

## Elemental Compositions of Spanish and Spanish-Colonial Majolica Ceramics in the Identification of Provenience

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*On the basis of the compositions obtained by neutron activation analysis two distinctive groups of pottery have been identified from the majolica sherds excavated from Spanish sites in the New World. The principal sites yielding majolica sherds analyzed in this project include Isabela, La Vega Vieja, Juandolio, and Convento de San Francisco in the Dominican Republic; Nueva Cadiz in Venezuela; and excavations in Mexico City at the Metropolitan Cathedral and for the Mexico City Metro transportation system. Concentrations of  $\text{Na}_2\text{O}$ ,  $\text{K}_2\text{O}$ ,  $\text{BaO}$ ,  $\text{MnO}$ ,  $\text{Fe}_2\text{O}_3$ ,  $\text{Rb}_2\text{O}$ ,  $\text{Cs}_2\text{O}$ ,  $\text{La}_2\text{O}_3$ ,  $\text{Sc}_2\text{O}_3$ ,  $\text{CeO}_2$ ,  $\text{Eu}_2\text{O}_3$ ,  $\text{HfO}_2$ ,  $\text{ThO}_2$ ,  $\text{Cr}_2\text{O}_3$ , and  $\text{CoO}$  are reported. Petrographic and x-ray diffraction results confirm the existence of two distinctive groups. Comparisons to known specimens indicate a Spanish source and a Mexican source.*

The study of tin-glazed ceramics (majolica) associated with Spanish colonization of the New World has identified the need for methods to classify the various types of majolica which have been excavated. One of the major efforts in this regard has been made by John M. Goggin (1). Goggin studied majolica from public and private collections and from field study and/or excavation in Cuba, Jamaica, Haiti, the Dominican Republic, Puerto Rico, St. Croix, Trinidad, Venezuela, Columbia, Yucatan, and Mexico. The system of type names which he assigned to ceramics with specific glaze decorations is now the basis for establishing the chronology and provenience of the majolica.

In the course of colonizing the New World the Spanish first brought majolica from Spain to the colonies, then in due course manufactured it there. Records reveal that between 1504 and 1555 some 2,805 ships sailed for the New World from Seville and the Canary Islands carrying cargos of weapons, provisions, metal, and household goods including "loza blanca" or white tableware. One of the types of majolica which is found in most of the early sites is a plain, white-glazed ceramic tableware which Goggin has named Columbia Plain. The glaze is made by adding tin oxide to a lead glaze. We have identified lead and tin by x-ray fluorescence in the glaze of four of our Columbia Plain sherds. Majolica types other than Columbia Plain have a colored design applied to a thin opaque white glaze or have a colored opaque glaze. Examples of these and other 16th century types are shown in Figures 1-4.

The tradition of majolica production in Spain is well documented. Its presence there dates back to before the 11th century. The types of majolica most frequently found in museum collections in Europe and the United States, however, are generally more decorative types than those excavated at the early Spanish sites in the New World. Nevertheless, we know that the types shown in Figures 1 and 2, Columbia Plain and Yayal-Blue-on-White, were used in Spain. These two types appear in paintings of Velasquez, Zurbaran, and Murillo, all of whom painted in Seville early in the 17th century. We know, therefore, that the majolica types excavated at the early 16th century Spanish sites in the New World were being produced in Spain. We also know that white tableware was being shipped to the New World. It is reasonable to conclude that majolica found in early sites in the New World may have originated in Spain.

### Sources of Specimens

The sites in the New World from which these sherds have been excavated are among the earliest to be inhabited by the Spanish. In the Dominican Republic there are four: Isabela, which was the first substantial settlement in the New World; La Vega Vieja, which appears to have been founded as early as 1495; the Convento de San Francisco, the first of several religious houses to be constructed in the city of Santa Domingo (for which the church was completed in 1555 and the monastery in 1556); and Juandolio, which was an early 16th century site. In Venezuela the site of Nueva Cadiz was the earliest Spanish settlement. It was founded about 1515 and was abruptly destroyed by an earthquake in 1545 and subsequently deserted. The sherds from Mexico come from excavations within Mexico City. These include archaeological excavations at the Metropolitan Cathedral and commercial excavation for the Metro

subway construction. We have analyzed a few additional sherds recovered from Guatemala, Panama, Peru, and Ecuador.

The sherds from the Dominican Republic and all but four of those from Venezuela were sent to us by Charles Fairbanks. Some of these sherds were excavated by John M. Goggin and are from the collection at the University of Florida. The sherds from Mexico City and from the sites in Panama, Guatemala, Ecuador, and Peru were provided by Florence and Robert Lister.

To determine whether any of the New World material was actually fabricated in Spain it is necessary, of course, to have material from Spain itself for comparison. However, little if any excavated majolica of the 16th or 17th century from Spain is available. According to F. Lister (2), there are no controlled excavations of 16th and 17th century sites in the area of Seville-Triana which she and others believe to have been the source of the majolica shipped to the New World. In 1975, F. Lister learned of some excavations which had taken place at the Carthusian monastery at Jerez, Spain. This monastery was constructed about the time of the Spanish penetration in the Caribbean. The sherds which we have analyzed are from among those found in a dump area outside of the monastery (2). They are of the types Columbia Plain and Yayal-Blue-on-White and therefore do correspond to types which are included with the excavated materials from the New World Sites. Admittedly this is a very small sampling of comparison material from Spain but the lack of archaeological excavations of sites which would have produced majolica of this period has made these sherds more valuable than they otherwise would be. It is hoped that future archaeological excavation in Spain will provide a more complete sampling of majolica of authentic Spanish origin which we can analyze.

#### *Experimental Methods and Results*

In this research we have attempted to obtain evidence of the origin of the sherds found in the New World through neutron activation analysis, x-ray diffraction, and petrographic analysis. Neutron activation analysis provided the concentrations of 15 constituent oxides for 178 sherds. These analyses were carried out in the Chemistry Department at Brookhaven National Laboratory using the standard procedures developed there which are reported by Abascal et al. (3). Six U.S. Geological Survey standard rocks designated AGVI, DTS-1, PCC-1, GSP-1, and G-2 were used as standards (4). The specific standards used for each element are reported by Bieber et al. (5).

Neutron activation analysis revealed that the compositions of all the specimens from the five early sites in the Dominican Republic and Venezuela closely agree and that they differ significantly from all of the

other specimens from New World sites. The individual data for these sherds are given in Table I, and in Table II the mean compositions of the sherds from the five sites are compared. The elements which do show some discernible variation among sites are the alkali metals rubidium and sodium. The alkali compounds tend to be water soluble and hence are susceptible to variations during burial.

Using the data in Table I for 40 Columbia Plain and Yayal-Blue-on-White sherds from Convento de San Francisco, Dominican Republic and for 41 sherds from Nueva Cadiz, Venezuela, we have calculated a value of  $t$  by dividing the difference between the means of the concentrations by the standard error of the difference between the means. We have calculated  $t$  for the oxides of rubidium, sodium, and cesium, and the numbers obtained are 5.6, 4.1, and 1.8, respectively. With this sample size the significance levels for the values of  $t$  for rubidium and sodium are greater than 99.5% and for cesium are greater than 90%. There is, therefore, a statistically significant difference between the concentrations of each of these three oxides for all of the sherds from Convento de San Francisco compared with those from Nueva Cadiz.

The Columbia Plain sherds from four different sites shown in Figure 1 have a different physical appearance. The physical condition of the glaze is consistent among sherds from a given site but is readily distinguishable in appearance from sherds from other sites. It appears that the glaze is altered to different degrees depending on different burial conditions. These differences in burial conditions could also have contributed to the variations in the alkali concentrations in the sherds. The basic similarity in composition of the sherds from all of these five sites strongly suggests, however, that they were manufactured from a single or at most a few closely related clays; that is to say, the similarity in composition would argue against a hypothesis that these wares were manufactured in each of the separate sites in which they were found. Indeed Goggin concludes that they were all probably of Spanish origin, most likely the Triana section of Seville (1).

The results of our analysis of the sherds from Spain which were supplied to us by F. Lister reveal that the composition of these sherds basically conforms to the same compositional pattern which characterizes the sherds from the five sites in the Dominican Republic and Venezuela. The data for the sherds from Jerez are given in Table III, and the mean compositions are given in Table II with the mean compositions for the sherds from sites in the New World. The agreement between the sherds found in the Dominican Republic and Venezuela and those found in Spain is excellent for 14 or 15 elements, the only exception being the component barium oxide. However, we frequently have observed erratic barium concentrations in related groups of sherds, and we tend to ascribe

Table I. Concentration of Oxides in Majolica from Spanish

| Specimen No.   | Na <sub>2</sub> O (%) | K <sub>2</sub> O (%) | BaO (%) | MnO (%) | Fe <sub>2</sub> O <sub>3</sub> (%) | Rb <sub>2</sub> O (ppm) | Cs <sub>2</sub> O (ppm) |
|--|-----------------------|----------------------|---------|---------|------------------------------------|-------------------------|-------------------------|
| <i>Columbia Plain, Convento de San Francisco, Dominican Republic</i> |                       |                      |         |         |                                    |                         |                         |
| SA01   | 0.84                  | 1.96                 | 0.040   | 0.073   | 4.8                                | 109                     | 5.7                     |
| SA02   | 0.85                  | 2.03                 | 0.039   | 0.067   | 5.0                                | 216                     | 6.6                     |
| SA03   | 0.93                  | 1.75                 | 0.041   | 0.075   | 4.9                                | 107                     | 6.0                     |
| SA04   | 1.20                  | 1.08                 | 0.035   | 0.077   | 5.2                                | 108                     | 6.8                     |
| SA5B   | 1.01                  | 0.91                 | 0.036   | 0.058   | 4.6                                | 110                     | 6.0                     |
| SO6A   | 0.79                  | 1.72                 | 0.050   | 0.068   | 4.9                                | 132                     | 5.4                     |
| SA07   | 0.87                  | 1.52                 | 0.042   | 0.063   | 4.9                                | 142                     | 6.2                     |
| SA08   | 1.05                  | 1.32                 | 0.041   | 0.069   | 5.0                                | 109                     | 6.8                     |
| SA9B   | 1.15                  | 1.06                 | 0.035   | 0.071   | 4.7                                | 95                      | 5.5                     |
| SA10   | 1.01                  | 1.10                 | 0.041   | 0.064   | 4.4                                | 122                     | 5.1                     |
| SB86   | 0.91                  | 1.77                 | 0.050   | 0.086   | 5.1                                | 82                      | 4.8                     |
| SB90   | 1.03                  | 1.19                 | 0.044   | 0.073   | 4.9                                | 92                      | 5.2                     |
| SB91   | 1.16                  | 1.12                 | 0.046   | 0.073   | 5.0                                | 79                      | 5.6                     |
| SB92   | 1.36                  | 0.98                 | 0.032   | 0.076   | 5.2                                | 83                      | 5.7                     |
| SB93   | 1.13                  | 1.14                 | 0.042   | 0.070   | 5.0                                | 96                      | 6.4                     |
| SB94   | 0.98                  | 1.27                 | 0.042   | 0.070   | 4.6                                | 85                      | 5.2                     |
| SB95   | 0.98                  | 1.07                 | 0.043   | 0.069   | 4.7                                | 84                      | 4.7                     |
| SB96   | 0.64                  | 2.28                 | 0.044   | 0.078   | 4.5                                | 85                      | 4.0                     |
| SB97   | 1.04                  | 1.44                 | 0.057   | 0.086   | 6.2                                | 69                      | 4.8                     |
| SB98   | 1.11                  | 1.56                 | 0.048   | 0.093   | 5.2                                | 81                      | 5.4                     |
| SB99   | 1.42                  | 0.90                 | 0.031   | 0.075   | 4.8                                | 80                      | 6.2                     |
| SC01   | 1.07                  | 1.56                 | 0.048   | 0.080   | 5.3                                | 74                      | 4.3                     |

*Columbia Plain, Nueva Cadiz, Venezuela*

|      |      |      |       |       |     |    |     |
|------|------|------|-------|-------|-----|----|-----|
| SA11 | 1.00 | 1.83 | 0.051 | 0.083 | 5.0 | 76 | 7.4 |
| SA12 | 0.67 | 2.05 | 0.063 | 0.077 | 4.9 | 72 | 6.1 |
| SA13 | 1.12 | 1.62 | 0.038 | 0.078 | 5.0 | 64 | 4.9 |
| SA14 | 1.06 | 1.14 | 0.054 | 0.083 | 5.3 | 63 | 7.7 |
| SA15 | 1.14 | 1.20 | 0.112 | 0.081 | 5.1 | 62 | 7.2 |
| SA16 | 1.09 | 1.62 | 0.048 | 0.077 | 5.0 | 79 | 8.1 |
| SA17 | 1.38 | 1.36 | 0.038 | 0.067 | 4.5 | 49 | 5.1 |
| SA18 | 1.00 | 1.28 | 0.055 | 0.077 | 5.1 | 48 | 7.2 |
| SA19 | 1.01 | 1.41 | 0.043 | 0.070 | 5.4 | 70 | 6.3 |
| SA20 | 1.29 | 1.02 | 0.048 | 0.076 | 5.3 | 48 | 6.6 |
| SB59 | 1.62 | 1.07 | 0.033 | 0.090 | 5.0 | 56 | 5.8 |
| SB60 | 1.77 | 1.05 | 0.062 | 0.081 | 5.5 | 44 | 6.1 |
| SB61 | 1.54 | 1.62 | 0.042 | 0.083 | 4.6 | 77 | 5.3 |
| SB63 | 1.56 | 1.78 | 0.042 | 0.081 | 4.7 | 75 | 5.9 |
| SB64 | 1.28 | 1.38 | 0.042 | 0.092 | 5.4 | 72 | 7.2 |
| SB65 | 1.58 | 1.06 | 0.055 | 0.080 | 4.9 | 70 | 5.1 |
| SB66 | 1.94 | 0.76 | 0.037 | 0.083 | 4.9 | 51 | 5.8 |
| SB67 | 1.84 | 0.99 | 0.053 | 0.085 | 4.4 | 48 | 4.1 |
| SB68 | 1.54 | 0.96 | 0.046 | 0.083 | 5.2 | 52 | 5.8 |

## American Sites in the Dominican Republic and Venezuela

| La <sub>2</sub> O <sub>3</sub> (ppm)                                 | Sc <sub>2</sub> O <sub>3</sub> (ppm) | CeO <sub>2</sub> (ppm) | Eu <sub>2</sub> O <sub>3</sub> (ppm) | HfO <sub>2</sub> (ppm) | ThO <sub>2</sub> (ppm) | Cr <sub>2</sub> O <sub>3</sub> (ppm) | CoO (ppm) |
|--|--------------------------------------|------------------------|--------------------------------------|------------------------|------------------------|--------------------------------------|-----------|
| <i>Columbia Plain, Convento de San Francisco, Dominican Republic</i> |                                      |                        |                                      |                        |                        |                                      |           |
| 39   | 19.2                                 | 79                     | 1.52                                 | 5.9                    | 11.3                   | 107                                  | 17.5      |
| 40   | 22.0                                 | 83                     | 1.49                                 | 6.5                    | 11.9                   | 112                                  | 15.8      |
| 40   | 20.0                                 | 81                     | 1.60                                 | 6.1                    | 12.1                   | 123                                  | 15.8      |
| 44   | 21.1                                 | 90                     | 1.74                                 | 6.5                    | 12.6                   | 126                                  | 16.7      |
| 39   | 18.4                                 | 80                     | 1.45                                 | 5.8                    | 11.6                   | 113                                  | 14.7      |
| 39   | 19.8                                 | 83                     | 1.52                                 | 5.8                    | 11.3                   | 93                                   | 16.8      |
| 42   | 19.9                                 | 85                     | 1.41                                 | 5.9                    | 11.9                   | 121                                  | 16.2      |
| 41   | 20.0                                 | 86                     | 1.58                                 | 6.0                    | 12.5                   | 112                                  | 16.3      |
| 39   | 19.5                                 | 81                     | 1.52                                 | 6.5                    | 11.4                   | 117                                  | 18.3      |
| 37   | 17.7                                 | 75                     | 1.38                                 | 6.2                    | 11.1                   | 107                                  | 15.0      |
| 47   | 20.6                                 | 133                    | 1.55                                 | 6.2                    | 12.5                   | 126                                  | 17.0      |
| 36   | 19.2                                 | 61                     | 1.38                                 | 5.4                    | 11.4                   | 114                                  | 15.1      |
| 37   | 20.0                                 | 86                     | 1.51                                 | 6.3                    | 12.2                   | 124                                  | 13.8      |
| 36   | 21.2                                 | 59                     | 1.58                                 | 5.8                    | 11.3                   | 129                                  | 16.2      |
| 37   | 20.5                                 | 69                     | 1.56                                 | 6.2                    | 11.8                   | 131                                  | 14.9      |
| 35   | 18.6                                 | 49                     | 1.35                                 | 5.7                    | 11.1                   | 111                                  | 14.3      |
| 34   | 19.1                                 | 90                     | 1.35                                 | 5.4                    | 10.1                   | 110                                  | 14.9      |
| 35   | 18.4                                 | 63                     | 1.37                                 | 6.0                    | 10.8                   | 99                                   | 15.0      |
| 39   | 22.0                                 | 88                     | 1.46                                 | 5.9                    | 11.8                   | 131                                  | 17.7      |
| 38   | 21.0                                 | 42                     | 1.72                                 | 6.6                    | 12.7                   | 122                                  | 17.1      |
| 35   | 19.5                                 | 73                     | 1.43                                 | 5.4                    | 11.1                   | 122                                  | 15.1      |
| 40   | 20.9                                 | 85                     | 1.61                                 | 6.9                    | 12.9                   | 116                                  | 17.8      |

*Columbia Plain, Nueva Cadiz, Venezuela*

|    |      |    |      |     |      |     |      |
|----|------|----|------|-----|------|-----|------|
| 37 | 20.4 | 80 | 1.66 | 5.5 | 11.8 | 111 | 17.1 |
| 38 | 19.8 | 84 | 1.56 | 6.3 | 11.7 | 113 | 17.0 |
| 37 | 20.0 | 82 | 1.47 | 5.6 | 11.5 | 113 | 15.4 |
| 39 | 21.0 | 83 | 1.47 | 5.6 | 11.2 | 113 | 16.1 |
| 40 | 20.7 | 81 | 1.64 | 5.5 | 11.9 | 117 | 16.0 |
| 37 | 20.1 | 82 | 1.66 | 5.6 | 11.9 | 110 | 16.7 |
| 40 | 18.0 | 79 | 1.41 | 5.7 | 10.5 | 98  | 13.3 |
| 39 | 20.5 | 88 | 1.50 | 5.9 | 11.8 | 111 | 15.6 |
| 38 | 20.0 | 82 | 1.59 | 5.6 | 11.0 | 111 | 14.1 |
| 36 | 22.0 | 87 | 1.67 | 6.0 | 12.6 | 125 | 16.0 |
| 36 | 20.0 | 79 | 1.60 | 6.1 | 11.8 | 120 | 16.7 |
| 40 | 22.4 | 89 | 1.71 | 5.9 | 12.7 | 134 | 16.2 |
| 35 | 19.5 | 77 | 1.65 | 5.5 | 10.9 | 115 | 18.3 |
| 38 | 19.6 | 86 | 1.68 | 6.3 | 13.0 | 115 | 16.9 |
| 38 | 21.9 | 83 | 1.64 | 5.2 | 12.1 | 132 | 17.5 |
| 36 | 20.2 | —  | 1.42 | 5.9 | 11.4 | 118 | 18.8 |
| 38 | 19.3 | —  | 1.61 | 6.2 | 11.8 | 123 | 15.3 |
| 32 | 17.4 | —  | 1.19 | 6.3 | 10.2 | 114 | 15.5 |
| 40 | 20.8 | —  | 1.74 | 6.2 | 13.0 | 133 | 14.7 |

Table I.

| Specimen No.  | Na <sub>2</sub> O (%) | K <sub>2</sub> O (%) | BaO (%) | MnO (%) | Fe <sub>2</sub> O <sub>3</sub> (%) | Rb <sub>2</sub> O (ppm) | Cs <sub>2</sub> O (ppm) |
|---|-----------------------|----------------------|---------|---------|------------------------------------|-------------------------|-------------------------|
| <i>Columbia Plain, Juandolio, Dominican Republic</i>                      |                       |                      |         |         |                                    |                         |                         |
| SA32  | 0.92                  | 2.20                 | 0.043   | 0.075   | 5.0                                | 81                      | 4.2                     |
| SA33  | 0.72                  | 2.08                 | 0.043   | 0.077   | 5.2                                | 83                      | 5.4                     |
| SA34  | 0.93                  | 2.10                 | 0.048   | 0.093   | 5.8                                | 89                      | 5.6                     |
| SA35  | 0.96                  | 1.87                 | 0.045   | 0.088   | 5.4                                | 76                      | 5.9                     |
| SA36  | 0.66                  | 2.60                 | 0.046   | 0.080   | 5.1                                | 98                      | 4.6                     |
| SA37  | 1.17                  | 1.47                 | 0.044   | 0.099   | 5.6                                | 116                     | 6.9                     |
| SA38  | 1.17                  | 1.53                 | 0.046   | 0.082   | 5.6                                | 80                      | 6.3                     |
| SA39  | 0.89                  | 1.41                 | 0.042   | 0.079   | 5.0                                | 92                      | 6.0                     |
| SA40  | 0.78                  | 1.78                 | 0.047   | 0.085   | 5.1                                | 93                      | 5.3                     |
| SA41  | 0.95                  | 1.73                 | 0.046   | 0.086   | 5.3                                | 85                      | 5.7                     |
| <i>Columbia Plain, La Vega Vieja, Dominican Republic</i>                  |                       |                      |         |         |                                    |                         |                         |
| SA50  | 0.67                  | 1.92                 | 0.056   | 0.098   | 5.6                                | 98                      | 5.8                     |
| SA51  | 0.45                  | 2.32                 | 0.052   | 0.072   | 5.7                                | 116                     | 5.6                     |
| SA52  | 0.24                  | 2.51                 | 0.045   | 0.076   | 4.7                                | 97                      | 3.9                     |
| SA53  | 0.69                  | 1.73                 | 0.046   | 0.078   | 5.2                                | 116                     | 4.9                     |
| SA54  | 0.49                  | 2.01                 | 0.065   | 0.088   | 5.8                                | 94                      | 5.2                     |
| SA55  | 0.68                  | 1.61                 | 0.055   | 0.084   | 5.8                                | 112                     | 6.1                     |
| SA56  | 0.49                  | 1.70                 | 0.055   | 0.093   | 5.7                                | 71                      | 3.6                     |
| SA57  | 0.59                  | 2.13                 | 0.060   | 0.076   | 5.8                                | 129                     | 5.8                     |
| SA58  | 0.51                  | 1.82                 | 0.049   | 0.064   | 4.7                                | 79                      | 4.4                     |
| SA59  | 0.61                  | 2.02                 | 0.056   | 0.088   | 5.6                                | 95                      | 5.2                     |
| <i>Yayal-Blue-on-White, Convento de San Francisco, Dominican Republic</i> |                       |                      |         |         |                                    |                         |                         |
| SA21  | 1.51                  | 1.23                 | 0.046   | 0.078   | 5.0                                | 59                      | 5.9                     |
| SA22  | 1.29                  | 1.20                 | 0.058   | 0.084   | 5.4                                | 96                      | 8.5                     |
| SA23  | 1.15                  | 1.49                 | 0.045   | 0.202   | 4.9                                | 68                      | 7.8                     |
| SB69  | 1.27                  | 1.20                 | 0.057   | 0.086   | 5.4                                | 99                      | 7.6                     |
| SB70  | 0.95                  | 1.50                 | 0.056   | 0.082   | 5.3                                | 80                      | 5.3                     |
| SB71  | 1.06                  | 1.52                 | 0.051   | 0.087   | 5.4                                | 74                      | 6.0                     |
| SB72  | 1.38                  | 1.46                 | 0.046   | 0.080   | 4.8                                | 98                      | 6.2                     |
| SB73  | 1.24                  | 1.41                 | 0.048   | 0.080   | 5.1                                | 91                      | 5.6                     |
| SB74  | 1.05                  | 1.52                 | 0.053   | 0.093   | 5.1                                | 58                      | 4.3                     |
| SB75  | 0.85                  | 1.67                 | 0.065   | 0.089   | 5.2                                | 73                      | 4.2                     |
| SB76  | 1.10                  | 2.32                 | 0.052   | 0.089   | 6.1                                | 111                     | 6.3                     |
| SB77  | 1.02                  | 1.87                 | 0.056   | 0.089   | 5.2                                | 67                      | 4.9                     |
| SB78  | 1.17                  | 1.42                 | 0.053   | 0.090   | 5.1                                | 58                      | 4.0                     |
| SB80  | 1.07                  | 1.96                 | 0.062   | 0.097   | 5.6                                | 67                      | 3.9                     |
| SB82  | 1.14                  | 1.09                 | 0.058   | 0.090   | 5.1                                | 59                      | 3.9                     |
| SB83  | 0.82                  | 1.75                 | 0.041   | 0.077   | 4.6                                | 70                      | 3.6                     |
| SB84  | 1.13                  | 1.10                 | 0.046   | 0.073   | 4.9                                | 73                      | 5.0                     |
| SB85  | 1.14                  | 1.52                 | 0.042   | 0.071   | 5.2                                | 98                      | 6.0                     |

Continued

| La <sub>2</sub> O <sub>3</sub> (ppm)                                      | Sc <sub>2</sub> O <sub>3</sub> (ppm) | CeO <sub>2</sub> (ppm) | Eu <sub>2</sub> O <sub>3</sub> (ppm) | HfO <sub>2</sub> (ppm) | ThO <sub>2</sub> (ppm) | Cr <sub>2</sub> O <sub>3</sub> (ppm) | CoO (ppm) |
|---|--------------------------------------|------------------------|--------------------------------------|------------------------|------------------------|--------------------------------------|-----------|
| <i>Columbia Plain, Juandolio, Dominican Republic</i>                      |                                      |                        |                                      |                        |                        |                                      |           |
| 47  | 20.4                                 | 84                     | 1.71                                 | 5.7                    | 12.6                   | 112                                  | 15.4      |
| 41  | 21.0                                 | 86                     | 1.67                                 | 6.0                    | 12.4                   | 110                                  | 15.2      |
| 46  | 22.9                                 | 95                     | 1.78                                 | 7.0                    | 14.9                   | 123                                  | 17.5      |
| 43  | 22.0                                 | 88                     | 1.71                                 | 6.2                    | 12.8                   | 124                                  | 16.2      |
| 34  | 21.0                                 | 85                     | 1.67                                 | 5.8                    | 11.6                   | 110                                  | 15.8      |
| 42  | 23.2                                 | 88                     | 1.61                                 | 5.4                    | 13.0                   | 124                                  | 16.8      |
| 44  | 23.0                                 | 93                     | 1.80                                 | 7.6                    | 14.1                   | 129                                  | 15.0      |
| 46  | 20.6                                 | 78                     | 1.62                                 | 5.7                    | 11.4                   | 114                                  | 14.6      |
| 40  | 20.4                                 | 82                     | 1.62                                 | 6.5                    | 11.9                   | 109                                  | 18.8      |
| 41  | 22.0                                 | 87                     | 1.67                                 | 5.5                    | 12.7                   | 121                                  | 16.5      |
| <i>Columbia Plain, La Vega Vieja, Dominican Republic</i>                  |                                      |                        |                                      |                        |                        |                                      |           |
| 46  | 22.0                                 | 96                     | 1.82                                 | 6.9                    | 13.5                   | 125                                  | 18.1      |
| 42  | 21.7                                 | 91                     | 1.69                                 | 6.1                    | 13.2                   | 126                                  | 17.3      |
| 44  | 19.1                                 | 92                     | 1.54                                 | 5.5                    | 14.4                   | 88                                   | 17.2      |
| 37  | 18.7                                 | 80                     | 1.50                                 | 5.4                    | 11.7                   | 114                                  | 14.9      |
| 46  | 24.0                                 | 94                     | 1.77                                 | 6.5                    | 14.6                   | 135                                  | 16.5      |
| 44  | 23.4                                 | 94                     | 1.77                                 | 6.1                    | 12.8                   | 134                                  | 15.5      |
| 43  | 22.5                                 | 94                     | 1.60                                 | 6.8                    | 13.5                   | 129                                  | 14.5      |
| 40  | 22.9                                 | 89                     | 1.62                                 | 6.6                    | 13.3                   | 133                                  | 13.9      |
| 40  | 19.0                                 | 84                     | 1.46                                 | 5.5                    | 11.5                   | 113                                  | 16.4      |
| 42  | 22.0                                 | 87                     | 1.71                                 | 6.3                    | 12.8                   | 126                                  | 16.4      |
| <i>Yayal-Blue-on-White, Convento de San Francisco, Dominican Republic</i> |                                      |                        |                                      |                        |                        |                                      |           |
| 32  | 19.5                                 | 83                     | 1.59                                 | 6.9                    | 10.9                   | 110                                  | 18.8      |
| 36  | 22.2                                 | 87                     | 1.61                                 | 5.6                    | 12.3                   | 123                                  | 19.3      |
| 40  | 19.5                                 | 80                     | 1.62                                 | 5.5                    | 11.9                   | 114                                  | 68.3      |
| 41  | 21.9                                 | —                      | 1.78                                 | 6.6                    | 13.1                   | 140                                  | 17.7      |
| 38  | 21.4                                 | —                      | 1.97                                 | 6.0                    | 11.8                   | 113                                  | 18.1      |
| 40  | 21.6                                 | —                      | 1.63                                 | 6.3                    | 12.2                   | 128                                  | 16.8      |
| 54  | 20.0                                 | —                      | 1.64                                 | 5.7                    | 11.6                   | 128                                  | 19.9      |
| 39  | 21.1                                 | —                      | 1.72                                 | 6.2                    | 11.7                   | 116                                  | 17.8      |
| 42  | 20.3                                 | —                      | 1.83                                 | 5.6                    | 12.7                   | 121                                  | 20.9      |
| 37  | 20.2                                 | —                      | 1.58                                 | 5.8                    | 11.9                   | 121                                  | 22.8      |
| 43  | 24.5                                 | —                      | 1.82                                 | 5.8                    | 13.6                   | 142                                  | 20.5      |
| 52  | 20.6                                 | —                      | 1.60                                 | 6.0                    | 11.8                   | 128                                  | 21.9      |
| 39  | 20.4                                 | —                      | 1.63                                 | 5.8                    | 12.6                   | 131                                  | 22.3      |
| 42  | 23.7                                 | —                      | 1.71                                 | 5.8                    | 13.0                   | 137                                  | 22.9      |
| 41  | 20.0                                 | —                      | 1.56                                 | 6.0                    | 11.4                   | 132                                  | 22.9      |
| 35  | 19.1                                 | 105                    | 1.30                                 | 4.7                    | 11.2                   | 110                                  | 18.5      |
| 35  | 19.7                                 | 56                     | 1.34                                 | 5.7                    | 11.0                   | 118                                  | 17.9      |
| 37  | 20.9                                 | 75                     | 1.58                                 | 6.3                    | 11.5                   | 132                                  | 15.8      |

Table I.

| Specimen No.  | Na <sub>2</sub> O (%) | K <sub>2</sub> O (%) | BaO (%) | MnO (%) | Fe <sub>2</sub> O <sub>3</sub> (%) | Rb <sub>2</sub> O (ppm) | Cs <sub>2</sub> O (ppm) |
|---|-----------------------|----------------------|---------|---------|------------------------------------|-------------------------|-------------------------|
| <i>Yayal-Blue-on-White, Nueva Cadiz, Venezuela</i>        |                       |                      |         |         |                                    |                         |                         |
| SA31  | 1.19                  | 1.42                 | 0.040   | 0.097   | 5.0                                | 65                      | 6.0                     |
| SA60  | 1.01                  | 1.30                 | 0.039   | 0.082   | 4.7                                | 56                      | 4.5                     |
| SA61  | 1.47                  | 1.59                 | 0.043   | 0.088   | 5.0                                | 57                      | 4.1                     |
| SA62  | 1.18                  | 1.36                 | 0.045   | 0.083   | 4.8                                | 52                      | 4.4                     |
| SA63  | 1.50                  | 1.06                 | 0.042   | 0.081   | 5.2                                | 58                      | 6.1                     |
| SA64  | 1.15                  | 1.23                 | 0.038   | 0.075   | 4.8                                | 52                      | 5.2                     |
| SB46  | 1.28                  | 1.38                 | 0.044   | 0.079   | 5.0                                | 70                      | 6.3                     |
| SB47  | 1.47                  | 1.47                 | 0.044   | 0.077   | 5.1                                | 72                      | 5.8                     |
| SB48  | 0.73                  | 1.41                 | 0.042   | 0.079   | 4.8                                | 77                      | 5.7                     |
| SB49  | 1.16                  | 1.54                 | 0.037   | 0.074   | 5.1                                | 58                      | 5.7                     |
| SB50  | 1.20                  | 1.34                 | 0.039   | 0.082   | 5.1                                | 63                      | 6.4                     |
| SB51  | 1.13                  | 1.55                 | 0.050   | 0.077   | 5.2                                | 82                      | 6.4                     |
| SB52  | 1.10                  | 1.35                 | 0.067   | 0.083   | 6.3                                | 52                      | 6.1                     |
| SB53  | 1.56                  | 1.55                 | 0.045   | 0.081   | 4.9                                | 61                      | 5.9                     |
| SB54  | 1.08                  | 2.00                 | 0.037   | 0.082   | 5.3                                | 73                      | 5.7                     |
| SB55  | 1.01                  | 1.39                 | 0.041   | 0.068   | 4.5                                | 71                      | 5.5                     |
| SB56  | 1.11                  | 1.14                 | 0.050   | 0.078   | 5.0                                | 67                      | 6.0                     |
| SB57  | 1.38                  | 1.21                 | 0.041   | 0.070   | 4.6                                | 64                      | 5.5                     |
| <i>Yayal-Blue-on-White, Juandolio, Dominican Republic</i> |                       |                      |         |         |                                    |                         |                         |
| SA46  | 0.94                  | 1.63                 | 0.046   | 0.090   | 5.7                                | 84                      | 5.3                     |
| <i>Caparra Blue, Nueva Cadiz, Venezuela</i>               |                       |                      |         |         |                                    |                         |                         |
| SA27  | 0.88                  | 2.21                 | 0.037   | 0.034   | 5.2                                | 110                     | 6.1                     |
| SA28  | 1.48                  | 1.31                 | 0.037   | 0.077   | 5.6                                | 63                      | 6.0                     |
| SA29  | 1.52                  | 1.04                 | 0.044   | 0.084   | 5.2                                | 57                      | 7.0                     |
| SA30  | 1.29                  | 1.24                 | 0.040   | 0.076   | 5.1                                | 52                      | 6.1                     |
| <i>Isabella Polychrome, Nueva Cadiz, Venezuela</i>        |                       |                      |         |         |                                    |                         |                         |
| SA47  | 1.30                  | 1.64                 | 0.037   | 0.072   | 4.9                                | 85                      | 5.4                     |
| SA48  | 1.44                  | 1.59                 | 0.044   | 0.076   | 5.3                                | 76                      | 5.8                     |
| SA49  | 1.25                  | 1.54                 | 0.039   | 0.076   | 5.4                                | 70                      | 5.7                     |
| <i>Isabella Polychrome, Juandolio, Dominican Republic</i> |                       |                      |         |         |                                    |                         |                         |
| SA42  | 1.10                  | 1.21                 | 0.046   | 0.087   | 5.3                                | 123                     | 6.3                     |
| SA43  | 0.76                  | 1.80                 | 0.039   | 0.081   | 4.8                                | 78                      | 4.9                     |
| SA44  | 0.91                  | 1.79                 | 0.051   | 0.089   | 5.7                                | 106                     | 6.1                     |
| SA45  | 1.05                  | 1.63                 | 0.046   | 0.088   | 5.4                                | 122                     | 6.7                     |
| <i>Isabella Polychrome, Isabella, Dominican Republic</i>  |                       |                      |         |         |                                    |                         |                         |
| SA24  | 1.08                  | 1.72                 | 0.041   | 0.086   | 5.4                                | 62                      | 4.9                     |
| SA25  | 1.00                  | 2.11                 | 0.063   | 0.091   | 5.6                                | 67                      | 7.5                     |
| SA26  | 1.00                  | 1.69                 | 0.058   | 0.079   | 5.3                                | 103                     | 6.4                     |

Continued

| <i>La</i> <sub>2</sub> O <sub>3</sub> (ppm)               | <i>Sc</i> <sub>2</sub> O <sub>3</sub> (ppm) | <i>Ce</i> O <sub>2</sub> (ppm) | <i>Eu</i> <sub>2</sub> O <sub>3</sub> (ppm) | <i>Hf</i> O <sub>2</sub> (ppm) | <i>Th</i> O <sub>2</sub> (ppm) | <i>Cr</i> <sub>2</sub> O <sub>3</sub> (ppm) | <i>Co</i> O (ppm) |
|---|---|--------------------------------|---|--------------------------------|--------------------------------|---|-------------------|
| <i>Yayal-Blue-on-White, Nueva Cadiz, Venezuela</i>        |   |                                |   |                                |                                |   |                   |
| 38  | 19.9  | 83                             | 1.70  | 5.8                            | 12.2                           | 111   | 26.7              |
| 39  | 18.9  | 74                             | 1.44  | 4.6                            | 10.4                           | 109   | 14.6              |
| 46  | 20.3  | 98                             | 1.59  | 6.5                            | 14.5                           | 114   | 19.0              |
| 39  | 19.4  | 83                             | 1.51  | 6.4                            | 11.7                           | 112   | 14.7              |
| 38  | 20.8  | 82                             | 1.53  | 6.0                            | 12.3                           | 118   | 16.4              |
| 36  | 19.4  | 80                             | 1.39  | 4.8                            | 11.2                           | 114   | 14.6              |
| 36  | 20.5  | 76                             | 1.55  | 5.9                            | 11.6                           | 119   | 20.7              |
| 36  | 20.3  | 81                             | 1.67  | 5.8                            | 11.8                           | 120   | 17.1              |
| 35  | 19.7  | 74                             | 1.61  | 5.1                            | 11.5                           | 117   | 16.6              |
| 36  | 20.5  | 78                             | 1.51  | 5.9                            | 11.5                           | 117   | 20.2              |
| 38  | 20.6  | 85                             | 1.65  | 5.2                            | 12.6                           | 122   | 20.3              |
| 38  | 20.4  | 83                             | 1.69  | 5.2                            | 12.2                           | 130   | 17.1              |
| 38  | 22.3  | 84                             | 1.62  | 5.5                            | 12.6                           | 134   | 20.6              |
| 37  | 19.6  | 79                             | 1.40  | 5.5                            | 11.8                           | 126   | 16.2              |
| 40  | 21.7  | 82                             | 1.68  | 5.6                            | 12.0                           | 131   | 19.9              |
| 34  | 18.2  | 73                             | 1.38  | 5.1                            | 10.8                           | 108   | 17.8              |
| 38  | 20.9  | 83                             | 1.69  | 5.7                            | 12.1                           | 121   | 16.5              |
| 35  | 18.3  | 77                             | 1.25  | 5.9                            | 11.1                           | 111   | 16.8              |
| <i>Yayal-Blue-on-White, Juandolio, Dominican Republic</i> |   |                                |   |                                |                                |   |                   |
| 46  | 23.9  | 99                             | 1.79  | 6.4                            | 14.0                           | 131   | 17.3              |
| <i>Caparra Blue, Nueva Cadiz, Venezuela</i>               |   |                                |   |                                |                                |   |                   |
| 45  | 22.8  | 86                             | 1.74  | 5.4                            | 12.4                           | 147   | 71.0              |
| 43  | 21.3  | 90                             | 1.64  | 6.8                            | 12.0                           | 124   | 29.8              |
| 41  | 21.2  | 83                             | 1.55  | 5.8                            | 12.5                           | 119   | 72.6              |
| 39  | 20.5  | 80                             | 1.66  | 6.4                            | 12.1                           | 121   | 36.3              |
| <i>Isabella Polychrome, Nueva Cadiz, Venezuela</i>        |   |                                |   |                                |                                |   |                   |
| 38  | 19.9  | 84                             | 1.47  | 5.6                            | 11.6                           | 114   | 14.9              |
| 43  | 20.7  | 92                             | 1.59  | 6.0                            | 13.5                           | 118   | 17.1              |
| 40  | 20.4  | 84                             | 1.39  | 5.6                            | 11.7                           | 115   | 23.6              |
| <i>Isabella Polychrome, Juandolio, Dominican Republic</i> |   |                                |   |                                |                                |   |                   |
| 40  | 21.7  | 81                             | 1.51  | 5.4                            | 12.1                           | 119   | 17.4              |
| 41  | 19.4  | 83                             | 1.36  | 5.7                            | 12.0                           | 109   | 17.7              |
| 51  | 23.4  | 90                             | 1.70  | 6.2                            | 12.9                           | 126   | 16.5              |
| 44  | 22.2  | 89                             | 1.78  | 6.2                            | 12.4                           | 124   | 17.3              |
| <i>Isabella Polychrome, Isabella, Dominican Republic</i>  |   |                                |   |                                |                                |   |                   |
| 38  | 20.7  | 83                             | 1.70  | 5.9                            | 12.4                           | 111   | 17.0              |
| 41  | 22.9  | 90                             | 2.13  | 6.9                            | 13.9                           | 127   | 20.6              |
| 36  | 20.9  | 82                             | 1.58  | 6.4                            | 11.5                           | 119   | 18.0              |

Table II. Mean Compositions of Majolica from Five Sites Spain; and from

| Oxides Determined                                 | Average Concentrations |               |               |
|---|------------------------|---------------|---------------|
|   | Convento de S.F.       | Juandolio     | La Vega Vieja |
| Sodium (Na <sub>2</sub> O) (%)                    | 1.06 ± 0.20            | 0.92 ± 0.16   | 0.57 ± 0.09   |
| Potassium (K <sub>2</sub> O) (%)                  | 1.42 ± 0.35            | 1.76 ± 0.34   | 1.91 ± 0.22   |
| Barium (BaO) (%)                                  | 0.046 ± 0.008          | 0.045 ± 0.002 | 0.054 ± 0.006 |
| Manganese (MnO) (%)                               | 0.080 ± 0.016          | 0.085 ± 0.006 | 0.082 ± 0.04  |
| Iron (Fe <sub>2</sub> O <sub>3</sub> ) (%)        | 5.1 ± 0.36             | 5.3 ± 0.30    | 5.5 ± 0.38    |
| Rubidium (Rb <sub>2</sub> O) ppm                  | 87 ± 24                | 92 ± 15       | 99 ± 20       |
| Cesium (Cs <sub>2</sub> O) (ppm)                  | 5.4 ± 1.1              | 5.6 ± 0.8     | 5.1 ± 0.9     |
| Lanthanum (La <sub>2</sub> O <sub>3</sub> ) (ppm) | 39 ± 4.0               | 43 ± 4.0      | 42 ± 3.0      |
| Scandium (Sc <sub>2</sub> O <sub>3</sub> ) (ppm)  | 20 ± 1.3               | 22 ± 1.3      | 22 ± 1.9      |
| Cerium (CeO <sub>2</sub> ) (ppm)                  | 77 ± 18                | 87 ± 5.4      | 90 ± 5.5      |
| Europium (Eu <sub>2</sub> O <sub>3</sub> ) (ppm)  | 1.56 ± 0.15            | 1.66 ± 0.12   | 1.66 ± 0.13   |
| Hafnium (HfO <sub>2</sub> ) (ppm)                 | 5.9 ± 0.44             | 6.0 ± 0.6     | 6.2 ± 0.5     |
| Thorium (ThO <sub>2</sub> ) (ppm)                 | 11.8 ± 0.73            | 12.7 ± 1.0    | 13.0 ± 1.0    |
| Chromium (Cr <sub>2</sub> O <sub>3</sub> ) (ppm)  | 121 ± 10               | 119 ± 8       | 126 ± 8       |
| Cobalt (CoO) (ppm)                                | 18.0 ± 4.7             | 16.5 ± 1.2    | 15.9 ± 1.4    |

\* Since we believe the data are logarithmically distributed, the geometric means are used, and for convenience of presentation the standard deviations are expressed

Table III. Concentration of Oxides in

| Specimen No.               | Na <sub>2</sub> O (%) | K <sub>2</sub> O (%) | BaO (%) | MnO (%) | Fe <sub>2</sub> O <sub>3</sub> (%) | Rb <sub>2</sub> O (ppm) | Cs <sub>2</sub> O (ppm) |
|----------------------------|-----------------------|----------------------|---------|---------|------------------------------------|-------------------------|-------------------------|
| <i>Columbia Plain</i>      |                       |                      |         |         |                                    |                         |                         |
| SC08                       | 0.60                  | 2.19                 | 0.078   | 0.066   | 4.7                                | 99                      | 5.7                     |
| SC09                       | 0.68                  | 1.82                 | 0.060   | 0.050   | 4.9                                | 101                     | 7.7                     |
| <i>Yayal-Blue-on-White</i> |                       |                      |         |         |                                    |                         |                         |
| SC03                       | 0.53                  | 2.37                 | 0.058   | 0.056   | 4.4                                | 90                      | 6.0                     |
| SC04                       | 0.80                  | 2.12                 | 0.088   | 0.068   | 4.9                                | 106                     | 7.9                     |
| SC05                       | 0.61                  | 1.69                 | 0.069   | 0.057   | 4.9                                | 100                     | 7.7                     |
| SC06                       | 0.97                  | 1.68                 | 0.095   | 0.059   | 5.3                                | 80                      | 4.5                     |
| SC07                       | 0.60                  | 2.13                 | 0.101   | 0.063   | 5.0                                | 84                      | 4.9                     |

in the Dominican Republic and Venezuela; from Jerez, Mexico City, Mexico\*

| Oxides Determined                                 | Average Concentrations |               |               |               |
|---|------------------------|---------------|---------------|---------------|
|   | Isabela                | Nueva Cadiz   | Jerez         | Mexico City   |
| Sodium (Na <sub>2</sub> O) (%)                    | 1.12 ± 0.05            | 1.26 ± 0.28   | 0.67 ± 0.14   | 1.69 ± 0.42   |
| Potassium (K <sub>2</sub> O) (%)                  | 2.08 ± 0.25            | 1.35 ± 0.29   | 1.98 ± 0.27   | 1.13 ± 0.40   |
| Barium (BaO) (%)                                  | 0.053 ± 0.012          | 0.045 ± 0.011 | 0.077 ± 0.017 | 0.053 ± 0.010 |
| Manganese (MnO) (%)                               | 0.085 ± 0.007          | 0.077 ± 0.011 | 0.060 ± 0.015 | 0.054 ± 0.019 |
| Iron (Fe <sub>2</sub> O <sub>3</sub> ) (%)        | 5.5 ± 0.15             | 5.0 ± 0.34    | 4.9 ± 0.26    | 4.1 ± 0.59    |
| Rubidium (Rb <sub>2</sub> O) ppm                  | 76 ± 21                | 63 ± 12       | 94 ± 10       | 54 ± 11       |
| Cesium (Cs <sub>2</sub> O) (ppm)                  | 6.2 ± 1.4              | 5.9 ± 0.9     | 6.2 ± 1.4     | 3.7 ± 1.3     |
| Lanthanum (La <sub>2</sub> O <sub>3</sub> ) (ppm) | 43 ± 2.7               | 38 ± 2.7      | 45 ± 1.9      | 22 ± 3.9      |
| Scandium (Sc <sub>2</sub> O <sub>3</sub> ) (ppm)  | 21 ± 1.2               | 20 ± 1.2      | 20 ± 0.9      | 16.5 ± 2.8    |
| Cerium (CeO <sub>2</sub> ) (ppm)                  | 85 ± 4.6               | 82 ± 4.8      | 80 ± 3.3      | 40 ± 8.6      |
| Europium (Eu <sub>2</sub> O <sub>3</sub> ) (ppm)  | 1.79 ± 0.28            | 1.56 ± 0.13   | 1.47 ± 0.05   | 1.22 ± 0.21   |
| Hafnium (HfO <sub>2</sub> ) (ppm)                 | 6.4 ± 0.5              | 5.7 ± 0.5     | 5.5 ± 0.4     | 4.5 ± 0.8     |
| Thorium (ThO <sub>2</sub> ) (ppm)                 | 12.6 ± 1.2             | 11.8 ± 0.8    | 11.7 ± 0.7    | 5.6 ± 0.7     |
| Chromium (Cr <sub>2</sub> O <sub>3</sub> ) (ppm)  | 119 ± 8                | 119 ± 10      | 122 ± 24      | 98 ± 27       |
| Cobalt (CoO) (ppm)                                | 18.5 ± 1.8             | 18.8 ± 6.7    | 15.4 ± 1.8    | 16.6 ± 14     |

as plus or minus one half of the total standard deviation spread of the groups as calculated logarithmically.

Majolica Sherds from Jerez, Spain

| La <sub>2</sub> O <sub>3</sub> (ppm) | Sc <sub>2</sub> O <sub>3</sub> (ppm) | CeO <sub>2</sub> (ppm) | Eu <sub>2</sub> O <sub>3</sub> (ppm) | HfO <sub>2</sub> (ppm) | ThO <sub>2</sub> (ppm) | Cr <sub>2</sub> O <sub>3</sub> (ppm) | CoO (ppm) |
|--------------------------------------|--------------------------------------|------------------------|--------------------------------------|------------------------|------------------------|--------------------------------------|-----------|
| <i>Columbia Plain</i>                |                                      |                        |                                      |                        |                        |                                      |           |
| 45                                   | 19.4                                 | 80                     | 1.55                                 | 5.0                    | 11.5                   | 111                                  | 15.2      |
| 44                                   | 19.0                                 | 81                     | 1.48                                 | 5.4                    | 11.0                   | 103                                  | 16.9      |
| <i>Yayal-Blue-on-White</i>           |                                      |                        |                                      |                        |                        |                                      |           |
| 42                                   | 18.4                                 | 76                     | 1.37                                 | 5.1                    | 11.4                   | 101                                  | 15.1      |
| 46                                   | 20.5                                 | 82                     | 1.49                                 | 6.1                    | 12.5                   | 127                                  | 14.9      |
| 44                                   | 20.0                                 | 80                     | 1.49                                 | 5.7                    | 12.1                   | 129                                  | 13.0      |
| 44                                   | 20.8                                 | 77                     | 1.46                                 | 5.3                    | 10.9                   | 180                                  | 14.8      |
| 48                                   | 20.6                                 | 86                     | 1.47                                 | 5.7                    | 12.5                   | 116                                  | 18.8      |

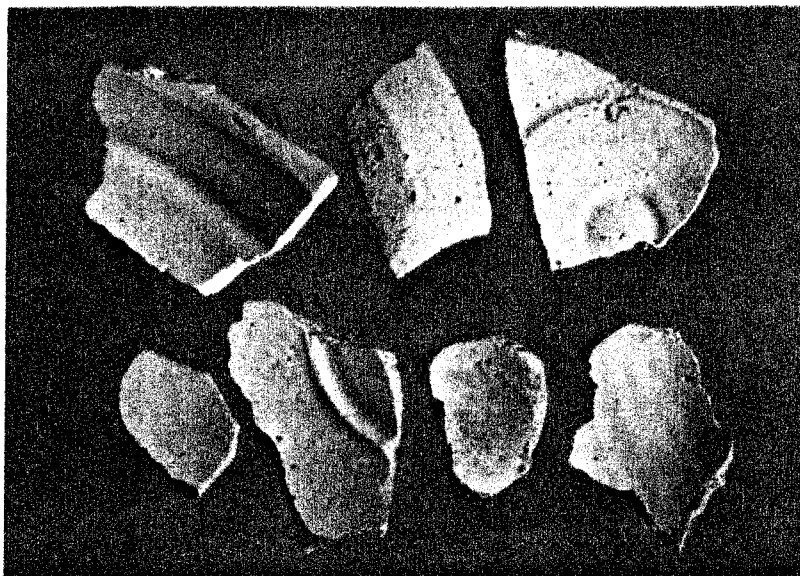


Figure 5. Columbia Plain (left to right): (top) SC11, SC12, and SC14; (bottom) SC15, SC18, SC20, and SC21 from excavations from the Metropolitan Cathedral in Mexico City



Figure 6. Abo Polychrome (left to right): (top) ST78-SA80; (bottom) SA81, San Luis Polychrome; SA82 and SA83 from the subway excavations in Mexico City

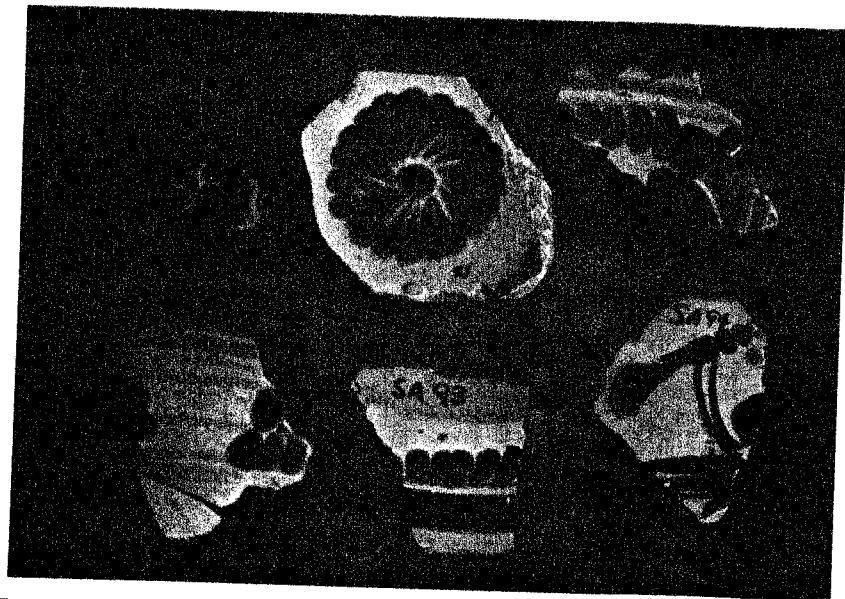


Figure 7. Puebla-Blue-on-White (left to right): (top) SA89-SA91; (bottom) SA92-SA94; San Elizario Polychrome from the subway excavations in Mexico City

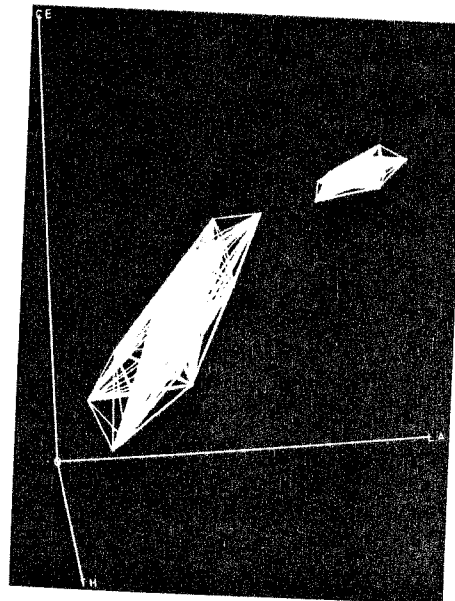


Figure 8. Computer projection of three-dimensional plot of cerium, lanthanum, and thorium oxide concentrations for Spanish Colonial majolica sherds. The data divides into a group having its source in Spain and a group whose source is Mexican, as discussed in the text.

these divergencies to the high probability that barium carbonate might be either deposited in or leached from sherds during burial along with calcium carbonate. Accordingly, we usually do not regard deviant barium values as being very significant. This agreement strongly indicates that the matching group of New World sherds and those found at Jerez came from related sources. Since it is historically and archaeologically very probable that these specimens found in the New World came from Spain and very improbable that the Jerez specimens were imported from the New World, a Spanish source, as postulated by Goggin and later authors, gains additional support from these data.

The data in Table IV are for sherds from the excavations of the Metro and the Metropolitan Cathedral in Mexico City. Approximately 100,000 majolica sherds were uncovered during the excavations for the Metro subway, and we have analyzed a small group of these. Recent excavations have been carried out at the Metropolitan Cathedral in mine-like tunnels and shafts as much as 40 ft below the cathedral floor. These excavations are below the central cathedral foundation which church records state was laid in 1573. There is no evidence of intrusions through this floor so that the excavated sherds can be dated as earlier than 1573. The compositions of these sherds can be distinguished readily from those of the sherds in Tables I and III. The lanthanum oxide, cerium oxide, and thorium oxide concentrations are the most distinctly different. Selected examples of the sherds in Table IV are shown in Figures 5-7. The sherds in Figure 5 are examples of Columbia Plain from the excavations of the Metropolitan Cathedral and those in Figures 6 and 7 are from the subway excavations.

In Figure 8 we have plotted the lanthanum oxide, cerium oxide, and thorium oxide concentrations for sherds excavated in the Dominican Republic and Venezuela and sherds from the Metro excavations using a computer system developed for this purpose at Brookhaven National Laboratory (8). On the basis of these three oxides there is a distinct separation between the sherds from the Dominican Republic and Venezuela and those from Mexico City. Unlike the sherds from the Dominican Republic and Venezuela, the sherds from Mexico City appear not to have originated in Spain, at least at that specific source. There is further evidence of this distinction between the two sets of sherds. X-ray diffraction analysis of the samples from Jerez and from the New World showed that the sherds from Jerez, the Dominican Republic, and Venezuela had intense quartz peaks whereas the sherds from Mexico City did not. This constitutes additional evidence that the majolica from Mexico City came from a different source than the majolica from the Caribbean sites.

The development of the ceramic industry in Mexico and its precedents in Italy and Spain have been discussed by Florence and Robert

Lister (9, 10). They and Gonzalo Lopez Cervantes (11) also refer to records of early pottery production in Mexico. There is, however, no complete record of the early history of the manufacture of ceramics by the Spanish in the New World, and the analyses discussed here will be used to better understand that history.

It is logical to consider whether the majolica sherds which were found in Mexico City could have been fabricated of local clay. Fortunately data on clays and related pottery from the Valley of Mexico has been collected at Brookhaven National Laboratory over many years. The ceramic material, which had previously been analyzed by Harbottle and Sayre in collaboration with other investigators, consisted of Precolumbian artifacts. The pottery and the clays from two archaeological sites within the Valley, Teotihuacan, and Tlatilco were all basically similar in composition, although the clays and pottery from the two separate sites could be differentiated through a subtle multivariate statistical analysis. It is likely that the entire Valley of Mexico is underlain with clay bed of moderately uniform trace impurity composition, and hence if the composition of the Mexico City majolica sherds was similar to that of ceramics and clay from Teotihuacan or Tlatilco, it would be probable that the majolica was fabricated from clays originating somewhere within the Valley of Mexico.

The data accumulated on clays and pottery from Teotihuacan are extensive, but only a few specimens from Tlatilco have been analyzed. Accordingly comparison was first made between the majolica and the Teotihuacan statistical group of analyses. The mean concentrations for these two groups of pottery are compared in Table V. Except for only two components, the calcium and cesium compounds, the concentrations of all measured components in the majolica were about three quarters of the levels in the Teotihuacan specimens. The components cesium oxide and calcium carbonate are among those that can be most readily affected during burial; cesium compounds tend to be water soluble and hence susceptible to migration through the action of soil water, and calcium carbonate can either be dissolved from or deposited into burial sherds depending upon the levels of carbon dioxide in the ground water to which they are exposed. Another cause of aberrant calcium concentrations in pottery is the occasional addition to the clay of crushed marble or other calcareous material as a temper. Therefore the difference between the calcium and cesium concentrations in the majolica and Precolumbian specimens does not rule out the possibility that they may have been formed from related clays, and the parallelism in the concentration of all other components in both sets of specimens suggests that they indeed might have had related common origins.



Table IV. Concentrations of Oxides in Matching

| Specimen No.   | Na <sub>2</sub> O (%) | K <sub>2</sub> O (%) | BaO (%) | MnO (%) | Fe <sub>2</sub> O <sub>3</sub> (%) | Rb <sub>2</sub> O (ppm) | Cs <sub>2</sub> O (ppm) |
|--|-----------------------|----------------------|---------|---------|------------------------------------|-------------------------|-------------------------|
| <i>Columbia Plain, Metropolitan Cathedral</i>                  |                       |                      |         |         |                                    |                         |                         |
| SC11   | 1.93                  | 1.61                 | 0.054   | 0.027   | 3.3                                | 59                      | 4.0                     |
| SC12   | 1.76                  | 1.60                 | 0.058   | 0.055   | 3.8                                | 51                      | 5.3                     |
| SC13   | 1.96                  | 1.26                 | 0.046   | 0.040   | 3.1                                | 44                      | 2.0                     |
| SC14   | 2.29                  | 2.02                 | 0.058   | 0.054   | 4.0                                | 80                      | 5.5                     |
| SC15   | 1.80                  | 0.72                 | 0.045   | 0.055   | 4.0                                | 50                      | 4.1                     |
| SC17   | 1.14                  | 0.98                 | 0.050   | 0.039   | 3.0                                | 42                      | 3.0                     |
| SC18   | 1.81                  | 2.00                 | 0.057   | 0.042   | 3.8                                | 73                      | 6.5                     |
| SC20   | 3.10                  | 1.75                 | 0.058   | 0.025   | 3.2                                | 62                      | 3.0                     |
| SC21   | 1.35                  | 1.26                 | 0.047   | 0.035   | 4.3                                | 67                      | 5.0                     |
| <i>Fig Springs, Polychrome, Metro Excavations, Mexico City</i> |                       |                      |         |         |                                    |                         |                         |
| SA65   | 1.64                  | 1.04                 | 0.058   | 0.038   | 3.8                                | 43                      | 2.7                     |
| SA66   | 1.46                  | 1.57                 | 0.043   | 0.039   | 5.3                                | 55                      | 4.6                     |
| SA67   | 1.38                  | 1.66                 | 0.052   | 0.047   | 3.6                                | 75                      | 3.9                     |
| <i>Los Angeles Polychrome, Metro Excavations, Mexico City</i>  |                       |                      |         |         |                                    |                         |                         |
| SA68   | 1.90                  | 0.93                 | 0.042   | 0.050   | 5.1                                | 57                      | 7.7                     |
| SA69   | 1.33                  | 1.67                 | 0.043   | 0.038   | 4.6                                | 80                      | —                       |
| SA70   | 1.12                  | 1.11                 | 0.052   | 0.038   | 3.9                                | 53                      | 7.0                     |
| <i>San-Luis-Blue-on-White, Metro Excavations, Mexico City</i>  |                       |                      |         |         |                                    |                         |                         |
| SA71   | 1.65                  | —                    | 0.042   | 0.052   | 5.0                                | 82                      | 4.0                     |
| SA72   | 2.40                  | 0.98                 | 0.062   | 0.099   | 4.5                                | 52                      | 3.0                     |
| SA73   | 1.20                  | 1.60                 | 0.044   | 0.052   | 4.5                                | 59                      | 5.4                     |
| <i>Puebla Polychrome, Metro Excavations, Mexico City</i>       |                       |                      |         |         |                                    |                         |                         |
| SA74   | 2.08                  | 0.75                 | 0.058   | 0.072   | 4.3                                | 43                      | 2.8                     |
| SA75   | 1.77                  | 1.62                 | 0.057   | 0.077   | 4.0                                | 53                      | 2.8                     |
| SA76   | 1.85                  | 2.05                 | 0.054   | 0.075   | 4.2                                | 40                      | 2.2                     |
| SA77   | 1.99                  | 1.06                 | 0.095   | 0.055   | 4.3                                | 57                      | 4.1                     |
| <i>Abo Polychrome, Metro Excavations, Mexico City</i>          |                       |                      |         |         |                                    |                         |                         |
| SA78   | 1.12                  | 1.14                 | 0.046   | 0.062   | 4.6                                | 55                      | —                       |
| SA79   | 1.44                  | 0.71                 | 0.061   | 0.071   | 3.8                                | 44                      | 3.2                     |
| SA80   | 1.73                  | 0.91                 | 0.058   | 0.083   | 4.1                                | 58                      | 2.6                     |
| SA81   | 1.60                  | 1.33                 | 0.064   | 0.086   | 4.0                                | 48                      | 2.2                     |
| <i>San Luis Polychrome, Metro Excavations, Mexico City</i>     |                       |                      |         |         |                                    |                         |                         |
| SA82   | 1.97                  | 1.20                 | 0.052   | 0.069   | 4.4                                | 53                      | 2.9                     |

Specimens of Majolica from Mexico City Sites

| La <sub>2</sub> O <sub>3</sub> (ppm)                           | Sc <sub>2</sub> O <sub>3</sub> (ppm) | CeO <sub>2</sub> (ppm) | Eu <sub>2</sub> O <sub>3</sub> (ppm) | HfO <sub>2</sub> (ppm) | ThO <sub>2</sub> (ppm) | Cr <sub>2</sub> O <sub>3</sub> (ppm) | CoO (ppm) |
|--|--------------------------------------|------------------------|--------------------------------------|------------------------|------------------------|--------------------------------------|-----------|
| <i>Columbia Plain, Metropolitan Cathedral</i>                  |                                      |                        |                                      |                        |                        |                                      |           |
| 36   | 19.5                                 | 57                     | 1.68                                 | 4.3                    | 8.3                    | 81                                   | 8.8       |
| 26   | 15.3                                 | 42                     | 1.16                                 | 4.1                    | 6.2                    | 71                                   | 12.0      |
| 18   | 11.3                                 | 27                     | 0.82                                 | 3.4                    | 4.9                    | 62                                   | 7.4       |
| 29   | 17.1                                 | 45                     | 1.26                                 | 4.2                    | 6.4                    | 76                                   | 12.4      |
| 23   | 15.9                                 | 37                     | 1.20                                 | 3.9                    | 5.2                    | 87                                   | 12.7      |
| 17   | 11.3                                 | 27                     | 0.86                                 | 2.9                    | 3.7                    | 60                                   | 10.3      |
| 28   | 15.7                                 | 43                     | 1.32                                 | 4.3                    | 6.4                    | 77                                   | 13.3      |
| 22   | 11.9                                 | 34                     | 0.96                                 | 3.9                    | 6.0                    | 59                                   | 9.2       |
| 25   | 12.3                                 | 39                     | 1.09                                 | 4.3                    | 6.6                    | 72                                   | 13.5      |
| <i>Fig Springs, Polychrome, Metro Excavations, Mexico City</i> |                                      |                        |                                      |                        |                        |                                      |           |
| 28   | 17.9                                 | 48                     | 1.51                                 | 4.7                    | 6.3                    | 78                                   | 9.7       |
| 25   | 22.0                                 | 41                     | 1.25                                 | 6.3                    | 5.5                    | 135                                  | 11.0      |
| 18   | 16.3                                 | 26                     | 1.07                                 | 4.7                    | 5.0                    | 86                                   | 36.0      |
| <i>Los Angeles Polychrome, Metro Excavations, Mexico City</i>  |                                      |                        |                                      |                        |                        |                                      |           |
| 22   | 20.7                                 | 35                     | 1.37                                 | 5.3                    | 5.9                    | 153                                  | 13.4      |
| 22   | 20.4                                 | 35                     | 1.22                                 | 5.7                    | 6.6                    | 123                                  | 15.7      |
| 19   | 17.8                                 | 31                     | 1.10                                 | 4.7                    | 5.1                    | 96                                   | 10.7      |
| <i>San-Luis-Blue-on-White, Metro Excavations, Mexico City</i>  |                                      |                        |                                      |                        |                        |                                      |           |
| 23   | 21.9                                 | 38                     | 1.31                                 | 5.8                    | 6.1                    | 112                                  | 14.4      |
| 26   | 16.6                                 | 62                     | 1.42                                 | 5.7                    | 7.0                    | 82                                   | 21.9      |
| 19   | 19.2                                 | 31                     | 1.10                                 | 4.9                    | 5.4                    | 114                                  | 12.3      |
| <i>Puebla Polychrome, Metro Excavations, Mexico City</i>       |                                      |                        |                                      |                        |                        |                                      |           |
| 123  | 16.0                                 | 50                     | 1.42                                 | 5.7                    | 5.5                    | 92                                   | 15.3      |
| 24   | 15.0                                 | 52                     | 1.32                                 | 5.1                    | 6.0                    | 82                                   | 13.8      |
| 23   | 15.4                                 | 50                     | 1.38                                 | 5.8                    | 5.6                    | 102                                  | 16.0      |
| 21   | 17.8                                 | 44                     | 1.33                                 | 5.0                    | 5.4                    | 133                                  | 20.8      |
| <i>Abo Polychrome, Metro Excavations, Mexico City</i>          |                                      |                        |                                      |                        |                        |                                      |           |
| 17.5   | 17.8                                 | 26                     | 1.01                                 | 4.9                    | 5.6                    | 116                                  | 11.3      |
| 21.9   | 14.0                                 | 43                     | 1.21                                 | 4.8                    | 5.0                    | 85                                   | 13.6      |
| 21.2   | 16.4                                 | 44                     | 1.33                                 | 4.8                    | 5.6                    | 85                                   | 13.0      |
| 21.6   | 15.1                                 | 45                     | 1.33                                 | 4.9                    | 5.4                    | 83                                   | 13.5      |
| <i>San Luis Polychrome, Metro Excavations, Mexico City</i>     |                                      |                        |                                      |                        |                        |                                      |           |
| 24.2   | 17.2                                 | 52                     | 1.47                                 | 5.6                    | 5.2                    | 95                                   | 14.5      |

Table IV.

| Specimen No.   | Na <sub>2</sub> O (%) | K <sub>2</sub> O (%) | BaO (%) | MnO (%) | Fe <sub>2</sub> O <sub>3</sub> (%) | Rb <sub>2</sub> O (ppm) | Cs <sub>2</sub> O (ppm) |
|--|-----------------------|----------------------|---------|---------|------------------------------------|-------------------------|-------------------------|
| <i>Castillo Polychrome, Metro Excavations, Mexico City</i>       |                       |                      |         |         |                                    |                         |                         |
| SA84   | 1.15                  | 1.26                 | 0.047   | 0.052   | 3.7                                | 51                      | 6.4                     |
| SA85   | 1.95                  | 0.47                 | 0.048   | 0.036   | 5.4                                | 50                      | 4.1                     |
| <i>Aucilla Polychrome, Metro Excavations, Mexico City</i>        |                       |                      |         |         |                                    |                         |                         |
| SA86   | 1.75                  | 0.76                 | 0.066   | 0.092   | 4.8                                | 44                      | 3.2                     |
| SA87   | 1.83                  | 0.87                 | 0.064   | 0.103   | 4.2                                | 47                      | 2.9                     |
| SA88   | 2.13                  | 0.72                 | 0.062   | 0.095   | 4.5                                | 45                      | 2.7                     |
| <i>Puebla-Blue-on-White, Metro Excavations, Mexico City</i>      |                       |                      |         |         |                                    |                         |                         |
| SA90   | 1.63                  | 1.42                 | 0.038   | 0.049   | 3.7                                | 52                      | 3.5                     |
| SA91   | 2.04                  | 1.66                 | 0.042   | 0.048   | 3.2                                | 59                      | 3.0                     |
| <i>San Elizario Polychrome, Metro Excavations, Mexico City</i>   |                       |                      |         |         |                                    |                         |                         |
| SA93   | 1.77                  | 0.70                 | 0.059   | 0.046   | 4.7                                | 51                      | 3.4                     |
| SA94   | 1.85                  | 0.95                 | 0.057   | 0.066   | 4.5                                | 53                      | 3.6                     |
| <i>San-Agustin-Blue-on-White, Metro Excavations, Mexico City</i> |                       |                      |         |         |                                    |                         |                         |
| SA95   | 2.14                  | 0.56                 | 0.062   | 0.058   | 4.9                                | 30                      | 2.5                     |
| SA96   | 2.14                  | 0.99                 | 0.060   | 0.028   | 4.4                                | 54                      | 3.3                     |
| <i>Huejotzingo-Blue-on-White, Metro Excavations, Mexico City</i> |                       |                      |         |         |                                    |                         |                         |
| SA99   | 2.09                  | 0.94                 | 0.062   | 0.070   | 4.3                                | 59                      | 4.1                     |
| SB01   | 1.77                  | 1.06                 | 0.042   | 0.053   | 3.9                                | 55                      | 4.9                     |
| <i>Unidentified Types, Metro Excavations, Mexico City</i>        |                       |                      |         |         |                                    |                         |                         |
| SA97   | 1.94                  | 1.36                 | 0.035   | 0.060   | 3.7                                | 56                      | 3.6                     |
| SA98   | 1.77                  | 1.11                 | 0.055   | 0.056   | 4.3                                | 64                      | 3.2                     |
| SB26   | 1.60                  | 1.22                 | 0.075   | 0.070   | 3.3                                | —                       | 2.9                     |
| SB32   | 0.74                  | 1.24                 | 0.055   | 0.095   | 3.7                                | 63                      | 3.5                     |
| SB36   | 1.62                  | 0.92                 | 0.038   | 0.055   | 4.9                                | 59                      | 9.4                     |
| SB37   | 1.49                  | 1.09                 | 0.056   | 0.040   | 3.5                                | 35                      | —                       |

The high calcium content in the majolica found in Mexico City—21.4% calculated as pure calcium carbonate compared with 5.9% in sherds of Teotihuacan—suggests that a calcium compound such as calcium or calcium magnesium carbonate may have been added to the majolica either as a temper or through deposition during burial. Petrographic examination of cross sections of representative Mexico City majolica sherds show heavy deposits of birefringent material with structures

Continued

| La <sub>2</sub> O <sub>3</sub> (ppm)                             | Sc <sub>2</sub> O <sub>3</sub> (ppm) | CeO <sub>2</sub> (ppm) | Eu <sub>2</sub> O <sub>3</sub> (ppm) | HfO <sub>2</sub> (ppm) | ThO <sub>2</sub> (ppm) | Cr <sub>2</sub> O <sub>3</sub> (ppm) | CoO (ppm) |
|--|--------------------------------------|------------------------|--------------------------------------|------------------------|------------------------|--------------------------------------|-----------|
| <i>Castillo Polychrome, Metro Excavations, Mexico City</i>       |                                      |                        |                                      |                        |                        |                                      |           |
| 16.2   | 15.3                                 | 31                     | 1.02                                 | 3.6                    | 5.2                    | 93                                   | 10.8      |
| 23.3   | 21.4                                 | 34                     | 1.36                                 | 5.0                    | 6.4                    | 156                                  | 12.4      |
| <i>Aucilla Polychrome, Metro Excavations, Mexico City</i>        |                                      |                        |                                      |                        |                        |                                      |           |
| 26.8   | 18.1                                 | 56                     | 1.60                                 | 5.1                    | 6.5                    | 96                                   | 16.5      |
| 24.8   | 15.8                                 | 50                     | 1.41                                 | 4.6                    | 5.5                    | 92                                   | 14.9      |
| 25.9   | 17.2                                 | 54                     | 1.50                                 | 5.1                    | 5.7                    | 105                                  | 16.0      |
| <i>Puebla-Blue-on-White, Metro Excavations, Mexico City</i>      |                                      |                        |                                      |                        |                        |                                      |           |
| 18.0   | 14.2                                 | 36                     | 1.04                                 | 3.5                    | 5.2                    | 94                                   | 12.2      |
| 17.2   | 11.5                                 | 28                     | 0.94                                 | 3.5                    | 4.6                    | 79                                   | 119.0     |
| <i>San Elizario Polychrome, Metro Excavations, Mexico City</i>   |                                      |                        |                                      |                        |                        |                                      |           |
| 20.7   | 18.3                                 | 40                     | 1.30                                 | 4.7                    | 5.2                    | 138                                  | 17.3      |
| 21.9   | 18.2                                 | 38                     | 1.36                                 | 4.2                    | 5.5                    | 129                                  | 16.5      |
| <i>San-Agustin-Blue-on-White, Metro Excavations, Mexico City</i> |                                      |                        |                                      |                        |                        |                                      |           |
| 20.3   | 20.1                                 | 38                     | 1.47                                 | 4.7                    | 5.4                    | 168                                  | 16.7      |
| 24.5   | 19.4                                 | 40                     | 1.37                                 | 4.8                    | 6.4                    | 148                                  | 38.0      |
| <i>Huejotzingo-Blue-on-White, Metro Excavations, Mexico City</i> |                                      |                        |                                      |                        |                        |                                      |           |
| 21.6   | 17.8                                 | 38                     | 1.36                                 | 4.3                    | 5.9                    | 118                                  | 25.5      |
| 19.4   | 15.8                                 | 36                     | 1.07                                 | 4.3                    | 5.7                    | 92                                   | 13.5      |
| <i>Unidentified Types, Metro Excavations, Mexico City</i>        |                                      |                        |                                      |                        |                        |                                      |           |
| 16.2   | 15.7                                 | 31                     | 1.04                                 | 3.4                    | 5.0                    | 105                                  | 190.0     |
| 18.1   | 17.6                                 | 36                     | 1.11                                 | 3.9                    | 5.6                    | 120                                  | 451.0     |
| 16.3   | 13.8                                 | 39                     | 1.07                                 | 3.2                    | 4.5                    | 86                                   | 11.1      |
| 15.7   | 15.2                                 | 40                     | 0.84                                 | 3.4                    | 5.2                    | 78                                   | 11.1      |
| 17.7   | 20.4                                 | 43                     | 1.24                                 | 4.3                    | 6.0                    | 180                                  | 13.9      |
| 20.5   | 16.4                                 | 41                     | 1.40                                 | 3.6                    | 5.6                    | 95                                   | 9.9       |

typical of secondary accumulations of carbonates lining the open spaces within the pottery structure. X-ray diffraction of samples from these specimens confirmed the presence of the mineral calcite within them. Inclusion of primary mineral calcite, that is, of crushed marble or the like added as temper, would have quite a different microscopic appearance, and little or no microscopic evidence of calcite added as a temper appears in the majolica specimens.

Table V. Comparison of Composition of Mexican Majolica with Precolumbian Ceramics

| Compounds  | Oxide Concentration |                                     | Ratio<br>Majolica/<br>Teotihuacan |
|--|---------------------|-------------------------------------|-----------------------------------|
|  | Majolica            | Teotihuacan<br>Clays and<br>Pottery |                                   |
| <i>Major Components (%)</i>  |                     |                                     |                                   |
| Na <sub>2</sub> O  | 1.69                | 2.42                                | 0.70                              |
| K <sub>2</sub> O   | 1.13                | 1.55                                | 0.73                              |
| BaO  | 0.053               | 0.078                               | 0.68                              |
| MnO  | 0.054               | 0.085                               | 0.64                              |
| Fe <sub>2</sub> O <sub>3</sub>   | 4.09                | 5.36                                | 0.76                              |
| CaO<br>Expressed as<br>CaCO <sub>3</sub>                               | 21.4                | 5.9                                 | 3.63                              |
| <i>Trace Components (ppm)</i>  |                     |                                     |                                   |
| Rb <sub>2</sub> O  | 53.6                | 59.1                                | 0.91                              |
| Sc <sub>2</sub> O <sub>3</sub>   | 16.5                | 20.4                                | 0.81                              |
| La <sub>2</sub> O <sub>3</sub>   | 21.6                | 25.6                                | 0.84                              |
| CeO <sub>2</sub>   | 39.4                | 55.0                                | 0.72                              |
| Eu <sub>2</sub> O <sub>3</sub>   | 1.2                 | 1.7                                 | 0.71                              |
| HfO <sub>2</sub>   | 4.5                 | 5.1                                 | 0.88                              |
| ThO <sub>2</sub>   | 5.6                 | 6.9                                 | 0.81                              |
| Ta <sub>2</sub> O <sub>5</sub>   | 0.8                 | 1.0                                 | 0.80                              |
| Cr <sub>2</sub> O <sub>3</sub>   | 97.9                | 109.2                               | 0.90                              |
| CoO  | 16.6                | 19.5                                | 0.85                              |
| Cs <sub>2</sub> O  | 3.8                 | 2.8                                 | 1.36                              |
| Mean ratio with CaCO <sub>3</sub> and Cs <sub>2</sub> O values deleted |                     |                                     | 0.78 ± 0.08                       |

Petrographic comparison of the Mexico City majolica with Teotihuacan sherds shows that except for the secondary deposition of carbonates, which is present in the majolica but absent in the Precolumbian sherds, the mineral composition of both sets of specimens is very similar. Both notably include hornblende and similar feldspars as inclusions, and both are low in quartz. Similarly, except for the calcite in the majolica, both sets of sherds show similar x-ray diffraction patterns. The mineralogical evidence, therefore, strongly suggests that both sets of sherds were made from closely related clays and that the compositional differences that exist between them are primarily the result of the accumulation of a secondary calcareous deposit within the majolica sherds during burial in the wet soil of Mexico City.

One would indeed expect the soil in central Mexico City, which to a great extent is the filled-in bed of Lake Texcoco, to be moist and hence conducive to carbonate deposition and the soil at Teotihuacan, which is

situated on relatively high ground at the northern edge of the Valley of Mexico, to be dry and hence not favorable for such deposition. The neutron activation analytical data are consistent with this hypothesis. If the secondary carbonate deposit were relatively free of the other elements determined, then its presence would simply dilute the concentrations of these other components by a constant factor. Table V indicates that the concentrations of 15 components in the majolica are related on the average to those in the Teotihuacan specimens by the nearly constant factor  $0.78 \pm 0.08$ . Therefore on the basis of relative rather than absolute concentrations there should be good agreement between the two groups of specimens.

The Brookhaven computer program ADSTAT can adjust sets of specimens by factors which bring them into closest relative agreement on the basis of a least squares fit in logarithms of concentrations. Using this program all specimens of both the Mexico City majolica and Teotihuacan were adjusted into best-relative-fit agreement with the mean concentrations of the majolica. In this adjustment the elements calcium, cesium, and cobalt were eliminated; calcium and cesium because of their previously noted inconsistency with other components, and cobalt because some majolica specimens were decorated with cobalt-colored glazes and there was some evidence of occasional contamination from these glazes. In this process, a fitting constant was calculated for each specimen which was used to adjust all components for that specimen. Hence the relative values for each specimen are left unaltered in the adjustment. If one then assumes that the adjusted Teotihuacan specimens constitute a log-normally distributed statistical group, it is possible to calculate the multivariate probability that each of the adjusted specimens might belong to this group. This is a so-called Mahalanobis distance calculation corrected for a finite group of specimens through Hotelling's  $T^2$  parameter.

Such a calculation showed that about half of the majolica specimens had a significant probability (i.e., greater than 5%) of belonging to the Teotihuacan group. This is a very sensitive test of agreement of specimens with a group which demonstrates that on a relative basis, for all elements other than calcium, cesium, and cobalt, there is a close agreement between the compositions of the two sets of specimens. We do not feel that this agreement proves that any or all of the majolica specimens were necessarily fabricated at Teotihuacan itself. It is historically unlikely that this would have occurred. It is more likely that the majolica was made from clays of a geological origin similar to those at Teotihuacan which in some instances are indistinguishable from Teotihuacan clays in their trace impurity patterns.

The usefulness of neutron activation analysis in assisting the archaeologist to establish the provenience of potsherd material is illustrated by

a consideration of the sherds in Figure 9. SB30 and SB31 were among the majolica sherds excavated in the Mexico City subway excavations. They have been identified by F. Lister (2) as late 16th century Italian. In Table VI the concentration of several oxides in these sherds are markedly different from their mean values in the majolica now considered to be of Mexican origin. (SB30 and SB31 have much higher concentrations of  $\text{CeO}_2$ ,  $\text{ThO}_2$ , and  $\text{Cr}_2\text{O}_3$  than most of the pottery from Mexico City.) Although F. Lister thought that SB33, also in Figure 9, was of Mexican origin, it also has a distinctive composition, having a much higher concentration of  $\text{HfO}_2$ .

Table VI. Nonmatching Majolica Sherds from

| Specimen No.  | $\text{Na}_2\text{O}$ (%) | $\text{K}_2\text{O}$ (%) | $\text{BaO}$ (%) | $\text{MnO}$ (%) | $\text{Fe}_2\text{O}_3$ (%) | $\text{Rb}_2\text{O}$ (ppm) | $\text{Cs}_2\text{O}$ (ppm) |
|---|---------------------------|--------------------------|------------------|------------------|-----------------------------|-----------------------------|-----------------------------|
| <i>Subway Excavations, Mexico City</i>                                      |                           |                          |                  |                  |                             |                             |                             |
| SB30  | 1.20                      | 1.97                     | 0.038            | 0.101            | 6.8                         | 108                         | 7.9                         |
| SB31  | 1.16                      | 2.31                     | 0.053            | 0.113            | 6.7                         | 88                          | 6.8                         |
| SB33  | 1.39                      | 2.41                     | 0.065            | 0.044            | 4.3                         | 88                          | 7.9                         |
| <i>Santiago de los Caballeros (Antigua), Guatemala</i>                      |                           |                          |                  |                  |                             |                             |                             |
| SB38  | 1.51                      | 0.92                     | 0.058            | 0.198            | 11.4                        | 44                          | 3.4                         |
| SB39  | 1.57                      | 0.58                     | 0.062            | 0.213            | 11.5                        | 40                          | 2.6                         |
| <i>Panama Viejo</i>   |                           |                          |                  |                  |                             |                             |                             |
| SB40  | 2.09                      | 2.94                     | 0.082            | 0.142            | 7.3                         | 151                         | 33.2                        |
| SB41  | 2.07                      | 2.93                     | 0.062            | 0.104            | 7.7                         | 146                         | 40.1                        |
| SB42  | 2.09                      | 2.40                     | 0.074            | 0.137            | 7.5                         | 193                         | 127.0                       |
| <i>Cuzco, Peru</i>  |                           |                          |                  |                  |                             |                             |                             |
| SB43  | 0.54                      | 3.97                     | 0.075            | 0.092            | 7.4                         | 128                         | 9.2                         |
| SB44  | 0.66                      | 4.46                     | 0.060            | 0.072            | 6.7                         | 137                         | 8.3                         |
| <i>Quito, Ecuador</i>   |                           |                          |                  |                  |                             |                             |                             |
| SB45  | 3.33                      | 1.49                     | 0.098            | 0.114            | 7.0                         | 54                          | 2.8                         |
| <i>Metropolitan Cathedral, Mexico City</i>                                  |                           |                          |                  |                  |                             |                             |                             |
| SC10  | 0.90                      | 2.57                     | 0.048            | 0.054            | 4.1                         | 146                         | 11.0                        |
| SC19  | 0.93                      | 3.35                     | 0.069            | 0.037            | 4.2                         | 167                         | 11.4                        |
| <i>Means and Means + and -95% Confidence Limit for Mexico City Majolica</i> |                           |                          |                  |                  |                             |                             |                             |
| Means   | 1.69                      | 1.13                     | 0.054            | 0.054            | 4.1                         | 54                          | 3.7                         |
| +0.05 limit   | 2.74                      | 2.24                     | 0.077            | 0.109            | 5.4                         | 81                          | 7.41                        |
| -0.05 limit   | 1.04                      | 0.57                     | 0.036            | 0.027            | 3.1                         | 36                          | 1.84                        |

We have analyzed two sherds from Guatemala, three sherds from Panama Vieja, two sherds from Cuzco, Peru, and one sherd from Quito, Ecuador. These sherds are shown in Figures 10 and 11. In all cases the sherds from each geographic location had matching compositions which were different from the compositions of the sherds from other geographic locations. These data are also given in Table VI. The fact that the elements cerium, europium, thorium, and chromium show major distinctions strongly suggests that these differences are not caused by burial. These elements are not subject to leaching in the manner that the alkali elements are. Finally, included in Table VI are sherds SC10 and SC19 (Figure

Mexico, Guatemala, Panama, Peru, and Ecuador

|   | $\text{La}_2\text{O}_3$ (ppm) | $\text{Sc}_2\text{O}_3$ (ppm) | $\text{CeO}_2$ (ppm) | $\text{Eu}_2\text{O}_3$ (ppm) | $\text{HfO}_2$ (ppm) | $\text{ThO}_2$ (ppm) | $\text{Cr}_2\text{O}_3$ (ppm) | $\text{CoO}$ (ppm) |
|---|-------------------------------|-------------------------------|----------------------|-------------------------------|----------------------|----------------------|-------------------------------|--------------------|
| <i>Subway Excavations, Mexico City</i>                                      |                               |                               |                      |                               |                      |                      |                               |                    |
| 30  | 30.2                          | 76                            | 1.39                 | 3.8                           | 11.5                 | 665                  | 38                            |                    |
| 31  | 29.7                          | 73                            | 1.49                 | 3.6                           | 10.9                 | 711                  | 41                            |                    |
| 21  | 20.9                          | 41                            | 1.24                 | 12.7                          | 6.9                  | 136                  | 165                           |                    |
| <i>Santiago de los Caballeros (Antigua), Guatemala</i>                      |                               |                               |                      |                               |                      |                      |                               |                    |
| 19  | 40.1                          | 53                            | 1.70                 | 5.4                           | 5.9                  | 41                   | 40                            |                    |
| 20  | 39.8                          | 56                            | 1.91                 | 5.5                           | 5.9                  | 41                   | 40                            |                    |
| <i>Panama Viejo</i>   |                               |                               |                      |                               |                      |                      |                               |                    |
| 46  | 28.6                          | 96                            | 1.69                 | 5.0                           | 16.8                 | 50                   | 24                            |                    |
| 34  | 34.1                          | 78                            | 1.56                 | 5.2                           | 14.0                 | 61                   | 29                            |                    |
| 31  | 32.0                          | 73                            | 1.77                 | 4.0                           | 16.8                 | 51                   | 29                            |                    |
| <i>Cuzco, Peru</i>  |                               |                               |                      |                               |                      |                      |                               |                    |
| 46  | 32.3                          | 99                            | 2.11                 | 6.3                           | 16.9                 | 138                  | 29                            |                    |
| 46  | 31.4                          | 99                            | 2.10                 | 6.4                           | 16.5                 | 137                  | 30                            |                    |
| <i>Quito, Ecuador</i>   |                               |                               |                      |                               |                      |                      |                               |                    |
| 42  | 21.0                          | 85                            | 1.73                 | 4.9                           | 11.6                 | 84                   | 23                            |                    |
| <i>Metropolitan Cathedral, Mexico City</i>                                  |                               |                               |                      |                               |                      |                      |                               |                    |
| 58  | 18.0                          | 104                           | 1.43                 | 7.9                           | 22.1                 | 68                   | 13.4                          |                    |
| 52  | 18.3                          | 158                           | 1.42                 | 6.6                           | 19.1                 | 70                   | 12.7                          |                    |
| <i>Means and Means + and -95% Confidence Limit for Mexico City Majolica</i> |                               |                               |                      |                               |                      |                      |                               |                    |
| 22  | 16.5                          | 39                            | 1.22                 | 4.5                           | 5.6                  | 98                   | 16.6                          |                    |
| 30.9  | 23.2                          | 61                            | 1.72                 | 6.4                           | 7.3                  | 167                  | 76.5                          |                    |
| 15.1  | 11.8                          | 26                            | 0.87                 | 3.1                           | 4.3                  | 57                   | 3.6                           |                    |

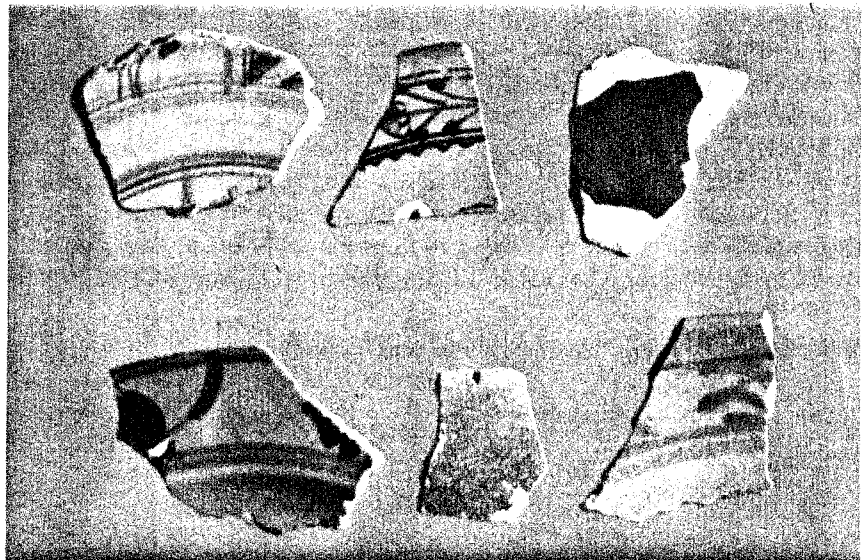


Figure 9. (left to right): (top) SB30, SB31, and SB32; (bottom) SB33 from the subway excavations in Mexico City; SB34 and SB35 from Maurica, Venezuela

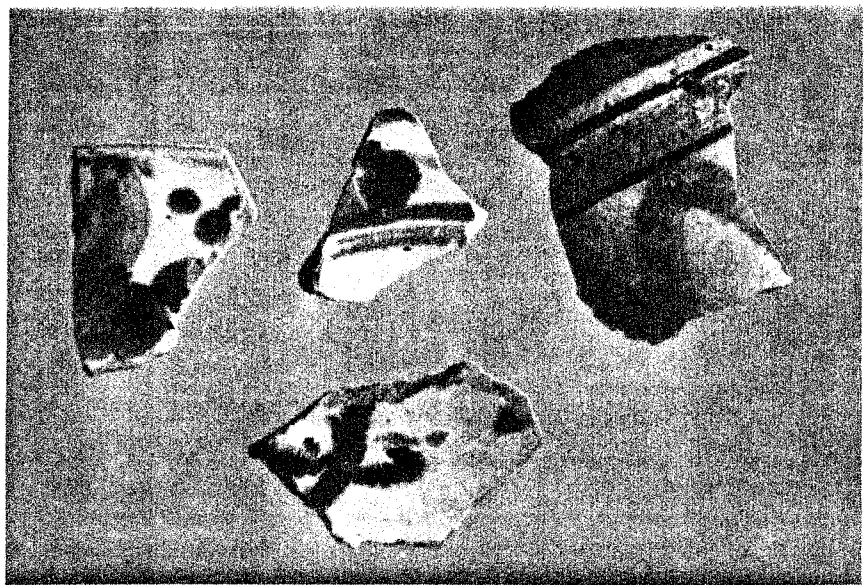


Figure 10. (left to right): (top) SB36–SB37 from the subway excavations in Mexico City; SB38 and (bottom) SB39 from Santiago de los Caballeros (Antigua), Guatemala

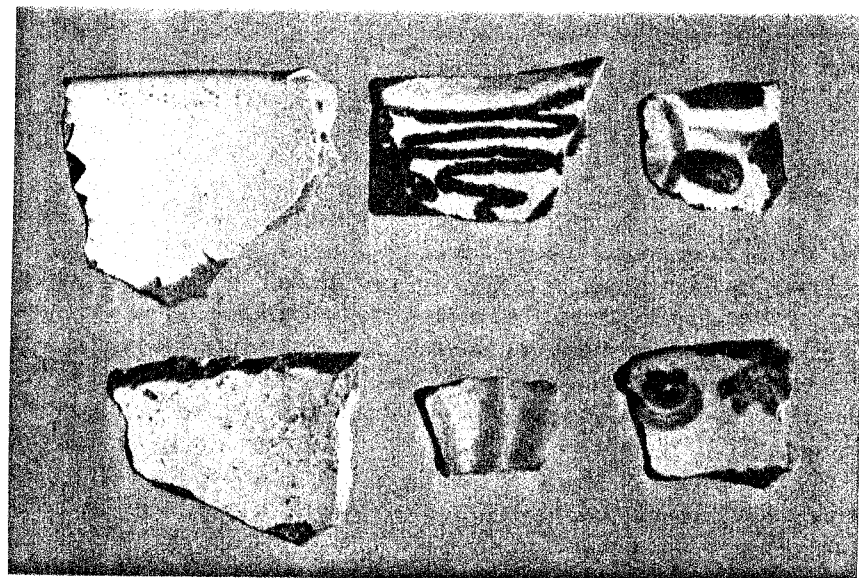


Figure 11. (left to right): (top) SB40, SB41, and SB42 from Panama Vieja; (bottom) SB43 and SB44 from Cuzco, Peru, SB45 from Quito, Ecuador

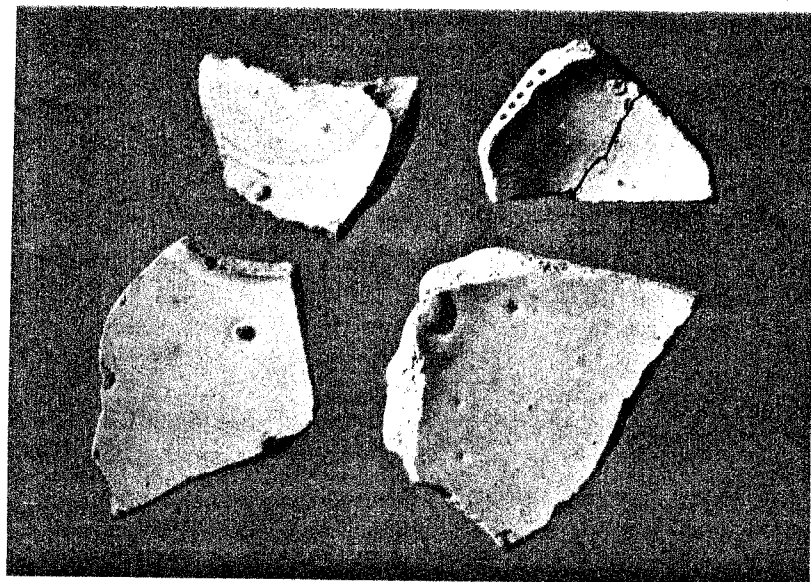


Figure 12. Columbia Plain (left to right): (top) SC10 and SC13; (bottom) SC19 and SC17 from excavations at the Metropolitan Cathedral in Mexico City

12) which are from the excavations at the Metropolitan Cathedral in Mexico City and are shown together with two additional sherds from that excavation. The compositions of sherds SC10 and SC19 do not match those of the other cathedral sherds. The appearance of the glaze is also not characteristic of the other cathedral sherds shown in Figure 5 and the two sherds to the right of Figure 12.

### Conclusions

We have been able to distinguish two distinctive groups of pottery among the majolica sherds excavated from Spanish sites in the New World. These distinctions are based on the examination and analysis of the paste portions of the sherds and have involved the combined use of neutron activation analysis, x-ray diffraction analysis, and petrographic examination. Preliminary investigations of the relationships of each of these two groups of sherds to sherds of known origin have also been undertaken. There is evidence to support a Spanish source for the sherds from sites in the Dominican Republic and Venezuela and a Mexican source for the sherds excavated in Mexico City.

We have been able to compare our samples to a small group of majolica sherds from Spain and to a reasonably large group of Precolumbian sherds from Teotihuacan. The majolica sherds from Caribbean sites agree in composition with the Spanish specimens, and those from sites in Mexico City have compositions sufficiently similar to the sherds from Teotihuacan, considering the secondary deposits of carbonates of calcium which are in the majolica sherds and not in the Precolumbian sherds. The presence of these deposits of carbonates of calcium in the majolica and their absence in the Precolumbian sherds was determined by petrographic examination and x-ray diffraction as well as by elemental analysis.

### Acknowledgments

We acknowledge the interest and encouragement of Richard Ahlborn (Smithsonian Institution) who brought this project to our attention and the further encouragement and generosity of our archaeological colleagues, Charles Fairbanks (University of Florida) and Florence and Robert Lister (Corrales, NM) who supplied well selected sherds for analysis. We also acknowledge the use of the data obtained from the analysis of the sherds from Teotihuacan provided to Brookhaven National Laboratory by Evelyn Rattray. We sincerely appreciate the assistance of Helen Warren (Santa Fe, NM) and William Melson, Martha Goodway, and James Blackman (Smithsonian Institution) for their important assistance in making the petrographic analyses. The contribution of Grover Moreland (Department of Mineral Sciences, Smithsonian Institution),

who prepared the thin-sections for petrographic examination, was an invaluable part of this investigation. We are grateful to Joan Mishara (Smithsonian Institution) for carrying out x-ray diffraction analysis and to Barbara Miller (now at the National Gallery of Art) for x-ray fluorescence analysis of the glazes. Finally, we acknowledge the helpfulness and willingness of the Brookhaven Reactor Group and the very capable technical assistance of Elaine Rowland. The research performed at Brookhaven National Laboratory was under contract with the U.S. Department of Energy and was supported by its Division of Basic Energy Sciences.

### Literature Cited

1. Goggin, J. M., "Spanish Majolica in the New World: Types of the Sixteenth to Eighteenth Centuries," Yale University Publications in Anthropology, No. 72, New Haven, 1968.
2. Lister, F., personal communication.
3. Abascal-M., R., Harbottle, G., Sayre, E. V., "Correlation between Terra Cotta Figurines and Pottery from the Valley of Mexico and Source Clays by Activation Analysis," in "Archaeological Chemistry," *ADV. CHEM. SER.* (1974) **138**, 86-87.
4. Flanagan, F. J., *Geochim. Cosmochim. Acta* (1969) **33**, 81.
5. Bieber, A. M., Brooks, D. W., Harbottle, G., Sayre, E. V., "Application of Multivariate Techniques to Analytical Data on Aegean Ceramics," *Archaeometry* (1976) **18**(1), 62.
6. Olin, J. S., Harbottle, G., Sayre, E. V., presented at Symposium on the Application of the Physical Sciences to the Study of Medieval Ceramics, University of California, March 1975.
7. Olin, J., Sayre, E., *Bull. Am. Inst. Conserv.*
8. Bieber, A. M., Jr., "Neutron Activation Analysis of Archaeological Ceramics from Cyprus," Ph.D. thesis, University of Connecticut, 1977.
9. Lister, F. C., Lister, R. H., "Majolica in Colonial Spanish America," *Historical Archaeology* (1974) **VIII**, 17-52.
10. Lister, F. C., Lister, R. H., "Non-Indian Ceramics from the Mexico City Subway," *El Palacio* (1975) **81**(2), Summer, 25-48.
11. Cervantes, G. L., "Colonial Ceramics in Mexico City," National Institute of Anthropology and History, Science Collection: Archaeology, No. 38, Mexico, 1976.

RECEIVED December 2, 1977.