CHEMICAL COMPOSITION OF LATE 18TH- AND 19TH-CENTURY GLASS BEADS FROM WESTERN NORTH AMERICA: CLUES TO SOURCING BEADS

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The Sullivans Island glass bead collection at the Smithsonian's National Museum of Natural History contains over 56,000 beads which date from the late 18th to the late 19th century. Excavated in the 1930s from a site on the Columbia River in the Plateau region of North America, this collection contains examples of most known bead varieties for this time period. Many of the beads conform to varieties that have been attributed to Bohemia, Venice, and China—three of the principal bead-producing centers at the time. One hundred and twenty-four beads were subjected to Laser-Ablation Inductively-Coupled Mass-Spectrometry (LA-ICP-MS) analysis at the Smithsonian's Materials Conservation Institute to see if the chemical composition of the glass could be correlated with a place of origin. The results revealed several distinct compositional groups, some of which could be linked to geographical areas.

INTRODUCTION

Now submerged beneath the waters of the Columbia River in Washington state, the burial site known as Sullivans Island was located at the head of a series of rapids called the Cascades (Cox 1832; Strong 1959). Smithsonian archaeologist Herbert Krieger conducted excavations on the island in 1934 while investigating sites threatened by the construction of the Bonneville Dam (Krieger 1935; Phebus 1978). The Sullivans Island site yielded over 56,000 glass trade beads along with thousands of other historic-period objects, including a high number of copper pendants, bracelets, and other ornaments. The beads and other objects indicate a date range from the late 18th to the late 19th century (Burgess 2004). This assemblage is one of a handful of major 19th-century bead collections from North America and is curated by the Department of Anthropology, National Museum of Natural History, Smithsonian Institution.

The breadth of the Sullivans Island collection is in part due to the location of the island at the Cascades, one of the richest salmon fisheries on the Columbia River. Strong (1959) found that phoenix buttons occur in greater quantities in productive fishing areas, and this holds true for other objects as well. In addition to being a major fishing area, the Cascades also formed a barrier to water travel. Since it was necessary to portage around the Cascades, the falls served as a crossroads for items moving up and down the river, both in prehistoric and historic times. A number of historical accounts refer to the difficulties encountered by fur traders and settlers when dealing with tribes during the long portage. Minor et al. (1985) write that in the early parts of the 19th century, no one could portage without having to hand over goods to the local inhabitants. In 1814, a Northwest Trading Co. group was attacked and lost an amazing amount of material which, according to Minor et al. (1985:60), would have accelerated cultural changes in the Cascades area. Goods obtained by the inhabitants through seizure or, later, through trade likely contributed to the wealth of Sullivans Island, and to the breadth of the bead collection. New settlers continued to use the portage through the 1850s, bringing with them "tons of clothing, tools, dishes and utensils" (Minor et al. 1985:60).

This assemblage of beads from a key area of the Plateau region contains varieties that have generally been ascribed to Venice, Bohemia, and Asia, namely China. Attributing geographical sources to archaeological glass beads has been a challenging proposition, but is an area where chemical-composition studies may prove helpful. Historical records have provided most of the known information on the sources of glass beads, but chemical analysis of glass has shown some promise in this area (Glascock and Speakman 2002). The majority of 19th-century glass beads were made in Venice, a major glassmaking center for centuries, with other varieties originating in Bohemia and other European locales. Glass beads were also made in Asia and certain varieties of glass beads are attributed to China (Francis 1986, 2002; Ross 1990, 2000). The bead varieties thought to be manufactured in China tend to appear frequently in the Pacific Northwest and in Alaska. The Sullivans Island collection from the Plateau region, which abuts the Pacific

Northwest, provides an opportunity to sample a substantial number of beads that have been tentatively attributed to each of the three major centers of bead production.

This study was undertaken to identify potential differences between the composition of 19th-century glass beads potentially from China and beads thought to be from European sources, namely Venice and Bohemia (Burgess and Dussubieux 2005). Glascock and Speakman (2002) have used laser ablation-inductively coupled-mass spectrometry (LA-ICP-MS) to identify the composition of Venetian and Bohemian glass beads. For the Sullivans Island study, the major research question was to see if glass attributed to China would exhibit a different chemical composition than glass thought to have a Venetian or Bohemian origin. It was hoped that the LA-ICP-MS analysis would provide some chemical markers that could help differentiate between various bead sources for this collection and for other 19th-century bead collections.

Chemical studies of glass trade beads have been undertaken in the recent past to help establish chemical chronologies for beads or determine the place of origin (Hancock et al. 1994, 1997, 1999; Hancock et al. 2000; Kenyon, Hancock, and Aufreiter 1995; Kenyon, Kenyon, Hancock, and Aufreiter 1995; Sempowski et al. 2000, 2001). Until recently, instrumental neutron activation analysis (INAA) was the technique most used because of its nondestructive nature. After a "cooling-down" period, beads could be returned to their owners unchanged (Glascock and Speakman 2002; Hancock et al. 1994, 1999). Despite its many advantages, INAA requires access to a reactor and the number of elements that can be identified using this technique is limited. Far more elements can be measured with LA-ICP-MS which is a multi-elemental analytical technique, allowing the identification of more than 50 major, minor, and trace elements in the glass with no visible damage (Gratuze et al. 2001). Temporally sensitive trace element patterns related to the use of copper as a coloring agent in turquoise-blue glass beads have been identified using this technique (Billeck and Dussubieux 2006).

In North America, chemical analyses have primarily been conducted on beads from archaeological sites located in Canada and the northeastern United States, although LA-ICP-MS studies have been performed on beads from western Missouri (Glascock and Speakman 2002) and from the Plains region (Billeck and Dussubieux 2006). The Sullivans Island study is one of the first to sample beads from the far-western region of North America. It is also one of the later historic-period collections to be studied in this manner.

Hancock et al. (1994) concluded that due to the dispersion of Italian glassmakers and the broad trade in raw materials in 17th-century Europe, the chemical composition of archaeological beads from the Great Lakes area could not be used to identify specific European sources of beads; e.g., Amsterdam or Venice, the two dominant sources of glass beads at the time. Nonetheless, since the Sullivans Island assemblage appeared to have beads that were likely from both Asia and Europe, it might be possible to at least distinguish between European and Asian glass compositions and, at best, between Chinese, Bohemian, and Venetian glass. Due to the wide range of elements measured with LA-ICP-MS and the high sensitivity of this technique, studies of the sources of ancient glass in South and Southeast Asia (Dussubieux and Gratuze 2004) and Africa (Robertshaw et al. 2003) have met with some success.

In his work with the Hudson's Bay Company's Fort Union and Fort Vancouver bead collections, Ross (1990, 2000) exhaustively reviewed historical records to assess bead sources, and identified Great Britain and China as distribution centers, and China, Bohemia, and Venice as likely manufacturing sources for the beads in 19th-century assemblages. Fort Vancouver, Washington, is located about 80 km downriver from Sullivans Island and, while a number of the highly ornate beads may not have come from the fort (Lester Ross 2004: pers. comm.), a large part of the collection was probably obtained directly or indirectly from the fort inventories.

Beads were selected for the Sullivans Island study based on descriptions and characteristics that have been linked to known bead manufacturing sites. Mold-pressed beads and drawn faceted beads are considered to have been made in Bohemia (Neuwirth 1994; Ross 1990, 2000, 2003; Ross with Pflanz 1989). Venice is assumed for the bulk of 19th-century beads since Venice was the dominant center of bead manufacture for this time period (Karklins 1985; Karklins with Adams 1990; Ross 1990, 2000). Although wound beads were produced in Venice, drawn beads were emphasized for this sample group. Beads from China are referred to as Canton or China beads in historical records and Ross (1990) found that these large, blue wound beads were imported in great quantities. Nineteenth-century beads also attributed to China include highly transparent/translucent, strongly pigmented wound specimens with a great number of air bubbles in the glass (Francis 1986; Liu 1995). Therefore, based on characteristics, comparison with other collections, and the archaeological and bead-research literature (Francis 1979a-b, 1986; Glascock and Speakman 2002; Karklins et al. 2002; Kenyon, Hancock, and Aufreiter 1995; Liu 1995; Ross 1990, 2000), three bead groups were identified for sampling: 1) beads that could potentially be attributed to Venice; 2) those attributed to Bohemia; and 3) varieties with characteristics attributed

to Chinese beads. It is worth emphasizing the hypothesized nature of these groups, especially the Venetian and Chinese ones.

These three groups were sampled using LA-ICP-MS to determine if the glass composition varied enough to distinguish between the three potential sources and to see if European glass could be distinguished from Asian glass. It was hoped that the major, minor, and trace elements that LA-ICP-MS can identify would provide a finer look at the glass composition than could be ascertained using INAA.

METHODOLOGY

The 56,343 glass beads from Sullivans Island had been classified (Burgess 2004) using the Kidd and Kidd (1970) system as expanded by Karklins (1985), and also following Ross (1990, 2000). Classification was based on manufacturing type, diaphaneity, shape, presence or absence of decoration, color, and size. Diaphaneity was recorded as transparent (tsp), translucent (tsl), or opaque (op). Length and least diameter were recorded, although size grading was done according to diameter size. Beads were separated into the following size groups: 0-2 mm = very small; 2-4 mm = small; 4-6 mm = medium; 6-10 mm = large; and 10+ mm = very large. Small, medium, large, and very large beads occurred within the sample groups.

A total of 128 beads representing 31 varieties were tested. Where possible, five beads from each variety were selected in order to test for consistency between visually identical beads. Color was controlled for, as much as possible, by trying to include beads of similar color across the sample groups, although both monochrome and polychrome beads were included. Different glass chemistries have been linked to the two principal pre-19th-century bead manufacturing techniques, with drawn beads primarily having a soda or soda-lime composition while wound beads have a potash-glass composition; lead occurs in both wound and drawn varieties (Hancock et al. 1996; Hancock et al 1997; Karklins et al. 2002). These findings were not supported by the Sullivans Island beads of probable Bohemian origin.

The compositional analyses were carried out at the Smithsonian Institution's Materials Conservation Institute in Suitland, Maryland, with a Perkin Elmer Elan 6000 Inductively Coupled Plasma-Mass Spectrometer (ICP-MS) connected to a Cetac LSX-200 *Plus* UV laser for direct introduction of solid samples. The single point analysis mode with a laser beam diameter of 100 μm was selected. The laser operated in Q-switch mode at 100% of the laser energy (4.8 mJ) and at a pulse frequency of 20 Hz. A 20 s pre-ablation time was set to ensure that possible surface contamination did not affect the results and to eliminate the transient part of the signal. A blank signal was acquired followed by three runs for each glass sample. The average signal corrected from the blank was considered for the calculation of concentrations.

To improve reproducibility of measurements, the use of an internal standard is required. The isotope Si^{30}, a major constituent of glass, was selected. The concentrations of the major and minor elements were calculated, assuming that the sum of their concentrations in weight percentage is equal to 100% (Gratuze 1999).

Quantitative results are obtained by comparing the signal intensity measured for a given element in a sample to the signal intensity for the same element in a standard reference material (SRM) with certified concentrations. NIST glass SRM 610 and Corning Glass A, B, C, and D (Brill 1999) were used to calculate major and minor element concentrations. Only SRM 610, using concentrations from Pearce et al. (1997), was used for the calculation of trace element concentrations expressed as ppm of elements.

Silica (SiO_2), alumina (Al_2O_3), soda (Na_2O), potash (K_2O), magnesia (MgO), lime (CaO), and iron oxide (Fe_2O_3) are considered the most relevant constituents for identifying glass ingredients and for classifying the different glass groups. All compositions were recalculated and reduced to these seven oxides (Brill 1999). Reduced concentrations will be in italics or indicated as such. In the case of lead glass, when lead was used as flux, there is some information loss in using reduced concentrations, but information on the composition of the base glass is still present. Oxide concentrations are reported in this study and are indicated by the use of the terms potash rather than potassium and lime rather than calcium.

THE RESULTS

Bohemian Beads

The glass beads that were identified as Bohemian for this study are mold-pressed specimens with ground facets and a biconical perforation; a mold-pressed bead with molded facets and a straight perforation; and drawn faceted beads (Pl. XA). The two blown-in-the-mold beads were inaccurately placed in the supposed Venetian group for sampling, but yielded glass compositions consistent with the other Bohemian beads. Neuwirth (1994) also attributes these beads to Bohemia.

Ross (2003) has dated the mold-pressed beads with ground facets and biconical perforations to the first half of the 19th century. Three such beads were sampled.

Mold-pressed beads with molded facets and a straight or cylindrical perforation are thought to date to the second half of the 19th century (Ross 2003). One bead of this type was sampled. The beads with ground facets and those with molded facets show different chemical compositions, which is particularly interesting since these bead varieties have temporal implications. The three beads with ground facets have a high-potash high-lime (K-Ca) glass type, which was the predominant glass encountered in the Bohemian beads. The one molded-facet bead that was sampled has a K-Al glass type. It is the only bead to have this composition in the entire study and it is one of the later beads in the collection.

The drawn faceted beads in the study have been frequently called "Russian blues," although this has long been considered a misnomer. These beads are frequently blue, but also occur in a variety of other colors. The sampled beads were dark blue, colorless, and green. The chemical composition of these beads, which was K-Ca glass, places them in the Bohemian group. They have six flat sides and a series of ground facets at either end. They occur throughout the 19th century and may first have appeared in the late 18th century (Ross 2000).

Most of the Bohemian beads have a high-potash high-lime or K-Ca composition (Table 1). The major constituent of this type of glass, after silica, is potash and the concentration ranges from 11.8 to 17.0%. The lime content is slightly lower and ranges from 6.1 to 10.6%. Soda concentrations can be as high as 2.3% but in most cases soda, as well as alumina and magnesia, concentrations are below 1.0%. This high concentration of potash and lime seems to indicate the use of forest-plant ashes as a flux (Turner 1956). The silica source is a sand with low quantities of alumina (< 1%).

Table 1. Average Reduced Composition for High-K-Ca Glass Attributed to Bohemia.

	Average and Standard Deviation
SiO_2	75.5 +/- 1.3%
Na_2O	1.1 +/- 0.5%
MgO	0.25 +/- 0.0.7%
Al_2O_3	0.6 +/- 0.1%
K_2O	13.9 +/- 1.2%
CaO	8.5 +/- 1.1%
Fe_2O_3	0.16 +/- 0.09%

There have been similar findings for 19th-century Bohemian glass and beads: potassium (K) and calcium (Ca) levels of 9.0-15.0% and 5.0-7.5%, respectively, for Bohemian glass, and 12.0-13.0% K and 5.4-8.0% Ca for 19th-century faceted beads (Kenyon, Kenyon, Hancock, and Aufreiter 1995; Ross 2000). Glascock and Speakman (2002) found that Bohemian beads had low magnesia (MgO less than 1000 ppm) and high potash.

The K-Ca glass beads are translucent and opaque dark blue, opaque white, transparent green, dark yellow, and colorless. Table 2 shows the elements or compounds involved as colorants. Dark blue beads are colored with cobalt. Arsenic is ubiquitous in the Bohemian glass but it seems that a correlation exists between nickel, an element only present in significant quantities in cobalt-blue beads, and arsenic.

Table 2. Coloring and Opacifying Elements in the High-Potash High-Lime Glass.

Color	Detected Elements	Coloring or Opacifying Agent	Comments	Reference
tsl. dark blue	Co	Co^{2+}	Association Co-Ni-As	Scholze 1980
op. dark blue	Co, P	Co^{2+}, $3Ca_3(PO_4)_2.CaF_2$ (bone ashes)	Association Co-Ni-As	Moretti and Hreglich 2005; Scholze 1980
tsl. white	P	$3Ca_3(PO_4)_2.CaF_2$ (bone ashes)		Moretti and Hreglich 2005
tsl. green	Cu, Pb	Cu^{2+} in a lead-containing glass		Scholze 1980
colorless	As, Mn	Oxidize Fe^{2+} into Fe^{3+}		Scholze 1980
tsl. dark yellow	Fe	Fe^{3+} - S_n^{2-}	Sulfur is not measured with LA-ICP-MS	Schreurs and Brill 1984

Bismuth is an element that has higher concentrations in cobalt-blue beads (Fig. 1). Cobalt associated with arsenic, nickel, and occasionally with bismuth is found in glass dating to the 16th century and onward, and probably originates from the mining district of Schneeberg (Erzgebirge) in Germany (Gratuze et al. 1996).

In addition to the K-Ca glass, three other glass types are identified in the present study among the mold-pressed beads attributed to Bohemia: 1) two red beads with ground facets are composed of lead-silica (Pb-Si) glass; 2) transparent yellow beads without facets are composed of high-soda high-lime or Na-Ca glass; and 3) white beads with molded facets are of a high-potash high-alumina or K-Al glass.

The mold-pressed red beads have a distinctive Pb-Si composition with slightly more than 50% lead and very small quantities of alkali. These beads also contain gold (44 ppm) which is used as a colorant and according to Weyl (1951:385), "the most desirable colour shades and the most intense colours are obtained in lead glasses because the higher the lead content, the more soluble is gold."

The transparent yellow mold-pressed beads with a Na-Ca composition contain soda (15%), lime (6.6%), and potash (2.5%). Other elements have concentrations below 1.0%. These beads are the only ones in the sample to contain uranium (209 ppm). Cadmium was also detected, along with significant quantities of zinc (3700 and 3600 ppm, respectively). These elements may have been added to the glass as colorants.

The white mold-pressed beads with molded facets have an Al-K composition. While alumina has a concentration lower than 1.0% in the other Bohemian glass beads, the white beads contain 16%. Potash concentrations range from 8.7 to 11.8%, soda has concentrations around 3%, and lime concentrations range from 0.13 to 2.8%. This composition seems to correspond to a type of glass described by Silverman

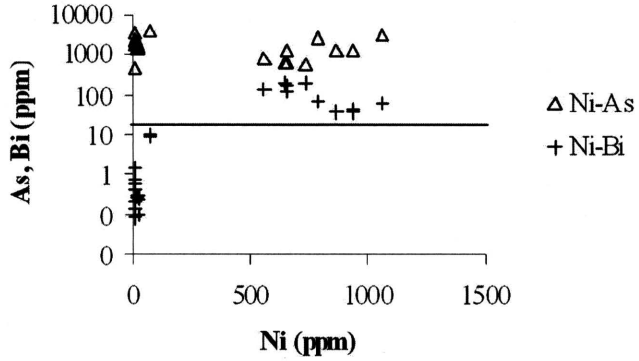

Figure 1. Nickel, arsenic, and bismuth concentrations in the high-potash high-lime glass.

(1918) which results from a mix of what was referred to as Russian potash, containing a small proportion of soda with no significant quantities of lime, as well as an alumina-rich ingredient. The introduction of the latter into this type of glass would have rendered it opaque. Alumina limits the solubility of other substances present in the glass batch (e.g., chlorides). Two of the five white beads have very low quantities of lime (0.1 and 0.2%) along with low quantities of phosphorus (< 1.0%). The presence of more phosphorus and lime in some beads of the white mold-pressed group may be due to the addition of bone ash (Moretti and Hreglich 2005), also used for opacifying purposes.

Of the two blown-in-the-mold beads, one is white with linear ridges, two annular ridges, and translucent blue stripes. The second one is a fragment of opaque purple-blue. These beads had been included in the group potentially from Venice, but their K-Ca composition is consistent with a Bohemian origin. Both contain phosphorus in quite high concentrations (more than 6%) and bone ash may have been used as an opacifier, as was also indicated for the white beads in the Bohemian group. Cobalt imparts a purplish-blue color and, as with the Bohemian dark blue beads, it is accompanied by arsenic, nickel, and bismuth, suggesting a common cobalt source.

Beads Attributed to Venice

The small, drawn, monochrome and polychrome beads that dominate 19th-century archaeological collections were the primary varieties selected for the sample group hypothesized to be from Venice (Pl. XB). For the most part, these heat-rounded beads are referred to as embroidery beads or, sometimes, seed beads, a label that is too often used incorrectly. They were typically used for beadwork and generally fall into the small (2-4 mm) size category. Opaque blue, opaque white, translucent red, translucent green, and colorless varieties (Kidd and Kidd type IIa) were sampled. Drawn red-on-white beads were included, as were opaque white-on-white beads. The latter are identical to the ones illustrated by Ross (1990:45, Table 5, Pl. I) which are listed as FOVA variety numbers 1040 and 1089. While red-on-white beads are sometimes called "whitehearts" or *cornaline d'Aleppos*, the term red-on-white is preferred and will be used here. Faceted drawn beads (Kidd and Kidd type IIf) of a reddish-purple color were sampled as well to see if they fell within the Bohemian or the Venetian glass compositions. These tumbled beads usually have one or two facets ground onto their surfaces.

The glass composition of the beads attributed to Venice showed more overall heterogeneity than the Bohemian

beads. Two different glasses were identified within the Venetian group—a non-lead glass and a lead glass (Table 3). The non-lead glass contains low quantities of lead, generally below 1.0%, which were probably unintentionally added to the glass along with another ingredient. Non-lead glass beads have a high-soda high-lime or Na-Ca composition with an average concentration of 12.4% soda and 8.7% lime.

The beads with non-lead compositions are all drawn and heat rounded: opaque turquoise-blue of small and medium size; translucent reddish-purple with ground facets; and the opaque white-on-white beads. Both the white and the blue varieties are among the most ubiquitous beads found on 19th-century sites. The turquoise-blue color is due to the presence of 1.4-1.7% of copper. The faceted reddish-purple beads have a glass composition that differs from the faceted beads attributed to Bohemia. The reddish-purple glass is colored with manganese, with concentrations ranging from 1.7 to 2.3%. Manganese occurs with a concentration of 0.12% or less in the other Na-Ca glass samples attributed to Venice.

The glass of the opaque white-on-white beads contains about 5% antimony. This element may have combined with calcium to produce calcium-antimony oxide, an opacifier (Rooksby 1962). This opacifier has been used in Venice in the production of white glass since the 15th century. Moretti and Hreglich (2005), and Sempowski et al. (2000) dated its occurrence in white beads found in northeastern North America to the beginning of the 17th century. Its appearance here, in a bead collection dating from the late 18th to the late 19th century, helps support the hypothesis that the opaque white-on-white beads were manufactured in Venice. In early North American bead collections, proving which beads were made in Amsterdam and which were made in Venice has been highly difficult. Since Amsterdam was no longer a bead-producing center during the time period covered by the Sullivans Island collection, it is not considered to be a potential source for any of the beads in this study.

Within the non-lead glass, potash has an average concentration of 6.0% and magnesia 1.8%. The potash and magnesia concentrations, higher than 1.5%, suggest the use of halophytic plant ash as a flux (Shortland et al. 2006). It has been established that Venetian glassmakers imported plant ash from the Levant (Verità and Toninato 1990). Lime could have been a constituent of the sand used, and high strontium concentrations (650 to 1450 ppm) and the correlated lime-strontium observed in Fig. 2 indicate that either the sand was taken from a coastal deposit or that crushed seashells were added to the glass batch to stabilize the glass (Freestone et al. 2002; Jackson 2005).

The other beads in the group thought to originate from Venice are composed of lead glass. Three varieties of drawn, monochrome heat-rounded beads contain lead: the opaque white beads, the translucent red beads, the translucent green beads, and the colorless beads. The drawn, polychrome, heat-rounded red-on-white beads are also in the lead-glass group.

Lead glass can be either potash- or soda-like. The lead glass encountered in these samples is referred to as Pb-(Na, K). Actually, no discrete groups appeared, but an inverse soda-potash concentration correlation was observed (Fig. 3). That kind of correlation has already been noted in non-lead Venetian glass by Šmit et al. (2005). Verità and Toninato (1990) explain this change in the soda concentrations of Venetian glass as a result of a decrease in the quality of the plant ash used, causing a diminution in its soda content. Alumina and magnesia concentrations are quite low (< 1.5%) and lime ranges from 0.1 to 6%. Strontium concentrations in this glass are always below 300 ppm.

Table 3. Reduced Average Compositions with Standard Deviations of Glass thought to be Venetian.

	Non-Lead High-Soda High-Lime Composition	Lead Glass
SiO_2	69.4 +/- 1.3%	74.9 +/- 3.4%
Na_2O	12.4 +/- 1.3%	6.0 +/- 2.8%
MgO	1.8 +/- 0.2%	0.7 +/- 0.3%
Al_2O_3	1.1 +/- 0.3%	0.6 +/- 0.3%
K_2O	6.0 +/- 1.5%	13.1 +/- 6.1%
CaO	8.7 +/- 1.5%	4.4 +/- 1.3%
Fe_2O_3	0.6 +/- 0.2%	0.4 +/- 0.4%

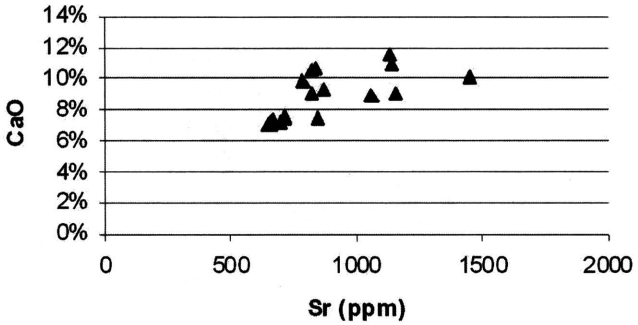

Figure 2. Sr-CaO graph for the high-soda high-lime glass attributed to Venice.

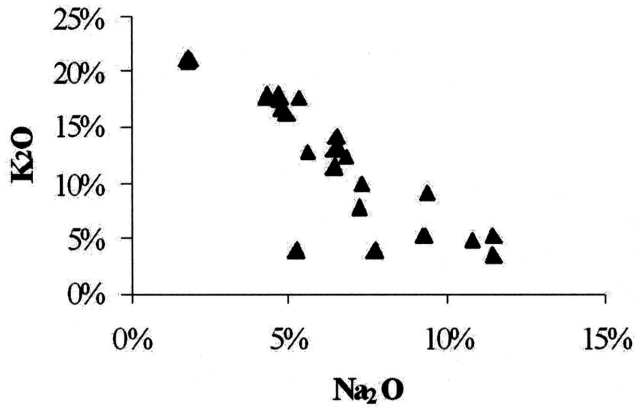

Figure 3. Soda and potash reduced concentrations in the lead-glass beads attributed to Venice.

Correlations between the composition of the beads and their color were observed throughout this study (Fig. 4). In the lead-glass group, the drawn green beads contain from 40 to 60% lead and no arsenic. They also contain copper in quantities higher than the other samples; 0.9% on average, while copper concentrations are lower than 0.1% in all the other beads. Copper ion Cu^{2+}, in a lead glass matrix, imparts a green color to glass (Scholze 1980). White beads not only contain lead but also arsenic in concentrations ranging from 2.6 to 5.7%. A lead-oxy-arsenate compound is a white opacifier that was identified in Venetian glass as early as the first half of the 18th century (Rooksby 1962). Moretti and Hreglich (2005) have reported a glass recipe dating to the end of the 17th century that involves the use of arsenic and lead oxide to produce an opalescent glass.

The translucent red and the colorless drawn beads have lower lead concentrations (<20%) and both varieties contain arsenic. Gold was detected in red glass in concentrations ranging from 122 to 294 ppm. Gold was detected in the transparent red Lycurgus Cup dating from the 4th century A.D. (Barber and Freestone 1990) and Hunt (1976) alleges that an Egyptian manuscript from the Greco-Roman era makes reference to red glass colored by gold. It was, however, only in the 17th century that a stable gold precipitate was discovered—the "purple of Cassius"—and successfully applied to the coloration of glass. The purple of Cassius consists of finely divided gold and colloidal stannic acid that gives a redder color to the glass in the presence of lead (Carbert 1980). The red beads contain lead but their tin concentrations are very low. Antimony concentrations are higher in the red beads compared to the other ones and it may have acted as a reducer instead of tin.

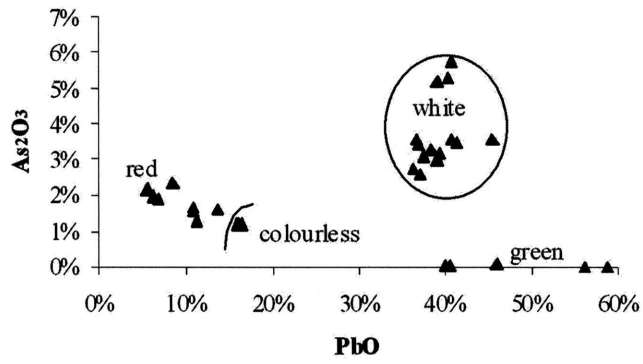

Figure 4. Lead and arsenic concentrations in the lead-glass beads attributed to Venice.

The colorless beads have a very specific composition; they have the highest concentration of potash (17.5%) and the lowest concentration of soda (1.5%). They also have extremely low concentrations of iron oxide (< 0.1%). Arsenic can also act as a decolorizer and is able to oxidize iron producing Fe^{3+} ions that have a very low coloring power. Lead may have been added to colorless glass because of the shine that it imparts.

Notably, the white glass in the drawn, heat-rounded IIa beads differs significantly from the white glass used for the Bohemian mold-pressed beads. The white Bohemian beads are composed of a potash-alumina glass, whereas the white Venetian glass has lead as the major element after silica, with potash and soda. Interestingly, the drawn opaque white-on-white beads in the sample group are non-lead beads, and have soda, lime, and potash as the major elements following silica. These small, multi-layered white beads and the drawn Kidd and Kidd IIa beads dominate collections during the period that Sullivans Island was occupied.

Red-on-White Beads

Two varieties of beads with white cores and red outer layers were included in the study: the drawn red-on-white beads, whose compositions fall into the lead-glass group defined for the Venetian material, and one wound-on-drawn bead.

Made by combining the two major bead-manufacturing processes (winding and drawing), the rare wound-on-drawn bead has a translucent red outer layer wound over a drawn, tubular, opaque-white core. Only three of these beads had been previously encountered in North America, and they were identified by Karklins (1985) and Sprague (2004: pers. comm.). Twelve additional wound-on-drawn beads were identified by Burgess (2002) in the Sullivans Island collection. Nothing was known about the source of these beads, although for the purposes of this study Venice was hypothesized due to their visual similarity to the common, drawn red-on-white beads.

The compositions determined for the red and white glasses differ from the ones defined for the beads attributed to Venice. The red is a lead glass containing almost 49% of this element, instead of about 10% as seen in the red glass attributed to Venice. Potash is about 7.5% (instead of about 16%), soda is 3.7% (instead of about 4.5%), and lime is less than 1.0% (instead of 3%). The glass contains gold (122 ppm) but does not contain a significant quantity of arsenic, contrary to the Venetian red-glass composition.

The white glass is non-lead with a high-potash high-lime composition which is very similar to the Bohemian glass with some slight differences. With a concentration of 3.6% soda, the white core does not fit within the range of concentrations determined for the Bohemian glass, which is 0.2-2.4%. The alumina (1.1%) and potash (10.2%) concentrations are very similar to the concentration of the same elements in Bohemian glass. The high quantity of phosphorus detected (about 5%) suggests the use of bone ash to opacify the glass.

Although the composition of the wound-on-drawn bead does not exactly match any of the compositions identified in this study, its makeup seems closer to Bohemian glassmaking tradition than to the Venetian one.

Beads Attributed to China

The manufacture of glass beads in China is of some antiquity. Elaborate glass beads were present by the late Zhou period (475-256 B.C.) and bead production there continued on and off over the following centuries (Francis 2002). Sprague (1990) received conflicting reports regarding Chinese bead production in general during a visit to the beadmaking center of Boshan.

According to Francis (2002), Chinese beads of non-lead glass are the primary kind encountered in the Americas. He describes some of them as being opaque blue, occurring from Alaska to Washington state, and links their widespread distribution to the ascent of Boshan as the main beadmaking center in China. These are among those described as "Canton beads" in historical records (Ross 1990). Other bead varieties which Francis (2002) describes as being Chinese have distinct colors, air bubbles in the glass, large perforations with clay present on their surface, and irregular outlines. Liu (1995) also lists the same traits as being signifiers of Chinese beads. For this study, the beads hypothesized to be from China were selected based on these descriptions, both the blue Canton-type beads, and the translucent, monochrome wound beads with numerous bubbles in the glass. Examples of both are present in the non-lead category.

The beads selected for analysis are all wound and fall into the large (6-10 mm) to very large (over 10 mm) size category (Pl. XC). Most are spherical (Kidd and Kidd type WIb) and occur in opaque turquoise blue, translucent pink, translucent red, translucent dark blue, and translucent green. A very large, oval, translucent dark blue bead (type WIc) was also included, as were some distinctive translucent red disk-shaped beads with flat ends and generally flat sides (type WId).

The beads hypothesized to be from China form a far more heterogeneous group than the beads attributed to either Venice or Bohemia. It should be noted that this heterogeneity could simply indicate that some of these beads could be from somewhere other than China since the selection process, while based on characteristics thought to be diagnostic, is nonetheless somewhat speculative. If broad compositional groups and subgroups are defined based on lead content and alkali or alkali-earth element concentrations, it appears that the compositions vary a lot within each subgroup, even for visually identical beads. The beads in the sample sort into four different subgroups based on composition: 1) a non-lead glass with a high-soda high-lime composition; 2) a lead glass that has a high-soda high-potash composition; 3) another lead glass with a high potash content (Table 4); and 4) a lead-barium glass represented by a single opaque blue bead.

Subgroup 1: Non-Lead High-Soda High-Lime Glass (Na-Ca)

The high-soda high-lime glass of the beads in this subgroup does not contain significant quantities of lead (< 0.14%). Average concentrations of soda and lime are close to 10% for both and potash concentrations are slightly lower on average (7.6%) and range from 2 to 12.5%. Magnesia is always less than 1.0%.

The high-soda high-lime or Na-Ca beads are opaque turquoise blue, transparent dark blue (both spherical and oval varieties), and transparent green. The turquoise-blue beads contain 0.8-0.9% copper. The opacity of these beads is not due to tin, nor antimony or arsenic since these elements occur in very low concentrations. One of the turquoise-blue beads has about 560 ppm of barium which is in the range of the concentrations exhibited by the glass beads in this group, the other beads have slightly higher concentrations, in the range 2000 to 6000 ppm. This element may have a role in the opacity of these beads. The translucent dark-blue beads contain cobalt. As with the Bohemian beads colored by cobalt, it seems that arsenic, nickel, and bismuth are associated with the coloring ingredient. The green

Table 4. Reduced Average Compositions with Standard Deviations of Three of the Four Chinese Glass Groups.

	Non-Lead High-Soda High-Lime Glass	High-Soda High-Potash Lead Glass	High-Potash Lead Glass
SiO_2	68.8 +/- 3.4%	74.9 +/- 1.0%	80.0 +/- 2.0%
Na_2O	10.9 +/- 2.3%	10.2 +/- 0.6%	1.8 +/- 0.8%
MgO	0.3 +/- 0.2%	0.6 +/- 0.4%	0.1 +/- 0.1%
Al_2O_3	1.6 +/- 0.8%	0.7 +/- 0.05%	1.0 +/- 0.8%
K_2O	7.6 +/- 2.9%	11.5 +/- 0.7%	14.5 +/- 1.1%
CaO	10.4 +/- 2.8%	1.8 +/- 0.4%	2.5 +/- 2.6%

beads are colored by chromium, present in the glass with concentrations in the range of 2000 to 3000 ppm.

Subgroup 2: High-Soda High-Potash Lead Glass (Pb-Na-K)

Five translucent red beads colored using gold (70-130 ppm) represent Subgroup 2. These are the distinctive, wound, disk-shaped beads with flat ends, somewhat resembling a tire. Lead concentrations are in the range of 20%. This glass is referred to as Pb-Na-K. Soda and potash concentrations average about 10%. The lime content is low and its concentration ranges from 1.5 to 2.3%.

Subgroup 3: High-Potash Lead Glass (Pb-K)

The beads of the third subgroup have lead concentrations ranging from 25.6 to 34.2% and potash has the highest concentration (14% on average) after silica and lead. This is referred to in this study as high-potash lead or Pb-K glass. Soda and lime concentrations vary respectively from 0.7 to 2.5% and 0.2 to 2.6%. Trace element concentrations seem to confirm the existence of distinctive compositions related to the use of different sources of ingredients as shown in Fig. 5 for protactinium and tantalum. Discrete groupings appear based on the color of the beads when plotting these two elements.

The Pb-K beads are the wound, spherical, translucent red, translucent pink, and opaque turquoise-blue varieties. The translucent pink beads contain gold (21 to 47 ppm) and slightly more tin than any other beads in this group (686 to 2594 ppm). This composition suggests that this glass is the only gold-containing glass of the beads sampled that may have been colored by the purple of Cassius. Two of the Pb-K beads are translucent red, but unlike the other red beads in this study, they do not contain significant concentrations of gold. These beads do contain uranium (12 and 13 ppm) which is an element that can produce a red color in a lead glass but the quantities of uranium seem quite low. Some elements not measured, like cadmium or selenium, may have been used to color these beads. Four of the Pb-K beads are opaque turquoise blue and contain copper but the opacifying agent remains a mystery. The opacity may be due to an element not measured using LA-ICP-MS, like fluorine, or to some physical properties of the glass rather than to a specific compound.

Subgroup 4: Lead-Barium Glass (Pb-Ba)

The four opaque turquoise-blue beads in the high-potash lead glass subgroup are visually identical to a fifth one that has a totally different composition. These five beads are often referred to as Canton beads (Ross 1990). Instead of being made of high-potash lead glass like the other four, one

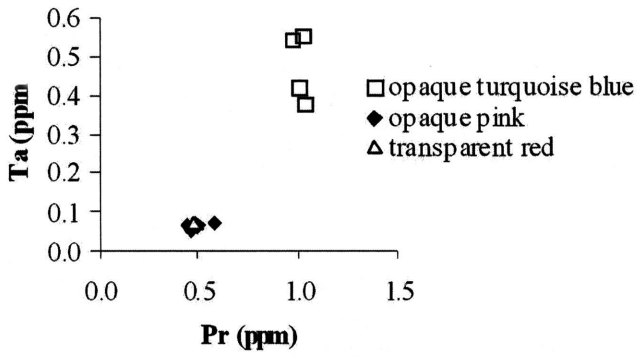

Figure 5. Protactinium and tantalum concentrations in the high-potash lead-glass samples.

of the turquoise-blue beads is made of a lead-barium glass with very little soda and potash and 5.8% of lime. Lead-barium glass is one of the earliest glasses manufactured in China, spanning the period from the 5th to 4th centuries B.C. until the 3rd century A.D. (Jiayao 1996; Brill 1999). The presence of barium in this bead is puzzling. Was barium still used at this late period or is the presence of barium a sign of glass recycling? Testing a larger sample of these visually-identical beads is required to resolve these questions.

The broad heterogeneity in the beads chosen for the supposed Chinese group is interesting. Historical records do in fact suggest some recycling of glass in China, and describe the use of broken European glass in glassmaking in late 18th-century Canton, a practice that had allegedly been abandoned by 1896 in favor of Chinese glassmaking (Francis 1986, 2002). In fact, the high-soda high-lime compositions in the sample sets attributed to China and Venice are quite similar when the major elements are compared (Fig. 6). The similarity could also mean that these beads originated from a European source and had nothing at all to do with China in terms of manufacture or distribution. Magnesia is, however, lower in these beads (< 1.0%) and trace elements exhibit different patterns (Fig. 7). A high-soda high-lime glass was also identified among the Bohemian beads, with very low strontium and cesium concentrations. Strontium and cesium levels are also reported in Fig. 7. For the Bohemian glass, the concentrations for these elements are extremely low. The results presented in Fig. 7 suggest the distinct presence of a third group of high-soda high-lime glass containing higher

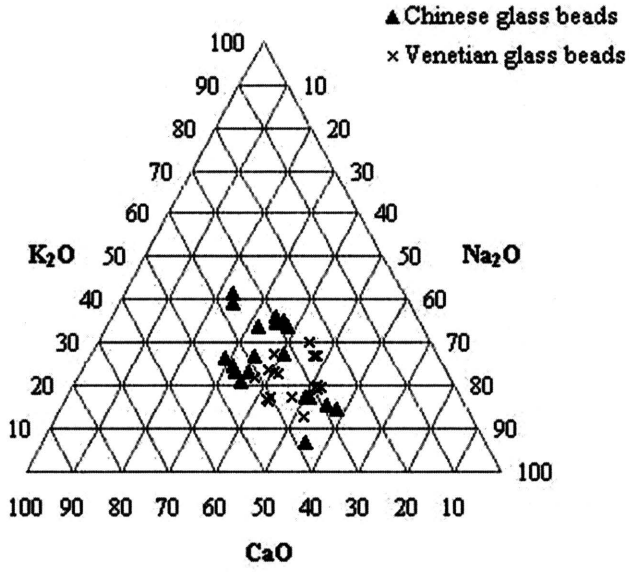

Figure 6. Ternary diagram including potash, lime, and soda concentrations in the high-soda high-lime glasses attributed to China and Venice.

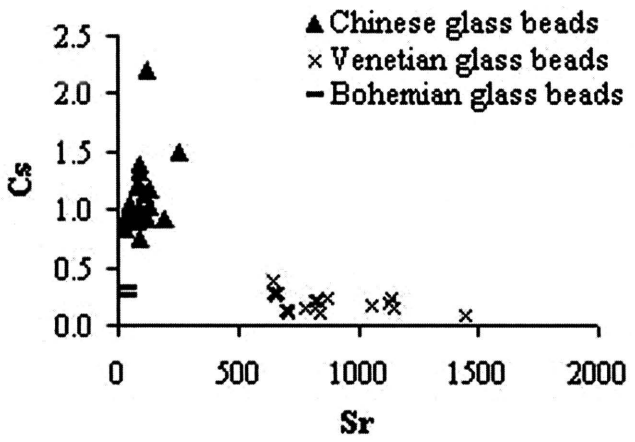

Figure 7. Cesium and strontium concentrations in the high-soda high-lime glasses attributed to China and Venice.

levels of strontium and cesium, that corresponds to some of the beads attributed to China. The number of samples for this third high-soda high-lime glass is very low (n = 2), however, and further analysis would be necessary to validate any conclusions.

DISCUSSION

The 128 beads for this study were selected and segregated into three groups according to their presumed place of manufacture: Bohemia, Venice, and China. Even though the beads were sorted based on descriptions and characteristics presented in the archaeological and bead literature, the division was still somewhat speculative. As expected, more groupings appeared following the LA-ICP-MS study (Table 5). Composition did seem to vary according to the suggested country of origin, especially for Bohemian glass, but was also affected by color and diaphaneity. Drawn beads are the most common 19th-century bead manufacturing type, and are generally followed in frequency by wound beads, and then by mold-pressed beads in collections of this time period. The Sullivans Island collection follows this general convention. The bead compositions generally sort into these manufacturing categories based on their place of origin: the molded beads from Bohemia, the drawn beads from Venice, and the wound beads from China. Part of this breakdown is due to sampling bias, however, since drawn beads were emphasized in the beads attributed to Venice. Wound beads were also made in Venice, especially the elaborate lampworked beads, and the oval, opaque white beads (Kidd and Kidd type WIc) included in this study yielded a composition typical of Venetian glass.

Eight significantly different compositions were identified. One of these, Na-Ca, was divided into three

Table 5. Chemical Composition Findings.

	Manufacture	Color	Size	Kidd & Kidd Type	Glass Type	Coloring/ Decolorizing/ Opacifying Element
Bohemia	Drawn, two rows ground facets	tsl. green	L	If	K-Ca	Cu, Pb
	Drawn, two rows ground facets	tsl. colorless	L	If	K-Ca	Mn, As
	Drawn, two rows ground facets**	tsl. dark blue	L	If	K-Ca	Co
	Drawn, two rows drawn facets	colorless outer layer; op. white core	M-L	IIIf	K-Ca	P
	Mold-pressed, ground facets	op. blue	L	MPIIa	K-Ca	Co, P
	Mold-pressed, ground facets	tsl. white	L	MPIIa	K-Ca	P
	Mold-pressed, ground facets	tsl. red	VL	MPIIa	Pb-Si	Au
	Mold-pressed, molded facets	op. white	L	MPIIa	K-Al	P
	Mold-pressed	tsl. dark yellow	L	MPIIa	K-Ca	Fe
	Mold-pressed, spherical, mold seam	tsp. yellow	L	MPIIa	Na-Ca	U, Cd
	Blown**	op. white w/ tsl. blue stripe	L	Blown in mold	K-Ca	P
	Blown**	tsl. purple blue*	VL	Blown in mold	K-Ca	Co, P
	Wound-on-Drawn*	tsl. red on op. white	L	Wound on drawn	K-Ca (white) Pb-Si (red)	P (white), Au (red)
Venice	Drawn	op. turquoise blue	S	IIa	Na-Ca	Cu
	Drawn	op. turquoise blue	M	IIa	Na-Ca	Cu
	Drawn	op. white	S	IIa	Pb-(Na,K)	As
	Drawn	tsp. colorless	S	IIa	Pb-(Na,K)	As
	Drawn	tsl. red	S	IIa	Pb-(Na,K)	Au
	Drawn	tsl. green	S	IIa	Pb-(Na,K)	Cu, Pb
	Drawn	op. white on op. white	M	IVa	Na-Ca	Sb
	Drawn	tsl. red on op. white	M?	IVa	Pb-(Na,K)	Au (red) As (white)
	Drawn, faceted	tsl. reddish purple	M	IIf	Na-Ca	Mn
	Wound	op. white	S/M	WIc	Pb-K	As
China	Wound	op. turquoise blue	VL	WIb	Pb-K Pb-Ba	Cu
	Wound	tsl. pink	L	WIb	Pb-K	Au

Table 5. Continued

	Manufacture	Color	Size	Kidd & Kidd Type	Glass Type	Coloring/ Decolorizing/ Opacifying Element
China	Wound	tsl. red	VL	WIb	Pb-K	Unknown
	Wound***	tsl. red	L	WId	Pb-Na-K	Au
	Wound	op. turquoise blue	L	WIb	Na-Ca	Cu
	Wound	tsl. dark blue	VL	WIc	Na-Ca	Co
	Wound	tsl. dark blue	VL	WIb	Na-Ca	Co
	Wound	tsl. green	VL	WIb	Na-Ca	Cr

* Hypothesized Bohemia, but composition suggests a Venetian origin

** Hypothesized Venice, but composition suggests a Bohemian origin

*** No source hypothesized, composition suggests a Chinese origin

subgroups with features that differed according to the production location. A correlation was observed between composition and provenance but, interestingly, no consistent correlation was found between the technique used to produce the beads and the composition of the glass, at least for glass from Bohemia. For example, the same K-Ca composition was identified for drawn, mold-pressed, and mold-blown Bohemian beads. On the other hand, five visually identical Chinese turquoise-blue beads belong to two different glass groups—one exhibits a lead-barium composition; the other a Pb-K composition. Thus, beads that look alike can have different compositions, at least for those made in Asia.

Several glass types were used at each of the three proposed manufacturing locales. They may have been produced by different glass manufacturing workshops or at slightly different periods. The presence of uranium in the Bohemian Na-Ca glass indicates that this glass was produced no earlier than the second half of the 19th century while the Bohemian K-Ca glass is part of an older tradition.

The Bohemian beads form the most homogeneous and consistent group in the study. Most of them are high-potash high-lime glass and correspond to a recipe involving forest-plant ash and lime sand. This supports Glascock and Speakman's (2002) LA-ICP-MS findings which, in addition to establishing that beads of similar color cluster together, found Bohemian beads to be much more homogenous in composition than Venetian beads. A Bohemian source for the varieties identified in the Sullivans Island collection is further supported by Neuwirth (1994). According to her research, all of these bead varieties—the mold-pressed, the drawn faceted, and the blown-in-the-mold beads—were manufactured in Bohemia.

Two glass types are identified for the Venetian beads: a high-soda high-lime glass and a lead glass with very variable concentrations of soda and potash. The glass type involving lead has a more heterogeneous composition, a finding that is consistent with Glascock and Speakman (2002). Venetian glass beads are the most commonly encountered beads on 19th-century North American archaeological sites, especially the small, drawn embroidery beads sampled here. The drawn faceted beads (Kidd and Kidd type IIf) were found to correspond to Venetian glass, although it had originally been hypothesized that they were made in Bohemia due to the presence of ground facets. Interestingly, Neuwirth (1994) states that beads made in Venice were sent to Bohemia for grinding as does Bussolin in 1847:

> Our beads are cut quite easily in Bohemia, and at very reasonable prices.... It should be noted that cut colored-crystal beads are also produced in Bohemia. The type of process, however, is very different and the product should not be confused with the beads produced in the Venetian factories (Karklins with Adams 1990:73; Ross 2000).

Some of the glass compositions encountered are related to achieving a specific color, especially for red glass. Red glass colored with gold was obtained using specific recipes

that involved the use of lead glass since the incorporation of this element seems essential to obtaining an intense ruby-red color. Four different recipes for red glass containing gold and lead were identified. Even though the composition of red glass varied according to the sample group, the red glass always contained lead; levels of more than 50% were detected in the Bohemian glass, while only 6% was present in the Venetian glass. The lead concentration in glass that also contained gold ranged from 18.6 to 32.8% in beads attributed to China. Concentrations of soda, potash, and lime varied too. The red glass in Venetian beads contained up to 2% arsenic, while only 24 and 27 ppm of arsenic were measured in the glass from Bohemia. Variable concentrations of antimony were also detected in the red glass according to the place of origin.

Lead in copper-containing glass has the property to impart a green color. The presence of high alumina concentration in the Bohemian Al-K glass resulted in an opaque glass.

It is interesting to note that different colorant traditions existed in Bohemia, Venice, and China. In Bohemia, white beads were mostly produced using a phosphorus-rich ingredient like bone ash. In Venice and China, however, phosphorus did not appear in especially high concentrations in white beads. Rather, arsenic, combined with lead, was used in China and Venice, and antimony was detected in some white Venetian beads. Chromium was used to produce green glass in China while copper and lead were preferred in Venice and Bohemia. The same cobalt, associated with nickel, arsenic, and sometimes bismuth, was used in dark-blue glass regardless of presumed source. Even in early periods cobalt pigment was traded over very long distances.

The composition of the red and white glasses from the rare wound-on-drawn bead was most surprising when compared to that of the glasses from the red-on-white drawn beads. While there were some differences, both glasses in the wound-on-drawn bead were close to the red and white Bohemian glass, but very different from the more common red-on-white beads which were similar to Venetian glass. Thus it appears that wound-on-drawn beads may have originated in Bohemia.

CONCLUSION

Compositional analysis studies of 19th-century glass beads are few and the beads from Sullivans Island provided the opportunity to investigate glass thought to be from the three major bead producing centers of this time period. A wide range of glass recipes were identified for the 128 beads sampled for this project. Some are specific to a given area (e.g., the high-potassium high-calcium glass of Bohemia) but others seem to have been used at different places, like the high-sodium high-calcium compositions identified for beads attributed to Bohemia, Venice, and perhaps China. Different trace element patterns within this glass suggest the use of different types of ingredients but further investigation is needed to define the significance of this recipe at each of the three production centers. Other possibilities also exist. It may be that beadmakers in different countries were copying each other or that some of the attributions regarding place of origin are incorrect. It is, of course, not possible to assert that different compositions automatically indicate different areas of manufacture, but this study was designed to take a broad look at a range of bead varieties produced in potentially different areas that will hopefully encourage additional studies, since comparative material for this time period and this technique is still scarce. While LA-ICP-MS analysis of the glass beads from Sullivans Island has answered certain questions, it has raised others. One of the strengths of this technique lies in its ability to identify trace elements, which may turn out to be subtle yet significant markers for identifying the places of origin of beads in future studies.

ACKNOWLEDGEMENTS

The authors would like to thank Pamela Vandiver, sponsor of Laure Dussubieux's Smithsonian fellowship; Smithsonian photographer Donald Hurlbert for bead photography; Roderick Sprague for ongoing encouragement and sage advice on bead collections from the Plateau; and William T. Billeck for comments on earlier drafts of this article.

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