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Chemical Characterization of the Roman Glass Finds from Muricelle Archaeological Site (Luzzi, Cosenza)

Anna Maria De Francesco [1], Antonio La Marca [2], Carmelo Colelli [3] and Donatella Barca [1,*]

[1] Dipartimento di Biologia, Ecologia e Scienze della Terra (DiBEST), Università della Calabria, 87036 Rende, Italy; anna_maria.defrancesco@unical.it
[2] Dipartimento Culture Educazione e Società (DiCES), Università della Calabria, 87036 Rende, Italy; antonio.lamarca@unical.it
[3] Soprintendenza Archeologia, Belle Arti e Paesaggio per la Provincia di Cosenza, 87100 Scalea, Italy; carmelocolelli81@gmail.com
* Correspondence: donatella.barca@unical.it

Abstract: This study reports the results obtained on seventeen glass finds collected from the archaeological site of Muricelle near Cosenza in Calabria (Italy), dated between the first and third century AD. Chemical characterization was undertaken by using an electron probe micro analyser (EPMA). The glasses showed a soda–lime–silica composition indicating the use of the natron as a flux. The comparison between the collected data and those on coeval finds available from the literature allowed us to subdivide the Muricelle glass into three compositional groups: (1) Mn_a, classified as Roman-Mn glasses; (2) Mn_b, which can be compared to the Mn-unintentionally coloured Roman glasses; and (3) Sb + Mn, obtained by the recycling of the Sb and Mn Roman-type glasses. These groups were different not only with respect to the contents and the types of decolourants but also for soda, lime, and aluminium contents, highlighting the use of different raw materials in the primary glass production and also indicating the procedures used in secondary glass production. The compositional characteristics suggested the Levant region as the primary glass source for the Mn_a and Mn_b groups. On the other hand, the Sb + Mn group was obtained through recycling, involving the melting and mixing of two types of primary glass (Sb-decolourised and Mn-decolourised) of Egyptian and Levantine origins, respectively.

Keywords: glass; Roman; Muricelle; chemical composition; EPMA

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1. Introduction

The present study focused on some glass finds collected during a surface survey conducted in the archaeological site of Muricelle in the territory of Luzzi, a village situated at the right side of the Crati river near Cosenza in Calabria (Italy) (Figure 1) [1].

In agreement with the numerous ceramic finds recovered there, the Muricelle site has been dated between the first century and the end of the second century AD [2].

The archaeometric study was carried out using an electron probe micro analyser (EPMA) that provided the chemical composition of seventeen glass fragments.

The analyses were aimed at establishing the nature and provenance of the raw materials and to understand the glass production technique [3–10].

Glass is essentially a mixture of three major components: silica sand which acts as network-former, an alkaline compound which acts as a flux, and lime which acts as a network-stabilizer. During the Roman times, the alkaline fraction originated prevalently from natron, a mixture of mineral compounds rich in sodium and abundant in some evaporitic basins, such as those of the Wadi El Natrun valley in Egypt [11]. Only in few cases, alkaline metal oxide was added as plant ash component richer in magnesia and potash [9].

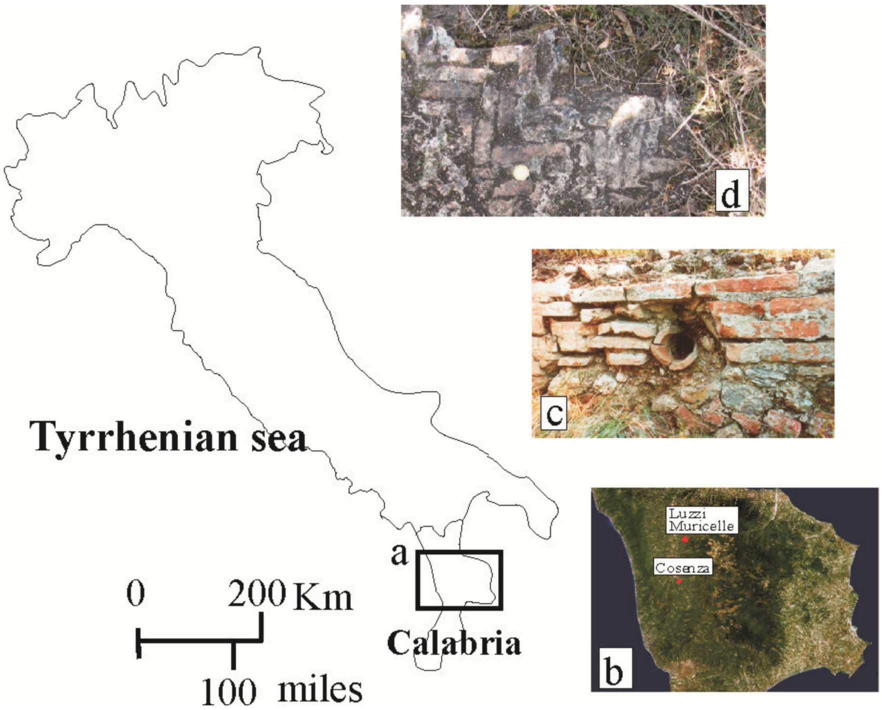

Figure 1. (**a**) Italy—Calabria; (**b**) Muricelle archaeological site; photos of (**c**) ruins of wall and (**d**) *opus spicatum* floor of the Roman *villa*.

Numerous researchers have dealt with the investigation of the glassmaking processes in the Roman times. The prevalent hypothesis is that large masses of raw glass were produced in a small number of primary centres, mainly in coastal areas of Egypt and in the Near East. Then, these slabs were broken up and distributed to secondary fabrication workshops to be re-worked [4,12].

In the eastern Mediterranean and in the Levant in the first millennium AD, five main groups of natron-type soda–lime–silica glass were recognised: Levantine I, Levantine II, Roman blue-green, HIMT (high iron, manganese, titanium), and Egyptian [4,9].

Roman glass has been the subject of many studies [5,13–18] which have improved the knowledge of these productions. Roman glass dating from the first to the third century AD has a very homogeneous composition, indicating that it was produced on a large scale [9]. In the same period, colourless glass reached its maximum production and its use declined at the end of the third century AD [16–18].

The studies have provided precious indications on the great care of the Roman glassmakers in the selection of materials, their ability in the production processes, and the addition of antimony or manganese as decolourant agents to obtain colourless glass [10,15,16,18].

Many archaeometric studies on ancient colourless glass [16–20], considering the contents of the two decolourants (Sb and Mn), distinguished four main groups: Roman-Sb, Roman-Mn, Roman mixed Sb + Mn, and the naturally colourless glass.

Roman-Sb and Roman-Mn refer, respectively, to the intentionally decoloured Sb and Mn glass groups, while the Roman mixed Sb + Mn was obtained from the recycling of the two end members [8,17,21,22]. The naturally colourless glass was produced with pure raw materials, without the addition of decolouring agents [16–18].

Previous studies individuated the compositional ranges of the two added decolourants [15,16,23]. The Sb-colourless glass group is characterized by high antimony content (Sb_2O_5 ranging from 0.3 to 0.8 wt% and MnO content less than 0.025 wt%); Mn-colourless glass by high manganese (MnO > 0.025 wt% and antimony below the detection limit); Sb + Mn colourless glass by the presence of both manganese (MnO > 0.025 wt%), and antimony (Sb_2O_5 > detection limit around 0.03 wt%). The naturally coloured glass (or Mn_unintentionally coloured glass) is characterized by generally low manganese (MnO < 1 wt%) and negligible antimony [16,23].

These four main compositional Roman groups differ not only by Sb and Mn contents but also different contents of SiO_2, Na_2O, CaO, and Al_2O_3 [17,22].

This research aimed at investigating the glass composition of the finds recovered in the archaeological area of Muricelle. By comparing the chemical analyses carried out on the vitreous samples with the known glass groups of the Imperial age, information on the glass production technology was obtained, thus allowing the correlation between typological analysis and chronological attribution.

2. Study Area

Muricelle is an archaeological area located 4 km north-west from the town of Luzzi (Cosenza). It stretches through about one hectare on the western slopes of the Sila massif (190 m a.s.l.) (Figure 1).

The location is particularly favourable to settlements due to the presence of water guaranteed both by the Ilice stream and by the Crati river which in ancient times was also an important communication route. The toponym probably derives from the presence of wall structures built with mortar and *cocciopesto*.

Today, only small traces of these structures are visible but throughout whole area, numerous artifacts have been found. The most ancient samples recovered in the Muricelle site are: a lithic spearhead dated to the prehistoric time, some ceramic fragments of the Bronze Age and Ancient Bronze Age [24,25], and a hoard of bronze axes (belonging to the so called "Cirò type") of the Iron Age [26,27]. However, the remains of a wall in *opus reticulatum* and a floor in *opus spicatum*, which most likely belonged to a Roman *villa*, attest the anthropic presence especially of the Roman imperial times.

In Muricelle, during the agricultural clearing, fragments of various materials have been recovered including pottery, coins, metallic, glass, and marble artefacts. The ceramic fragments belonged to common ware, amphorae, oil lamps, and fine pottery which are well documented (Italic and African sigillata, thin-walled). These ceramic artifacts testify the frequentation of the *villa* in the first centuries of the Roman imperial age (1st–2nd century AD) and to a lesser extent, up to the Late Antiquity, when the site was probably abandoned [1,2,24]. No black gloss pottery or other materials dating back to the Republican period have been found [24]. The presence of vitreous objects was reported for the first time by Marchese [26] who referred to the discovery of glass fragments of a jar and of "a very large bell-shaped glass jar with two handles and two figured faces" as richly decorated [26].

Overall, the quantity and quality of the archaeological finds confirm that Muricelle was a rural Roman *villa*, well-placed within a circuit of exchange and trade.

3. Materials

In the present study, seventeen glass finds recovered in the archaeological site of Muricelle were analysed (Figure 2). The glass finds unfortunately were small in size because they were recovered following the ploughing of the land. The repertoire consisted of a limited number of shapes, mainly cups, plates, and bottles. After a preliminary examination, the samples were catalogued and subjected to a morphological analysis in order to identify their likely form, and they were compared with known shapes in the main contexts of Italy and the Mediterranean. However, because of the fragmentation of glass finds, some of them lacked diagnostic elements that would allow their attribution.

Only six vitreous fragments could be classified as being parts of various containers (cups, plates, and bottles) or of decorations (Table 1); for the remainings, we proposed only a possible shape attribution.

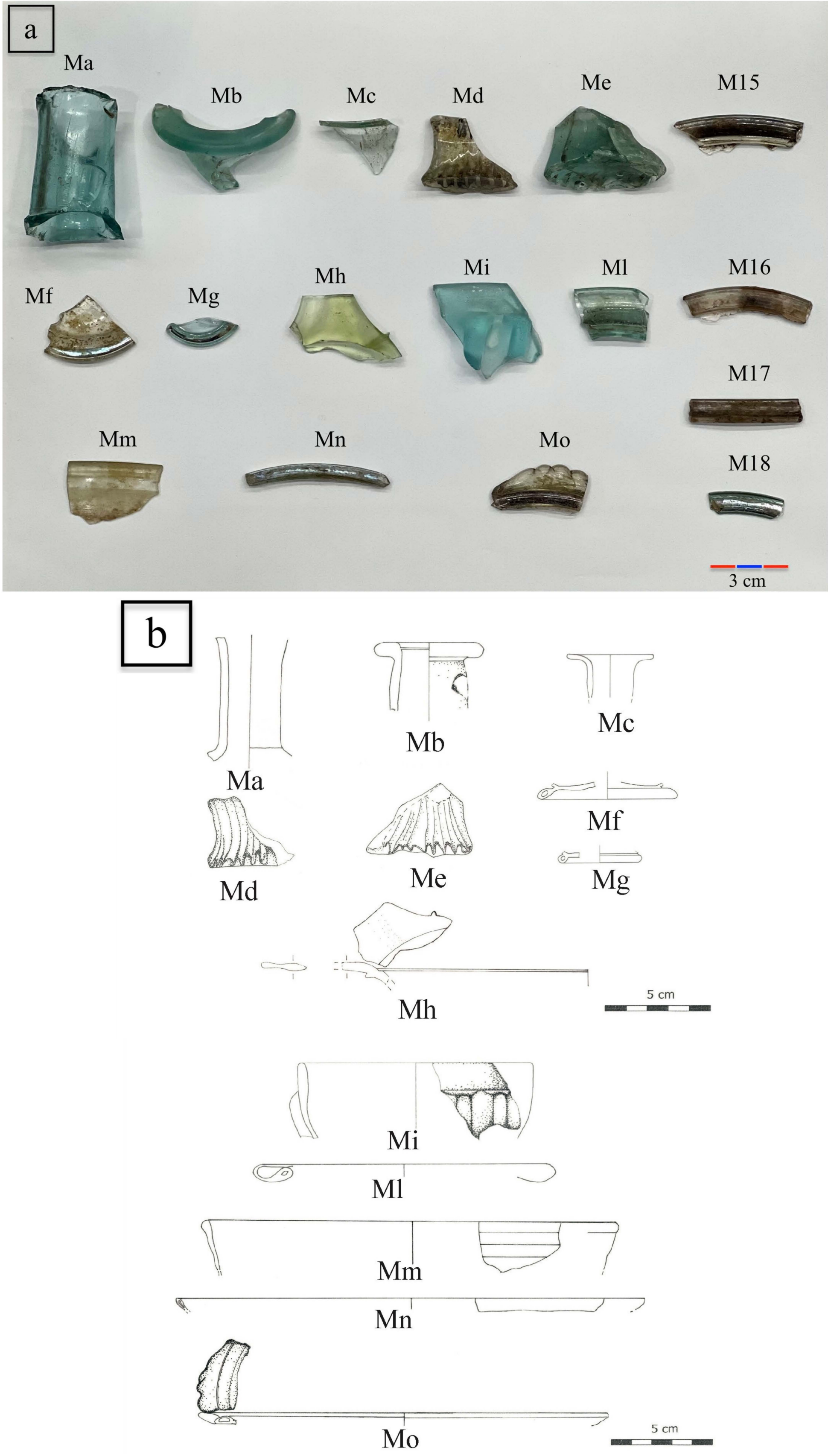

Figure 2. (**a**) Photograph of Muricelle glasses analysed in this study; (**b**) possible shape attribution of the vitreous finds.

Table 1. Typological analyses of Muricelle vitreous finds (N.I. = not identified; ? = dubious type or chronology).

Sample	Shape	Type	Chronology	Colour
Ma	Bottle	N.I.	N.I.	Light blue green
Mb	Bottle	Isings 50/51 [28]	1st–3rd century AD [29]	Light blue green
Mc	*Bombilium*	Isings 28A ? [28]	1st–3rd century AD ?	Light blue
Md	Bottle	Isings 50/51 [28]	1st–3rd century AD [29] Similar to those recovered in Pompei [30]	Light green
Me	Bottle	Isings 50/51 [28]	1st–3rd century AD [29] Similar to those recovered in Pompei [31]	Light blue green
Mf	Glass	Isings 34 ? [28]	1st–3rd century AD ?	Colourless-light blue
Mg	Cup	Isings 47 ? [28]	First half of 2nd century AD ? Similar to those recovered in Settefinestre (Grosseto—Italy)	Light blue
Mh	Open shape	Isings 75 ? [28]	1st–3rd century AD ?	Light green
Mi	Cup	Isings 3 [28]	1st AD similar to those recovered in Pompei [31]	Light blue green
Ml	Jug	Isings 67A ? [28]	1st–3rd century AD ?	Light blue green
Mm	Cup or Plate	Isings 18 [28]	1st–3rd century AD	Colourless
Mn	Plate	Isings 97c ? [28]	1st–3rd century AD ?	Light blue
Mo	Cup	Isings 43 [28]	Second half of 1st century AD—first half of 2nd century AD [32] Similar to those recovered in Poggio del Molino (Populonia Piombino LI)	Light green
M15	Cup	Unknown	N.I	Colourless
M16	Cup	Unknown	N.I	Colourless
M17	Cup	Unknown	N.I	Colourless
M18	Cup	Unknown	N.I	Light blue green

The finds can be divided as follows: nine fragments of cup or plate, four bottles, one jug, one *bombilium*, one open shape, and one glass as indicated in the Table 1, where the colour and other features are also reported. Concerning the colours (Table 1), the majority of Muricelle glasses had a light blue green colour, typical of the Roman times, and some were light green, light blue, or colourless.

The best-preserved specimens suggested a chronology between the first and second centuries AD as bottles (samples Mb, Me, and Md) and cups (Mi, Mn, and Mo). This dating agrees with the ceramics study [24], confirming that the occupation of the Muricelle site occurred mainly in the first and second century AD.

4. Methods

Before proceeding to the analytical phase, a small fragment of about 3 mm $\times$ 3 mm was collected from each sample and cleaned by an ultrasonic bath in order to eliminate any residual dust [33]. Polished cross-sections were obtained, embedding the glass fragments in epoxy resin blocks, then transversely cut to enable unaltered glass surface analysis. The surface was finally polished with diamond pastes and then coated with a graphite layer [34] to be studied by electron probe micro analyser with wavelength dispersive spectrometer (EPMA-WDS). To determine major element concentrations, EPMA analyses were carried out using JEOL-JXA 8230 (JEOL Ltd., Tokyo, Japan) equipped with 5 WDS Spectrometers: LDE (artificial layered dispersive element), TAP (thallium acid phthalate), PET (PEnTaerythritol), LiF (lithium fluoride), and crystals. The EPMA was used under the following operating conditions for chemical analyses: acceleration voltage 15 kV, probe current 10 nA, and a defocused beam of 10 μm to avoid the loss of alkali. Furthermore, an acquisition time of 15 s was selected to avoid the loss of Na and ensure the correct determination of other element concentrations. The reliability of the results was verified by measuring the SRM610 by NIST (National Institute of Standards and Technology)-certified reference for materials and glass (Table 2). The detection limits under the specified working conditions was 0.01 wt%.

Table 2. Analyses of NIST SRM610 standard glass. Comparison between certified and measured values.

	NIST-SRM610		
	Certified Values (1)	**Measured Values**	**Accuracies (2)**
SiO_2	72	71.8	0.28
TiO_2	0.07	0.069	1.43
Al_2O_3	2	2.12	−6.00
FeO	0.059	0.054	8.47
CaO	12	11.8	1.67
Na_2O	14	14.3	−2.14
K_2O	0.056	0.057	−1.79
MnO	0.059	0.056	5.08
PbO	0.05	0.049	2.00
CuO	0.056	0.052	7.14
Sb_2O_5	0.055	0.053	3.64
SnO_2	0.051	0.049	3.92

(1) Certified values of Ti, Fe, K, Mn, Pb, Cu, Sb, and Sn expressed in ppm were converted to wt% of oxides.
(2) Accuracy = ((measured value − certified value)/certified value) × 100.

The analyses were conducted on a selected area (about 100 μm^2) in the glass core which was free of bubbles and other inclusions.

5. Results and Discussion

Table 3 lists the composition of major and minor element concentrations in wt% of oxides determined by EPMA. Each value represents the mean of the analyses executed on three different points.

All samples were soda–lime–silica glass for which the SiO_2 content varied from 68.72 to 71.35 wt%, and Na_2O from 16.29 to 19.62 wt%. The results showed that, consistently with the chronology obtained from the archaeological investigations on the Muricelle site [24,30], they fit perfectly into the Roman glass composition [3,5,15,17,18,22,34,35].

All vitreous fragments showed values of both MnO and K_2O below 1.50 wt%, as the typical Roman glass production, indicating natron as the unique flux employed [3,11,19,36].

The archaeological studies conducted on the Muricelle site suggest that the *villa* and the surrounding area were active in the imperial period [24,30]; therefore, to understand the origin of raw materials and the technology of production employed in the Muricelle glass, the data were compared with the productions of that period (first–third century AD).

As stated above, the main compositional glass groups dominating the Roman imperial period (first–third century AD) were: Roman naturally coloured blue green, Sb-decolourised, Mn-decolourised, and Sb + Mn decolourised glasses related to the different decolouring agent used during the glassmaking production [15,17,18,21,22].

Regarding the Muricelle glasses, they were divided into three groups: (1) Mn_a characterised by high MnO content, formed by only three samples (Md = 1.38 wt%; M15 = 1.47 wt%; and M16 = 1.43 wt%); (2) Mn_b with MnO varying from 0.13 wt% to 0.56 wt% and Sb below the detection limit (samples: Ma, Mb, Mg, Mh, Mi, and Ml); and (3) Sb + Mn, decoloured glass, which was the largest group consisting of eight samples (Mc, Me, Mf, Mm, Mn, Mo, M17, and M18). In this last group, MnO varied from 0.30 wt% to 0.74 wt% and Sb_2O_5 from 0.13 wt% to 0.71 wt% (see Table 3).

Table 3. Major element concentrations in wt% of oxides determined by EPMA. For each oxide, we calculated the standard deviation (StDev) of three analyses.

Group	Sample	SiO_2	TiO_2	Al_2O_3	FeO	MgO	CaO	Na_2O	K_2O	MnO	PbO	CuO	Sb_2O_5	SnO_2
Mn_b	Ma	71.04	0.07	2.48	0.32	0.49	7.44	16.48	0.59	0.30	n.d.	0.02	n.d.	0.07
	Ma_StDev	*0.07*	*0.06*	*0.04*	*0.05*	*0.03*	*0.09*	*0.10*	*0.03*	*0.03*	-	-	-	*0.02*
Mn_b	Mb	71.35	0.06	2.49	0.37	0.52	7.00	16.99	0.52	0.22	0.03	0.03	n.d.	0.06
	Mb_StDev	*0.93*	*0.04*	*0.06*	*0.07*	*0.02*	*0.17*	*0.39*	*0.03*	*0.08*	*0.02*	-	-	*0.01*
Sb + Mn	Mc	69.26	0.07	2.32	0.39	0.54	6.15	19.26	0.67	0.71	0.12	0.03	0.33	0.02
	Mc_StDev	*0.66*	*0.03*	*0.08*	*0.04*	*0.05*	*0.09*	*0.51*	*0.01*	*0.09*	-	*0.004*	*0.06*	-
Mn_a	Md	69.78	0.03	2.32	0.35	0.55	7.38	16.29	0.50	1.38	0.10	n.d.	n.d.	n.d.
	Md_StDev	*0.39*	*0.04*	*0.08*	*0.09*	*0.02*	*0.11*	*0.05*	*0.02*	*0.11*	*0.03*	-	-	-
Sb + Mn	Me	68.72	0.12	2.31	0.37	0.61	6.59	18.48	0.62	0.46	0.13	0.05	0.13	0.05
	Me_StDev	*0.43*	-	*0.05*	*0.08*	*0.02*	*0.12*	*0.55*	*0.01*	*0.01*	*0.03*	-	*0.08*	*0.03*
Sb + Mn	Mf	69.80	0.08	2.13	0.36	0.53	5.70	19.62	0.52	0.42	0.14	n.d.	0.71	0.02
	Mf_StDev	*0.30*	*0.03*	*0.03*	*0.04*	*0.03*	*0.05*	*0.29*	*0.07*	*0.05*	*0.05*	-	*0.12*	*0.01*
Mn_b	Mg	70.45	0.03	2.51	0.33	0.55	8.15	16.71	0.67	0.36	0.25	n.d.	n.d.	0.11
	Mg_StDev	*0.55*	-	*0.01*	*0.09*	*0.02*	*0.18*	*0.18*	*0.03*	*0.04*	-	-	-	-
Mn_b	Mh	70.16	n.d.	2.33	0.27	0.49	8.17	17.52	0.43	0.13	0.05	0.04	n.d.	0.05
	Mh_StDev	*0.35*	-	*0.07*	*0.04*	*0.02*	*0.02*	*0.20*	*0.02*	*0.02*	*0.02*	-	-	-
Mn_b	Mi	70.56	0.06	2.56	0.40	0.55	7.84	17.44	0.58	0.19	0.08	0.02	n.d.	0.06
	Mi_StDev	*0.24*	*0.04*	*0.04*	*0.06*	*0.03*	*0.07*	*0.11*	*0.02*	*0.00*	-	-	-	-
Mn_b	Ml	70.78	0.05	2.63	0.39	0.66	7.62	16.69	0.57	0.56	n.d.	0.01	n.d.	0.02
	Ml_StDev	*0.25*	*0.03*	*0.01*	*0.11*	*0.02*	*0.10*	*0.19*	*0.01*	*0.03*	-	-	-	*0.004*
Sb + Mn	Mm	70.23	0.11	2.06	0.41	0.52	5.82	19.26	0.63	0.35	0.02	n.d.	0.55	0.04
	Mm_StDev	*0.21*	*0.08*	*0.06*	*0.06*	*0.04*	*0.07*	*0.07*	*0.02*	*0.06*	-	-	*0.05*	-
Sb + Mn	Mn	70.33	0.07	2.33	0.34	0.65	6.54	18.41	0.65	0.61	0.11	0.03	0.24	n.d.
	Mn_StDev	*1.29*	*0.04*	*0.08*	*0.11*	*0.04*	*0.17*	*0.56*	*0.05*	*0.00*	*0.08*	*0.01*	*0.07*	-
Sb + Mn	Mo	69.00	0.09	2.25	0.49	0.69	5.84	18.43	0.75	0.55	0.16	0.02	0.43	0.03
	Mo_StDev	*0.36*	*0.02*	*0.05*	*0.09*	*0.01*	*0.13*	*0.19*	*0.01*	*0.04*	*0.05*	-	*0.02*	-
Mn_a	M15	69.70	0.06	2.49	0.20	0.62	7.30	17.70	0.54	1.47	0.08	n.d.	n.d	0.14
	M15_StDev	*0.15*	*0.04*	*0.03*	*0.02*	*0.00*	*0.06*	*0.12*	*0.04*	*0.05*	*0.08*	-	-	-
Mn_a	M16	69.53	0.07	2.39	0.20	0.59	7.26	16.59	0.60	1.43	n.d.	n.d.	n.d.	n.d.
	M16_StDev	*0.45*	*0.04*	*0.04*	*0.03*	*0.01*	*0.04*	*0.47*	*0.04*	*0.04*	-	-	-	-
Sb + Mn	M17	69.70	0.07	2.16	0.42	0.63	5.96	19.24	0.80	0.30	0.10	n.d.	0.60	n.d.
	M17_StDev	*0.50*	*0.09*	*0.02*	*0.03*	*0.03*	*0.02*	*0.20*	*0.02*	*0.04*	*0.05*	-	*0.09*	-
Sb + Mn	M18	69.57	0.06	2.30	0.45	0.72	6.68	18.18	0.98	0.74	0.15	0.04	0.19	0.03
	M18_StDev	*0.30*		*0.04*	*0.08*	*0.05*	*0.17*	*0.18*	*0.05*	*0.05*	*0.14*	-	*0.02*	*0.02*

In the Al_2O_3 versus MnO/(MnO + Sb_2O_5) diagram (Figure 3), the data obtained on the studied glasses were compared with a selection of: (1) glasses from the Archaeological Museum of Adria [5], (2) glasses of Carthage [8], (3) vitreous finds of Coppergate, York [15], (4) colourless glasses from the Roman ship *Iulia Felix*, wrecked off the town of Grado (Udine—Italy) [17], and (5) Mn-decoloured Roman vitreous objects and a naturally coloured raw glass chunk from Pompeii [34].

In Figure 3, the antimony free glasses (groups Mn_a and Mn_b) plot in the upper part of the diagram is consistent with the bibliographic data of the Roman manganese-decolourised glasses [5,8,15,17,34].

The other studied Muricelle samples were distinct from the antimony free glass groups (Figure 3). Considering the presence of variable quantities of Sb, this group showed a remarkable affinity with most of the Sb + Mn glass of bibliographic Roman mixed antimony and manganese glass [5,8,15,17]. They contained both antimony and manganese, with MnO/(MnO + Sb_2O_5) ratios varying from 0.34 to 0.80. This result is a consequence of the recycling of glass, which has been a particularly widespread practice during the imperial period, as suggested by previous studies [15,17,18,21,22].

The Muricelle glass groups, other than the contents and the type of decolourants, also differed from each other for soda, lime, and aluminium; however, they showed quite similar potash, magnesium, and iron oxide contents.

In the diagrams CaO versus Na_2O and CaO versus Al_2O_3 (Figure 4a,b, respectively), the three Muricelle groups plotted in distinct areas. In detail, the Mn_a and Mn_b samples were richer in CaO (>7 wt%) and poorer in Na_2O (from 16.29 to 17.70 wt%) than the Sb + Mn glasses which, on the contrary, showed a maximum CaO content of 6.59 wt% and

Na_2O varying from 18.18 to 19.72 wt%. Concerning Al_2O_3, all three groups showed similar concentrations (2.06–2.63 wt%).

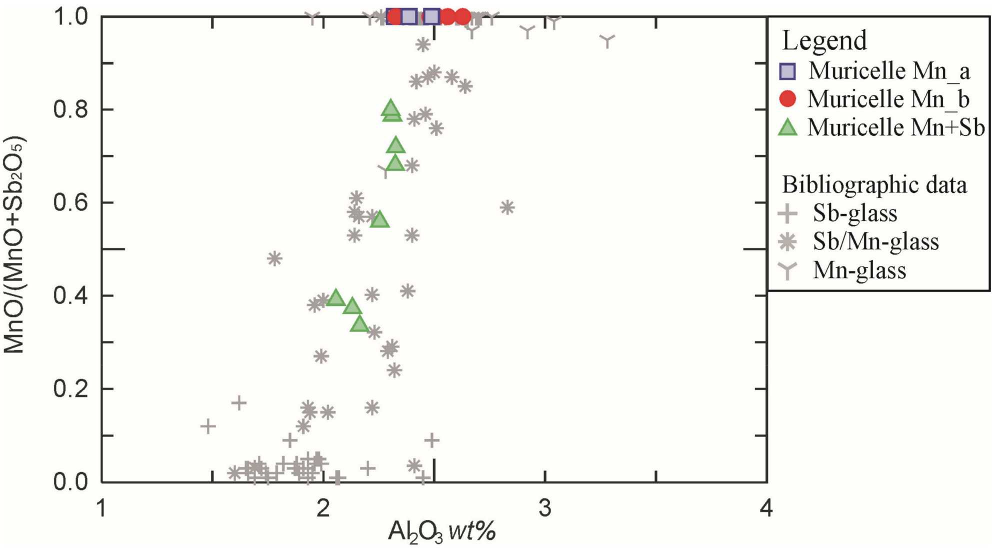

Figure 3. Al_2O_3 versus $MnO/(MnO + Sb_2O_5)$ diagram. The bibliographic data refer to coeval Roman glasses [5,8,15,17,34].

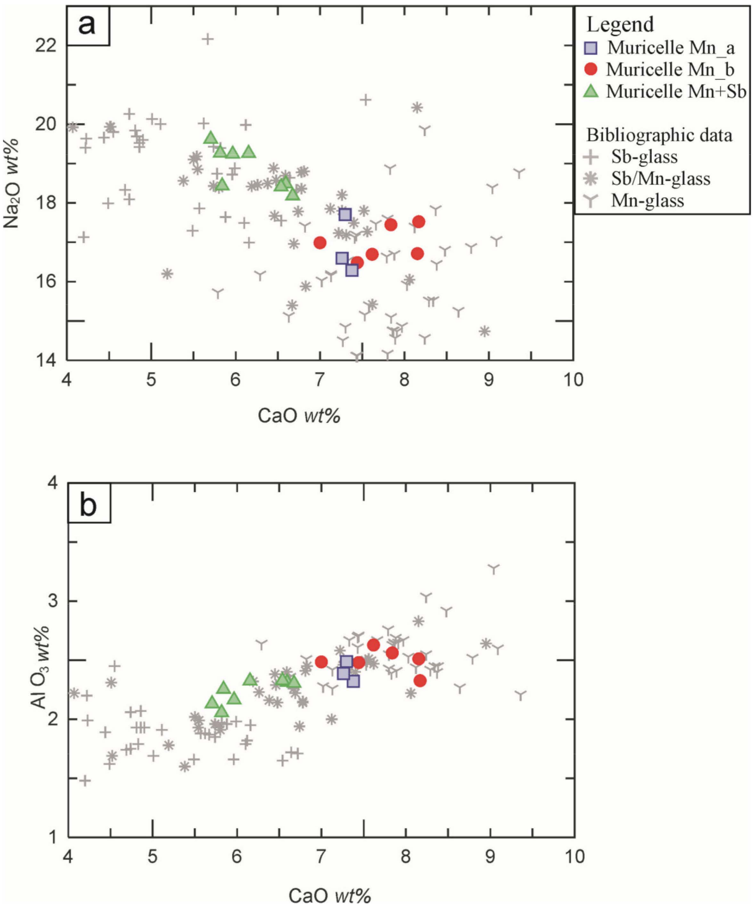

Figure 4. (**a**) CaO versus Na_2O; (**b**) CaO versus Al_2O_3. The bibliographic data refer to coeval Roman glasses [5,8,15,17,34].

In the diagrams of Figure 4a,b, the comparison between the Muricelle glasses and the bibliographic data confirmed the similarity of Mn_a and Mn_b groups with the Mn glass of Gallo et al. [5], Schibille et al. [8], Jackson and Paynter [15], Silvestri et al. [17], and De Francesco et al. [34]. At the same time, the Sb + Mn samples overlapped with the Sb + Mn glasses of Gallo et al. [5], Schibille et al. [8], Jackson and Paynter [15], and Silvestri et al. [17].

As for the raw materials used for the glass production, some major and minor elements such as CaO and Al_2O_3 content and even iron and titanium are indicative of the care selection of sand [3] and above all, the presence of impurity such as calcite, feldspar, clay minerals, and heavy minerals. The CaO contents observed in the Muricelle glass groups highlighted the use of sands with a variable content of calcium during the glassmaking production.

The FeO versus MnO plot (Figure 5) shows the low values of FeO (below 0.5 wt%) for all the analysed glasses, which together with the high value of SiO_2 (Table 3) confirms the use of selected sands. The MnO content, as stated above, had variable values; particularly, the high concentrations (around 1.5 wt%) of MnO in the Mn_a group indicated the deliberate addition of this element as a decolouring agent [19] and the probable use of pyrolusite (MnO_2), the most common manganese minerals [26], which was a very widespread technique in the Roman period to neutralize the colour due to the iron oxides naturally present in the raw materials [18,19,37]. The Mn_b and Sb + Mn groups showed similar contents of Fe, confirming that the iron content was low for all Muricelle glasses, highlighting the absence of intentional addition. On the other hand, the MnO contents > 0.025 and ≤ 1 wt% suggested the possibility of unintentional addition of this element through a recycling process [8,15,21].

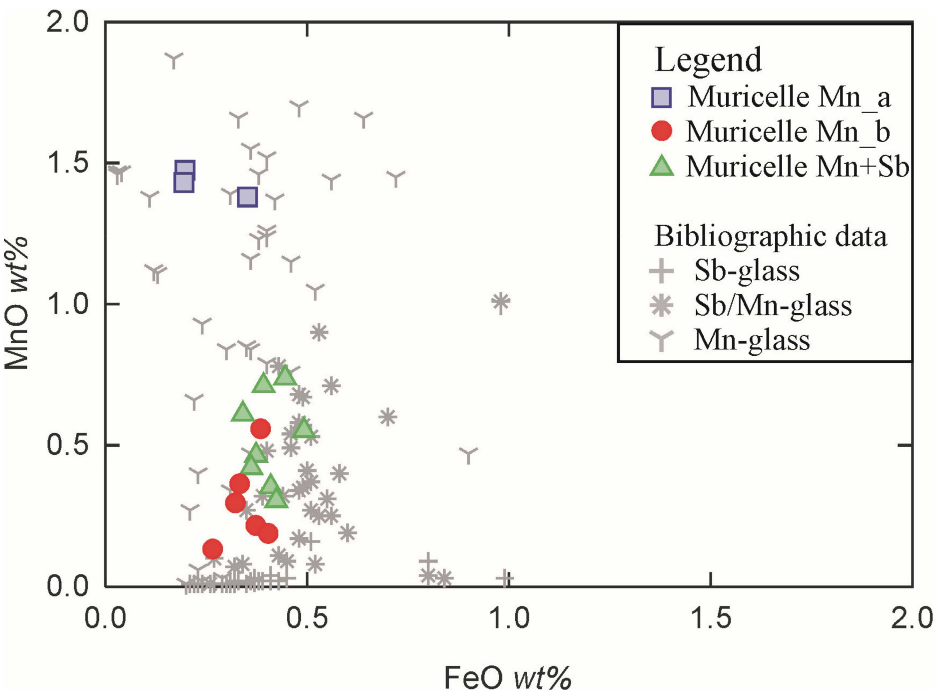

Figure 5. FeO versus MnO diagram. The bibliographic data refer to coeval Roman glasses [5,8,15,17,34].

In all samples of Mn_b and Sb + Mn groups and in the M15 sample of Mn_a group, CuO and/or PbO and/or SnO were detected in amounts higher than 100 ppm; this also suggested the production through recycling involving coloured glass scraps added in the re-melting process [5,15,17,19].

Further information on the raw materials and their origin could be obtained from the diagram Al_2O_3/SiO_2 versus TiO_2/Al_2O_3 (Figure 6), as these oxides represent the quartz content (SiO_2), the feldspar content (Al_2O_3), and the heavy minerals content (TiO_2) [5,8,17,22].

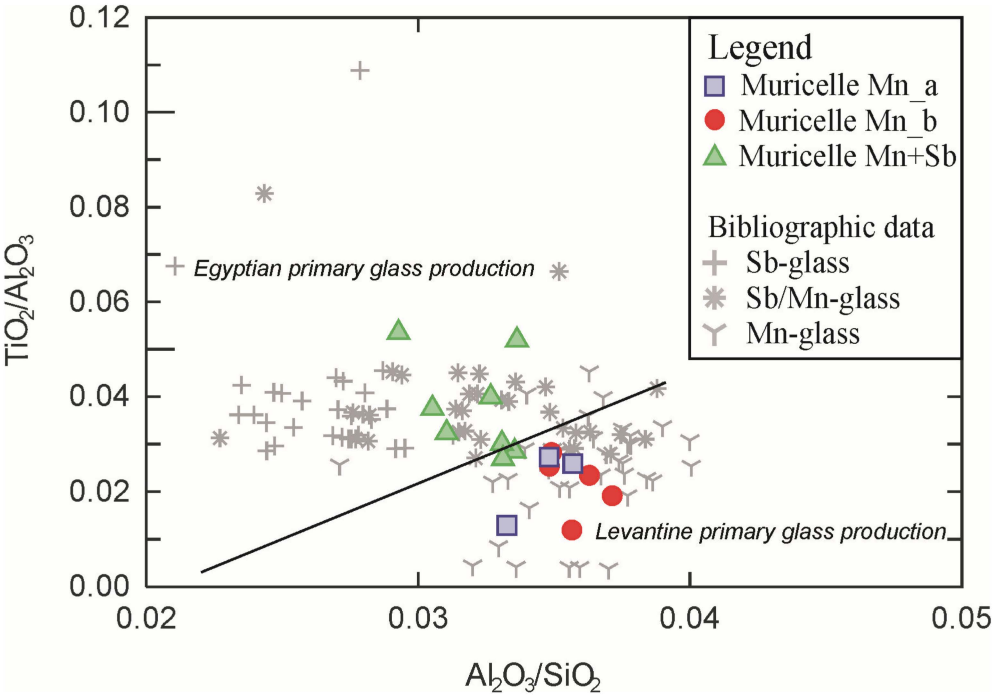

Figure 6. Al_2O_3/SiO_2 versus TiO_2/Al_2O_3. The bibliographic data refer to coeval Roman glasses [5,8,15,17,34].

Analysing Figure 6, we observed that the Muricelle glasses perfectly fit the coeval bibliographic data [5,8,15,17,34].

All samples of Mn_a and Mn_b groups overlapped with the Roman-Mn glass. On the contrary, no fragments belonging to the Roman-Sb group were recognised. The Sb + Mn Muricelle group, as stated above, included recycled glasses. In particular, the Sb + Mn samples plot between the two bibliographic end members (Roman-Sb and Roman-Mn) reflected the proportion of the two glass types during the re-melting process. All the recycled glass samples plotted in the central area of the diagram, close to the coeval bibliographic data of the Roman-Sb + Mn group [5,8,15,17].

Based on the Al_2O_3/SiO_2 and TiO_2/Al_2O_3 ratios, Schibille et al. [10] proposed a subdivision of primary glasses into two provenances from Egypt and Levant. In detail, Schibille et al. [10] assigned to the Roman-Sb glasses an Egyptian origin and to the Roman-Mn glasses a Levantine coast origin. The Mn_a and the Mn_b Muricelle glasses were produced mainly using Levantine sands; on the other hand, for the glasses from the Sb + Mn group, it is more complicated to define the raw materials provenance because they are recycled and therefore represent a mixture of the two end members (i.e., Sb and Mn colourless glass).

6. Conclusions

The chemical characterization carried out by EPMA on seventeen light coloured glass finds dated first to the third century AD from the archaeological site of Muricelle near Cosenza in Calabria (Italy) highlighted different compositions attributable both to employed raw materials and production technologies. Although all the studied finds were soda–lime–silica glasses produced with natron as flux, the chemical composition and the comparison with coeval productions revealed the presence of three groups. The samples of Mn_a and Mn_b groups were classified as Roman-Mn and Mn-unintentionally coloured Roman glasses, respectively; on the other hand, the Sb + Mn group included samples obtained through recycling Mn- and Sb- Roman glass. The first two groups (Mn_a and Mn_b) showed a remarkable compositional homogeneity, especially for calcium, sodium, and aluminium contents but were distinguished by the content of MnO added as a decolourant in the Mn_a group. The Sb + Mn group, unlike the other two, contained antimony and showed different chemical concentrations in calcium, sodium, and aluminium, suggesting the use of different raw materials in the primary glass centres. In all samples, except for the Md and M16 glasses from Mn_a group, the presence of Cu, Sn, and Pb oxides in amounts higher than 100 ppm suggested the recycling involving the inclusion of coloured glass in the cullet.

Considering the compositional characteristics and the differences among the analysed glasses, despite the recycling, the samples pertaining to Mn_a and Mn_b groups could be considered as being produced mainly from glass originating in primary Levantine workshops, while the samples from the Sb + Mn group resulted from recycling glass manufactured in both Levantine and Egyptian primary workshops.

These new data improve the knowledge about the Muricelle site, and they are in agreement with the results on coeval finds from different Roman archaeological sites dated to the first–third century AD. Moreover, the different raw material provenance of the Muricelle glasses indicated that the site was commercially active and played a leading role within the middle valley of Crati River during the imperial age.

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