Ceramics Final Report:
A Comparison of Ceramic Types from Montserrat

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Kristie Chin
Katherine Harrington
Peter Johnson

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I. Introduction and Archaeological Background

Montserrat is a small island in the Lesser Antilles, approximately 30 miles from Antigua (Fig. 1). Like many islands in this area, Montserrat is volcanic, and it gained notoriety in the late 20^{th} Century when the dormant Soufriere Hills volcano became suddenly active on July 18, 1995. Many residents have been forced to evacuate and roughly half of the island is now uninhabitable (Ryzewski et al. 2010: 3). The volcano remains active to this day, which lends archaeological investigation on Montserrat a certain urgency; Trants, one of the earliest known Saladoid sites in the Caribbean and thus a key to understanding the spread of the Saladoid people through the Antilles, was destroyed by pyroclastic flow in February 2010 (Ryzewski et al. 2010: 6). This paper will present the results of an investigation of a Post-Saladoid ceramic assemblage from a recently discovered site at Valentine Ghaut. Using a combination of traditional and archaeometric techniques, we have examined samples of four different fabrics. This study has allowed us to draw some conclusions regarding production method and technical properties, within the social context of the Post-Saladoid period.

These sherds (along with lithic, shell, and bone materials) were collected at Valentine Ghaut, on the northwestern coast of Montserrat, during the January 2010 field season of the Survey and Landscape Archaeology of Montserrat Project (SLAM), led by Krysta Ryzewski and John Cherry of Brown University. A total of 76 sherds were collected from the surface of Feature 11, a midden pit which appears to be approximately 80 cm deep (Fig. 2) (Ryzewski et al. 2010: 26). Based on morphological and decorative comparisons with ceramics from nearby islands, the SLAM team tentatively dated the assemblage to the early Post-Saladoid period (See especially rim profiles in the "Mill

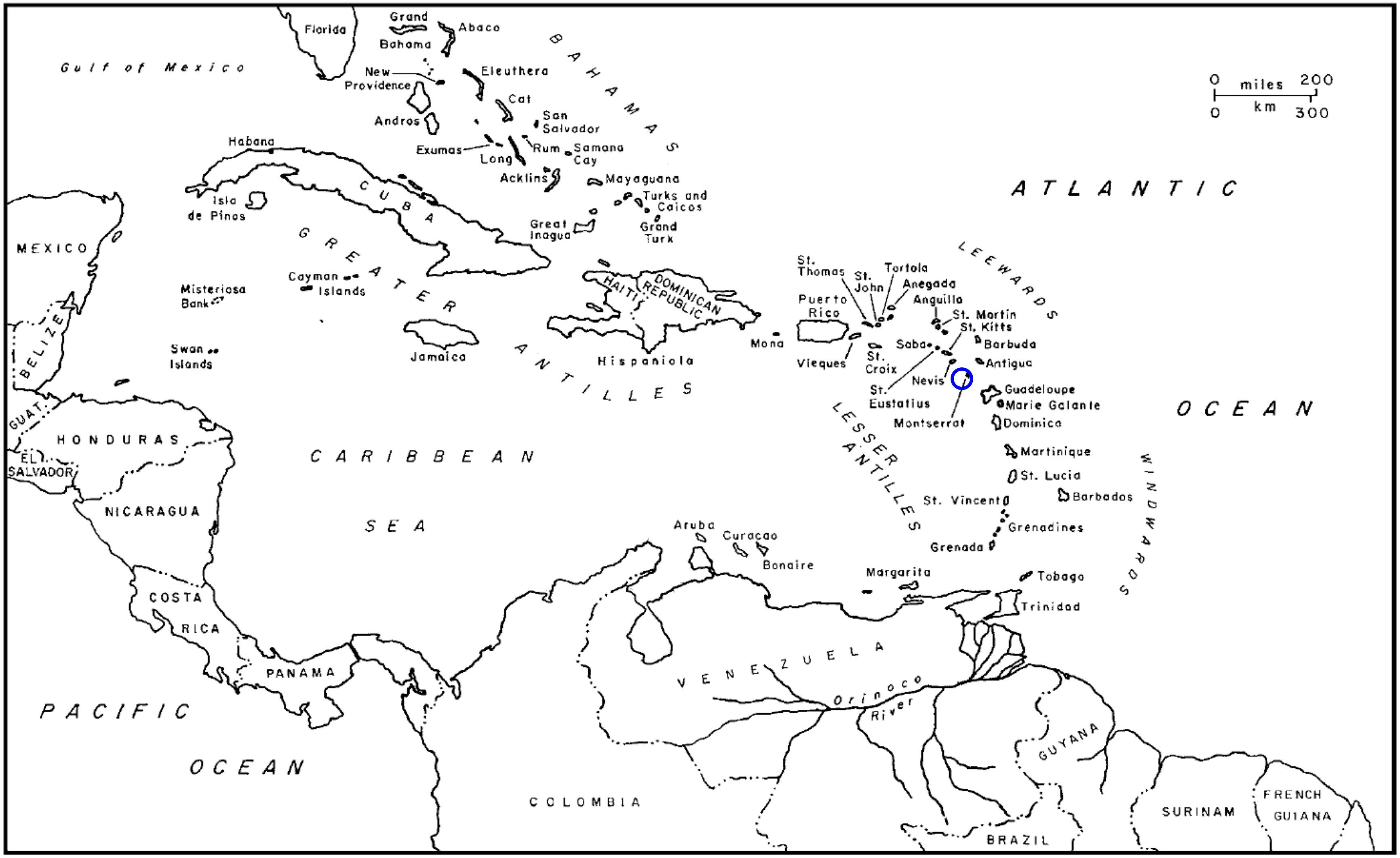


Figure 1: Map of the Caribbean. Montserrat indicated. (Keegan 2000: 137)

Reef" and "Mamora Bay" periods on Antigua, Rouse and Morse 1999: 31-43). Subsequently, two fish bones collect at Feature 11 underwent radiocarbon dating, which confirmed a Post-Saladoid date. One bone dated to cal. AD 680-780 and the other to cal. AD 1020-1060 or 1080-1150 (Tamers and Hood 2010: 3-4). The further investigation of the site, as well as the tests conducted in this project, are of particular importance since Valentine Ghaut is currently one of only two accessible prehistoric sites on the island of Montserrat. All other known prehistoric sites have been destroyed by volcanic activity or development (Ryzewski et al. 2010: 27, Krysta Ryzewski personal communication).

Figure 2: Feature 11, a midden deposit (Ryzewski et al. 2010: 26).

The term Saladoid refers to a group of people thought to have migrated into the Lesser Antilles from northern South America in the last centuries B.C.E. The precise mechanism of this migration is not well understood at this time, but it seems likely that migration was gradual and intermittent (Haag: 242; Wilson 2007: 59-60, 70). Though most previous archaeological work has focused primarily on middens, the few Saladoid towns excavated indicate that Saladoid people lived in small villages comprised of circular huts, generally facing onto a central plaza, thought to be a communal space which also often served as a cemetery (Wilson 2007: 88-89; Keegan 2000: 141, 144). They cultivated manioc and other plants, and hunted and fished (Wilson 2007: 86-87; Fitzpatrick and Keegan 2007: 34). Trade and communication networks among the islands were well developed, and there is strong evidence of continued contact with the South American mainland (Hofman et al. 2007: 247, 249, 262; Keegan 2000, 144).

Typically, archaeologists recognize the presence of Saladoid people by one of two distinctive ceramic decorative styles: white paint on red slip (WOR) or zone-incised-crosshatched (ZIC). Burnished and slipped pottery is also common, as are undecorated vessels (Petersen and Watters 1995: 135; Wilson 2007: 67). In contrast, Post-Saladoid ceramic assemblages typically contain far fewer examples of elaborately decorated vessels (Hofman and Hoogland 2004: 50). The assemblage from Valentine Ghaut consists of griddle fragments (typically assumed to function to bake cassava bread), undecorated plain-ware sherds, and fragments of slipped and burnished bowls or other open shapes.

Although the Post-Saladoid Period has not received as much archaeological attention as the Saladoid Period and the period of initial European contact, the available evidence suggests that Caribbean societies experienced a variety of social changes during this time (Hofman et al. 2007: 254; Hofman 1995: 236; Keegan 2000: 151). This is particularly evident in the Greater Antilles, where there was a radical shift from a presumably egalitarian society to a socially stratified society (Keegan 2000: 145; Hofman and Hoogland 2004: 47, 49). Evidence of such changes is not as clear in the Lesser Antilles, but similar social transformations may have been occurring (Hofman 1995: 237-

239; Hofman and Hoogland 2004: 47). Trade networks were circumscribed, and local pottery styles dominate (Hofman et al. 2007: 252-253, 262). The amount of decorated wares decreases, as does symbolic representations (Hofman and Hoogland 2004: 50). Additionally, evidence from some sites suggests a change in diet to a greater reliance on terrestrial resources, with an accompanying decrease in marine animal size, possibly due to over exploitation (Fitzpatrick and Keegan 2007: 35). Climatic evidence points to a series of dramatic shifts between hyper arid and wetter conditions, which may be related to some cultural changes (Keegan 2000: 145, 146). There is also a change in the nature of burials, from communal cemeteries in the plaza of circular villages to burial within homes. Hofman and Hoogland suggest that this may indicate the development of ancestor worship (Hofman and Hoogland 2004: 51, 53).

Through the 1980s, Caribbean archaeology remained in a cultural-historical framework, largely concerned with clarifying stylistic pottery chronologies. Since the 1990s, however, archaeological research in the area has diversified and expanded (Hofman et al. 2008 a). Recent studies in Caribbean archaeology have utilized a wide range of traditional and scientific methods, and have added nuance to our understanding of prehistoric life.

II. Sample Selection:

The assemblage was visually inspected and divided into four subgroups based on fabric appearance, surface decoration, and, when possible, function. First, identifiable griddle fragments were separated from the general assemblage. Griddles are recognizable by their incised, flat surfaces. Second, a single sherd thinner and lighter in color than the other sherds was separated. The fabric of this sherd appeared to have fewer inclusions than the others. Since this sample was unique, we felt that it was necessary to examine it more closely in order to determine how it differed from the other sherds. The two remaining groups constituted the majority of the sherds: coarse wares with many sandy inclusions, and burnished and/or red-slipped wares. One sample was selected from each of these categories. Sample 1 is a small fragment of a griddle with a series of incisions on one surface. Sample 2 is an undecorated sherd with a comparatively thin wall. Sample 3 is a small, undecorated rim sherd with a very coarse, sandy fabric containing more inclusions than of Sample 2. Finally, Sample 4 is burnished and partially slipped. Pictures of the samples and the assemblage can be found in the Appendix. From this point forth, the samples will be referred to as the griddle fragment, pale piece, sandy sherd, and slipped/burnished sample.

Though only four samples were selected--due primarily to time and financial constraints--they were carefully chosen as representative samples to provide sufficient information about each of their respective categories, and to allow us to adequately compare and contrast the samples with one another. A series of archaeometric techniques, including optical microscopy, X-ray fluorescence (XRF), thin-section petrography, and scanning electron microscopy with energy dispersive spectroscopy (SEM-EDS), were carried out on each of the samples. Knowledge of the physical properties of each of these four samples will add to the understanding of their production, the role they played within their society, and ultimately the cultures surrounding the ceramics.

III: Research Questions:

Our objective was to compare different samples with one another in order to observe and, when possible, to quantify the similarities and differences of four ceramic types. Through this analysis, we hoped to determine the correlation between the nature of the fabric and surface treatment and functionality. By carrying out technical analyses, we determined the chemical composition of the samples, the nature of the inclusions, and the temperatures required to fire the materials. This scientific information can help us answer questions about certain production processes and, potentially, vessel use. Though our small sample size limits our ability to draw definitive conclusions, we have attempted to situate our findings in the social context of the Late Saladoid Period.

IV: Experimental Procedure:

Taking an approach similar to Hofman et al. 2008, we combined scientific techniques with more traditional investigations. First, we conducted XRF analysis, taking multiples points on each sample, to measure the elemental composition of the sherds in a semi-quantitative manner. Subsamples were then cut from each sherd, mounted for SEM-EDS and prepared for thin-section petrography. Preliminary optical microscopy was used during the sample preparation process to gain a basic visual understanding of each of the fabrics at high resolution. SEM-EDS was used to study the microstructure and porosity of the samples, and look for further evidence of production, particularly organic and shell inclusions that could be used to provide evidence for firing conditions or temper materials. Thin-section petrography was also used to examine and identify inclusions in addition to characterizing their size and shape. This combination of scientific techniques will provide data used to analyze the fabric, surface treatment, inclusions, physical properties, and achieve a deeper understanding of the production techniques and cultures involved with the ceramics.

A. X-Ray Fluorescence

Apparatus:

An Innov-X Alpha 4000 Handheld XRF, as seen in Figure 3, was used to carry out non-destructive elemental analysis. It is an energy dispersive model that features proven SiPin Diode Detector technology. It combines X-ray tube expertise, multiple beam filtering capability, and an HP iPAQ pocket PC to detect up to 27 elements with reliable precision (Innov-X Systems, Inc., 2007).

Figure 3: Innov-X Alpha 4000 Handheld XRF

The XRF shoots x-rays at the sample, depicted as the green hv in Figure 4, and the high energy x-rays are able to ionize elements; essentially, they eject an electron from the inner valence shell of a given atom. When this happens, the atom becomes unstable, forcing another electron to drop down from an outer valence shell. As a result, energy is emitted, illustrated by the red hv, in the form of either ‘K’ or ‘L’ transitions.

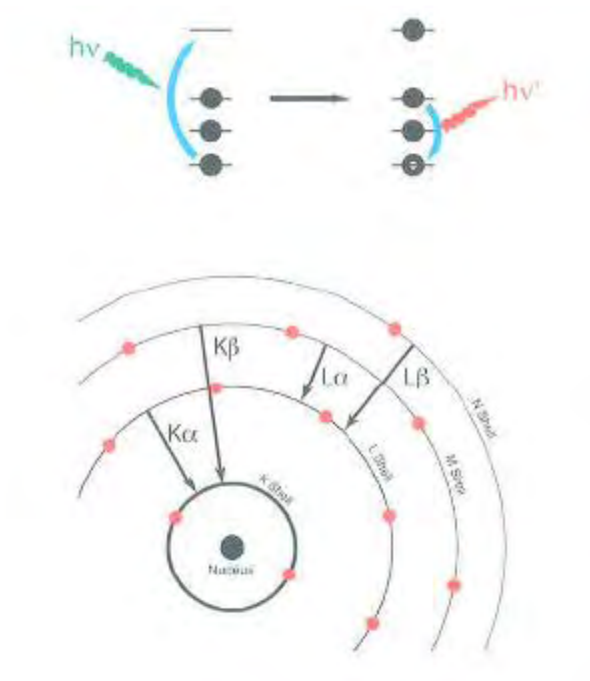


Figure 4: XRF Emission Lines

The emission lines are characteristic for each element. When the detector in the XRF encounters a certain emission line, it counts the number of emissions per second at that energy level and converts it into a concentration of the corresponding element. Sediment standards with known values of heavy metals in them are used to ensure that the XRF is calibrated correctly.

Procedure:

The XRF was configured to analyze ice cores and was thus attached to a track. The track was first covered with foam padding, and then the sample was placed on top, inside of a plastic bag to ensure the protection of the lens. The XRF was adjusted mechanically to lower the lens as close as possible to the surface without touching the sample. Once in place, the XRF was triggered and two 90-second iterations were made. The first pass detected lighter elements below atomic number 18 (P, S, Cl), while the second detected heavier elements (K, Ca, Ti, Cr, Mn, Fe, Co, Ni, Cu, Zn, As, Se, Rb, Sr, Zr, Mo, Ag, Cd, Sn, Sb, I, Ba, Hg, and Pb). Major elements, such as Al and Si, were not detected by the XRF because of air absorption; the secondary radiation from lighter elements is of relatively low energy and has low penetrating power, causing its long wavelength to be severely attenuated when the beam passes through air for any distance. Three or four points on the surfaces of each of the samples were irradiated by the XRF, covering an area of about 1 mm^2 for each selected point. The particular points chosen for each of the sample measurements can be found in the Appendix.

B: SEM-EDS

<u>Apparatus:</u>

We used a LEO 1530 VP SEM, a high-resolution SEM equipped with a field emission source. Resolution of 1.5 nm is obtainable and the machine has imaging capabilities up to 50,000x magnification. This instrument has excellent capability for low-voltage imaging, which is particularly important for examining insulating materials, like ceramics. The SEM is coupled to an EDS.

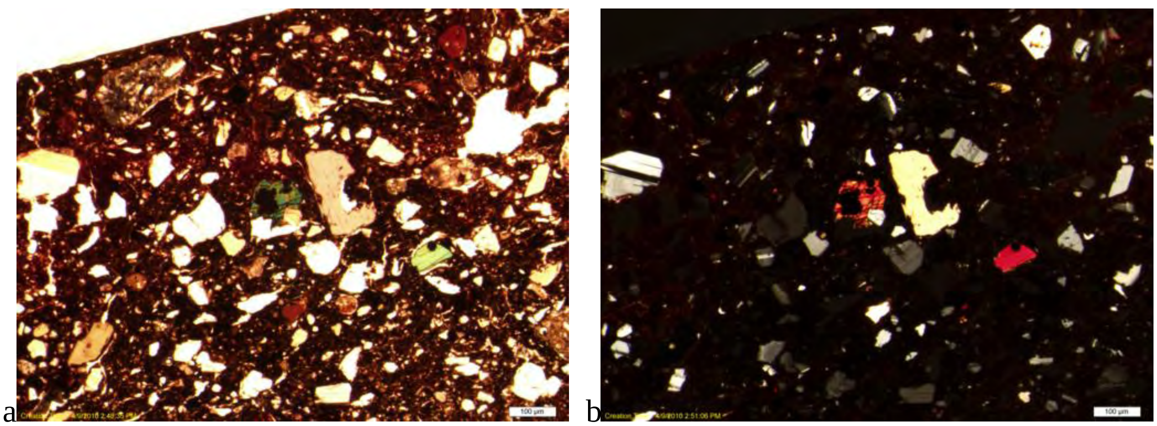
<u>Procedure:</u>

The selected ceramic fragments were cut to size using a diamond-blade circular saw, cleaned using distilled water, and dried. They were then mounted within 1.25" diameter resin blocks and allowed to set. The samples were each sanded down to expose the surface using 200, 400, and 600 grains, polished using the 30 and 6 micron grit. Finally, the samples were depressurized and carbon coated. Before entering the SEM, the sample blocks were taped with copper tape, making contact with each end of the ceramic material. Then, the samples were mounted within the SEM chamber. Before looking at the samples, the chamber had to depressurize, taking up to an hour in some cases because of the porosity of the ceramics. Afterwards, we were able to take pictures of the ceramics and then add the EDS, which allowed us to take analytical compositional measurements of data points, area regions, and line transects.

C: Thin-Section Petrography

Apparatus:

Thin-sections of the four samples were examined using a polarizing petrographic microscope. A petrographic microscope allows light to pass through the sample from beneath. The sample can be examined with transmitted light or polarized light. The sample rests on a rotatable stage. Examining the sample in plane-polarized or cross-polarized light while rotating the stage allows the identification of minerals by their optical properties (Peterson 2003: 10).



a b

Figure 5: Thin-section of slipped/burnished sample under a) transmitted light and b) cross-polarized light, demonstrating how optical properties can be used to identify minerals. In this case, the green mineral in the center of the image can be identified as amphibole based on its color and fractures (Stephen Parman, personal communication).

Procedure:

Small sawn fragments of each sample left over from SEM-EDS sample preparation were chosen. Each fragment was impregnated with a resin over heat. The sawn surface was ground by hand, and then attached to a slide. The top of the fragment was cut down to 1-2 mm, and then ground mechanically and by hand. When the samples reached a thickness of approximately 30 microns, they were polished. After polishing, the samples were examined with a petrographic microscope.

V. Results and Discussion

XRF:

The data found through XRF will be closely tied to the findings of SEM-EDS and thin-section petrography. Once the analyses are integrated with one another, these results will be related back to the social and cultural contexts of the ceramics. XRF analysis investigates the elemental signature of pottery, which is mostly influenced by the composition of raw clay materials, but heavily depends on the influence of larger clastic inclusions in the bulk reading. Therefore, the elemental characterization of ancient ceramics may give information about source materials, manufacturing recipes, and fabric types (Tite, 2008). It should be noted that the interpretation of the results is not a straightforward task, since the abundance ratios of some elements can be altered as a result of adding or subtracting any number of materials intentionally or inadvertently. Further, XRF is only a semi-quantitative and surface technique. The data combined with related archeological information are expected to help draw conclusions about the ceramic technology, potential sources of the clays, and other materials used for their production.

According to Padilla et al. 2006, the group of elements that concentrate in the clay fraction and whose concentrations vary largely among different geological formations includes barium, the rare earth elements (REE), yttrium, scandium, manganese, iron, chromium, hafnium, zirconium, and thorium. The elements commonly associated with the temper fraction in the ceramic fabric are mainly sodium, potassium, rubidium, cesium, calcium, strontium, and titanium. Furthermore, the alkali and alkali-earth elements are associated with feldspars, which are the most abundant minerals in the earth's crust. Drawing upon Padilla et al. 2006, Terenzi et al. 2010, and several other ceramic XRF analyses, the elements chosen to focus on are potassium, calcium, titanium, manganese, iron, sulfur, copper, zinc, rubidium, strontium, zirconium, and barium.

Prudence Rice, among other sources, was consulted to determine the potential causes and effects of each these elements. Inclusions can be present as natural impurities in clays or as the result of potters deliberately adding temper in order to achieve better properties in forming and consistency of the material (Padilla et al., 2006). These oxides usually have larger ionic radii than silica and weaken the bonds in the network; they also have the important role of lowering the very high melting point of silica, which is normally 1710^{o}C (Rice 1987, 99).

Potassium has been considered a member of the potassium mica clay group or as part of the nonplastic inclusions. The addition of quartz sand temper can contribute to potassium-bearing minerals such as muscovite, biotite and, of course, potassium feldspar (Munita et al., 2000). Potassium fluxes include potash feldspars, such as Custer and G-200, Cornwall stone, volcanic ash, and pearl carbonate. Potassium fluxes have greater durability than soda fluxes and may be used as a color modifier.

Calcium most commonly occurs in various forms of calcium carbonate ($CaCO_3$), such as limestone, calcite, and shell, or as calcium sulfate, such as gypsum ($CaSO_4$). Lime or calcium may occur naturally in clays, and then the clay is described as calcareous or marly. Calcium may sometimes be added to clays, for example, in the form of animal bone ash or calcium triphosphate ($Ca_3[PO_4]_2$). Calcium carbonate has a very distinctive property that affects its usefulness in fired clay bodies: calcite decomposes on firing at about 870^{o}C (Rice 1987, 97).

Titanium, present as either TiO_2 or as titanate of iron, may contribute a light tan or cream color in iron-free clays (Rice 1987, 406). **Manganese** may be present as reddish-brown or blackish-brown flecks or nodules in clays, particularly those from swampy areas. It is rarely found in sufficient quantities (ca. 10% or more) as a natural constituent of clays to cause an overall black color in the paste, but nodules or concentrations may be ground and mixed with a binder for use as a paint (Rice 1987, 406). Iron produces reddish hues when fired in an oxidized environment and is closely linked with feldspar, the most common compound in the earth's crust (Rice 1987, 406).

Sulfides (pyrites, marcasite) and **sulfates** (gypsum) may also influence the color of fired clays. As soluble salts, these materials migrate through the capillaries of the wares as they dry and concentrate on the surfaces, forming a brownish or whitish scum or efflorescence that looks like a white slip after firing (Rice 1987, 406). **Copper** oxide can be used as a flux former, and hence, as an appropriate sintering aid (Derling et al., 2001). **Zinc** oxide acts as a flux at high temperatures. However, it will vaporize in a reduction atmosphere, resulting in highly toxic fumes. **Rubidium** feldspar can be used in ceramics to greatly increase their insulating capacity (Applications of Rubidium.). **Strontium** carbonate can be used very much like calcium carbonate, increasing a ceramic's resistance to crazing and scratches. It should be noted also that calcium and strontium occur together in limestone (Rice 1987, 420). **Zirconium** oxide is an intermediate used to increase the viscosity of the ceramic and strengthen the fabric (Rice 1987, 99).

Barium carbonate is used as a flux at high temperatures (Rice 1987, 99).

The results of the semi-quantitative XRF analysis on ceramics are presented in Table 1, where the elemental composition of each sample has been averaged over the measurements taken on different surface points. The unslipped and slipped/burnished points were averaged separately, and their percent difference is displayed below. The formula used to calculate the percent difference is as follows:

$$\% \text{ Diff} = \left|\frac{x_1 - x_2}{(x_1 + x_2)/2}\right| \times 100 \tag{1}$$

Outliers for the griddle fragment in potassium and zinc were discarded to prevent skewing of the data. The reported errors are the standard deviations of the three XRF acquisitions for each fragment and the symbol "BDL" refers to element contents below the spectrometer detection levels. The minority species are expressed as atoms, and not as oxides, to account for the fact that elements in archaeological ceramics can be found in several chemical forms, depending on both raw clay type and firing conditions. Minor elements are expressed in weight percentages while trace elements are expressed in parts per million to make the data more readable; it enables a better comparison of elements of similar magnitude, comparable to the usage of scientific notation. The conversion from one to the other is given below:

Concentration (wt%) = Concentration (ppm) / 10,000

Table 1: XRF Results for Significant Elements

Minor Elements (wt%)																		
	K			Ca			Ti			Mn			Fe			S		
Griddle	0.67	±	0.10	2.07	±	0.41	0.43	±	0.06	0.07	±	0.04	4.66	±	0.84	BDL	±	BDL
Sandy	0.66	±	0.02	1.85	±	0.13	0.35	±	0.03	0.07	±	0.02	4.73	±	0.06	BDL	±	BDL
Pale	0.62	±	0.06	1.53	±	0.14	0.41	±	0.02	0.03	±	0.00	4.25	±	0.45	BDL	±	BDL

Unslipped	0.62 ± 0.03	2.53 ± 0.05	0.33 ± 0.01	0.21 ± 0.00	4.10 ± 0.06	0.97 ± 0.16
Slipped/Burnished	0.77 ± 0.03	1.41 ± 0.03	0.40 ± 0.01	0.19 ± 0.00	6.06 ± 0.09	BDL ± 0.39
% Difference	22.01 ± 8.63	56.91 ± 41.39	19.12 ± 11.81	10.18 ± 6.54	38.66 ± 37.70	200.00 ± 85.18
Trace Elements (ppm)						
	Cu	Zn	Rb	Sr	Zr	Ba
Griddle	27.33 ± 4.93	67.00 ± 7.07	20.33 ± 8.50	245.67 ± 26.31	84.00 ± 6.24	324.00 ± 140.87
Sandy	7.67 ± 13.28	40.67 ± 4.51	13.33 ± 2.08	192.00 ± 9.17	87.00 ± 9.54	261.00 ± 87.74
Pale	7.25 ± 14.50	44.50 ± 2.52	17.25 ± 2.22	245.00 ± 6.78	84.00 ± 6.00	205.75 ± 139.43
Unslipped	18.00 ± 14.00	46.00 ± 4.50	16.00 ± 1.00	256.50 ± 4.50	78.00 ± 3.00	249.00 ± 44.50
Slipped/Burnished	32.50 ± 7.50	50.50 ± 5.00	14.00 ± 1.00	207.50 ± 4.00	85.00 ± 3.00	309.00 ± 50.50
% Difference	57.43 ± 60.47	9.33 ± 10.53	13.33 ± 0.00	21.12 ± 11.76	8.59 ± 0.00	21.51 ± 12.63

Examining Table 1, one can see that sulfur was only found in the unslipped region of the slipped/burnished piece. Using EDS, the sulfur was found as part of FeS_2, more commonly known as pyrite. Impurities, such as carbon black, organic matter, sulfides, etc., commonly present in natural clays and other raw materials are responsible for many defects in ceramic products by acting as reducing agents for iron (III) ions during firing. Small amounts (generally lower than about 1 wt%) of pyrite (the main sulfide present in ceramic raw materials) can cause "black core," affecting the rate of oxidation of the core after it has been formed, while, when carbon was present in the clay sulfur was maintained in the sulfide state and iron was stabilized in low oxidation states (Corradi, 1996). The slipped/burnished sample does indeed have a black core, however this effect may also occur due to organic inclusions not fully burning out (Rice 1987, 334). The interpretation of the analytical results leading to establishing different compositional groups and understanding the presence of some temper inclusions should be aided by the study of the geology of the territories where the communities were settled (Padilla et al., 2006). Several sources of sulfur on the island are available to explain its presence (Atkinson, 2000). Soufrière Hills, French for "Sulfur" Hills, is in the southern region. This toponame indicates that this geographical feature is strongly associated and known for its mineral deposits. Figure 6 shows locations of known sulfur, though it is likely that sulfur may be dispersed throughout the entire region.

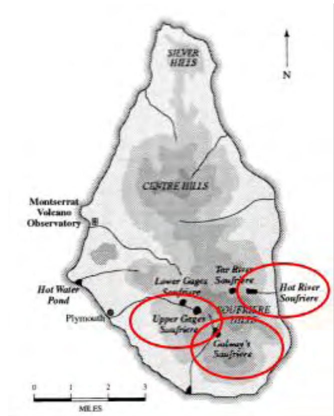


Figure 6: Known Regions of Sulfur Deposits (Atkinson, 2000)

A local source of sulfur is also known to exist within a short distance of the site. The outcrop can be seen in Figure 7.

Figure 7: Local Sulfur Outcrop (Photo courtesy of Krysta Ryzewski)

The Zinc content in the griddle fragment was significantly higher than in the other pieces. Zinc oxide acts as a flux at high temperatures above 750°C, which corresponds with our knowledge of the griddle fragment being exposed to high temperatures as a cooking ware and visual confirmation of lime spalling due to heat on another griddle fragment in the assemblage. The presence of calcium may occur for a variety of reasons.

It may occur as natural inclusions in the clay or be added as temper. If calcium were present at high ratios, considered to be 20-30%, it is more often associated as added temper in the form of shell (Rice 1987, 410). However, the calcium content for all fragments was far below this range, therefore making its presence due to natural occurrence far more likely. Two possible sources are from lime and/or shell. On the whole, the main type of rock on Montserrat is dark-spotted, yellow, fine-grained limestone with fragments of calcareous algae; therefore, it is possible that it occurs naturally in the clay as eroded fragments of limestone. The site is also located near a beach, making shell an equally likely prospect. A small shell-like inclusion was found during SEM in the slipped/burnished fragment, and can be seen in Figure #. This confirms shell as one source of calcium; however, it does not eliminate the possibility of limestone as a source of calcium. Shell has a coefficient of thermal expansion very similar to clays, and because shell fractures into broad plates with large surface areas per unit volume, these properties provide a barrier to crack propagation; in other words, the use of shell increases the durability and strength of a ceramic (Rice 1987, 410).

The high presence of iron in the slipped portion of the slipped/burnished sample indicates that an iron oxide slip was most likely used. Slip glazes are high in iron and clay, which gives them their name (Rice 1987, 99). The high presence of iron in all fragments may be as a result of the soils of Montserrat. The black sand beaches of many volcanic islands, including Montserrat, consist of grains of olivine and magnetite, derived from the erosion of volcanic rocks (Papers 19). Olivine is a magnesium iron silicate, $(MgFe)_2SiO_4$, and has manganese as one of the most common additional elements. Magnetite is a ferromagnetic mineral, Fe_3O_4, which is composed of one part wustite and one part hematite. The red color of the ceramics may be linked to the presence of hematite, which forms by oxidizing firings at temperatures above 750^{o}C (Terenzi et al., 2006).

The overall known weight percentages were also found for the four fragments and are tabulated in Table 2. The known matrix (wt%) is the sum of all measured weight fractions for each of the samples.

Table 2: Known Weight Percent of Samples from XRF Analysis

	Known Matrix (wt%)		
Griddle	8.39	±	1.58
Sandy Fabric	7.82	±	0.20
Pale Fabric	7.00	±	0.74
Slipped/Burnished	9.06	±	0.38
Average	**8.07**	±	**0.87**

The two most abundant elements in clays are aluminum and silicon, neither of which is detected by the XRF. To find the weight percent not detected by the XRF, the known weight percent can be subtracted from 100:

$$\text{Unknown wt\%} = 100 - \text{Known wt\%} \tag{3}$$

This number closely reflects the percentage of hydrated aluminosilicates of varying composition and free silica. These are formed through the weathering process of complex aluminosilicates, which causes them to become hydrolyzed, forcing the alkali and aklkaline earth ions to form soluble salts and leach out. Table 2 suggests a slightly higher abundance of aluminum and silicon compounds in the sandy and pale fragments and a slightly lower abundance in the slipped/burnished sample (Terenzi et al., 2010).

These aluminum and silicon compounds are most likely particles of feldspar. Feldspar is the most abundant mineral in the earth's crust, constituting 39% of the rock-forming minerals of the surface. Feldspar is an alumina silicate; that is, it consists of SiO_2 and Al_2O_3, with the relative proportion of SiO_2 varying roughly from 43% to 65%. Three additional elements – potassium, sodium, and calcium – are also present in feldspars in differing proportions. These elements are responsible for the division of the mineral family into potash or alkali feldspars (containing potassium; e.g. orthoclase and microcline), soda-lime feldspars (containing different relative quantities of sodium and calcium; e.g. albite, oligoclase, andesine, labradorite, bytownite, and anorthite), and lime feldspars (containing caclcium; e.g. anorthite). These last, the soda-lime feldspars, are collectively called plagioclases. Alkali feldspars (orthoclase and albite) have relatively high percentages of silica and are characteristic of more acid rocks, occurring with quartz in granites, for example, whereas the calcic or plagioclase feldspars are characteristic of the more basic rocks (diorites and basalts) (Rice 1987, 97). The high potassium contents in conjunction with SEM-EDS results, to be discussed later, verify that the ceramics are of an alkaline feldsparic nature, though plagioclase feldspar does occur as inclusions in some of the samples. The presence of potassium, sodium, and calcium in feldspars and in the clays that develop from them can also help determine the firing characteristics of clays, also to be discussed later.

The composition of any ceramic fragment depends on the types and proportions of clay paste and mineral, rock, grog, and organic tempers used in its manufacturing. It is well established that pottery can be grouped based on similarities or dissimilarities derived from chemical data. Relative ratios are one method used to understand the chemical makeup of ceramics, which can in turn be used establish fabric groups or determine provenance. Four abundance ratios were calculated to compare the relative quantities of elements to one another. Abundance ratios are used to establish patterns and determine if tempers were added in specific ratios, if the ceramics were made of similar clay compositions, or if natural reasons are primarily responsible for random variation. The average values and standard deviations can be found in Table 3.

Table 3: Abundance Ratios

Abundance Ratios				
	Fe/Ca	Ca/K	Mn/Fe	K/Ti
Griddle	2.27 ± 0.28	3.19 ± 1.19	0.015 ± 0.007	2.30 ± 1.33
Sandy Fabric	2.57 ± 0.19	2.79 ± 0.27	0.015 ± 0.004	1.89 ± 0.13
Pale Fabric	2.79 ± 0.08	2.45 ± 0.11	0.006 ± 0.000	1.51 ± 0.14
Slipped/Burnished	2.99 ± 1.61	2.97 ± 1.38	0.041 ± 0.019	1.89 ± 0.21

The Fe/Ca ratio is used by Grifa et al. 2009 and Terenzi et al. 2006 to differentiate between table and cooking wares. Grifa et al. 2009 determined that cooking wares will exhibit a Fe/Ca ratio of 3.3 ± 1.3, while table wares will exhibit ratios of 0.6 ± 0.1. Though these ratios are calculated for their sites in Cuma and Miseno, Italy, the griddle fragment, a known cooking ware, certainly falls within their postulated range. Since the other fragments exhibit similar Fe/Ca ratios, it could be said that these ceramics were able to withstand heat; though no conclusions as to their official function as either cooking or table wares can be drawn. The Mn/Fe ratio is associated with hardness. Lower manganese to iron ratios indicate that the ceramic is harder without being any more brittle. Manganese may also be responsible for the brown color of the ceramics while iron may be responsible for the reddish hue. The Mn/Fe ratio for the pale fabric is significantly lower, while the ratio for the slipped/burnished piece is significantly higher than the griddle and sandy fabrics, which exhibit equal ratios. Therefore the pale piece can be considered significantly harder, yet equally brittle.

Finally, a ternary diagram was generated for Zirconium, Strontium, and Rubidium to visually express a relative ratio. Other plots could be easily generated for provenance studies or for group establishments; other examples of this technique include Fe-Ca-K (Terenzi et al., 2006) and Ba-Cu-Zn (Adan-Bayewitz et al., 2009). The ratios found in this study are compared with those of majolica tiles found in Puebla, Mexico (Cruxent et al., 2003) in Figure 8.

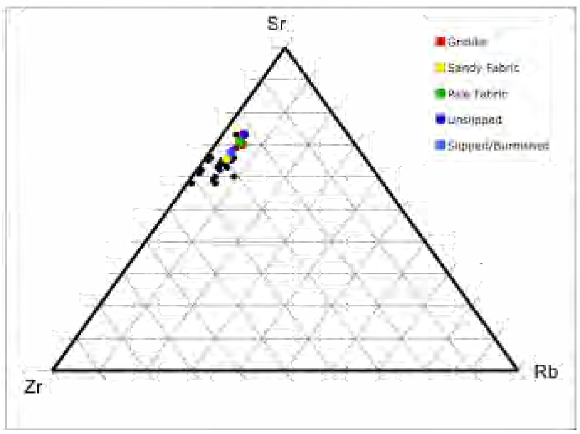


Figure 8: Zr-Sr-Rb Ternary Diagam

Each vertex represents a 100% proportion of the corresponding element. Each tick mark represents an increment of 10%. The black circles correspond to Mexican majolica tiles (Cruxent et al., 2003). The elements rubidium, strontium, and zirconium were chosen because they are considered to be part of the principal components used to analyze relative mass fractions of the clay materials found in Caribbean countries. They are also primary trace elements used to compare ceramic compositions globally: Greece (Pappalardo et al., 2004), Italy (Terenzi et al., 2006), and Galilee (Adan-Bayewitz, D., 2009). The ratios found in this study correlate well with those found in Puebla, Mexico. There is no reason at this point to believe that there is a direct link. A close examination of other elements may reveal greater discrepancies, but it is still interesting to note their similarity. Both sets of ceramics contain greater fractions of strontium. The high levels of strontium can indicate that large amounts of volcanic rock were being eroded and the sediment was finding its way into the ceramics. It should be noted here that both Puebla, Mexico and Valentine Ghaut, Montserrat are in close proximity to volcanoes. Puebla, Mexico is surrounded on all four sides by Popocatépetl, Iztaccíhuatl, La Malinche, and Pico de Orizaba volcanoes; while, Soufrière Hills Volcano is located in the southern region of Montserrat. Though no comparisons can be made in terms of production techniques or cultures due to vast distances and time spans between the Post-Saladoid (ca. 700 A.D) ceramics of Montserrat and majolica tiles (ca. 1650 A.D.) of Puebla, Mexico, it is important to acknowledge their similar geographical relationships to volcanoes and understand how this may have affected the chemical composition of their respective ceramics.

Although initially the pale fragment was thought to belong to a different fabric class based upon visual examination, there was nothing in particular that distinguished its chemical composition from the other samples among the detectable elements. Though certain differences in composition among the samples do exist, a larger sample size would be required to definitively state that they are in fact separate fabric types. Elements such as sulfur, calcium, and iron were used to hypothesize sources of materials used in the production process. Zinc and the Fe/Ca ratio were used to characterize the fragments' relationship with heat, relying heavily upon comparisons to the griddle as a known cooking ware. The color of the fragments was evaluated using sulfur, iron, and manganese to characterize their yellow, red, or brownish hues, respectively. Finally, the Zr-Sr-Rb relative ratio was used to compare the ceramic fragments of this study to another representative example of volcanic, Caribbean archaeology.

SEM/EDS:

Our research questions revolved around comparing fabric types and processes of production. Both SEM and EDS were valuable methods utilized to determine a range of possible firing temperatures and therefore carryout these objectives. It is visually obvious that all of the ceramics were low-fired, below 1000˚C. They are terracottas, as are virtually all unglazed prehistoric pottery is (Rice 82). However SEM and optical microscopy images were able to narrow the range because all of the samples showed that organic inclusions were present. EDS confirmed this with a compositional data makeup of carbon and oxygen of such inclusions (Figure 9).

Element	Weight%	Atomic%
C K	28.78	35.02
O K	71.08	64.94
Mn K	0.14	0.04
Totals	100.00	

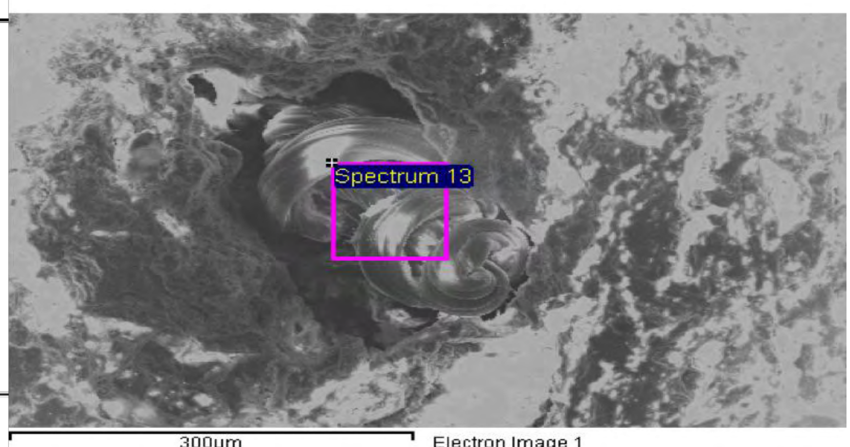


Figure 9: SEM-EDS of Pale Sample and an Organic Inclusion

More precisely, the firing range was probably between 600˚C-800˚C, as organic inclusions do not begin to volatilize until temperatures above 600˚C and more frequently above 750˚C (Rice 88).

SEM images also revealed a large amount of pores and voids within the samples. Organics, which can take the form of fibers, seeds, or other microbotanicals, whether intentionally added or not, leave such voids that are “casts” of the original (Rice 351). And therefore, porosity increases as organic matter is burned out. High porosity is a desirable property in low fired earthenware cooking pots because larger pores function to arrest the propagation of cracks that are generated by differential expansion of heated exterior and interior wall surfaces (Reid 63). So, whether intentional or not, the inclusion of such organic materials in the clay did aid in giving the ceramics properties to withstand differential exposure to heat used in cooking, however it is not definitive if these samples were indeed used for cooking. Additionally, fibrous materials can reduce shrinkage and strengthen the clay matrix (Hofman et al. 2008b: 27).

Organic material may have been added intentionally, to obtain a highly porous fabric. Alternatively, the organics may have entered the ceramic body inadvertently, as part of the production process. There are examples of Saladoid vessels preserving the imprint of basketry or other textiles, possibly as a result of the potter using a basket as base or guide while forming the vessel. Most vessels preserving this type of impression are of an earlier period than our assemblage (Hofman et al. 2008a: 6-7). However, if the potter smoothed the surface of the vessel after it was removed from basket, the impression need not have remained. Alternatively, the clay could have been wedged or otherwise worked on a woven mat, resulting in incorporation of organic matter in the clay matrix. Fine organic matter can naturally occur in raw clay sources in the Caribbean (see Isendoorn et al. 2008: 17 for organic matter in Saban clays). The organic inclusions in our samples are rather large, however, and are therefore less likely to be naturally occurring inclusions.

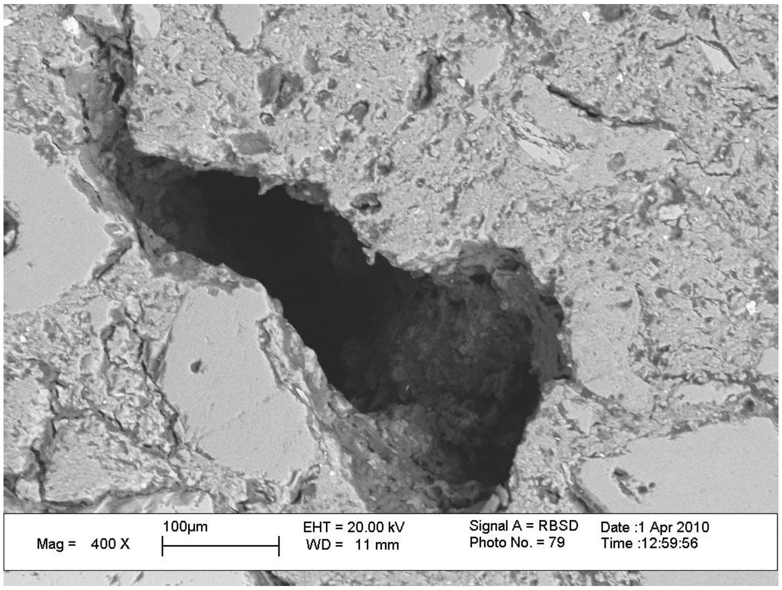


Figure 10: SEM image of a Pore in the Slipped/Burnished Sample

SEM images, accompanied with EDS compositional results, have allowed us to also examine the nature of both the fabric and inclusions of the ceramics. All of the fabrics of the ceramics were shown to be feldsparic in nature, meaning that they consist primarily of SiO_2 and Al_2O_3 with possible trace elements of calcium, potassium, and sodium. It is likely that they are specifically alkali feldspars because of the high percentages of silica and presence of acidic rocks, like quartz or albite (Rice 35). All of the samples exhibited fabrics that contained around a 50 wt% Oxygen content, 30 wt% Silica content, and 10wt% Aluminum content, appearing in the forms of SiO_2 and Al_2O_3. Feldspar may also exist either as minor or prominent constituents in clay bodies, and is not a constituent found in every ceramic (Grim 229). There are some locations of feldspar present on the island of Montserrat. Both the Soufriere Hills and Centre Hills regions contain phenocrysts of feldspar which suggest that it is possible that the clays were sourced locally on the island (MacGregor 241). When feldspar is present in a ceramic body, the thickness of the grains causes the ceramic to resist thermal impact, similar to pores. And therefore, the presence of feldspar, whether intentional or not, does show that these ceramics could be exposed to heat (Garcia et. al 9). Other inclusions detected by SEM-EDS in the samples consisted of iron and boron, both of which act as fluxes in the body of ceramics at low temperatures around 800˚C.

SEM-EDS was also able to detect certain production characteristics of the slipped and burnished sample. The sample has an iron oxide slip, which appeared on the EDS as a 10wt% difference in the level of iron. Also, a micrograph reveals some of the nature of the process of burnishing and slipping the ceramic (Fig. 11). When a ceramic is burnished, the platelets on the surface are compacted and all face and reflect light in the same way (Turner116). The figure shows this compaction beneath the slip. Unfortunately, we were not able to obtain a transect composition through the slipped surface with the EDS. This result could have clarified the nature of the slip attachment. According to Rice, low fired ceramics were often burnished after slipping to aid in slip adhesion (Rice 1987: 150). This technique may have been used on the slipped and burnished sample. Although the slip appears to lie over a flat, burnished surface in some areas of this image, in others it appears to have been forced into the surface of the clay body. Based on visual inspection, the un-slipped portion of the sherd appeared to be lightly burnished or smoothed, while the slipped portion was more heavily burnished, so the vessel may have been burnished twice, before and after it was slipped.

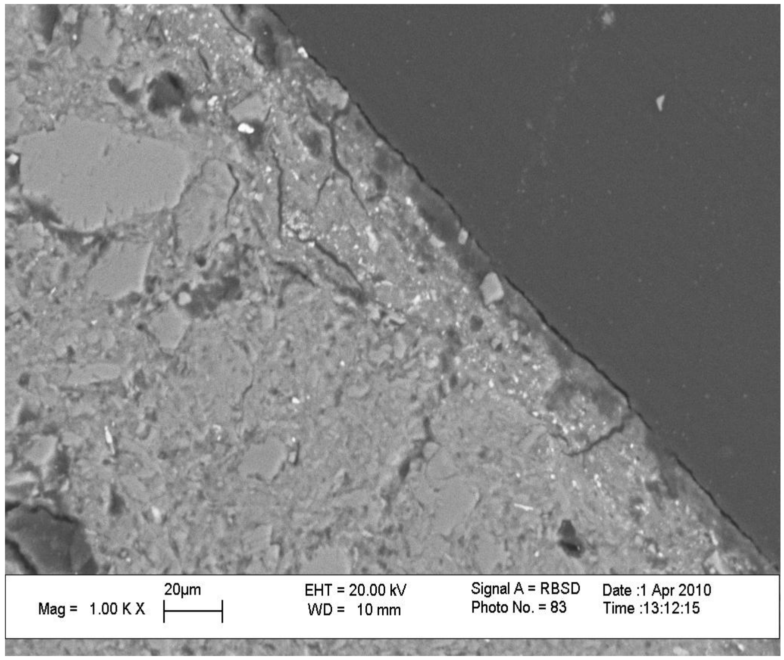


Figure 11: Cross-section of the Slipped/Burnished Sample Showing the Compaction of Clay by Burnishing and the Slip on Top

C: Thin-Section Petrography

Thin-section petrography is a useful method for identifying inclusions and characterizing their size, shape, and alignment. However, identification of minerals requires an extensive knowledge of geology. For this reason, we enlisted the aid of two geologists at Brown University, William Collins and Stephen Parman. William Collins helped us with sample preparation and Stephen Parman identified volcanic inclusions in the thin-sections. Due to time constraints we were only able to obtain expert opinions on two of the samples, the slipped and burnished sherd and the sandy sherd.

Volcanic minerals were found in both samples. In the slipped and burnished sample, Stephen Parman identified a large, angular inclusion of plagioclase feldspar (Fig. 12) (Stephen Parman, personal communication). The striated structure forms during successive eruptive event. The record of these events is unique and such a mineral could potentially be matched with the volcano that produced it (Stephen Parman, personal communication).

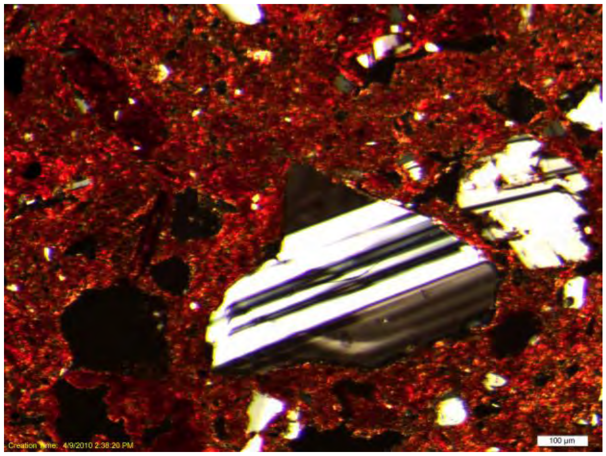


Fig. 12: Plagioclase feldspar in slipped and burnished sample

The sandy sample contained an inclusion of andesitic lava (Stephen Parman, personal communication) (Fig. 13). The matrix of the inclusion is composed of the andesitic magma that cooled quickly after eruption, while the mineral crystals within the larger inclusion were present within the liquid magma prior to the eruption. These minerals are primarily feldspars, and possibly pyroxene and amphibole. The circular black areas are gas bubbles formed during the eruption (Stephen Parman, personal communication). The volcanoes of Montserrat have primarily produced andesitic lavas (Donahue et al. 1990: 230). This correlation suggests that the vessel could have been made on Montserrat. However, volcanic inclusions in ceramics from limestone islands like Barbuda and Anguilla indicate that ceramic vessels, or at least raw materials, were traded among the islands (Hofman et al. 2008a: 11; Fuess et al. 1991: 29). Thus, we cannot rule out the possibility that this vessel was produced on another volcanic island in the region.

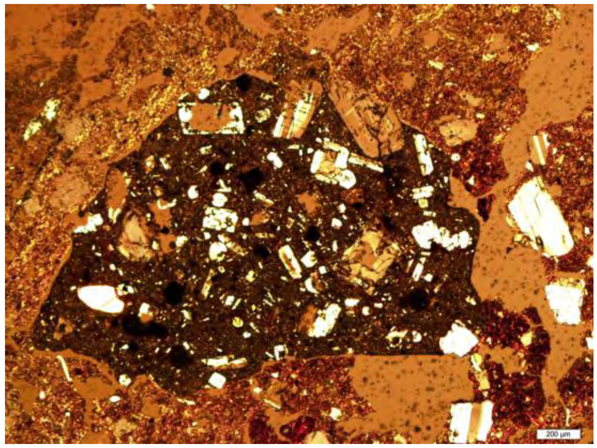


Fig. 13: Inclusion of andesitic lava in sandy sample.

In a study of 44 sherds from various islands in the Lesser Antilles Donahue et al. found that the primary inclusion in most sherds was volcanic minerals, generally in the form of relatively sand-sized, angular grains. Plagioclase feldspar and volcanic rock fragments were the most common constituents (Donahue et al. 1990: 240, 243). This study included five Saladoid sherds from Montserrat, all of which had inclusions of plagioclase feldspar and volcanic rock fragments. Additionally, these sherds also contained a small amount of carbonate material, much like our sherds (see discussion of shell fragment found with SEM, above). Donahue et al. suggest that the vessels were intentionally tempered with volcanic beach sand, based on the presence of carbonate material and the angular nature of the inclusions (Donahue et al. 247). Rounded grains are more often interpreted as naturally occurring inclusions, while angular grains are considered to be less likely to occur in a weathered clay bed. Angular grains can also result from deliberate crushing of temper material. However, determining intentionality is extremely difficult, and angular grains can still occur in primary clays (Rice 1987: 410-411, Donahue et al. 1990: 252). Still, the combination of shell and angular volcanic grains is suggestive of a sand temper.

The Saladoid period thin-sections from Trants and Radio Antilles were compared to historic period, slave-made vessels, and the inclusions appeared to be quite similar, possibly indicating some continuity in vessel manufacture (Donahue et al. 1990: 246-248). Our samples may fit into this tradition. Isendoorn describes improvement in the workability of some Saban clays upon the addition of fine sand (Isendoorn et al. 2008: 17). This may also be true of clays on Montserrat, though further investigation of local clay sources would be required to confirm this theory.

Thin-section petrography can also show evidence of surface treatment. Burnishing compacts the clay matrix and aligns the particles (Rice 1987: 138). This can be seen in the images below. Although there are many internal pores, the surface compaction and slip would have made this vessel less permeable than the other samples (Rice 1987: 232). Although it is difficult to suggest functionality from a single sherd, this reduced permeability could potentially be related to the vessel's function. Notably, this sherd was slipped and burnished on both the interior and exterior.

a b

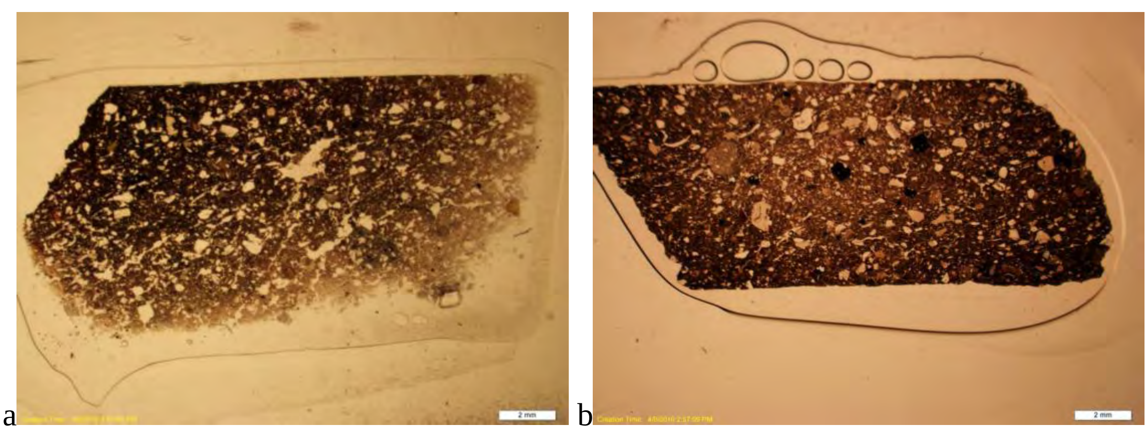


Fig. 14: Thin sections of slipped and burnished (a) and sandy (b) samples. Note the layer of compaction at the top of the burnished sample. Large inclusions are also absent from the slipped and burnished surface. No such compaction or inclusion sorting is evident on the sandy sample.

VI. **Remarks**

One of the issues confronted during the investigation was the small sample size. Having only four samples severely limited our ability to make broader claims about the ceramic types. What is true for our slipped and burnished sample, for example, may not be characteristic of slipped and burnished vessels from Valentine Ghaut in general.

It was difficult to conduct SEM-EDS because it took an excessively long time for the sample chamber to depressurize due to off-gassing from the samples. The slipped and burnished sample took particularly long to depressurize. Furthermore, the SEM was not configured correctly to allow a line composition with the EDS during one of our sessions.

In order to perform the petrography, it was necessary to rely heavily upon other scholars in the geology department. Petrography requires an extensive knowledge of the properties and appearance of minerals, and can take years to master.

XRF is only capable of taking surface readings, meaning that we were unable to characterize the interior composition of the samples with this method. Further, the selection of sampling points lead to outliers that were not characteristic of the fabric overall. Additionally, XRF is a semi-quantitative technique and can only detect certain elements.

VII. **Areas of Future Analysis**

Previous scientific analyses of material from Montserrat are rather limited, though studies exist of other islands in the area. The results established here leave room for future analyses of this assemblage. Tests should be carried out on a much larger sample size in order to make conclusive statements about the different fabric types. Furthermore, samples of clay and sand can be taken from the island in order to carry out a more thorough and comparative provenance study. Neutron activation analysis (NAA) could assist in this effort. An experience petrographer could create a fabric type collection and more fully elucidate the nature of inclusions. To our knowledge, our own study brings the total of analyzed ceramic thin-sections from Montserrat to a total of 9 (4 from our study, 5 from Donahue et al.'s study).

VIII. **Conclusions**

Though certain differences in composition, level of porosity, and number of inclusions exist among our samples, a larger sample size would be required to definitively state that they are in fact separate fabric types. Still, we have detected some trends (Fig. 15). The griddle fragment was much more porous than the other fragments, although all the samples had a high degree of porosity. The griddle was so porous that focusing was extremely difficult to conduct optical and scanning election microscopy. Although we initially thought that the pale fragment belonged to a different fabric class based upon visual examination, there was nothing in particular that distinguished its chemical composition from the other samples. In thin-section, it appears to have fewer inclusions than the other samples, though this has not been quantified. The sandy sample appears to have slightly more non-plastic inclusions than the other samples, including a higher number of organic inclusions. This vessel may have been lower fired than the others based on the number of organic inclusions that survive. As mentioned above, although the slipped and burnished sample has a high degree of porosity in the interior of the sherd, the slipping and burnishing potentially would have made it less permeable than the other samples.

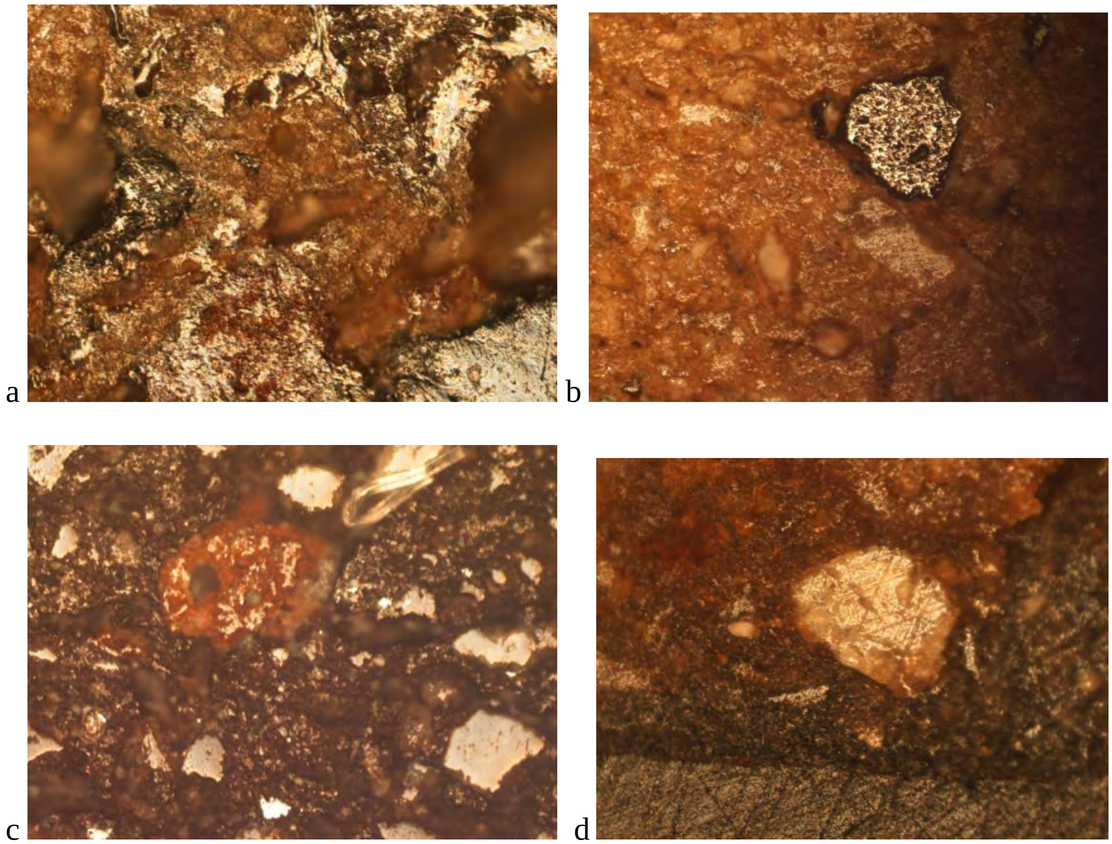


Figure 15: a) Highly porous griddle fragment. b) Light colored fabric of pale sherd with fewer large inclusions. c) Sandy fabric with many inclusions, including an organic fiber. d) Compacted burnished edge of slipped and burnished sample.

High iron contents found in the slipped portion of the slipped/burnished sample indicate that an iron oxide slip was used to produce the reddish hue. SEM and petrography show that the clay matrix was compacted during burnishing and the slip was added afterwards. The only other surface treatment evident in the ceramics was the incisions on the griddle. We believe this surface was used to cook food. Griddles from other sites and islands are typically described as having a flat, sometimes burnished, cooking surface and an unfinished bottom surface, which may bear the impression of finger prints or reeds. A “scratched” surface is only occasionally described. (Leiden Codebook for Ceramics 2009: 12; Petersen and Watters 1995: 135; Rouse and Morse 1999: 29). The griddle fragments in the assemblage from Feature 11 do have one flat surface, but the other surface is not unfinished. Rather, it is deliberately incised with a series of parallel lines. Descriptions of the flat cooking surfaces of griddles at other sites have led to speculation that the incised surface of our griddles was not the cooking surface. However, lime spalling occurs on the flat surface of at least one fragment, but not the incised surface, suggesting that this side was exposed to a heat source. Further, we have found a join between an incised griddle fragment and a likely base or leg that would require the incised surface to be the cooking surface, according to Leiden griddle shape 6. Alternatively, the joining fragment could be reconstructed as a wall or rim, akin to shape 3, though the angle between our fragments is much closer to 90 degrees. During the Post-Saladoid Period, trade networks seem to have been circumscribed and there was a flourishing of local pottery styles. Perhaps the griddle fragments from Valentine Ghaut represent a local style or innovation. Starch residue studies on future excavated griddle fragments, similar to those conducted by Suarez and Jimenez could clarify which surface was used for cooking (Suarez and Jimenez 2008).

Our investigations have also allowed us to draw some conclusions about the provenance, production, and functionality of these vessels. Montserrat can be considered a possible origin based upon calcium, feldspar, sulfur, and volcanic contents. Calcium may have been derived from either lime or shell sources. Shell and sand inclusions were found in the samples. These sherds were found near a beach; the levels of calcium detected during XRF indicate that lime or shell may have been present in the fabric. In fact a shell inclusion was found during SEM. Furthermore, optical micropscopy, petrography, and visual inspection show high levels of sand inclusions. All of the ceramics were shown to have a feldsparic fabric. There are two feldsparic pyroclasts on the island. Though sulfur was only detected in one sample, local outcrops in addition to deposits surrounding Sufrrere Hills exist. Volcanic materials were found in all of the samples. Further a volcanic inclusion found in the sandy sample during petrography was formed from andesitic magma, which is consistent with the volcanoes on Montserrat.

The fragments are hypothesized to be able to withstand heat based upon their high porosity, Fe/Ca ratios, contents of fine feldspar particles, and close comparison to the griddle fragment, a known cooking ware. All of the samples were fairly porous, particularly the griddle fragment. High porosity is desirable in vessels that are differentially exposed to heat because they arrest small cracks. The griddle fragment is known to be a cooking ware, and though no function can be assigned to the other ceramics, their mechanical properties are such that they would have been able to withstand heat. This hypothesis is also supported by the Fe/Ca ratios through XRF. The fine particles of feldspar found in the fabrics also indicate that they are capable of resisting thermal impacts.

Firing temperatures are thought to have been in the range of 600-800^{o}C based upon evidence of organic inclusions found during optical microscopy and SEM. Although pores can arrest small cracks, they do weaken the overall strength of the fabric. The high porosity of these fabrics, in conjunction with their high inclusion content and low firing temperature would have resulted in relatively friable vessels. Hofman, et al. conclude that vessels of similar fabrics from other islands would have had short life spans (Hofman et al. 2008b: 26). Thus, the need to replace broken vessels means that ceramic production was likely a reasonably regular activity, even among small communities.

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X: Appendix (note: please ignore numbering scheme)

Figure 6: Griddle - Top

Figure 7: Griddle - Bottom

Figure 8: Griiddle – Fabric

Figure 9: Griddle - Edge

Figure 10: Pale – Interior

Figure 11: Pale - Exterior

Figure 12: Pale– Fabric

Figure 13: Pale– Edge

Figure 14: Sandy – Interior

Figure 15: Sandy - Exterior

Figure 16: Sandy – Fabric

Figure 17: Sandy - Edge

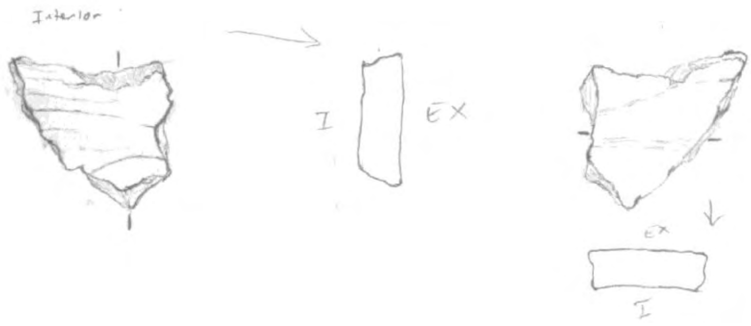
Figure 18: Slipped/Burnished – Interior

Figure 19: Slipped/Burnished - Exterior

Figure 20: Slipped/Burnished – Fabric

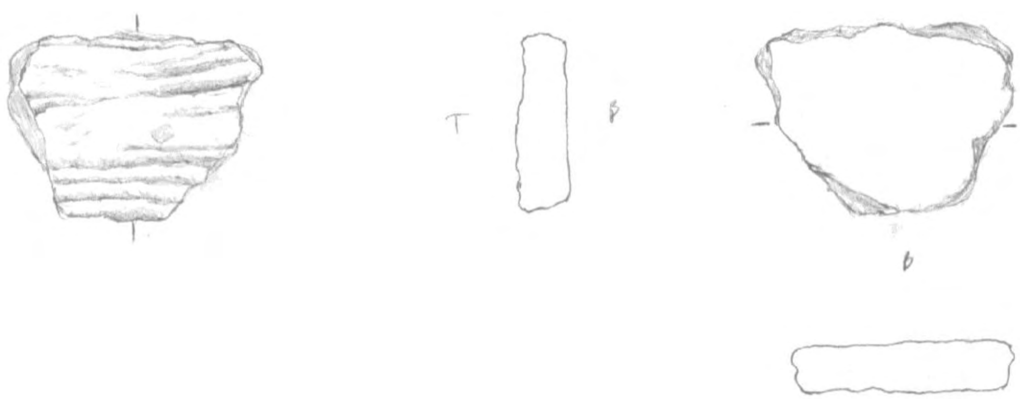
Figure 21: Slipped/Burnished - Edge

PALE FABRIC

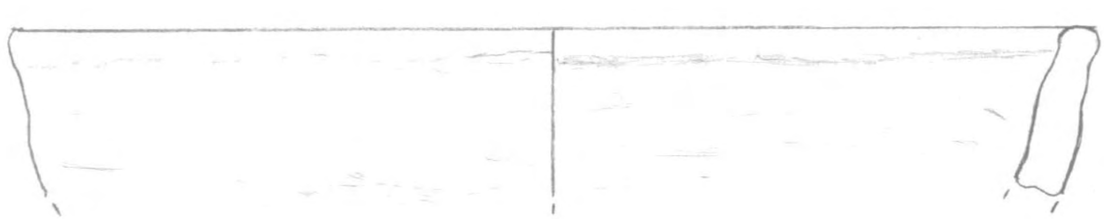


GRIDDLE

TOP



SANDY FABRIC



SLIPPED/BURNISHED