, since these are of the utmost importance for the glassmaking potential of the sand. The compositional variation in the sand deposit will be compared to the observed variation in the composition of ancient natron glass to determine whether or not a single sand deposit can be seen as a homogenous source of silica for glass production. Additionally the variation in the isotopic composition of Sr and Nd will be determined, since these isotopic signatures are thought to provide important information about the geological origin of the raw materials used and the primary provenance of ancient glass (Brems, Ganió and Degryse 2014; Brems et al. 2013a, b; Degryse and Schneider 2008; Degryse et al. 2014; Freestone et al. 2003; Wedepohl and Baumann 2000).

## Methods

A present-day beach sand was chosen in the Basilicata Region (SE Italy). Beach sands in this area are mostly

derived from Pliocene–Pleistocene sedimentary rocks and are potentially suitable for natron glass production (Brems and Degryse 2014b; Brems et al. 2012b). The studied beach sand deposit is located in Lido Quarantotto, between the mouths of the Basento and the Cavone Rivers, approximately 87 km south of Bari ( $N40^{\circ}19'30''$   $E016^{\circ}48'25''$ ). Over a distance of 600 m along the coastline, a sand sample was taken every 15 m (samples LQ 1 to LQ 40 taken from NE to SW). Additionally, samples were taken every 3 m along a profile perpendicular to the coast line (samples LQ 20/1 to LQ 20/12 taken from SE to NW). This resulted in a total of 52 sand samples. The sample locations are shown in Fig. 3. About 2 kg of sediment sample was collected from the upper 10 cm of sand, representing the contemporaneous sedimentation layer.

In the laboratory, all samples were oven dried. Major and minor element analysis of all sand samples were obtained using Inductively Coupled Plasma – Optical Emission Spectrometry (ICP–OES) after fusion with lithium metaborate ( $LiBO_2$ ). Precision is better than 5%. Accuracy is better than 10%. The amounts of volatile elements were determined via loss on ignition (L.O.I.) measurements. Details about the analytical procedures used can be found in Brems et al. (2012b).

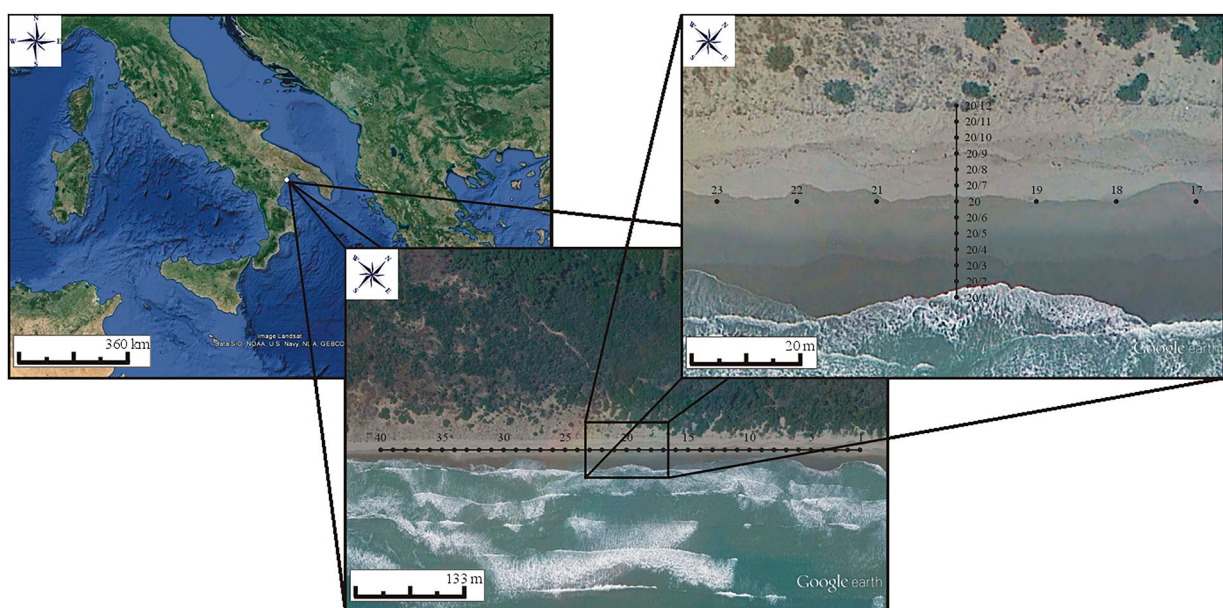
A selection of the sand samples were analysed for Sr and Nd isotope ratios. Along the coastline, a sample was analysed every 75 m (i.e. 9 samples in 600 m). Along the profile perpendicular to the coast, a sample was analysed every 9 m (i.e. 5 samples in 36 m). After separating Sr and Nd from the sample solutions using sequential extraction procedures,  $^{87}Sr/^{86}Sr$  and  $^{143}Nd/^{144}Nd$  isotope ratios were measured using Multi Collector – Inductively Coupled Plasma – Mass Spectrometry (MC–ICP–MS) (De Muynck et al. 2009; Ganio et al. 2012).

## Results and discussion

The results of the major and minor element analyses, the L.O.I. measurements, and the Sr–Nd isotopic ratios of the analysed beach sands are listed in Table 1 and Table 2 shows the correlation matrix for these geochemical properties. The major and minor elements in the beach sands analysed can be roughly assigned to 3 mineral groups: (1) (alumino-) silicates, (2) carbonates, and (3) heavy and accessory minerals.

(Alumino-) silicates such as quartz and feldspar are mainly composed of  $SiO_2$ ,  $Al_2O_3$  and  $K_2O$ . As a result, these elements all show moderate to strong positive correlations (correlation coefficients  $r$  between 0.74 and 0.97; Table 2).  $Na_2O$  also readily occurs in alkali feldspar. This usually results in a strong positive correlation between  $K_2O$  and  $Na_2O$ . According to the correlation matrix (Table 2) these elements only have a correlation coefficient of 0.73. A  $K_2O$ – $Na_2O$  biplot (Fig. 4) shows that two of the sand samples do not follow the general trend and have relatively elevated  $Na_2O$  concentrations. These two samples (LQ 20/3 and LQ 20/4) were taken in a local depression on the beach along the profile perpendicular to the coastline. Here the sand was permanently wet. The excess  $Na_2O$  originates from the salty seawater. If these two samples are excluded from the dataset,  $K_2O$  and  $Na_2O$  would have a correlation coefficient of 0.92 (Fig. 4).

Carbonate minerals such as calcite, aragonite and dolomite are the main source of CaO and MgO. Also Mn occurs as divalent cations and can be incorporated in carbonates. When heated to temperatures of 1000° C, carbonate minerals decompose with the release of  $CO_2$ . Hence they are the major contributor to the L.O.I. fraction. Consequently, CaO, MgO and MnO



**Figure 3** Sampling locations: 40 sand samples were taken along the coastline with a spacing of 15 m and 12 samples were taken perpendicular to the coastline with a spacing of 3 m (Google Earth).

**Table 1** Results of the elemental analyses, L.O.I. measurements and Sr-Nd isotopic analyses. All results for the major and minor elements and L.O.I. are in Wt.%. (nd. = not determined; SD = standard deviation)

	LQ 1	LQ 2	LQ 3	LQ 4	LQ 5	LQ 6	LQ 7	LQ 8	LQ 9	LQ 10	LQ 11	LQ 12	LQ 13
SiO <sub>2</sub>	68.38	67.59	68.83	66.08	65.97	65.98	70.70	69.29	67.63	67.45	67.79	67.44	69.54
Al <sub>2</sub> O <sub>3</sub>	4.07	3.84	4.00	3.84	3.75	3.92	4.27	4.22	3.81	3.76	3.95	3.77	4.06
K <sub>2</sub> O	1.15	1.05	1.14	0.98	0.98	1.09	1.28	1.22	1.03	1.01	1.10	1.04	1.16
Na <sub>2</sub> O	0.81	0.77	0.79	0.74	0.74	0.76	0.86	0.85	0.76	0.73	0.76	0.74	0.79
CaO	13.79	14.27	13.60	14.47	14.48	12.93	12.46	13.48	14.02	14.20	14.13	14.44	13.58
MgO	1.48	1.59	1.43	1.83	1.72	1.43	1.31	1.47	1.60	1.71	1.52	1.54	1.42
MnO	0.10	0.10	0.10	0.12	0.12	0.10	0.10	0.10	0.11	0.12	0.10	0.11	0.10
Fe <sub>2</sub> O <sub>3</sub>	1.58	1.63	1.59	1.97	1.95	1.77	1.61	1.67	1.77	2.00	1.68	1.66	1.64
TiO <sub>2</sub>	0.15	0.19	0.18	0.42	0.41	0.34	0.20	0.22	0.32	0.55	0.22	0.23	0.19
P <sub>2</sub> O <sub>5</sub>	0.03	0.02	0.03	0.05	0.05	0.05	0.04	0.04	0.05	0.11	0.04	0.04	0.03
LOI	11.67	12.24	11.70	12.57	12.56	11.44	10.63	11.55	12.07	12.29	12.05	12.25	11.57
<sup>87</sup> Sr/ <sup>86</sup> Sr	0.71073	nd.	nd.	nd.	0.71039	nd.	nd.	nd.	nd.	0.71048	nd.	nd.	nd.
2s	0.00010	nd.	nd.	nd.	0.00009	nd.	nd.	nd.	nd.	0.00012	nd.	nd.	nd.
<sup>143</sup> Nd/ <sup>144</sup> Nd	0.512141	nd.	nd.	nd.	0.512120	nd.	nd.	nd.	nd.	0.512092	nd.	nd.	nd.
2s	0.000061	nd.	nd.	nd.	0.000058	nd.	nd.	nd.	nd.	0.000057	nd.	nd.	nd.
ε <sub>Nd</sub>	-9.70	nd.	nd.	nd.	-10.10	nd.	nd.	nd.	nd.	-10.66	nd.	nd.	nd.
	LQ 14	LQ 15	LQ 16	LQ 17	LQ 18	LQ 19	LQ 20	LQ 21	LQ 22	LQ 23	LQ 24	LQ 25	LQ 26
SiO <sub>2</sub>	68.44	71.56	71.05	67.92	73.06	70.22	71.31	69.27	73.03	70.08	68.55	71.87	71.08
Al <sub>2</sub> O <sub>3</sub>	4.03	4.08	4.22	4.21	4.24	4.09	4.35	4.32	4.77	4.48	4.34	4.64	4.61
K <sub>2</sub> O	1.11	1.20	1.25	1.17	1.28	1.18	1.31	1.27	1.53	1.31	1.24	1.48	1.40
Na <sub>2</sub> O	0.79	0.81	0.85	0.82	0.88	0.82	0.87	0.84	1.06	0.87	0.84	0.92	0.90
CaO	13.83	12.76	13.21	14.38	11.76	13.43	12.69	13.35	11.13	13.16	14.03	11.85	12.25
MgO	1.51	1.33	1.31	1.51	1.20	1.40	1.27	1.39	1.10	1.38	1.46	1.17	1.28
MnO	0.10	0.10	0.09	0.10	0.09	0.10	0.09	0.09	0.08	0.09	0.10	0.09	0.09
Fe <sub>2</sub> O <sub>3</sub>	1.75	1.60	1.56	1.73	1.49	1.56	1.51	1.56	1.48	1.62	1.64	1.47	1.63
TiO <sub>2</sub>	0.25	0.22	0.16	0.19	0.13	0.14	0.12	0.13	0.11	0.14	0.14	0.11	0.17
P <sub>2</sub> O <sub>5</sub>	0.04	0.03	0.03	0.03	0.01	0.04	0.02	0.03	0.02	0.03	0.03	0.03	0.04
LOI	11.71	10.57	10.94	12.14	10.06	11.49	11.18	11.58	9.52	11.43	12.07	10.34	10.40
<sup>87</sup> Sr/ <sup>86</sup> Sr	nd.	0.71094	nd.	nd.	nd.	nd.	0.71099	nd.	nd.	nd.	nd.	0.71125	nd.
2s	nd.	0.00013	nd.	nd.	nd.	nd.	0.00009	nd.	nd.	nd.	nd.	0.00012	nd.
<sup>143</sup> Nd/ <sup>144</sup> Nd	nd.	0.512139	nd.	nd.	nd.	nd.	0.512156	nd.	nd.	nd.	nd.	0.512144	nd.
2s	nd.	0.000053	nd.	nd.	nd.	nd.	0.000056	nd.	nd.	nd.	nd.	0.000080	nd.
ε <sub>Nd</sub>	nd.	-9.74	nd.	nd.	nd.	nd.	-9.39	nd.	nd.	nd.	nd.	-9.64	nd.

	LQ 27	LQ 28	LQ 29	LQ 30	LQ 31	LQ 32	LQ 33	LQ 34	LQ 35	LQ 36	LQ 37	LQ 38	LQ 39
SiO <sub>2</sub>	71.70	66.95	71.82	70.87	70.00	70.72	72.33	69.85	71.22	72.02	70.39	69.66	69.25
Al <sub>2</sub> O <sub>3</sub>	4.58	4.34	4.65	4.27	4.31	4.23	4.28	3.97	4.17	3.98	3.79	3.95	4.01
K <sub>2</sub> O	1.45	1.24	1.46	1.26	1.29	1.27	1.31	1.17	1.26	1.13	1.12	1.15	1.12
Na <sub>2</sub> O	0.89	0.84	0.90	0.86	0.88	0.85	0.86	0.79	0.83	0.83	0.77	0.79	0.80
CaO	12.16	14.55	12.07	12.84	12.79	12.96	12.16	13.30	12.85	12.81	13.40	13.47	14.04
MgO	1.22	1.46	1.22	1.25	1.21	1.23	1.18	1.26	1.22	1.21	1.23	1.34	1.38
MnO	0.09	0.10	0.09	0.09	0.09	0.09	0.09	0.09	0.09	0.09	0.09	0.09	0.10
Fe <sub>2</sub> O <sub>3</sub>	1.56	1.67	1.54	1.52	1.46	1.51	1.42	1.46	1.46	1.42	1.41	1.49	1.60
TiO <sub>2</sub>	0.12	0.13	0.13	0.11	0.11	0.11	0.12	0.11	0.11	0.12	0.11	0.14	0.14
P <sub>2</sub> O <sub>5</sub>	0.03	0.03	0.05	0.02	0.03	0.04	0.04	0.03	0.03	0.04	0.04	0.03	0.01
LOI	10.29	12.71	10.33	10.81	10.85	10.99	10.41	11.12	10.98	10.77	11.11	11.42	11.75
<sup>87</sup> Sr/ <sup>86</sup> Sr	nd.	nd.	nd.	0.71100	nd.	nd.	nd.	nd.	0.71103	nd.	nd.	nd.	nd.
2s	nd.	nd.	nd.	0.00010	nd.	nd.	nd.	nd.	0.00012	nd.	nd.	nd.	nd.
<sup>143</sup> Nd/ <sup>144</sup> Nd	nd.	nd.	nd.	0.512145	nd.	nd.	nd.	nd.	0.512144	nd.	nd.	nd.	nd.
2s	nd.	nd.	nd.	0.000062	nd.	nd.	nd.	nd.	0.000052	nd.	nd.	nd.	nd.
ε <sub>Nd</sub>	nd.	nd.	nd.	-9.63	nd.	nd.	nd.	nd.	-9.64	nd.	nd.	nd.	nd.
	LQ 40	LQ 20/1	LQ 20/2	LQ 20/3	LQ 20/4	LQ 20/5	LQ 20/6	LQ 20/7	LQ 20/8	LQ 20/9	LQ 20/10	LQ 20/11	LQ 20/12
SiO <sub>2</sub>	73.43	73.91	76.94	75.04	70.04	70.56	70.53	68.90	67.86	68.02	67.99	67.33	69.72
Al <sub>2</sub> O <sub>3</sub>	4.33	4.96	4.76	4.90	4.45	4.28	4.12	4.02	4.03	3.70	3.63	3.67	3.59
K <sub>2</sub> O	1.30	1.63	1.62	1.66	1.34	1.25	1.21	1.11	1.11	0.99	0.97	1.01	1.01
Na <sub>2</sub> O	0.89	0.96	0.96	1.33	1.41	0.90	0.89	0.80	0.86	0.72	0.74	0.74	0.71
CaO	11.45	10.61	9.39	9.70	12.14	12.65	12.98	13.53	14.15	14.27	14.42	14.48	13.75
MgO	1.14	1.01	0.87	1.02	1.39	1.31	1.38	1.59	1.61	1.65	1.54	1.43	1.44
MnO	0.08	0.07	0.07	0.07	0.09	0.09	0.09	0.10	0.11	0.11	0.11	0.10	0.10
Fe <sub>2</sub> O <sub>3</sub>	1.48	1.45	1.37	1.36	1.51	1.50	1.50	1.68	1.70	1.67	1.65	1.51	1.54
TiO <sub>2</sub>	0.13	0.10	0.09	0.10	0.13	0.13	0.13	0.22	0.23	0.26	0.26	0.16	0.20
P <sub>2</sub> O <sub>5</sub>	0.01	0.04	0.03	0.02	0.04	0.03	0.02	0.04	0.04	0.05	0.03	0.04	0.04
LOI	9.59	8.93	8.17	8.96	11.35	10.82	10.86	11.64	12.14	12.18	12.11	12.21	11.60
<sup>87</sup> Sr/ <sup>86</sup> Sr	0.71124	0.71178	nd.	nd.	0.71106	nd.	nd.	nd.	nd.	0.71045	nd.	nd.	0.71051
2s	0.00008	0.00010	nd.	nd.	0.00012	nd.	nd.	nd.	nd.	0.00008	nd.	nd.	0.00013
<sup>143</sup> Nd/ <sup>144</sup> Nd	0.512116	0.512137	nd.	nd.	0.512139	nd.	nd.	nd.	nd.	0.512144	nd.	nd.	0.512134
2s	0.000064	0.000055	nd.	nd.	0.000043	nd.	nd.	nd.	nd.	0.000064	nd.	nd.	0.000050
ε <sub>Nd</sub>	-10.19	-9.78	nd.	nd.	-9.74	nd.	nd.	nd.	nd.	-9.65	nd.	nd.	-9.84

**Table 2** Correlation matrix for the main and trace elements, L.O.I.,  $^{87}\text{Sr}/^{86}\text{Sr}$  and  $\epsilon_{\text{Nd}}$  values of the analysed sand samples.

	SiO <sub>2</sub>	Al <sub>2</sub> O <sub>3</sub>	K <sub>2</sub> O	Na <sub>2</sub> O	CaO	MgO	MnO	Fe <sub>2</sub> O <sub>3</sub>	TiO <sub>2</sub>	P <sub>2</sub> O <sub>5</sub>	LOI	$^{87}\text{Sr}/^{86}\text{Sr}$	$\epsilon_{\text{Nd}}$
SiO <sub>2</sub>	1.00												
Al <sub>2</sub> O <sub>3</sub>	0.74	1.00											
K <sub>2</sub> O	0.84	0.97	1.00										
Na <sub>2</sub> O	0.58	0.73	0.73	1.00									
CaO	-0.92	-0.81	-0.90	-0.71	1.00								
MgO	-0.92	-0.74	-0.85	-0.54	0.88	1.00							
MnO	-0.92	-0.76	-0.86	-0.66	0.90	0.96	1.00						
Fe <sub>2</sub> O <sub>3</sub>	-0.78	-0.50	-0.62	-0.47	0.66	0.87	0.89	1.00					
TiO <sub>2</sub>	-0.66	-0.58	-0.63	-0.45	0.52	0.77	0.80	0.91	1.00				
P <sub>2</sub> O <sub>5</sub>	-0.43	-0.34	-0.36	-0.28	0.32	0.48	0.51	0.61	0.73	1.00			
LOI	-0.95	-0.76	-0.86	-0.58	0.97	0.91	0.91	0.70	0.56	0.37	1.00		
$^{87}\text{Sr}/^{86}\text{Sr}$	0.90	0.96	0.98	0.52	-0.98	-0.93	-0.91	-0.72	-0.70	-0.47	-0.94	1.00	
$\epsilon_{\text{Nd}}$	0.34	0.35	0.38	0.22	-0.24	-0.44	-0.51	-0.71	-0.78	-0.68	-0.22	0.30	1.00

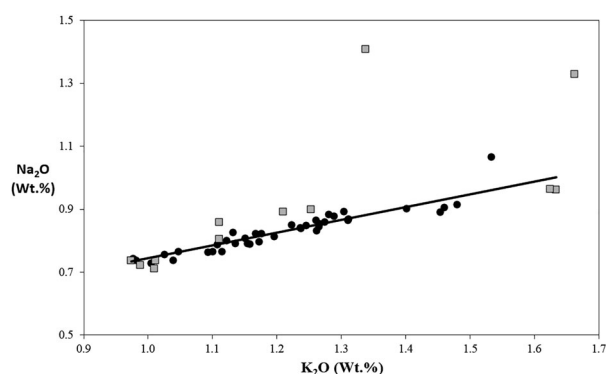
concentrations and the L.O.I. fraction are all strongly correlated in the sands analysed ( $r$  between 0.88 and 0.97; Table 2).

Heavy and accessory minerals such as magnetite, ilmenite, apatite, etc. contain the major part of the Fe<sub>2</sub>O<sub>3</sub>, TiO<sub>2</sub> and P<sub>2</sub>O<sub>5</sub>. Fe<sub>2</sub>O<sub>3</sub> and TiO<sub>2</sub> are strongly correlated ( $r = 0.91$ ; Table 2). The correlation between P<sub>2</sub>O<sub>5</sub> and the other elements is less good (Table 2). The correlation between MgO and Fe<sub>2</sub>O<sub>3</sub>, and between MnO and Fe<sub>2</sub>O<sub>3</sub> suggests that MgO and MnO are not exclusively related to the carbonate fraction of sand, but they are also brought in with the heavy minerals.

Negative correlations between elements attributed to the silicate minerals and the carbonate and heavy minerals are related to variations in the relative proportions of these different mineral groups in the sand (Table 2 and Fig. 5).

### Homogeneity of the major and minor elemental composition

Average compositions and standard deviations for the major and minor elemental compositions of the beach sands are presented in Table 3. In the same table the



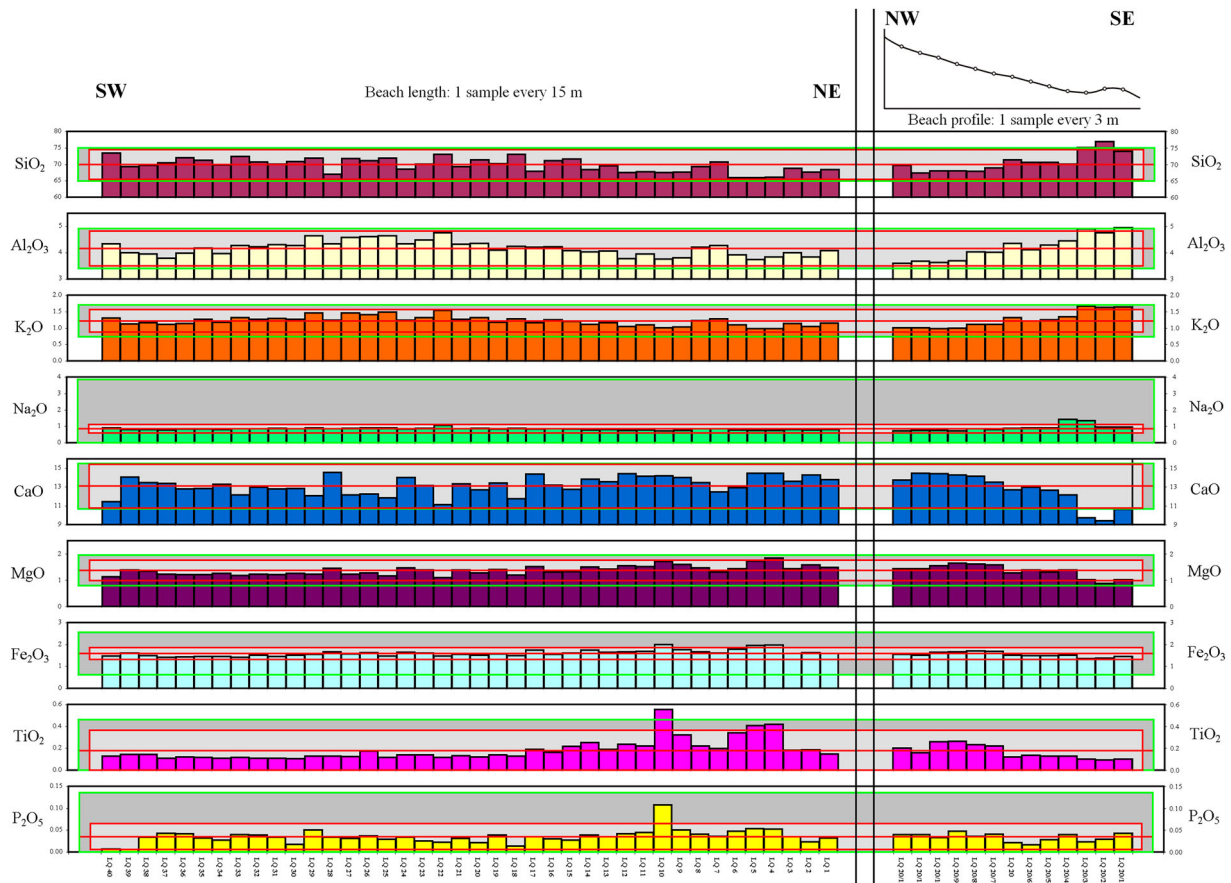
**Figure 4** Na<sub>2</sub>O vs. K<sub>2</sub>O plot for the beach sands analysed. The black circles correspond to the samples taken along the coastline. Grey squares correspond to samples taken along the profile perpendicular to the coast. The trendline ( $r = 0.92$ ) is calculated after the removal of the two samples with the highest Na<sub>2</sub>O concentration from the dataset.

mean values and standard deviations of Roman natron glass are also shown for comparison (Foster and Jackson 2009). The standard deviation is a good measure for the variation in the dataset.

The variation in major and minor elemental composition of the analysed beach sands is shown in Fig. 5. The results are shown from SW to NE along the coastline and from NW to SE along the profile perpendicular to the coast. For every element the variation (average  $\pm$  two times standard deviation, thus representing 95% of the dataset; Table 3) in the sand samples is shown in red. The variation in geochemical composition of Roman glass (four times standard deviation) is indicated by the green box (values given by Foster and Jackson (2009); Table 3).

For all of the major and minor elements the variation, as indicated by the standard deviation, is smaller in the sand deposit than in Roman glass. For SiO<sub>2</sub>, Al<sub>2</sub>O<sub>3</sub> and CaO the difference is relatively small. For K<sub>2</sub>O, MgO, Fe<sub>2</sub>O<sub>3</sub>, TiO<sub>2</sub> and P<sub>2</sub>O<sub>5</sub> the difference in variation is larger. Na<sub>2</sub>O concentrations show a much wider range in Roman natron glass than in beach sand. This can be attributed to the fact that relatively large quantities of soda were added to the glass batch to lower the melting temperature. The absolute concentrations and possible variation in MnO content is an order of magnitude larger in Roman natron glass than in beach sands analysed in this study (Table 3). This can be explained by the fact that MnO was regularly added to a batch of natron glass as a decolourising agent (Henderson 1985; Sayre 1963; Sayre and Smith 1961). The background level of MnO that can be attributed to the sand raw material is about 0.1% (Brems and Degryse 2014a, b, c; Brems et al. 2012b).

Several different glass groups have been identified in literature, such as Levantine 1, Levantine 2, Egypt 1, Egypt 2 and HIMT (Foy et al. 2003; Freestone 2006; Freestone, Gorin-Rosen and Hughes 2000; Freestone, Greenwood and Gorin-Rosen 2002; Freestone, Wolf and Thirlwall 2005; Nenna, Vichy and Picon, 1997). In Fig. 6 the variation in CaO and Al<sub>2</sub>O<sub>3</sub> concentrations for these glass types is compared to those found in the beach sands analysed in this study. The extent of



**Figure 5** Histograms showing the elemental composition of the beach sands analysed. The red boxes show the average composition  $\pm 2$  times the standard deviation of the sand samples. The green boxes represent the compositional ranges of Imperial Roman natron glass (4 times the standard deviation centred around the average of the sand compositions; Foster and Jackson, 2009). All values are in Wt.%.

the variation in this single sand deposit is comparable to that within the different glass groups. This indicates that sand from a single stretch of beach is relatively homogenous, but it also explains very well the compositional variations seen in ancient glass groups.

### Sr-Nd isotopic composition

The isotopic composition of Sr in natron glass is generally considered to be a proxy for the source of lime (Freestone et al. 2003; Wedepohl and Baumann 2000). When the lime entered the glass in the form of Holocene seashell, the isotopic composition of Sr in the glass reflects that of modern seawater which has a  $^{87}\text{Sr}/^{86}\text{Sr}$  ratio of approximately 0.7092 (Banner 2004). When the lime was derived from older limestone, the signature reflects that of seawater at the time the limestone was deposited, possibly modified by diagenesis. However, the  $^{87}\text{Sr}/^{86}\text{Sr}$  ratio in glass

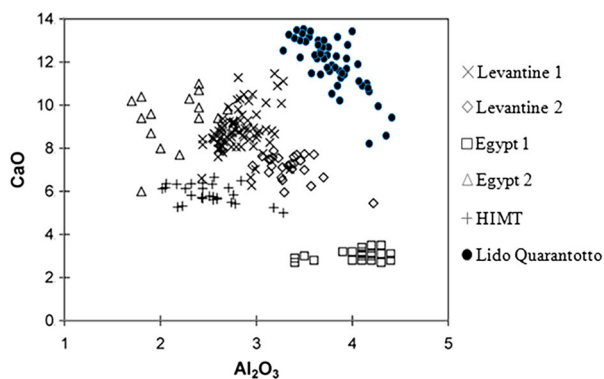
can be significantly influenced by the silicate fraction of the sand raw materials (Brems, Ganio and Degryse 2014; Brems et al. 2013a; Degryse et al. 2006b; Freestone et al. 2003). As a result, the  $^{87}\text{Sr}/^{86}\text{Sr}$  ratio in natron glass does not always provide a clear indication of the lime source. It could also be indicative for the source of the sand raw material, with ratios close to or below the present-day seawater signature pointing to an eastern Mediterranean origin and higher ratios suggesting an origin in the west (Brems, Ganio and Degryse 2014; Brems et al. 2013a).

The beach sands analysed in this study show a range of  $^{87}\text{Sr}/^{86}\text{Sr}$  isotope ratios between 0.71039 and 0.71178 (Table 1). The  $^{87}\text{Sr}/^{86}\text{Sr}$  ratio in sands from a single beach appears to vary considerably due to local variations in the relative proportions of carbonates and silicates (mostly feldspar). This is indicated by the strong correlations between the Sr

**Table 3** Comparison between the average composition and variation in geochemical composition of beach sand (this study) and Roman glass as given by Foster and Jackson (2009). (SD = standard deviation)

	SiO <sub>2</sub>	Al <sub>2</sub> O <sub>3</sub>	K <sub>2</sub> O	Na <sub>2</sub> O	CaO	MgO	MnO	Fe <sub>2</sub> O <sub>3</sub>	TiO <sub>2</sub>	P <sub>2</sub> O <sub>5</sub>	LOI
Mean sands	69.97	4.17	1.22	0.85	13.08	1.37	0.09	1.59	0.18	0.03	11.19
SD sands	2.29	0.33	0.17	0.12	1.17	0.19	0.01	0.14	0.09	0.01	0.98
Mean glass	69.54	2.59	0.75	16.63	7.48	0.59	0.73	0.62	0.13	0.12	/
SD glass	2.53	0.38	0.24	1.50	1.18	0.29	0.74	0.48	0.14	0.05	/

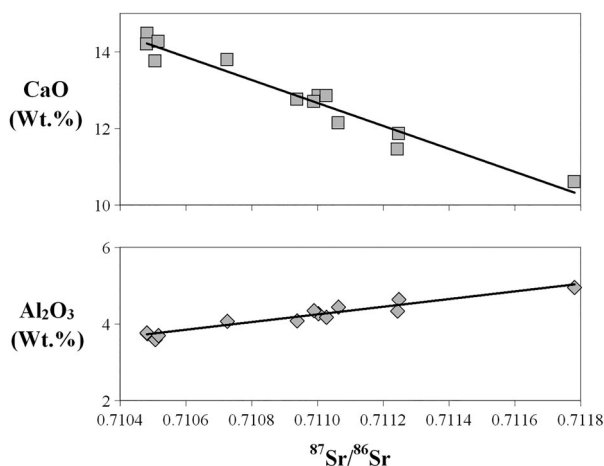




**Figure 6** CaO vs. Al<sub>2</sub>O<sub>3</sub> plot for a number of glass groups identified in literature (Freestone, 2006 and references therein) and the sand samples from Lido Quarantotto analysed in this study. All values are in Wt.%.

isotopic signature and the CaO and Al<sub>2</sub>O<sub>3</sub> concentrations ( $r = -0.98$  and  $0.96$  respectively; Fig. 7). Carbonate minerals derived from seashells and recycled limestone are a source of relatively non-radiogenic Sr (i.e. high CaO and low Al<sub>2</sub>O<sub>3</sub> contents and  $^{87}\text{Sr}/^{86}\text{Sr}$  ratios of 0.7092 or lower (Banner 2004)). Geologically old feldspar and minor mica derived from recycled Pliocene–Pleistocene sedimentary rocks provide relatively radiogenic Sr with high  $^{87}\text{Sr}/^{86}\text{Sr}$  ratios. Furthermore, these minerals have low CaO and high Al<sub>2</sub>O<sub>3</sub> contents. The measured  $^{87}\text{Sr}/^{86}\text{Sr}$  ratios in the sand deposit are a mixture of these two sources.

The isotopic composition of Nd varies throughout the earth's crust because of the differing nature and geological age of the rocks. The  $^{143}\text{Nd}/^{144}\text{Nd}$  isotope ratio and  $\epsilon_{\text{Nd}}$  values of sediments are usually a good indicator for their geological (and often geographical) provenance (Banner 2004). Nd in natron glass originates from the non-quartz mineral fraction of the silica raw material and hence can provide valuable information about the geological and possibly geographical provenance of the raw materials used (Brems, Ganio and Degryse 2014; Brems et al. 2013b;

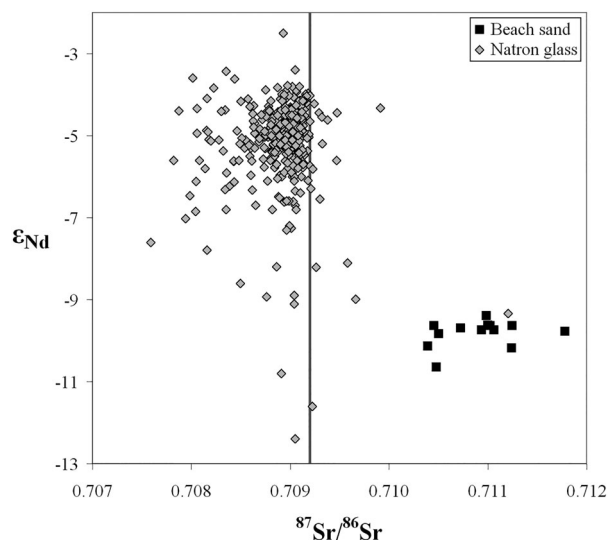


**Figure 7** CaO vs.  $^{87}\text{Sr}/^{86}\text{Sr}$  and Al<sub>2</sub>O<sub>3</sub> vs.  $^{87}\text{Sr}/^{86}\text{Sr}$  plots for the beach sands analysed.

Degryse and Schneider 2008; Degryse et al. 2014). The use of the isotopic signature of Nd as a proxy for the source of silica in glass is, however, not always straightforward because of the possible overlap of signatures from different suppliers (Brems, Ganio and Degryse 2014; Brems et al. 2013b).

The  $^{143}\text{Nd}/^{144}\text{Nd}$  isotope ratios in the analysed beach sands range from 0.51209 to 0.51216 (Table 1). This corresponds to  $\epsilon_{\text{Nd}}$  values between -10.7 and -9.4. All Nd isotopic signatures are identical within analytical error (2 standard deviations).  $\epsilon_{\text{Nd}}$  values do not show a strong correlation with the concentration of any of the major or minor elements, indicating that Nd does not originate from one specific mineral. This shows that the isotopic composition of Nd is a geochemical property capable of characterising a source of sand raw materials.

Degryse and co-workers compiled a substantial database of Sr-Nd isotopic signatures of natron glass from both primary production sites and consumption sites (Degryse and Schneider 2008; Degryse et al. 2006b, 2008, 2009, 2014; Ganio et al. 2012b, 2012c). The results of the Sr-Nd isotopic analyses of the beach sands are compared to this database in Fig. 8. The great majority of the natron glasses have  $\epsilon_{\text{Nd}}$  values higher than -7. These relatively high values are most likely indicative of a primary origin in the Eastern Mediterranean (Brems et al. 2012a; Degryse and Schneider 2008; Degryse et al. 2014).  $\epsilon_{\text{Nd}}$  values lower than -7 would suggest the glass was produced using raw materials from the Western part of the Mediterranean (Brems, Ganio and Degryse 2014; Brems et al. 2013b; Degryse and Schneider 2008; Degryse



**Figure 8** Sr-Nd isotopic composition of the beach sands analysed (blue squares) and natron glass (432 analyses from glass dating between the 1<sup>st</sup> cent. BC and the 9<sup>th</sup> cent. AD compiled from Brems et al. (2012a), Degryse and Schneider (2008), Degryse et al. (2006b, 2008, 2009, 2014) and Ganio et al. (2012b, 2012c)). The vertical line indicates the  $^{87}\text{Sr}/^{86}\text{Sr}$  ratio of present-day seawater.

et al. 2014). From the  $\epsilon_{\text{Nd}}$  vs.  $^{87}\text{Sr}/^{86}\text{Sr}$  plot (Fig. 8), it is clear that Nd isotopic signatures in sand samples from a single beach are relatively homogenous in comparison to the spread in ancient natron glass. This is very encouraging for the use of Nd isotopes as a provenance indicator of archaeological glass. The variation in Sr isotopic signatures in subsamples from a single beach sand deposit, however, is fairly large compared to that in a substantial database of natron glass from more than 30 archaeological sites across the extents of the Mediterranean world and the Roman Empire (Degryse et al. 2014). This important local variation in Sr isotope ratio within a single sand deposit is troublesome for its use for the provenance determination of natron glass. However, this large discrepancy does not mean that Sr isotopes can no longer be used in glass studies. The melting of a vast volume of sand in a tank furnace will definitely result in at least some degree of homogenisation in the geochemical characteristics of the glass. Therefore it will still be possible to distinguish different glass groups and define regional signatures, i.e. east vs. west, which together with other geochemical data (Nd isotopes and trace elements) can provide valuable information on the origin of ancient glass artefacts. However, pinpointing the exact location of a primary glass production centre based on the Sr-Nd isotopic signature of a glass artefact alone will never be possible.

## Conclusions

To determine the primary provenance of natron glass, a complete geochemical characterisation of any potentially suitable sand raw materials and of raw glass from any identified primary production centres is required. To account for overlap in geochemical signatures of different sources, information about the variation in these properties is important. In this study we investigated the magnitude of this possible variation along the stretch of a single beach in the Basilicata Region (SE Italy).

The major and minor elements in the beach sands can be roughly assigned to 3 mineral groups: (1) (alumino-) silicates, mainly composed of  $\text{SiO}_2$ ,  $\text{Al}_2\text{O}_3$ ,  $\text{K}_2\text{O}$  and  $\text{Na}_2\text{O}$ ; (2) carbonates, the main source of  $\text{CaO}$  and  $\text{MgO}$ ; and (3) accessory minerals containing the major part of the  $\text{Fe}_2\text{O}_3$ ,  $\text{TiO}_2$  and  $\text{P}_2\text{O}_5$ . Negative correlations between elements are related to variations in the relative proportions of these different mineral groups in the sand. The variation in major and minor elemental composition of beach sand on a local scale was found to be smaller than the variation in Roman natron glass. Therefore, a single sand deposit can be seen as a relatively homogeneous source of silica for glass production.

Nd isotopic signatures are fairly homogenous within a single source of sand. The isotopic composition of Sr in a single sand deposit can vary considerably due to local variations in the relative proportions of carbonates and silicates (mostly feldspar). This has

to be taken into consideration when attempting to use these isotopic signatures for the provenance determination of Roman natron glass.

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## ORCID

Dieter Brems  <http://orcid.org/0000-0003-2181-9392>

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