

# Blue Decoration on Glass Objects: Unravelling Origins and Significance in Late Medieval – Early Modern Portugal

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## Abstract

Late medieval and Early modern Portuguese archaeological contexts have brought to light several colourless objects with blue thread application. Usually related to cylindrical glasses and pedestal goblets, this decoration can be found in several features: one or more lines applied horizontally or in a spiral, frills forming festoons or blue lines combined with a ribbed decoration. The presence of these objects reveals a connection with the upper strata of society, as they were considered luxurious and could be identified as imported, raising questions about their provenance. This study examines 20 fragments of glass objects adorned with blue decoration from five Portuguese archaeological sites in Lisbon, Almada, and Setúbal, dating from the 14<sup>th</sup> to 16<sup>th</sup> centuries. Through a combination of micro particle-induced X-ray emission ( $\mu$ -PIXE) and inductively coupled plasma mass spectrometry with laser ablation (LA-ICP-MS), the chemical composition of the glass was examined. Stylistic investigations and analytical techniques shed light on how the raw materials used in objects with this type of decoration differ between older and more recent artefacts, indicating changes in society's taste and the raw materials used, hence the place of production. The chemical composition of the analysed fragments, which falls within the soda-silica-lime group, gives clues to their origin and leads to a debate about whether these objects are imported or locally produced. This interdisciplinary approach combining archaeological findings, stylistic analysis and chemical analysis, offers an understanding of the cultural and technological dynamics that shaped glass production in medieval and Renaissance Europe.

**Keywords:** archaeological glass, 14<sup>th</sup>- 16<sup>th</sup> centuries, Portugal,  $\mu$ -PIXE, LA-ICP-MS

## 41 1. Introduction

42 Recent studies of late medieval and early modern materials from archaeological excavations in Portugal  
43 have uncovered several glass objects characterised by their applied blue decoration, dating from the 14<sup>th</sup>  
44 to 16<sup>th</sup> century. The use of blue decoration on colourless glass objects has been documented in France  
45 as early as the 13<sup>th</sup> century (Foy & Sennequier, 1989, p. 266-267), and in the following century in Spain,  
46 Italy and France (Capellà Galmés, 2015; Fossati & Mannoni, 1975; Foy & Sennequier, 1989). Beakers  
47 and goblets with applied blue decoration have been produced since the late 14<sup>th</sup> century in Venice  
48 (Pause, 2000; Gallo & Silvestri, 2012), which became the main European production centre from the  
49 15<sup>th</sup> century onwards. Venetian innovations in technology and manufacturing were appreciated  
50 throughout Europe for the refinement of their products, which were exported to various courts. In  
51 several countries, glass began to be produced *à la façon de Venise* from the 16<sup>th</sup> century onwards, using  
52 local raw materials, often employing expatriate Venetian glassmakers (see, for example: de  
53 Rochebrune, 2004; Doménech, 2004; Henkes, 1994). Documentary information and some  
54 archaeological studies lead to the assumption that this phenomenon existed in Portugal as well  
55 (Coutinho, Medici, Alves, *et al.*, 2016; Lima *et al.*, 2012). Unfortunately, there is a lack of  
56 archaeological evidence in the country that indicates local production.

57 Considering the glass assemblages which these specimens belong to, they are mostly ordinary glass  
58 used for serving or consuming liquids, without decoration and with common shapes. It is therefore  
59 possible that these objects with blue decoration might have been luxury items and, in some cases,  
60 imports. Nevertheless, it is known that in the Middle Ages the colour blue is associated with prestige  
61 (Pastoureau, 2002)<sup>1</sup>. This research will combine the study of the archaeological context with the  
62 stylistic analysis of the glasses and their chemical characterisation, to use them as a basis for the  
63 discussion on provenance.

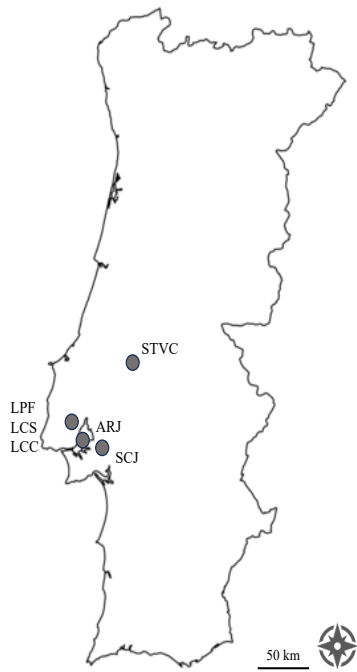
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<sup>1</sup> On several occasions, glass objects discovered in medieval and late medieval Portuguese excavations have been identified in coeval Portuguese and European works of art. These identifications have often provided valuable insights into the functions of the objects and the social contexts in which they were used (Medici, 2014; Valente, 1950).

In Coutinho, Medici, Coentro, *et al.*, 2016, the recognition of this type of glass is proposed in the painting from the Portuguese painters Vasco Fernandes and Francisco Henriques, 'Última Ceia', from the collection of the Museu Nacional Grão Vasco, Viseu, Portugal, and dated between 1501-1506. Looking closely at the painting through the advanced features of Google Art & Culture, a new interpretation emerges. Examination of the objects reveals details that reconsider the initial perception of a decoration with applied blue threads. Instead, it appears that the beaker has a distinct rim, characterised by an outward fold.

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## 2. Archaeological contexts and materials

The study focuses on 20 glass objects (Fig. 2, Tab. 1), from different archaeological excavations across Portugal (Fig. 1), that share a common characteristic: the presence of blue decorations applied to colourless bodies. Overall, the stylistic analysis identified these vessels as related to the consumption of liquids, belonging to beakers, pedestal beakers and goblets.

Among the oldest finds, three fragments dating from the 14<sup>th</sup>-15<sup>th</sup> centuries were excavated in Travessa das Capuchas in Santarém (STCV) and Rua da Judiaria in Almada (ARJ). These sites revealed silos reused as cesspits filled with various materials and, among the others, fragments of glass objects (Boavida *et al.*, 2013; Barros & Henriques, 2003). Furthermore,

in Praça da Figueira in Lisbon (LPF), a fragment of colourless glass with an opaque blue frill was brought to light in the layers related to a domestic dwelling in Rua da Betesga (Banha da Silva, 2018). This also dates back to the 14<sup>th</sup>-15<sup>th</sup> century and probably belonged to a high-ranking residence, a hypothesis put forward on the basis of the other materials found in the excavation (Cámara del Río, 2015).

Most of the fragments (=15) date back to the 16<sup>th</sup> century. One (LPF0038) was found during the excavation of the Hospital Real de Todos-Os-Santos in Praça da Figueira (LPF) in a layer associated with the reconstruction of a section of the hospital that occurred at the end of the 16<sup>th</sup> century (Bargão *et al.*, 2018). A group of objects was unearthed during excavations along the riverbank of the Tagus River in Lisbon, in the areas of Largo do Corpo Santo (LCS) and Campo das Cebolas (LCC). These finds are connected to a large dump site formed by piling up debris from demolitions caused by the earthquakes of 1531 or 1551 (Alves *et al.*, 2001, Manso & Abrantes Garcia, 2019). This area, known as *Ribeira Velha*, located near the banks of the Tagus River, was an intermediate space between the city and the waterways for communication and trade. The objects recovered likely reflect the daily life of the noble palaces built nearby, as well as Lisbon's commercial activities with Europe and the rest of the world (Manso & Abrantes Garcia, 2019). Additionally, four colourless fragments with blue decorations, dated to the late 16<sup>th</sup> to early 17<sup>th</sup> century, were found in a dump area outside the Convento de Jesus in Setúbal (SCJ) (Cândido, 2009).



Figure 1, Fragments considered in this work (Drawings: ARJ © T. Medici; STCV, LPF, LCS and SCJ © A. Cristoforetti; LCC © B. Farinha).

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102 Among the fragments analysed in this study (Fig. 2, Tab. 1), the most prevalent decorative feature is  
 103 the application of horizontal blue threads on the body of the vessel (n = 10). This decorative is identified  
 104 in items dating to the 14<sup>th</sup> century (STCV0005) and continues to appear in fragments from the 15<sup>th</sup>  
 105 (ARJ0999) and 16<sup>th</sup> centuries (LPF0038, SCJ0060, LCS1027, LCS0924, LCS1059, LCS0286,  
 106 LCC0029, LCC0013). This continuity highlights the enduring appreciation for this adornment across  
 107 these centuries. Objects with the same decorations are found in the production of Altare glassblowers,  
 108 present in the south of France between the 15<sup>th</sup> and 16<sup>th</sup> centuries (Foy & Sennequier, 1989) and in  
 109 Majorca (Capellà Galmés, 2015) and Italy, on vessels dated between the late 14<sup>th</sup> and early 15<sup>th</sup> century

110 from Savona (Ventura, 2001) and Monte Lecco, Liguria (Fossati & Mannoni, 1975), and in the south,  
111 in Apulia (Catacchio, 2020).

112 ARJ0452 and LPF0006, dated to the 15<sup>th</sup> century, show a frill decoration applied on a colourless wall  
113 adorned with regularly spaced vertical ribs. ARJ0452 conserves a complete profile of a goblet with a  
114 pedestal made from a single gather of glass, with a truncated conical bowl and a pushed-in base (Medici,  
115 2005). Given the presence of the same decorations and its curvature, it is proposed that LPF0006  
116 belonged to same shape. This applied blue festoon decoration is recognisable in the base of a colourless  
117 goblet dated to the 14<sup>th</sup> century, decorated with an applied blue frill, of Majorcan production or under  
118 the crown of Aragon (Capellà Galmés, 2015).

119 SCJ0058 is characterised by a blue thread decoration applied to the rim and an undecorated body. A  
120 similar fragment, the dating or provenance of which are unknown, was found in Beja (Praça Miguel  
121 Fernandes) (Medici, 2014). Comparisons of this type of decoration are attested in Spain, dated to the  
122 14<sup>th</sup>-15<sup>th</sup> centuries (Medici *et al.*, 2009) and in Italy between the 14<sup>th</sup> and 16<sup>th</sup> centuries in Pavia (Corpus  
123 Pavia, 2004), Venice (Gallo & Silvestri, 2012; Pause, 2000) and Altare (Ventura, 2001). Furthermore,  
124 the production of objects with this decoration is attested in France from the 14<sup>th</sup> century onwards (Foy  
125 & Sennequier 1989) and in Austria in the first half of the 16<sup>th</sup> century (Tarcsay, 2018).

126 SCJ0078 belongs to a colourless cylindrical beaker with blue thread applied to the rim and a body  
127 decorated with densely spaced twisted ribs, resulting from mould-blowing. Notably, two direct  
128 comparisons from Beja, Portugal, dated between the 14<sup>th</sup> and 15<sup>th</sup> centuries, exhibit similar  
129 characteristics (Coutinho, Medici, Coentro, *et al.*, 2016).

130 LCS0883, LCS0836, and SCJ0059 have, in addition to a blue thread applied to the wall approximately  
131 one centimetre below the rim, an oblique ribbed decoration probably obtained *mezza stamptatura*,  
132 invented in Murano (Mentasti Barovier & Tonini, 2019) and widespread in several European *façon de*  
133 *Venice* productions (Philippart & Mergenthaler, 2011; Liefkes, 2004; Doménech, 2004). In Portugal,  
134 an example of goblets (16<sup>th</sup> century) with this same decoration was found in Carnide, in Largo do Coreto  
135 (Boavida & Medici, 2018).

136 LCC0019 is part of a beaker with a truncated conical body and pushed-in base, decorated with a pinched  
137 thread applied. The body is adorned with a blue thread applied to the body of the vessel, enamelled  
138 spots near the rim and vertical ribs alternating with regular spaces, made using the *mezza stamptatura*  
139 technique typical of 16<sup>th</sup> century Venetian production. Objects with similar features have been found in  
140 Venetian production starting from the 14<sup>th</sup> century (Pause, 2000) and in *façon de Venice* glass produced  
141 in Antwerp in the second half of the 16<sup>th</sup> century (Veeckman, 2002). Beakers with comparable

142 decorative motifs, identified as Venetian imports dated between the 15<sup>th</sup> and 16<sup>th</sup> centuries, have been  
 143 unearthed in the Tirol region (Tarcsay, 2024) .

144 *Table 1, Fragments under study, divided based on their decoration*

<i>Fragment</i>	<i>Part preserved</i>	<i>Shape</i>	<i>Decoration</i>	<i>Age Century (CE)</i>
ARJ0452	Wall with rim and pedestal base with tubular ring base	Pedestal goblet		15 <sup>th</sup> cent.
STCV0005	Part of wall with rim	Beaker		
LPF0038	Part of wall with rim	Beaker		
LCS0286	Wall		Horizontal threads applied on the body	16 <sup>th</sup> cent.
LCS1059	Wall			
LCS1027	Part of wall with rim	Beaker		
LCS0924	Wall	Beaker (?)		
SCJ0060	Part of wall with rim	Beaker		
ARJ0099	Part of wall with rim and pedestal base with tubular ring base	Pedestal goblet	Horizontal frill applied on the body	15 <sup>th</sup> cent.
LPF0006	Part of wall with blue frill applied	Pedestal goblet (?)		14 <sup>th</sup> -15 <sup>th</sup> cent.
LCC0010	Part of wall with rim	Beaker	Horizontal thread applied on the rim	16 <sup>th</sup> cent.
SCJ0058				
SCJ0078	Part of wall with rim	Beaker	Horizontal thread applied on the rim and oblique pattern of ribs	16 <sup>th</sup> cent.
LCC0019	Part of rim with wall and base	Beaker	Horizontal thread applied on the body and vertical pattern of ribs	16 <sup>th</sup> cent.
LCS0883				
LCS0836				
SCJ0059	Part of wall with rim	Beaker (?)	Horizontal threads applied on the wall and oblique pattern of ribs	16 <sup>th</sup> cent.
LCC0022				
LCC0029				
LCC0013				

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### 147 3. Experimental

148 To determine the chemical composition of the glass under investigation, evaluate the raw materials  
 149 used, and propose hypotheses regarding their origin, all the fragments described (Tab. 1; Fig. 2) were  
 150 analysed using micro particle-induced X-ray emission ( $\mu$ -PIXE). Based on their composition, a  
 151 selection of samples was then subjected to laser ablation inductively-coupled-plasma-mass  
 152 spectrometry (LA-ICP-MS) to obtain trace and rare-earth elements (REE). Small samples, ranging from  
 153 2 to 4 mm<sup>2</sup>, were dry-cut from the fragments using a diamond wire, then embedded in epoxy resin and  
 154 polished with SiC sandpapers down to a 4000 mesh. This procedure avoids obtaining information on  
 155 the surface corroded layer then allowing accurate analysis of the bulk glass. Sampling focused on both  
 156 the body glass and the decoration. However, in some cases, sampling the applied thread would have  
 157 compromised the integrity of the fragment, being then avoided.

158 Quantitative results were obtained via  $\mu$ -PIXE, an ion-beam technique based on detecting X-ray  
 159 radiation induced by a particle beam with energy in the range of a few MeV and typically generated by  
 160 an electrostatic accelerator. The analyses were conducted in collaboration with the Instituto Superior  
 161 Técnico, Universidade de Lisboa (C2TN-IST/UL), using an Oxford Microbeams OM150 scanning

162 nuclear microprobe setup. All samples were initially irradiated with a low-energy 0.7 MeV proton beam  
163 to detect lighter elements such as Na, Mg and Al. For quantifying elements with higher atomic numbers  
164 higher than the one of Fe a 2 MeV proton beam was employed. To validate the results, Corning Museum  
165 of Glass standard CMOG B was also analysed.

166 The  $\mu$ -PIXE results were processed using the GUPIXWIN v3.0 software to obtain oxide concentrations,  
167 expressed in weight percent (Tab. 2, Supplementary Tab. 1).

168 LA-ICP-MS was performed at the National Centre of Scientific Research (CNRS) in Orleans, France.  
169 This technique is frequently used in the analysis of archaeological glass, allowing to quantify the major,  
170 minor, trace and rare-earth elements (REE), important indicators of the provenance of sediments and  
171 sedimentary rocks (Gratuze, 2013; Brems & Degryse, 2014). LA-ICP-MS set-up consists of a  
172 Resonetics M50E excimer laser working at 193 nm coupled with a Thermo Fisher Scientific ELEMENT  
173 XR mass spectrometer. The excimer laser was operated at 5mJ with a repetition rate of 10 Hz. The  
174 beam diameter can be adjusted from 20  $\mu$ m to 100  $\mu$ m. A pre-ablation time of 15 s is set in order to  
175 eliminate the transient part of the signal which is then acquired for 27s corresponding to 9 mass scans  
176 from lithium to uranium. Calibration for glass was carried out using NIST610 and Corning B, C, and D  
177 reference materials (Gratuze, 2013). The detection limits range from 0.1% to 0.01% for major elements  
178 and from 20 to 500 ng/g for others. The composition is calculated as the average of two ablations  
179 performed in different sample areas, and the results are reported in Tab 2 (Supplementary Tab. 2). To  
180 ensure accuracy, glass reference standards Corning A, B, C, D and NIST SRM 612 were regularly  
181 analysed as unknown samples throughout the analytical sequence.

Table 2, Composition of the analysed samples determined by  $\mu$ -PIXE (wt%) (left) and LA-ICP-MS ( $\mu\text{g/g}$ ) (right). <dl= below detection limit; bg= body glass; d= decoration  
The groups division is explained in the text.

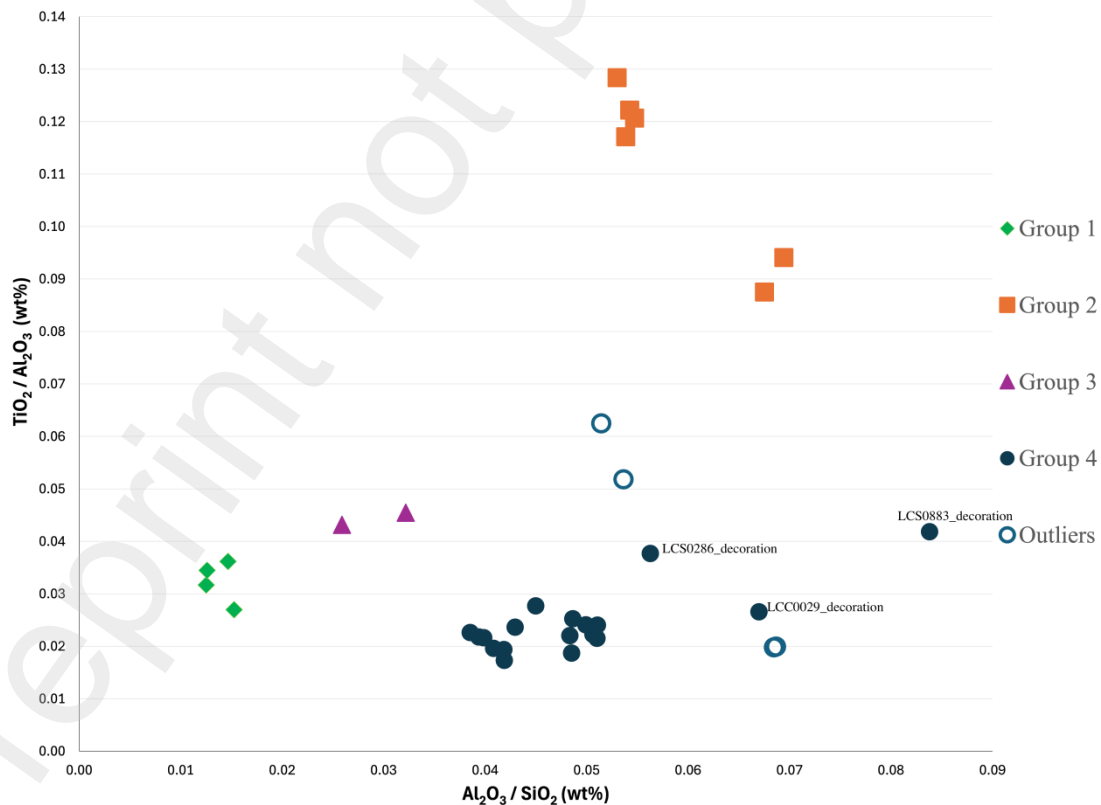
Sample	wt%														$\mu\text{g/g}$																		
	Na <sub>2</sub> O	MgO	Al <sub>2</sub> O <sub>3</sub>	SiO <sub>2</sub>	P <sub>2</sub> O <sub>5</sub>	SO <sub>3</sub>	Cl	K <sub>2</sub> O	CaO	TiO <sub>2</sub>	MnO	Fe <sub>2</sub> O <sub>3</sub>	CoO	CuO	Ni	Zn	Pb	Sn	Bi	As	In	Rb	Y	Sr	Zr	Ba	La	Ce	Hf	Th			
Group 1	LCC0019_bg	15.6	1.5	0.9	72.4	0.51	0.30	0.89	2.54	5.03	0.03	0.24	0.19	<dl	0.01	5.63	22.9	35.2	39.2	0.15	3.30	<dl	11.9	1.36	274	7.6	86	1.57	2.73	0.19	0.37		
	LCC0019_d	14.7	1.5	1.1	72.2	0.13	0.30	0.69	2.69	5.09	0.04	0.24	1.05	0.19	0.20	1269	91.5	283	188	0.33	36.58	<dl	16.7	1.63	268	10.8	109	1.95	3.72	0.26	0.56		
	SCJ0058_bg	13.0	1.5	1.1	72.0	0.14	0.20	0.97	2.28	4.45	0.03	0.38	0.26	0.01	0.01	6.79	18.0	120	125	4.25	6.50	<dl	26.3	1.64	270	12.2	175	2.12	3.51	0.33	0.61		
	SCJ0058_d	14.2	1.8	0.9	69.0	0.16	<dl	0.88	2.28	6.20	0.03	0.44	0.53	0.15	0.04	414	43.7	1527	1567	1388	1387	<dl	25.2	1.96	318	13.5	187	2.48	4.08	0.35	0.68		
Group 2	ARJ0452_bg	18.3	2.4	3.5	64.7	0.51	0.08	1.13	3.29	4.03	0.43	0.30	1.01	<dl	0.01	8.33	67.5	25.7	2.3	0.03	1.57	0.01	19.3	10.65	132	271.2	118	13.95	28.57	7.53	4.40		
	ARJ0452_d	16.8	2.2	3.5	65.1	0.49	<dl	1.18	3.48	4.22	0.44	0.30	1.63	0.25	0.26	511	109	39.3	3.3	0.03	76.88	0.01	20.1	11.33	134	281.9	144	14.73	28.96	7.43	4.39		
	ARJ0099_bg	17.9	2.1	3.5	65.0	0.45	<dl	1.01	3.31	4.00	0.41	0.28	1.11	<dl	<dl																		
	ARJ0099_d	17.4	2.4	3.5	64.5	0.44	<dl	1.03	3.61	4.17	0.43	0.29	1.48	0.19	0.21	520	121	31.6	3.1	0.04	71.67	0.01	24.4	11.29	145	295.2	148	15.78	30.70	7.46	4.58		
	LPF0006_bg	19.6	3.6	4.2	60.8	0.61	0.12	1.21	2.70	4.83	0.40	0.34	1.15	<dl	<dl	11.5	93.6	9.85	1.4	0.03	2.34	0.02	17.1	10.89	226	248.1	232	14.66	28.23	6.16	4.38		
LPF0006_d	17.9	3.1	4.0	60.0	0.57	<dl	1.06	3.02	5.43	0.35	0.41	2.14	0.26	0.31	934	160	9.23	2.4	0.03	76.62	0.04	19.4	11.22	224	254.3	242	15.54	30.17	6.51	4.75			
Group 3	SCJ0059_bg	16.8	4.4	1.8	68.9	0.26	0.16	1.05	1.45	3.48	0.08	0.84	0.51	<dl	<dl	12.61	75.1	12.9	20.6	0.01	1.52	<dl	13.6	3.43	330	31.6	232	14.66	28.23	0.82	1.42		
	SCJ0060_bg	16.3	2.8	2.1	65.3	0.44	0.27	0.92	2.16	7.14	0.10	0.38	0.48	<dl	0.01																		
Group 4	LPF0038_bg	15.0	3.2	2.6	67.1	0.33	0.08	0.93	2.41	6.14	0.06	0.52	0.61	<dl	0.01	11.1	59.5	53.3	76.2	3.36	9.03	<dl	27.4	3.46	337	25.4	242	15.54	30.17	0.69	1.56		
	LPF0038_d	14.8	3.0	2.6	67.4	0.32	0.08	1.02	2.50	6.18	0.06	0.51	0.66	0.09	0.01	164	66.7	74.4	133.9	205.28	339.05	<dl	28.1	3.38	325	24.8	242	4.40	7.96	0.70	1.60		
	LCS0924_bg	18.1	4.0	2.6	65.5	0.29	0.09	1.10	1.64	5.17	0.05	0.33	0.36	<dl	0.01																		
	LCS1059_bg	16.8	3.6	2.8	67.0	0.48	0.07	1.02	1.94	4.86	0.05	0.63	0.50	<dl	0.01																		
	LCS1059_d	16.6	3.6	2.8	66.3	0.46	0.08	0.94	1.95	4.95	0.07	0.64	0.92	0.10	0.10																		
	LCS0286_bg	19.0	3.7	2.6	64.8	0.32	0.06	1.10	1.73	5.30	0.06	0.40	0.43	<dl	0.01	9.09	41.7	2.80	33.5	0.01	0.83	<dl	25.0	3.68	326	27.6	268	4.46	8.01	0.69	1.55		
	LCS0286_d	16.2	3.5	3.5	62.9	0.49	0.08	0.34	3.21	6.79	0.13	0.62	1.19	0.15	0.12	494	104	36.3	211.0	0.05	77.01	<dl	50.1	3.88	352	47.6	357	7.55	14.15	1.29	2.54		
	LCC0029_bg	18.0	3.2	3.2	63.7	0.37	0.11	1.04	1.95	6.29	0.07	0.55	0.62	<dl	0.01																		
	LCC0029_d	15.6	1.0	4.2	63.4	0.70	0.64	0.22	1.13	9.19	0.11	0.26	0.38	0.05	3.12																		
	LCC0010_bg	17.2	3.5	3.2	66.2	0.52	0.13	0.97	2.29	3.73	0.06	0.62	0.56	<dl	<dl																		
	LCC0010_d	16.5	2.8	3.3	63.8	0.49	0.17	1.03	2.16	7.11	0.08	0.54	0.98	0.08	0.05																		
	SCJ0078_bg	14.8	3.1	2.7	65.7	0.40	<dl	0.83	2.40	7.78	0.05	0.41	0.34	0.01	<dl																		
	LCS0883_bg	18.1	3.4	3.0	65.6	0.35	0.07	1.15	1.80	5.44	0.08	0.30	0.51	<dl	<dl																		
	LCS0883_d	17.7	3.2	5.0	60.2	0.96	0.11	0.66	3.30	4.28	0.21	0.80	1.96	0.17	0.25																		
	LCS0836_bg	17.6	3.6	3.2	65.6	0.32	0.08	1.16	1.81	5.31	0.08	0.28	0.49	<dl	0.01																		
	LCC0013_bg	17.5	3.6	2.8	65.6	0.29	<dl	1.14	1.86	5.59	0.05	0.39	0.42	<dl	0.01																		
LCC0013_d	15.1	2.8	3.1	61.9	0.25	0.28	0.86	2.41	7.50	0.07	0.46	2.33	0.07	0.21																			
LCC0022_bg	16.7	2.7	3.2	64.5	0.51	0.15	1.12	2.21	7.13	0.08	0.56	0.66	<dl	0.01	13.1	49.5	32.8	64.5	0.04	1.91	<dl	26.3	4.82	385	29.8	380	6.54	11.41	0.81	1.92			
LCC0022_d	17.1	3.4	3.2	66.8	0.44	0.08	1.01	2.35	3.76	0.07	0.62	0.60	0.06	<dl	250	64.5	41.0	65.7	0.04	5.16	<dl	30.4	4.56	392	29.6	372	6.77	11.64	0.73	1.89			
Outliers	LCS1027_bg	16.3	1.5	4.3	62.8	0.53	0.08	1.06	5.26	6.01	0.09	0.98	0.55	0.01	0.01	11.6	76.2	20.0	3.8	0.02	14.44	0.01	58.0	7.45	282	66.9	238	7.86	13.95	1.50	2.64		
	LCS1027_d	16.6	1.5	4.3	62.1	0.53	0.10	1.00	5.15	5.82	0.09	0.92	0.99	0.13	0.11	782	124	31.2	4.7	0.03	37.76	0.01	56.5	7.33	281	68.1	249	7.78	14.01	1.49	2.64		
	STCV0005_bg	18.9	3.3	3.2	62.2	0.56	0.17	1.07	2.32	5.95	0.20	0.34	1.30	<dl	0.01	11.9	89	10.8	1.1	0.06	1.76	0.09	10.4	7.06	163	86.6	93	9.21	17.71	2.20	2.63		
	STCV0005_d	18.7	3.7	3.3	61.2	0.59	0.17	0.81	2.60	6.26	0.17	0.33	1.82	0.19	0.13	35.4	446	238	55.0	0.11	8.64	44.33	14.2	7.21	169	93.4	116	9.39	17.90	2.40	2.71		
CMOG B	16.5	0.9	4.2	62.6	0.71	0.59	0.16	1.01	8.40	0.11	0.25	0.35	0.04	2.89	NIST SRM 612	40.9	38.5	34.3	37.6	31.9	34.8	37.0	32.3	37.8	67.7	40.2	38.5	39	38.8	36.6	40.0		
St. D.	0.3	0.1	0.1	0.2	0.01	0.04	0.03	0.01	0.10	0.01	<0.01	<0.01	<0.01	0.07	St. D.	1.21	1	2.15	1.89	0.6	1.22	0.86	1.26	1.22	1.25	0.49	0.61	1.8	1.5	1.50	0.8		
Reference (Adlington, 2017)	17.0	1.0	4.3	62.3	0.82	0.49	0.16	1.00	8.56	0.09	0.25	0.34	0.04	2.66	Reference (Jochum et al., 2011)	38.8	39.1	38.6	38.6	30.2	35.7	38.9	31.4	38.3	78.4	37.9	37.9	36.0	38.4	36.7	38.6		

182 **4. Results**

183 The results of the analytical study reported in Tab. 2 show that the largest component, silica, ranges  
184 from 59.9% to 72.4%. The presence of high contents of Na<sub>2</sub>O (12.9 - 19.6 wt%), in combination with  
185 MgO (1.4- 4.3 wt%), K<sub>2</sub>O (1.4- 5.2 wt%) and relatively low P<sub>2</sub>O<sub>5</sub> (0.2- 0.4 wt%), and approximately 1  
186 wt% of chlorine suggests the use of soda-rich coastal plant ashes, commonly employed as fluxing agents  
187 in Mediterranean glassmaking in the time period considered (Šmit *et al.*, 2009). Almost all blue  
188 decorations analysed have a composition similar to that of the body glass in terms of silica sources and  
189 flux. Minor differences are probably due to coloration, indicating that the same glass was used for both  
190 the vessel body and the decoration. However, there are exceptions for fragments LCS0286, LCC0029,  
191 LCS0883 that will be discussed below.

192  
193 **4.1 Silica sources**

194 The mineralogy of sand can provide valuable information about the origin of silica sources as it carries  
195 mineral impurities such as kaolinite, feldspar, zircon, monazite, rutile, and iron oxides (Brems &  
196 Degryse, 2014). In Figure 3 the relationship between the quartz content (SiO<sub>2</sub>), the feldspar (Al<sub>2</sub>O<sub>3</sub>),  
197 and the heavy minerals contribution (TiO<sub>2</sub>) of the glassmaking sands is shown. The plot allows to  
198 identify 4 compositional groups, and two outliers (LCS1027 and STVC0005, both body and  
199 decoration).



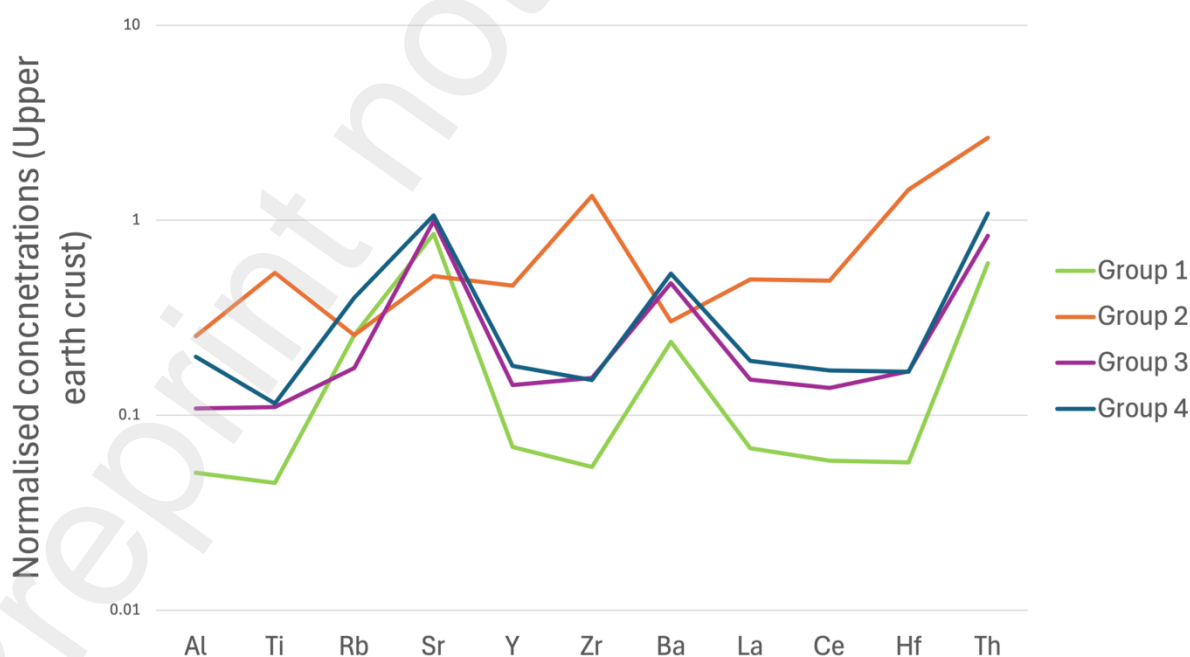
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201

Figure 3, Binary plot of Al<sub>2</sub>O<sub>3</sub> vs. TiO<sub>2</sub>, both normalized to the SiO<sub>2</sub> content.

202 Group 1 was produced from a relatively pure silica source with alumina levels close to 1 wt% and  
 203 titanium below 0.04 wt%. This group includes two objects from the 16<sup>th</sup> century: LCC0019 has a  
 204 horizontal thread applied to the body with a vertical pattern of ribs, while the second SCJ0058 features  
 205 a blue thread applied to the rim. Group 2 contains most of the fragments from the period between the  
 206 14<sup>th</sup> and 15<sup>th</sup> centuries. They consistently show silica sources rich in TiO<sub>2</sub> (0.35-0.44 wt%) and Al<sub>2</sub>O<sub>3</sub>  
 207 between 3.4 and 4.2 wt%. Furthermore, these fragments contain iron levels around 1 wt%, indicating  
 208 that the silica sources used were rich in impurities. It includes fragments with frills applied to the body  
 209 (ARJ0452 and LPF0006) and ARJ0099 with a horizontal blue thread applied.

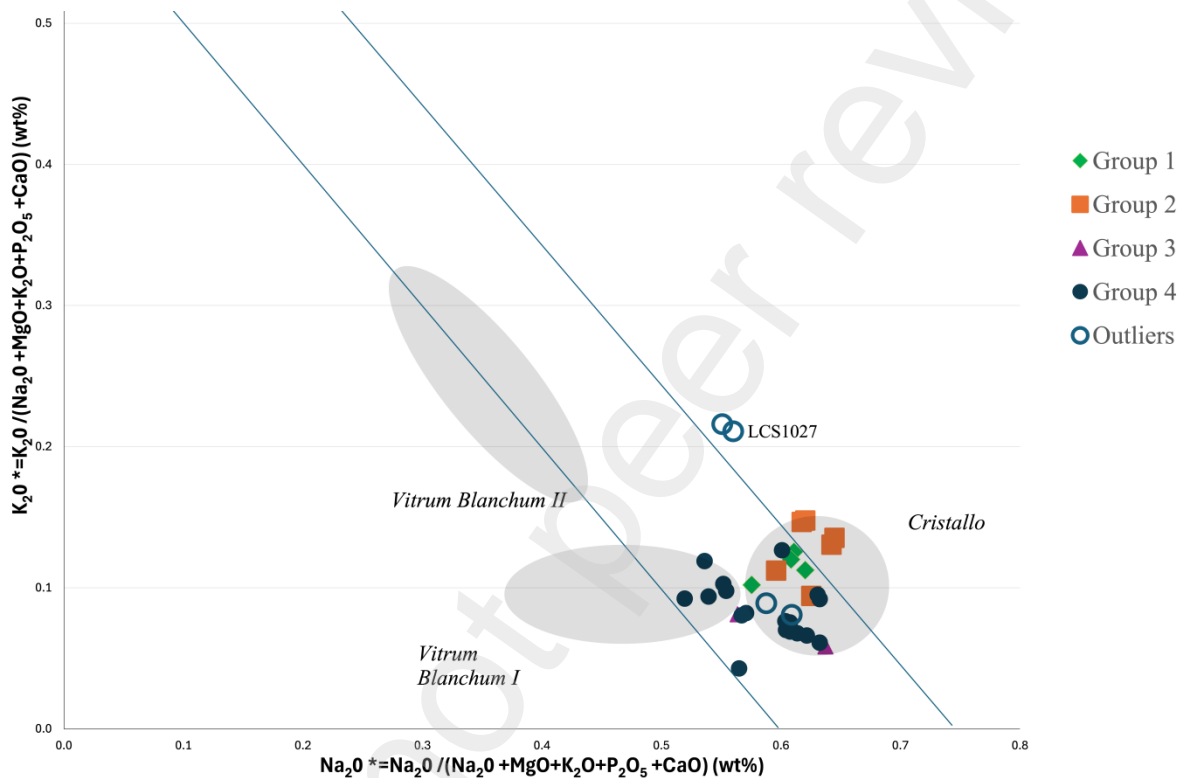
210 SCJ0059 with horizontal threads applied to the wall and oblique pattern of ribs and SCJ0060 which  
 211 has horizontal decoration applied on the wall, fall into Group 3 and are distinguished by alumina  
 212 concentrations of 1.78 wt% and 2.10 wt% and titanium levels of 0.08 wt% and 0.09 wt% respectively.  
 213 Group 4 comprises most of the samples analysed and is characterised by TiO<sub>2</sub> levels below 0.07 wt%  
 214 and varying levels of alumina (2.58 wt% to 6 wt%). All objects in this group are dated to the 16<sup>th</sup> century  
 215 and most of them have one or more horizontal blue threads applied to the body of the object (SCJ0059,  
 216 SCJ0060, LPF0038, LCS0924, LCS1059, LCS0286, LCC0029, LCS0883, LCC0013, LCC0022). This  
 217 cluster also includes LCC0010 with a blue line applied on the rim and SCJ0078, which has an oblique  
 218 rib motif impressed on the body along with the rim decoration. Although the blue threads LCC0029,  
 219 LCS0286 and LCS0883 show different concentrations compared to the body glass, they have been  
 220 included in the group and identified in Fig. 3 as the body glass and its corresponding decoration are part  
 221 of the same object.



222  
 223 *Figure 4, Contents of selected trace and REE, normalized to Earth's upper crust, in logarithmic scale.*

224 Figure 4 shows the average of each previously identified group within a normalised plot of trace element  
 225 to earth crust (Wedepohl, 1995). It can be observed that the groups present distinct trace element  
 226 patterns. Groups 3 and 4 are very close except for Al and Rb. The important discrepancy in Al, and  
 227 the difference in the ratio of Al/Ti underlined in Fig. 3 leads to the proposal that the objects originate  
 228 from different silica sources. Group 1 differs in lower trace element concentrations, confirming the use  
 229 of a silica source low in impurities.

230 4.2 Flux agents



231  
 232  
 233 Figure 5, Binary plot of  $\text{Na}_2\text{O}^*$  vs.  $\text{K}_2\text{O}^*$ .  $\text{Na}_2\text{O}^*$  and  $\text{K}_2\text{O}^*$  values are obtained through the division of the respective oxide  
 234 by every component introduced by the ash ( $\text{Na}_2\text{O}$ ,  $\text{MgO}$ ,  $\text{P}_2\text{O}_5$ ,  $\text{K}_2\text{O}$  and  $\text{CaO}$ ). The two correlation lines represent limit  
 235 concentrations for the use of purified ash ( $\text{Na}_2\text{O}^* + \text{K}_2\text{O}^* = 0.75$ ) or of unpurified ash ( $\text{Na}_2\text{O}^* + \text{K}_2\text{O}^* = 0.6$ ) (Cagno *et al.*,  
 236 2012).

237  
 238 All fragments fall under the soda-silica-lime type, thus with the use of plant ash to decrease the melting  
 239 point of silica. To differentiate the types of fluxes used in glass production, Figure 5 compares the soda  
 240 and potash values obtained by dividing the respective oxides with the sum of all components introduced  
 241 into the ash. Two correlation lines are considered in medieval and post-medieval glass:  $\text{Na}_2\text{O}^* + \text{K}_2\text{O}^* = 0.6$   
 242 for the use of natural ash and  $\text{Na}_2\text{O}^* + \text{K}_2\text{O}^* = 0.75$  for purified ash (Cagno *et al.*, 2012). Plant  
 243 ash purification, introduced into the Murano glass production process in the 15<sup>th</sup> century, comprises  
 244 several steps to remove water-insoluble substances, including iron, calcium and magnesium. This  
 245 meticulous process was conducted on ashes imported from the Levant, which were utilized in the

246 production of Venetian *crystallo* glass (Moretti & Hreglich, 2013). Conversely, unpurified ashes refer  
247 to those that have not undergone this treatment.

248 All fragments belonging to Group 1 and 2 arrange themselves along the inverse correlation line of glass  
249 made using purified ashes and are close to the *crystallo* boundaries, which have a flux with low  
250 potassium concentrations and a higher soda content, comparable to that used in the Venetian glass  
251 production (Šmit *et al.*, 2009; Verità, 2013). Fragments of Group 3 and 4 fall both in the *crystallo* area  
252 and close or inside the boundaries of *vitrum blanchum I* (Šmit *et al.*, 2009). LCS1027 stands out as  
253 outlier, positioned on the line indicating a purified ash but with high potassium content.

#### 254 255 4.3 Colourants and decolourants

256 The results reported in Tab. 2 for the decorations reveal that the blue glass was created by incorporating  
257 cobalt as a colouring agent into the base glass, with concentrations ranging from 0.07 to 0.26 wt%.  
258 Additionally, cobalt is found in correlation with copper (0.01 to 0.31 wt%) and iron oxides (0.66 to 2.14  
259 wt%), elements naturally associated with the mineral ore used to produce the colorants, which were  
260 integrated into the glass batch. As mentioned earlier, the majority of the body glass and its associated  
261 blue decorative elements exhibit very close compositions, suggesting that they were likely produced  
262 using the same base glass to which a colouring agent was added. The blue decoration of LCS0286,  
263 LCC0029, and LCS0883, have higher levels of alumina and titanium indicating that the raw materials  
264 used for the blue glass were different and richer on impurities.

265 Examining the composition of the body glass in the analysed objects it can be observed that the MnO  
266 content in most samples, from approximately 0.3% to 0.85%, aligns with the typical range used for  
267 decolorizing glass (Moretti & Hreglich, 2013). However, LCS1027 stands out with a higher  
268 concentration of 0.98 wt%, likely due to impurities in the silica source. In contrast, LCC0019 shows a  
269 rather lower concentration of 0.24 wt%, which may be attributed to the fact the discolorations process  
270 in Venice needed only small amounts of manganese added to the molten glass (Verità *et al.*, 2024).

271

## 272 5. Discussion

273 The fragments analysed in this study were compared with results from literature regarding objects from  
274 the same period found in other European contexts to identify potential compositional relationships (Fig.  
275 6). Fragments SCJ0058 and LCC0019 (Group 1) align with 16<sup>th</sup> century Venetian *vitrum blanchum*  
276 productions, which involved the use of sands with low TiO<sub>2</sub> (< 0.07 wt%) and Al<sub>2</sub>O<sub>3</sub> contents (< 1.5  
277 wt%) (Verità, 2013). In Fig. 4 Group 1 stands out for its lowest trace elements, further highlighting the  
278 purity of the silica source. Specifically, the Zr and Hf content is often used in the literature to  
279 differentiate between Venetian and *façon de Venise* glass. Venetian glass typically exhibits low Zr  
280 levels, generally under 20 µg/g, and Hf concentrations below 1 µg/g (Cagno *et al.*, 2012; Coutinho,  
281 Medici, Alves, *et al.*, 2016; De Raedt *et al.*, 2001). The alkaline component of Group 1 indicates that  
282 these objects were made using purified ashes close to the *crystallo* type. It is proposed that objects in

283 this group were produced using pure ashes similar to those brought to Venice from the Levantine region  
284 and with pure silica source, therefore advancing the hypothesis of a Venetian import.

285

286 Within Group 2, ARJ0099 and ARJ0452 exhibit overlapping characteristics with a fragment unearthed  
287 in Beja, Portugal, dating to the 15<sup>th</sup> century with close titanium and aluminium contents ( $\text{Al}_2\text{O}_3$  3.4 wt%;  
288  $\text{TiO}_2$ : 0.45wt%) (Coutinho, Medici, Coentro, *et al.*, 2016). This fragment features a blue decoration  
289 applied to the rim and an oblique ribbed pattern on a colourless body. Given the strong similarity in the  
290 values related to the silica sources and fluxing agents, it is suggested that these fragments were produced  
291 in the same production centre yet to be identified.

292

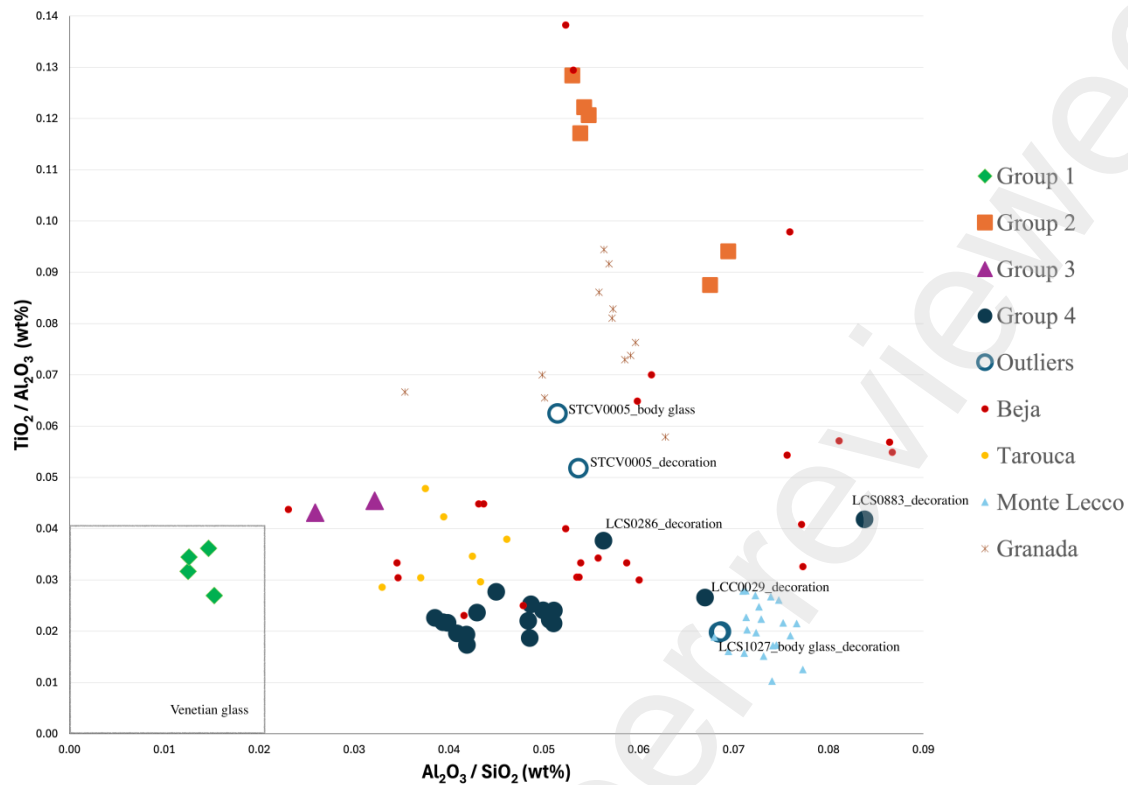
293 The ratios of  $\text{Al}_2\text{O}_3/\text{SiO}_2$  versus  $\text{TiO}_2/\text{Al}_2\text{O}_3$  of Group 3 and 4 align with *façon de Venise* objects from  
294 Portuguese excavations in Beja and the Monastery of São João de Tarouca (16<sup>th</sup>-17<sup>th</sup> centuries)  
295 (Coutinho, Medici, Alves, *et al.*, 2016). When compared with other known European production centres  
296 such as Antwerp, Amsterdam and London (De Raedt *et al.*, 2001; Hulst & Kunicki-Goldfinger, 2017;  
297 Janssens *et al.*, 2013) the compositions show discrepancies, implying that the fragments here studied  
298 were not imported from these locations.

299 All fragments belonging to Group 3 and 4, except for the blue decorations, have iron oxide  
300 concentrations below 0.7 wt%, suggesting that the raw materials, rich in feldspars, were carefully  
301 selected (Coutinho *et al.*, 2022). For these groups two production technologies have been identified  
302 using both unpurified and purified plant ashes (Fig. 5). These procedures may indicate an attempt to  
303 produce objects following Venetian techniques using locally available materials.

304

305 Considering the two outliers for LCS1027, both the body glass and decoration differ from the main  
306 group of fragments dated to the 16<sup>th</sup> century due to their higher alumina levels (4.30 wt%). Its impurity  
307 concentrations and the high potassium (5.15–5.25 wt%) align with analyses conducted on glass dated  
308 to the 15<sup>th</sup> century found in the glasshouse of Monte Lecco, in the Genoese Apennines. Comparable  
309 sodium and potassium oxide levels have been identified in Granada, where it was suggested that local  
310 plants rich in sodium were purified and used in production (Coutinho *et al.*, 2021). It cannot be excluded  
311 that the same process was used to produce this object. In Fig. 6, STCV0005 shows similarities in  
312 mineralogical characteristics with two objects from the production centre in Granada. Unlike the objects  
313 from the Spanish context, however, the fragment studied here was produced using purified ashes.

314



316  
 317 *Figure 6, Binary plot of weight ratio of  $Al_2O_3/SiO_2$  vs. weight ratio of  $TiO_2/Al_2O_3$  for the samples considered in this work*  
 318 *together with the concentrations of glass unearthed in Spain, in Granada (Coutinho *et al.*, 2021), and in Portugal in Beja*  
 319 *(14<sup>th</sup>-16<sup>th</sup> cent.) (Coutinho, Medici, Alves, *et al.*, 2016; Coutinho, Medici, Coentro, *et al.*, 2016) and in Monastery of São*  
 320 *João de Tarouca (16<sup>th</sup>-17<sup>th</sup> cent.) (Coutinho, Medici, Alves, *et al.*, 2016). The areas marked as Venetian glass refers to the*  
 321 *cristallo and vitrum blanchum groups, considering values reported in Verità, 2013.*  
 322  
 323

324 Cobalt was identified as the element responsible for the colouring in all the blue glass analysed. A study  
 325 on the mining locations used to obtain cobalt oxides enables the identification of possible areas of origin  
 326 (Gratuze *et al.*, 1996). To investigate the colouring technology, the elements in  $\mu\text{g/g}$  related to the cobalt  
 327 ore are plotted in Fig. 7. The co-presence of zinc, lead, and indium, suggests that STCV0005 can be  
 328 dated between the 13<sup>th</sup> and 15<sup>th</sup> centuries, and that the cobalt ore used for its blue decoration is likely  
 329 originating from the Erzgebirge mines near Freiberg, as outlined by Gratuze *et al.*, 1996.

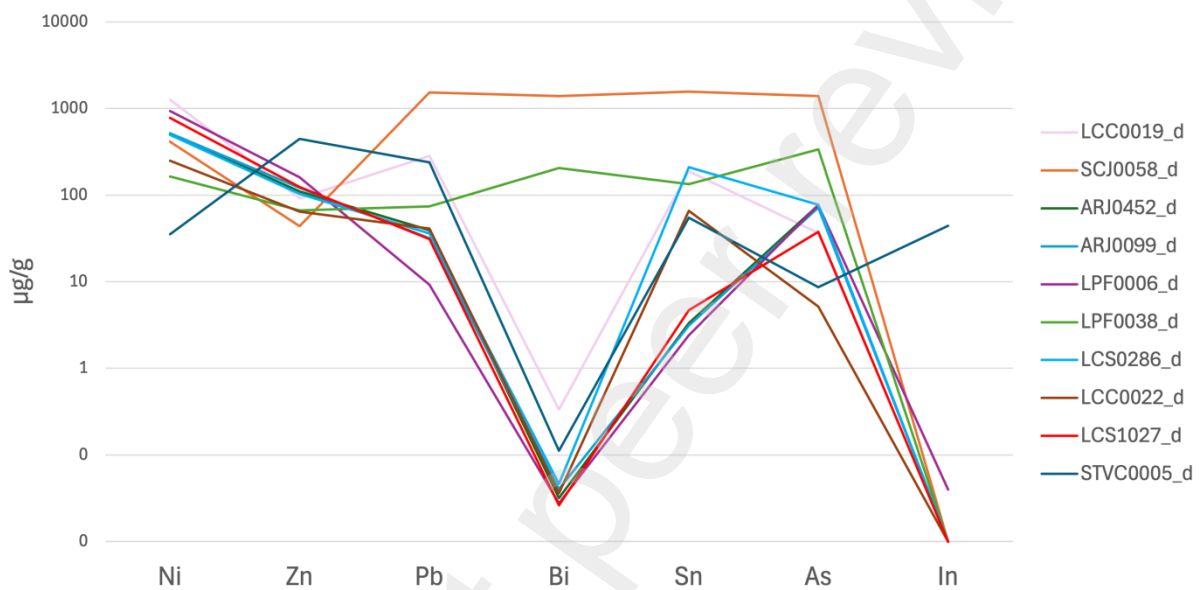
330 With reference to ARJ0452, ARJ0099 and LPF0006, Fig. 7 shows that the concentrations of cobalt-  
 331 related elements have almost overlapping patterns. This similarity suggests a common origin for these  
 332 objects and strengthens the hypothesis that they were produced in same production centre, as the  
 333 composition of the body glass is very similar as well.

334 Elements such as arsenic and bismuth help in the more precise dating of the objects. Their presence in  
 335 the ore has been documented starting from 1520, when a shift occurred in the methodology of  
 336 processing cobalt, which involves sorting and roasting the minerals in a reverberatory oven (Zucchiatti  
 337 *et al.*, 2006). The detection of larger amounts of these two elements in LPF0038 and SCJ0058 allows  
 338 these objects to be dated to the second part of the 16<sup>th</sup> century. Furthermore, the presence of arsenic,

339 nickel, and bismuth is consistent with cobalt ore extracted from the Schneeberg mines in the Erzgebirge,  
340 Saxony (Gratuze *et al.*, 1996).

341 Additionally, LCC0019 and SCJ0058 exhibit relatively high levels of tin (188–1567  $\mu\text{g/g}$ ) and lead  
342 (283–1527  $\mu\text{g/g}$ ) in the blue decoration, compared to the other objects analysed. These elements were  
343 detected in Renaissance Venetian blue *crystallo* and *vitrum blanchum* samples (Verità *et al.*, 2024;  
344 (Verità & Biron, 2021), where their presence was believed to be due to the addition of unselected cullet  
345 which probably didn't affect the final blue colour of the glass (Verità & Biron, 2021). This comparison  
346 further reinforces the Venetian origin proposed for these two objects.

347



348  
349 *Figure 7, Contents of elements related to cobalt ore, in  $\mu\text{g/g}$ .*

350

351

352

353

354

## 6. Conclusions

355 The study of glass objects featuring applied blue decoration, spanning from the 14<sup>th</sup> to the 16<sup>th</sup> century,  
356 provides significant insights into production techniques, raw material selection, and the origins of these  
357 items. From a stylistic perspective, the blue decoration was found in Portuguese excavations on  
358 tableware used for drinking, including beakers, goblets, and pedestal goblets, all with colourless bodies.  
359 Among the decorations observed, the applied blue horizontal thread stands out as the only consistent  
360 decorative element across the entire period. The consistent use of these horizontally applied blue threads  
361 highlights their popularity across different time frames and production centres.

362 Considering the results of  $\mu$ -PIXE and LA-ICP-MS analysis, the objects dated to the 14<sup>th</sup>/15<sup>th</sup> centuries  
363 exhibit higher levels of impurities, which suggests the use of less refined silica sources. It is proposed  
364 that the objects belonging to Group 2 likely originate from the same production site. Specifically,

365 fragments ARJ0099 and ARJ0452 share compositional characteristics, such as elevated titanium  
366 (around 0.4 wt%) and alumina (3.45-3.50 wt%), closely resembling a 15<sup>th</sup>-century beaker found in Beja,  
367 Portugal, which also features blue decoration applied to the rim. The strong similarities in the silica  
368 sources and flux agents suggest that they may have been produced within the same glassmaking  
369 workshop. The fragments dated to the 16<sup>th</sup> century are identified as Venetian in origin (Group 1) or  
370 *façon de Venise* (Group 3 and 4). The presence of Venetian-imported goods indicates that Portuguese  
371 commercial activities were connected with the Venetian Lagoon during this period, reflecting the  
372 demand of the Portuguese high society for luxury items that were in fashion at the time. For Group 3  
373 and 4, their origin likely traces back to an unknown production centre shared with other *façon de Venise*  
374 objects dated between the 16<sup>th</sup> and 17<sup>th</sup> centuries, recovered from Portuguese excavations. The evidence  
375 suggests two distinct production technologies: one utilising unpurified plant ashes and the other using  
376 purified plant ashes, indicating efforts to replicate Venetian techniques with locally available materials.  
377 Additionally, the fragment LCS1027, characterised by its higher alumina content and impurity profile,  
378 might be linked to the glass factory located in the Monte Lecco area. The compositional data are  
379 consistent with 14<sup>th</sup>/15<sup>th</sup>-century production remains from this region, indicating a potential exchange  
380 of goods with the area of Genoa, in Italy.

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### 386 387 **Author contributions**

388 Conceptualization: A.C., T.M. and I.C.; data curation: A.C and I.C.; formal analysis: L.C.A. and N.S.;  
389 investigation: A.C., L.C.A. and N.S; methodology: I.C. and A.C; project administration: A.C and I.C;  
390 resources: I.C. and A.C.; supervision: T.M, I.C. and C.V.; validation: A.C.; visualization: A.C.; writing  
391 – original draft: A.C.; Writing – review & editing: A.C., T. M., C.V., L.C.A., N.S. and I.C.

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### 397 398 **Declaration of competing interest**

399 The authors declare that they have no known competing financial interests or personal relationships that  
400 could have appeared to influence the work reported in this paper.

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