Pb Isotopic Composition of Panamanian Colonial Majolica by LA-ICP-MS

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Javier G. Iñañez, Jeremy J. Bellucci, Juan Guillermo Martín, Richard Ash, William F. McDonough, and Robert J. Speakman

Abstract

Panamá Viejo, founded in 1519 by the Spanish explorer Pedrarias Dávila, was the first permanent European settlement on the Pacific Ocean, and became a city, by royal decree, in 1521. Shortly after its creation, the city became an important base for trade with Spain. In 1671, the English pirate Henry Morgan waged an attack on Panamá Viejo, which resulted in a fire that destroyed the entire city. A new settlement built a few miles west, called Casco Antiguo or San Felipe, is now the historic district of modern Panama City. The Pb isotopic compositions of the glazes on the surface of sixteenth to seventeenth century majolica pottery sherds from Panama Viejo and Casco Antiguo (both in Panama), and Lima (Peru) were determined via non-destructive laser ablation multi-collector ICP-MS (LA-MC-ICP-MS). The contrast in Pb isotopic compositions in the glazes on ceramics recovered in different locations demonstrate that early majolica pottery production during this period used Pb obtained from the Andes. However, the Pb used in later majolica production in Panama is of Spanish origin. After Panamá Viejo was burned to the ground, Panamanian majolica production ended.

J.G. Iñañez (✉)
Grupo de Investigación en Patrimonio Construido (GPAC), Universidad del País Vasco/Euskal Herriko Unibertsitatea (UPV/EHU), CIEA Lascaray, Avda. Miguel de Unamuno, 3, 01006 Vitoria-Gasteiz, Álava, Spain

IKERBASQUE, Basque Foundation for Science, Maria Diaz de Haro 3, 6 floor, 48013 Bilbao, Bizkaia, Spain
e-mail: javier.inanez@ehu.eus

J.J. Bellucci
Department of Geology, University of Maryland, College Park, MD 20742, USA

Department of Geosciences, Swedish Museum of Natural History, Stockholm 104 05, Sweden
e-mail: Jeremy.bellucci@gmail.com

J.G. Martín
Universidad del Norte, Barranquilla, Colombia
e-mail: jgmartin@uninorte.edu.co

R. Ash • W.F. McDonough
Department of Geology, University of Maryland, College Park, MD 20742, USA
e-mail: rdash@umd.edu; mcdonough@umd.edu

R.J. Speakman
Center for Applied Isotope Studies, The University of Georgia, 120 Riverbend Rd., Athens, GA 30602, USA
e-mail: archsci@uga.edu

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19.1 Introduction

19.1.1 The Old City of Panama or Panamá Viejo

After the expedition of Christopher Columbus to the Caribbean side of the current Republic of Panama in 1502, the Spanish monarchy believed it was imperative to explore these new territories, and assigned this task to Alonso de Ojeda and Diego de Nicuesa. The mainland was divided from Cabo de la Vela to Uraba's Gulf, as "Nueva Andalucia," and from Uraba's Gulf to the west, as "Castilla de Oro," respectively. These explorations, little more than raids by today's standards, aimed to conquer and colonize the mainland, and led to the founding of San Sebastián de Urabá in 1509 (now called Necoclí-Colombia). This settlement was subsequently destroyed by the indigenous people of this region and a year later the Spaniards founded Santa María la Antigua del Darién, near the Tanela river (now called Acandí-Colombia), which became the first settlement with the title of city in continental America (Martín 2009).

After his arrival as governor of "Castilla del Oro," Pedrarias decided to move Santa María la Antigua to the shores of the Pacific Ocean, a strategic location in which he could carry out a campaign of conquest (Martín 2009). Panama Viejo was founded on August 15th, 1519, in a native village under the command of Cori, and served as the first Spanish port on the Pacific coast of the Americas. Although the Spanish later established other Pacific Coast ports, Panama City remained one of the largest ports in the Pacific, in part due to the traffic of wealth looted from the Inca Empire. Likewise, all goods from Europe passed through this port for redistribution to the South American continent (Mena 1998). 152 years later, in 1671, the English pirate Henry Morgan attacked the city, leading to its destruction and final abandonment. This attack prompted the relocation of the city to what is now known as Casco Antiguo or San Felipe (Fig. 19.1). The new city of Panama, founded in 1673, was partly a reflection of the destroyed city. The layout—*traza*—of the new city followed the traditional rules of Spanish urban design in which the Plaza Mayor served as a point of reference for the central distribution and location of buildings within the city (Castillero 1994, 2004a, b; Mena 1984, 1992). However, "the *traza* of San Felipe is unthinkable without its walls, the need was a crucial aspect of the move" (Tejeira 2001:87).

19.1.2 Majolica and Spanish Production

Majolica is an earthenware ceramic generally characterized by a creamy light-buff colored ceramic paste and an opaque white tin-lead glaze covering the entire outer surface of the vessel. Perhaps the most characteristic feature of majolica pottery lies in the metallic oxide decorations that were applied on top of the tin-lead white glaze coat. The opaque white glaze is usually achieved after dipping the bisque ceramic into a soupy suspension made out of sand (e.g., quartz), tin and lead to the ceramic biscuit, and then fired again in the kiln. Lead plays an important role during the glaze maturation since it acts as a flux, decreasing the temperature needed for melting SiO_2, resulting in a bright and transparent/translucent glaze (Tite et al. 1998).

According to the historical majolica making tradition and extant written sources and current scientific literature, this opaque glaze is commonly achieved by the addition of a fine fraction of tin oxide (SnO_2) particles, likely cassiterite, the most common mineral source of Sn in nature. Thus, Sn might had been incorporated into the glaze mixture suspension by two different processes: the most common process was likely a frit, which is a raw mixture of Sn, Si and Pb minerals that had to be fused and then quenched, forming a glassy fine-grained compound, which was ground afterwards and added to water in order to form the glaze suspension; or as finely ground particulates added to the glaze suspension. During the cooling stage in the kiln after firing, cassiterite crystals grow within the glaze

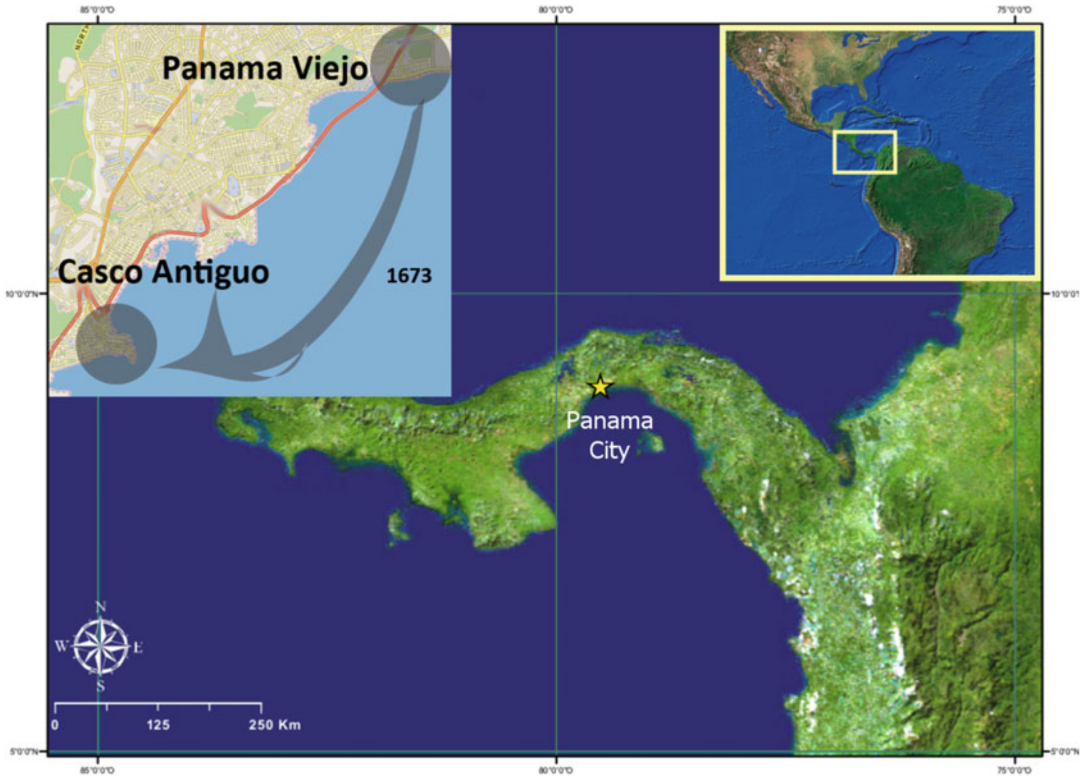

Fig. 19.1 Map of the new and old cities of Panama

into micrometric crystals and small crystalline aggregates. The appropriate small size of newly formed cassiterite particulates, along with extant small quartz and feldspar inclusions, as well as any bubbles that may result from the firing process, absorb, scatter, and/or reflect incident light, thereby giving the transparent glaze a whitish appearance. This white opacity makes a perfect canvas on which to apply chromatic decoration, which is normally applied to the outer surfaces of the glaze coat (Iñañez 2007; Molera et al. 1999; Tite et al. 2008).

Although the first evidence of opacified glazed pottery can be traced to the Middle East as early as the fifth century BC, evidence for ceramic production showing the general features described above remain unclear until the ninth century AD (Hill et al. 2004; Mason and Tite 1997). Following the known historical occurrences, majolica technology shows a clear link to the Islamic *Al-Andalus* (the Cordoba Caliphate, and subsequently *taifas* or Islamic petty kingdoms) during the medieval period on the Iberian Peninsula. It is generally considered that from the tenth century AD onwards majolica technology became widespread throughout the entire Iberian Peninsula, even in the New Christian kingdoms and principalities of the North and Northeast, and reached the rest of Western Europe soon after. By the sixteenth century, Spanish majolica production flourished as Italian-influenced decorative styles diffused into the Iberian Peninsula, incorporating new chromatic choices to the potter's palette such as yellow, usually combined with the more traditional blue, black and green colors (e.g. Iñañez 2007 and references therein).

Many majolica production centers were fully functional in the Castile and Aragon kingdoms during the period of Spanish colonial presence in the Americas. The workshops from cities like Seville, Talavera, Manises, Muel or Barcelona, just to mention a few, might be considered as representative of a genuine proto-industrial activity, which supplied not only their immediate hinterland, but also reaching distant markets. Regarding the colonial trade towards the American market, there is one production center that stood above the rest in terms of quantity and importance—the city of Seville. This city, occupying the riverbanks of the Guadalquivir River on the southern Iberian Peninsula, served as both the departure point and the final destination for most of the Spanish galleons that traded with the Americas in the so called "*Carrera de Indias*," the official (and only allowed) armored convoy of ships from Castile to the Americas. For more than 200 years, the vast traffic of commodities that resulted from the emergence of the new colonial markets was supervised by *Casa de la Contratación*, a bureau of trade established in Seville in 1503. It is, therefore, not surprising that both the importance of, and eventual decline, of Sevillian ceramic manufactures are directly linked to the endurance of the rigid monopolistic trade established by the Castilian crown and the prevalence of the *Casa de la Contratación* in the city.

19.1.3 Archaeology of Panamanian Colonial Majolica

Panama was a territory of the Viceroyalty of Peru, and due to the rigid protectionist economy established by the Castilian Crown, it was able to trade only within this colonial administrative region but generally not to other political administrative regions, such as the Viceroyalty of New Spain, although illicit trade was regularly present, as evidenced in written sources (Glave 2000; Jamieson 2001; Stein and Stein 2002). Panamanian ceramic workshops took advantage of this legal situation and traded their products southwards following the coastline and into the nearby hinterland with cities in Ecuador (Cuenca), Colombia (Popayán) (Therrien et al. 2002) and most importantly Peru (e.g., Lima, the capital of the Viceroyalty of Peru, and Moquegua) (Iñañez et al. 2012; Rice 1997).

The development of historical archaeology in Panama is relatively recent, as is the study of Panamanian majolica (Rovira and Martín 2008). The first archaeological investigations were initiated in the 1960s, and continued intermittently until the present day. From these archaeological excavations in Panama, specifically in Panama Viejo, where kiln related evidence was found (Long 1964), emerged a defined set of ceramic types referred to as "Panamanian majolica." Panamanian ceramics visually appear different because of a "brick-red paste that makes it unmistakable at first sight" (Rovira 1997). A typological classification of Panamanian majolica serves as a chronological indicator and trade marker in colonial America and is defined basically by three types: Panama Plain, Panama Blue on White, and Panama Polychrome (Goggin 1968; Long 1967; Rovira 1997).

The first type, as defined by Long (1967), and assumed to be the earliest, is Panama Plain, which is characterized by thick enamel and in some cases white or greenish tinges. Other scholars have also noted possible technological influence from the Hispano-Moresque tradition, such the marks on the surface of plates that resulted from the use of tripods during firing, and the occurrence of flat-bottomed dishes without borders (Rovira 1997). Panama Blue on White has similar characteristics to a Talavera pottery tradition—Ichtuknee Blue on White—with some obvious American-influenced designs such as corn plant motifs, as well as motifs related to the Chinese porcelain tradition (Deagan 1987; Long 1967; Rovira 1997). Panama Polychrome occurs infrequently in excavated archaeological contexts at Panama Viejo (Goggin 1968). This type has different designs using brown, blue and green colors. In some cases it is possible to find variants that have yellow on their enamel (Rovira 1997).

According to Rovira (1997), Panama Plain ceramics occur earliest in the Panamanian majolica production sequence and have features that are reminiscent of "archaic" majolica and manufacturing techniques similar to those found in the Hispano-Moresque tradition. It is in the mid-seventeenth century when the production of

Panamanian Blue on White and Polychrome majolica purportedly started; related in turn with the decline of European types in the studied archaeological contexts at Panama Viejo.

19.1.4 Pb Isotopic Studies of Colonial Ceramics and Provenance Studies of Panamanian Majolica

Until multi-collector-inductively coupled plasma-mass spectrometry (MC-ICP-MS) gained popularity for Pb isotopic analyses, the traditional method for the measurement of Pb isotope ratios was via thermal ionization mass spectrometry (TIMS). TIMS provides high analytical accuracy and precision, although at the cost of relatively slow and arduous sample preparation (for further discussion see Stos-Gale and Gale (2009), and references therein). Recently, other laboratories have developed novel approaches to Pb isotope analysis seeking alternatives to TIMS, such as EDTA (Ethylenediaminetetracetic acid) extraction and different ICP-MS configurations (see Reslewic and Burton 2002). The use of magnetic sector ICP-MS (Woolard et al. 1998), or quadrupole ICP-Q-MS (Marzo et al. 2007) are among the different alternatives chosen by researchers. Unfortunately, none of these techniques can achieve the analytical precision that TIMS or MC-ICP-MS have demonstrated for Pb isotopic measurements. Currently, studies utilizing high-precision Pb isotope ratios characterization conducted by MC-ICP-MS report excellent agreement with data acquired by TIMS (Baker et al. 2006). Moreover, MC-ICP-MS coupled with laser ablation offers the ability to efficiently generate a large, statistically significant data set more quickly than solution analyses.

In recent years, Pb isotopic analysis of glazed ceramics has gained popularity in archaeometrical studies, although still not as recurrent as its use for analysis of artifacts made of bronze, copper, silver, or glass (e.g. Degryse et al. 2009; Ponting et al. 2003; Shortland 2006; Stos-Gale and Gale 2009; Stos-Gale et al. 1997; Thibodeau et al. 2007; Yener et al. 1991). Among the first works of this nature one can cite those published by Brill et al. (Brill and Wampler 1967; Brill et al. 1987), and, especially relevant to the present study, Joel et al. (1988), an early work focused on the Pb isotopic fingerprinting of majolica pottery in the Americas. Since these initial publications, several more projects on Pb isotope provenance of glazed ceramics have been conducted. For example, it is worth noting the analyses of Islamic glazes by Mason et al. (1992) and Islamic and Hispano-Moresque ceramics by Resano et al. (2008), ceramics from the El Paso area by Pingitore et al. (1997), Rio Grande glazed ceramics by Habicht-Mauche et al. (2000, 2002), majolica from eighteenth century New Spain presidios by Reslewic and Burton (2002), and Mexican and Spanish colonial ceramics by Iñañez et al. (2010).

The first provenance studies of Panamanian majolica were conducted by Vaz and Cruxent (1975) employing thermoluminescence to discriminate between different Spanish colonial production centers in the Caribbean. Olin et al. (1978) included three ceramics found in Panama Viejo in their large chemical study of Spanish and Colonial Spanish majolica conducted by instrumental neutron activation analysis (INAA), which showed a different chemical composition than those studied from other areas. More recently, Jamieson and Hancock (2004) conducted chemical analyses by INAA on a set of ceramics collected in Cuenca, Ecuador, including a set of ceramics found in the same site and archaeologically identified as Panamanian. Soon after, Rovira et al. (2006) reported the chemical characterization by INAA of Panamanian majolica and other ceramic types unearthed at the site of Panama Viejo and two clay samples. Recently, Iñañez et al. (Iñañez and Martín 2011; Iñañez et al. 2012) reported the chemical and technological characterization by INAA and scanning electron microscopy (SEM) of Panamanian ceramics unearthed during recent archaeological excavations at the sites of Panamá Viejo and Casco Antiguo, as well as the convent of Santo Domingo in Lima, Peru. This study included over-fired ceramics and kiln related materials, such as clay spurs, confirming the local origin of

Panamanian ceramics, in agreement with Rovira et al. (2006).

19.2 Goals and Sample

The Pb isotopic composition of the glaze coating of majolica pottery can provide constrains on its source. The main goal of this study is to establish the origin of the Pb used in the manufacturing of Panamanian majolica, taking into account available archaeological evidence and the particular historical circumstances at Panamá Viejo and Casco Antiguo. Majolica technology required significant amounts of Pb and Sn in order to obtain the white glazed enamel characteristic of this ware. However, although quite common in the Earth's crust, Pb is not ubiquitous, so it has to be mined and traded from where Pb minerals are found abundantly. During the early stages of Spanish settlement in the Americas, almost every artifact and commodity was imported from the Iberian Peninsula, including Pb. However, around the mid-sixteenth century, the occupation of the Americas by Spaniards was extensive, and many important Au and Ag mines were being exploited. Therefore, the supply of Pb to Panamanian workshops was oriented towards the southern areas of the viceroyalty of Peru instead of Spain. In addition, Pb is often related to Ag in sulfide ore deposits, so ancient miners used to extract galena (PbS) and other sulfides to obtain Ag in significant quantities. Thus, studying the origin of Pb in American metallic and glazed artifacts can provide significant information regarding trade within the colonial market. In addition, assessing the use of Pb isotopic analysis by LA-MC-ICP-MS as a tool to study the provenance of colonial archaeological material in a nearly non-destructive fashion, consequently providing a powerful technique for cultural heritage studies, is also among the goals of the present work.

In order to achieve these objectives, 30 majolica Pb-glazed ceramics from Panama and Lima, Peru, were analyzed by laser ablation multicollector inductively coupled plasma mass spectrometry (LA-MC-ICP-MS) (Table 19.1). As seen in Fig. 19.2, typical Panamanian majolica decorations range from plain white glazed coats to various geometric motifs produced in blue on white, green and black on white, or even polychrome patterns. The samples investigated in this study include (1) 15 previously studied ceramics from Panama Viejo, which have been identified as of Panamanian origin according to the chemical composition of their clay pastes and archaeological evidence that included studies of kiln related materials (Iñañez et al. 2012; Rovira et al. 2006); (2) five ceramics excavated at Casco Antiguo, the new city built after the destruction and abandonment of Panamá Viejo in 1671; and (3) ten majolica ceramics that date to the sixteenth to seventeenth centuries and recovered at the Convento de Santo Domingo, Lima, which have been recently identified as Panamanian, likely from Panamá Viejo, according to the chemical analysis of their clay pastes (Iñañez et al. 2012). Additionally, this study also incorporates extant majolica and non-tin-lead glazed Pb isotopic data from sixteenth to eighteenth centuries Spanish ceramic production centers (Iñañez et al. 2010 and references therein; Joel et al. 1988), as well as Andean Pb ores (Gunnesch and Baumann 1984; Gunnesch et al. 1990; Kontak et al. 1990; Mukasa et al. 1990; Sangster et al. 2000; Tilton et al. 1981), and Spanish Pb ores (Arribas and Tosdal 1994; Canals and Cardellach 1997; Hunt 2003; Santos Zalduegui et al. 2004; Tornos and Chiaradia 2004; Velasco et al. 1996). These studies, combined with the data obtained by this study, will put Panamanian majolica Pb isotopic data into an interpretable context, and allow us to determine whether or not Panamanian majolicas used American or Spanish Pb for their glazed coatings.

Mexican Pb analyses have not been included in this study because of historical and geological reasons. The fact that some Pb ores from the Andes and Central Mexico share similar metallogenetic ages provides some overlap in their Pb isotopic signatures. Additionally, for historical reasons, one has to bear in mind that the main Mexican Ag and Au mines during Spanish colonial times, like Zacatecas, Guanajuato,

Table 19.1 Pb isotopic values of the Majolica ware analyzed from Panama and Peru

Sample	Decorations	Origin location	Chronology	Pb provenance	$^{208}Pb/^{204}Pb$	$2\sigma_{(mean)}$	$^{207}Pb/^{204}Pb$	$2\sigma_{(mean)}$	$^{206}Pb/^{204}Pb$	$2\sigma_{(mean)}$	$^{207}Pb/^{206}Pb$	$2\sigma_{(mean)}$	$^{208}Pb/^{206}Pb$	$2\sigma_{(mean)}$
OP0012	Green and black on white	Panama Viejo	1519–1671	Andes	38.776	0.0108	15.679	0.0030	18.756	0.0034	0.836	0.00013	2.068	0.00059
OP0018	Blue, green and black on white	Panama Viejo	1519–1671	Andes	38.837	0.0057	15.690	0.0016	18.796	0.0016	0.835	0.00007	2.067	0.00048
OP0019	Blue, green and black on white	Panama Viejo	1519–1671	Andes	38.836	0.0075	15.686	0.0026	18.788	0.0024	0.835	0.00006	2.068	0.00018
PVM002	n/a	Panama Viejo	1519–1671	Andes	38.828	0.0483	15.680	0.0027	18.801	0.0104	0.834	0.00029	2.065	0.00181
PVM003	n/a	Panama Viejo	1519–1671	Andes	38.971	0.0440	15.670	0.0084	18.841	0.0062	0.832	0.00068	2.068	0.00325
PVM006	n/a	Panama Viejo	1519–1671	Andes	38.763	0.1258	15.656	0.0388	18.789	0.0381	0.833	0.00028	2.063	0.00265
PVM008	n/a	Panama Viejo	1519–1671	Andes	38.853	0.0108	15.684	0.0049	18.802	0.0111	0.834	0.00033	2.066	0.00067
PVM013	n/a	Panama Viejo	1519–1671	Andes	38.807	0.1088	15.667	0.0411	18.777	0.0513	0.834	0.00014	2.066	0.00030
PVM020	Plain white	Panama Viejo	1519–1671	Andes	38.943	0.0188	15.712	0.0102	18.846	0.0091	0.834	0.00016	2.067	0.00041
PVM026	Blue on white	Panama Viejo	1519–1671	Andes	38.945	0.0091	15.718	0.0045	18.841	0.0061	0.834	0.00007	2.067	0.00047
PVM031	Blue on white	Panama Viejo	1519–1671	Andes	38.827	0.0291	15.676	0.0107	18.799	0.0100	0.834	0.00018	2.066	0.00073
PVM032	Blue on white	Panama Viejo	1519–1671	Andes	38.810	0.0180	15.671	0.0068	18.795	0.0102	0.834	0.00017	2.065	0.00075
PVM033	Blue on white	Panama Viejo	1519–1671	Andes	38.923	0.0253	15.714	0.0067	18.840	0.0053	0.834	0.00014	2.066	0.00089
PVM042	Polychrome	Panama Viejo	1519–1671	Andes	38.916	0.0272	15.718	0.0051	18.810	0.0039	0.836	0.00032	2.069	0.00169
PVM043	Polychrome	Panama Viejo	1519–1671	Andes	38.929	0.0103	15.725	0.0035	18.816	0.0039	0.836	0.00005	2.069	0.00030
PVM045	Plain white	Panama Casco Antiguo	1673–1750	Spain	38.606	0.0310	15.663	0.0076	18.503	0.0079	0.847	0.00040	2.087	0.00175
PVM046	Blue on white	Panama Casco Antiguo	1673–1750	Andes	38.661	0.0099	15.670	0.0029	18.654	0.0051	0.840	0.00009	2.072	0.00043
PVM050	Blue on white	Panama Casco Antiguo	1673–1750	Andes	38.838	0.0186	15.677	0.0066	18.786	0.0081	0.835	0.00007	2.068	0.00051

(continued)

Table 19.1 (continued)

Sample	Decorations	Origin location	Chronology	Pb provenance	$^{208}Pb/^{204}Pb$	$2\sigma_{(mean)}$	$^{207}Pb/^{204}Pb$	$2\sigma_{(mean)}$	$^{206}Pb/^{204}Pb$	$2\sigma_{(mean)}$	$^{207}Pb/^{206}Pb$	$2\sigma_{(mean)}$	$^{208}Pb/^{206}Pb$	$2\sigma_{(mean)}$
PVM052	Polychrome	Panama Casco Antiguo	1673–1750	Spain	38.615	0.0427	15.657	0.0082	18.483	0.0083	0.847	0.00056	2.090	0.00236
PVM056	Polychrome	Panama Casco Antiguo	1673–1750	Spain	38.597	0.0213	15.662	0.0072	18.502	0.0077	0.847	0.00009	2.087	0.00045
LIM002	Plain white	Lima, Peru	16th–17th cent.	Andes	39.035	0.0679	15.694	0.0274	18.868	0.0315	0.832	0.00016	2.069	0.00032
LIM024	Green and yellow on white	Lima, Peru	16th–17th cent.	Spain	38.576	0.0107	15.647	0.0031	18.496	0.0065	0.846	0.00019	2.086	0.00098
LIM037	Green on white	Lima, Peru	16th–17th cent.	Andes	38.811	0.0128	15.670	0.0023	18.791	0.0024	0.834	0.00022	2.066	0.00091
LIM038	Green on white	Lima, Peru	16th–17th cent.	Andes	38.671	0.0233	15.626	0.0082	18.686	0.0065	0.836	0.00024	2.069	0.00104
LIM040	Green and yellow black on white	Lima, Peru	16th–17th cent.	Andes	38.737	0.0124	15.669	0.0049	18.663	0.0063	0.840	0.00018	2.076	0.00079
LIM041	Green and yellow black on white	Lima, Peru	16th–17th cent.	Andes	38.825	0.0178	15.671	0.0072	18.800	0.0079	0.834	0.00017	2.066	0.00059
LIM054	Green on white	Lima, Peru	16th–17th cent.	Andes	38.802	0.0213	15.660	0.0070	18.925	0.0071	0.827	0.00011	2.050	0.00045
LIM056	Plain white	Lima, Peru	16th–17th cent.	Andes	38.880	0.0252	15.642	0.0102	18.814	0.0106	0.831	0.00011	2.066	0.00063
LIM062	Blue on white	Lima, Peru	16th–17th cent.	Spain	38.551	0.0076	15.638	0.0043	18.487	0.0059	0.846	0.00013	2.086	0.00047
LIM063	Blue on white	Lima, Peru	16th–17th cent.	Andes	38.778	0.0213	15.659	0.0064	18.787	0.0059	0.834	0.00020	2.065	0.00108

Fig. 19.2 Examples of plain *white*, *blue* on *white* and polychrome majolica found in Panama (from *left* to *right* and from *top* to *bottom*: PVM020, PVM006, PVM031, PVM042)

Pachuca and Sombrerete, are located in Central and North Mexico. Because of their geographic location and the rigid monopolistic control exhibit by Spaniards towards trade between colonial viceroyalties, metals mined in Mexico were generally carried by ground transport to the ports of Veracruz and Acapulco to be shipped to Spain or to the Spanish colonies in Asia (Castillo and Lang 1995; Lacueva Muñoz 2010). Therefore, Mexican influence on Pb supply in Panama had little historical significance and, given the geological constraints, consequently Mexican Pb isotopic signatures have been ruled out of the examples presented here in order to provide less cluttered representations of isotopic data.

19.3 Analytical Methods

Lead has four isotopes, ^{208}Pb, ^{207}Pb, ^{206}Pb, and ^{204}Pb; ^{204}Pb is invariant in nature, whereas ^{208}Pb, ^{207}Pb, ^{206}Pb are daughter products of the decay of ^{232}Th, ^{235}U, and ^{238}U, respectively. Therefore, variation in the Pb isotopic composition of a material is a function of its initial U, Th and Pb concentrations, the starting Pb isotopic composition, and the time-integrated growth of radiogenic Pb. Due to dissimilarity in the chemical behavior of U, Th, and Pb, the Pb isotopic composition of different materials can vary widely in nature. These natural variations, therefore, make the Pb isotopic system an ideal candidate for constraining the potential provenance of geologic materials and the archaeological materials derived from them (e.g. Brill and Wampler 1967; Pollard et al. 2007; Pollard 2009; Shortland 2006; Stos-Gale and Gale 2009 and references therein).

All analyses were conducted at the University of Maryland College Park Plasma Laboratory following the methodology reported by Iñañez et al. (2010). Pb isotopic compositions were determined *in situ* via LA-MC-ICP-MS employing a New Wave UP-213 laser system and a Cetac Aridus desolvating nebulizer system coupled to a Nu Plasma multiple-collector ICP-MS. Before reaching the plasma torch, the He gas from the laser ablation cell was combined with an Ar and N_2 gas flow from the Aridus nebulizer via a T-junction. During each analytical session, ultra-pure 18 MΩ (milli-Q) water was flushed through the Aridus ensuring only Ar and N_2 reached the plasma (see Iñañez et al. 2010 for gas flow settings). Laser ablation MC-ICP-MS analysis of majolica glazes has several benefits when compared to traditional thermal ionization mass spectrometry (TIMS) or MC-ICP-MS analyses. These benefits are: (1) minimally destructive analysis, which preserves samples for future investigations; (2) rapidity—each analysis takes ~2 min, which allows the collection of far more data and in turn generates statistically significant datasets; and (3) the precision of LA-MC-ICP-MS on glazes that contain wt% Pb (as the ceramics in this study), approaching that of TIMS or solution MC-ICP-MS.

Before each sample analysis, an on-peak background was taken for 45 s with the laser on and shuttered. The Nu Plasma time-resolved software was used to establish the average of the background for each analysis and to calculate each ratio using the background corrected signals for each time-resolved measurement. Typical ablation spectra were collected for ~60 s. The isobaric interference of ^{204}Hg on ^{204}Pb was monitored by measuring the background-corrected ^{202}Hg signal and corrected for using the natural isotopic abundances of each Hg isotope, ^{202}Hg/^{204}Hg = 0.2299 (de Laeter et al. 2003). The total Hg interference was insignificant (<1 part per 10,000) during our analyses due to the large amount of Pb (≥30 wt% in all cases) and negligible amount of Hg (≤μg/g) in the analyzed ceramic glazes. In addition, matrix effects are limited when using laser ablation systems with wavelengths in the deep UV (213 and 193 nm) and with nanosecond pulse durations.

Isotopic fractionation corrections were performed using the Exponential Law and NIST SRM610 values from Baker et al. (2004) by means of standard-sample bracketing (e.g., Jochum et al. 2006; Kent 2008; Paul et al. 2005; Simon et al. 2007). The fractionation factor was determined using the ^{208}Pb/^{206}Pb ratio measured in NIST SRM 610. Block analyses consisted of two standard measurements (NIST SRM610) before and after six sample measurements.

Replicate analyses of NIST 610 during each day of analyses yielded an external precision of 0.4 % on ^{20x}Pb/^{204}Pb ratios and 0.2 % on ^{20x}Pb/^{206}Pb (with x being 6, 7 or 8, as appropriate). Typical internal and external precisions for ^{20x}Pb/^{204}Pb of ceramic glazes, based on triplicate analyses of an individual sample, are 0.05 % and <0.1 %, respectively (Iñañez et al. 2010). All errors in this study are reported as internal $2\sigma_{mean}$ on an individual analysis. Iñañez et al. (2010) performed triplicate analyses on similar samples and showed that the internal precision is roughly the same as the external precision due to the high concentrations (30–50 wt%) of Pb in the glazes. All analyses were conducted on untreated ceramics on the white glazed area of the coated vessel. Small sherds of around 1 cm^2 of each sample were gently wiped with ethanol and mounted together on an aluminum microscopy wafer using a double-side sticker. This setup was used to avoid repeated manipulation of the laser chamber to introduce new samples. Reference materials were also placed inside the laser chamber at the same time. No other pre-treatment was necessary, as laser pre-ablation removed any possible superficial contamination on the samples.

19.4 Results and Discussion

The Pb isotopic compositions for the Panamanian ceramics discussed above are illustrated in Fig. 19.3 and reported in Table 19.1. Andean Pb isotopic compositions are clearly distinct from that of European produced Spanish majolica—a consequence of the different geological sources of Spanish and American materials, albeit there is some limited compositional

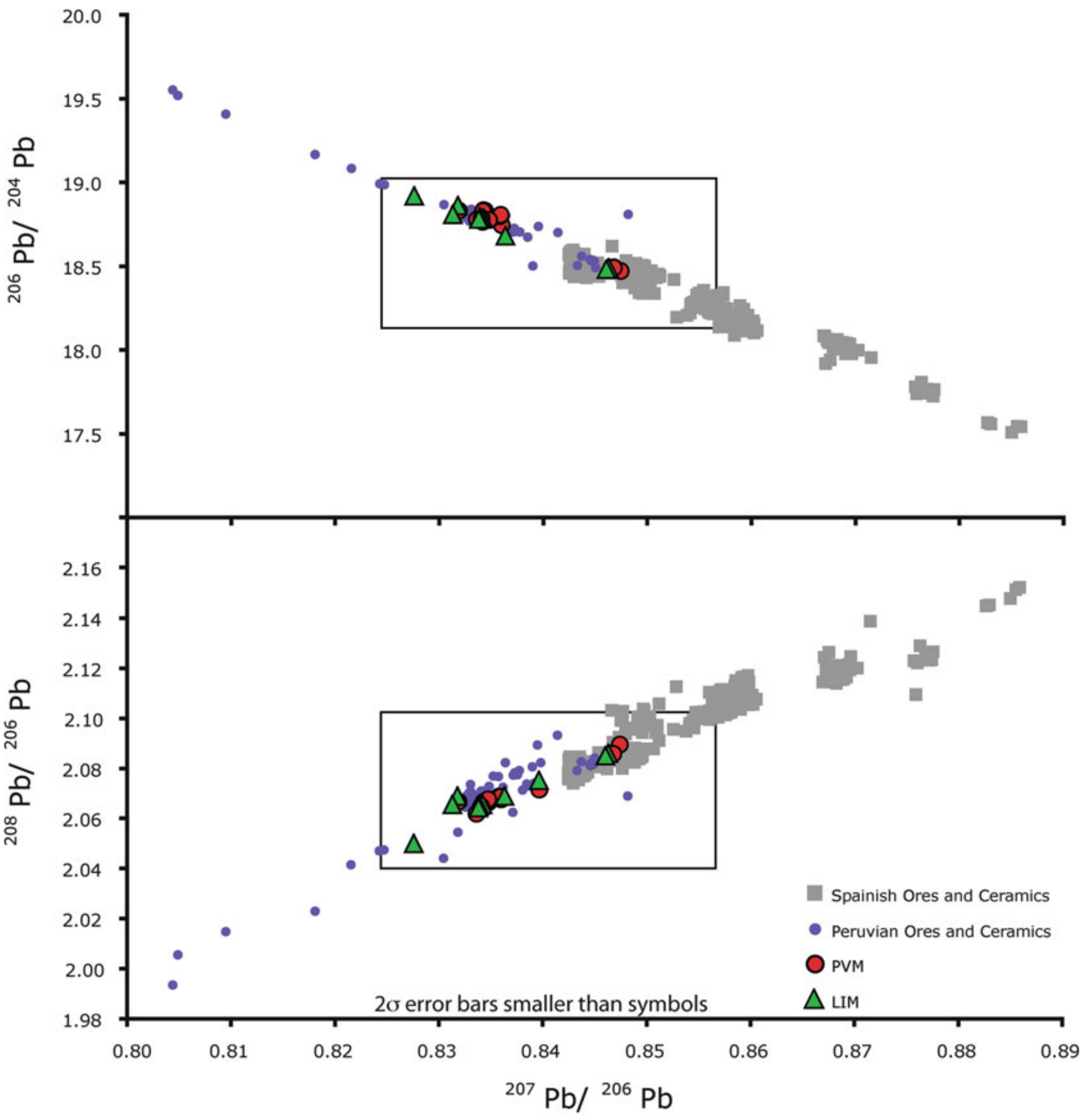

Fig. 19.3 Pb isotopic ratios of Panamanian, Mexican and Spanish ceramics, and lead deposits

overlap. Figure 19.4 represents an enlarged view of the measured Pb isotopic compositions, shading the composition for each historical ore area and projecting the Panamanian ceramics onto these shaded regions.

The Panamanian ceramics found in Panama and in Lima have similar Pb isotopic compositions, which corroborates further observations (Iñañez et al. 2012), that the ceramics found in Lima (PVM) were Panamanian products. Pb isotopic compositions for most of the ceramics found in Panama and Lima (PVM and LIM) overlap with Andean Pb isotopic ratios measured in ore samples. Furthermore, the Pb ratios of three ceramics from Panama Casco Antiguo (PVM045, PVM052 and PVM056) show higher $^{208}Pb/^{206}Pb$ and $^{207}Pb/^{206}Pb$ ratios and lower $^{206}Pb/^{204}Pb$ ratios (Fig. 19.4), exhibiting similar Pb isotopic compositions to Spanish ores and artifacts. Two of the Panamanian ceramics found in Lima (LIM) show Pb isotopic compositions similar to the Panamanian outliers and likely were produced

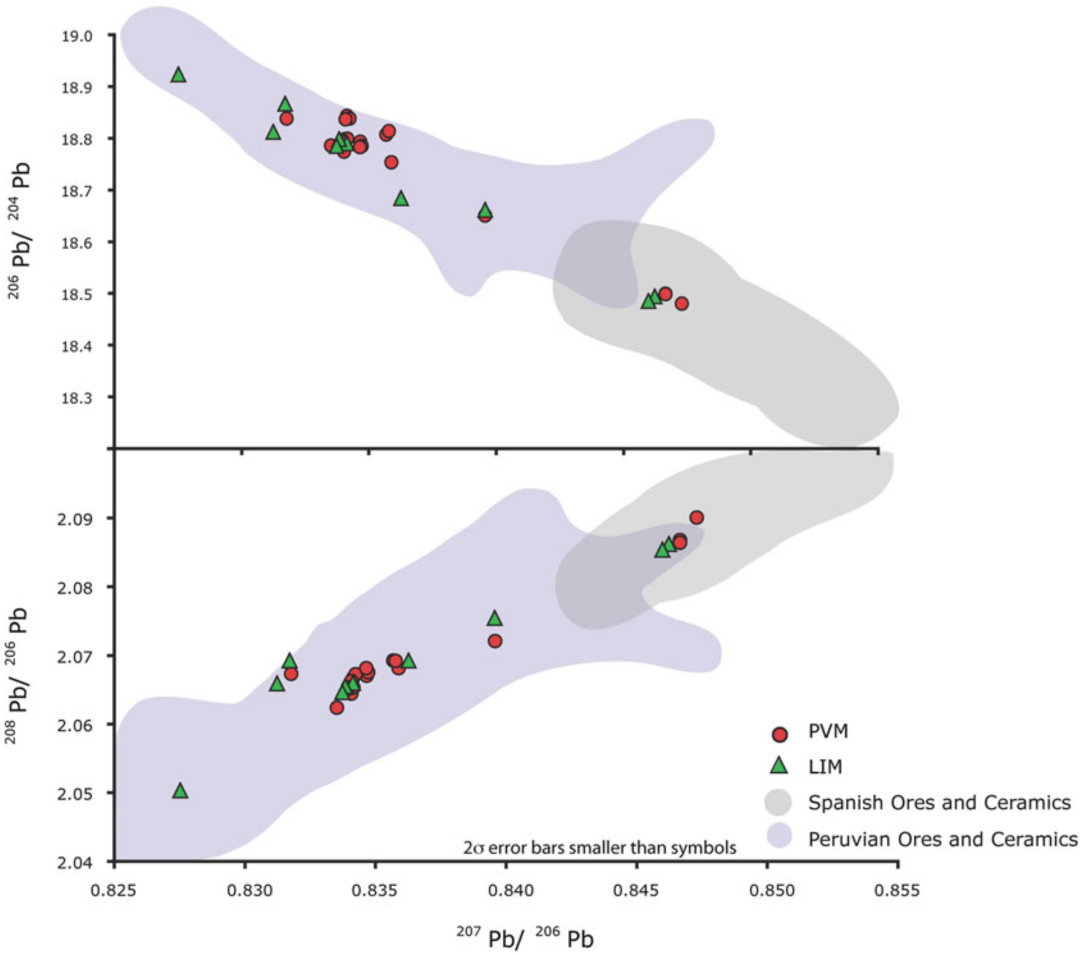

Fig. 19.4 Pb isotopic ratios of Panamanian ceramics. *Grey symbols* correspond to Spanish and Peruvian lead deposits and ceramics

from Spanish Pb. Interestingly, the outlier ceramics from Panama, with white and polychrome decorations respectively, were unearthed at the Casco Antiguo site, chronologically the later of the contexts studied. Unfortunately, the two ceramics traded to Lima, one with blue on white and the other with green and yellow on white decoration, can only be dated approximately to the seventeenth century, which may coincide with the earlier dates attributed to Casco Antiguo.

According to the data presented here, sixteenth and early seventeenth century majolica from Panama Viejo was manufactured using Pb that was most likely obtained from the Andes. This conclusion is in harmony with the idea that Panama Viejo played a leading role during the early colonial period and was considered a commercial hub in the Spanish colonial market due to its strategic location. At that time, Panama Viejo was considered one of the most important ports on the Pacific Ocean, serving as a pivotal connecting point from the territories recently conquered by the Spaniards in South America—the subsequent Viceroyalty of Peru and their important supplies of metals—to Mexico, the Caribbean, and Spain.

Nonetheless, three out of five ceramics analyzed from Casco Antiguo and two out of the ten Panamanian ceramics from the archaeological site of convent of Santo Domingo in Lima, the new capital of the Viceroyalty of Peru, have Pb isotopic compositions compatible with Spanish, not Andean, sources. Unfortunately, the chronological attribution for these ceramics has not been established accurately. However, it is possible to argue a *post quem* date of the first decade of the seventeenth century for these ceramics, when written sources provide data about tiles ordered and traded from Spain to Lima for decorating the convent, with the supply ship stopping by in Panama on the way (Rovira 2002).

One possible interpretation of our results is that as a result of the attack and sack of the city by the English pirate Morgan, which resulted in the relocation of the city 8 km southwest of its original location, the provisioning of Pb was largely supplied by new sources. Thus, at least during the first years of life of the new city, the utilization of Spanish Pb by the pottery industry may imply a shift of the Pb supply and/or a recycling process, perhaps by scavenging objects found in the ruins of or in the vicinity of the destroyed city, instead of relying on the provisioning by trade from South America. However, the archaeological surveys conducted so far at Casco Antiguo do not provide evidence regarding majolica production after the relocation of the city in 1673.

Historical written documentation provides another piece of evidence to help solve this archaeological puzzle. The government of Felipe II (second half of the sixteenth century) was known to have continued the economic and fiscal reforms established by his father, Carlos I. Among the fiscal reforms implemented by the king, the so called "*Siete Rentillas*" (seven little charges) duty is of great importance for studying Pb supply during the sixteenth to eighteenth centuries in Spain and the colonial market. This is because this duty taxed seven products of little profit to the state: black powder, sulfur, mercury, sealing wax, playing cards, iron-based pigments and, importantly, Pb (Torrente 1835). Some of these products were likely exported from the Viceroyalty of Peru. It is no coincidence that the trade of Pb from South America is linked to the beginning of extensive mining activity in Peru around 1545. Thus, Andean Pb easily arrived to Panama, becoming the primary ore used in the Panama Viejo majolica workshops, in agreement with the isotopic data from this study.

However, around the mid-seventeenth century, a deep economic crisis hit the Viceroyalty of Peru. This crisis had multiple parallel causes—perhaps the most relevant to this study is the decline in the amount of precious metals mined at Peruvian mines, which includes indirectly the quantity of Pb extracted and later traded to Panama. The purported decline in the amount of metals mined at the Andean mines resulted from several causes—the depletion of the main metallic ore veins that had been mined since the mid-sixteenth century, and the demographic decline of the native Andean population were perhaps the most significant. The native population formed the main workforce in the mine districts at that time, working under two similar types of exploitive labor conditions: forced labor (*encomienda*) or very low waged labor (*repartimiento*), both resulting in very strenuous work conditions, which ultimately greatly contributed to the decimation of the native population. In addition, a new fiscal reform that increased tax pressure fostered illicit trade by colonial market agents, who constantly broke the once rigid monopoly established by the Spanish crown and often traded with the British, French, and Dutch. During this crisis situation, the precious metals and Pb trade from Peru northwards to Panama significantly reduced in volume, and Spanish imports may have increased in importance as a result (Andrien 2011). These historical data are in agreement with the results presented here.

Furthermore, the Panamanian majolica found in Lima show a *post quem* dating to the first decades of the seventeenth century. Thus, a relative chronological assignment for Lima ceramics having a Pb isotopic signature compatible with Spanish Pb origin can be demonstrated. Based on the Pb isotopic compositions measured here and the historical and archaeological evidence known to date, these ceramics were likely manufactured in Panama after the mid-seventeenth century and transported to Peru. Consequently, and according to the current archaeological knowledge of

Panamanian majolica production, we can argue that the *ante quem* chronology for these ceramics must be around the date of 1671, when the city, and likely also its ceramic industry, was destroyed. As a final remark, we attribute the Pb used in the sixteenth and early seventeenth century Panamanian majolica to an Andean Pb ore source. However, around the second third of the seventeenth century, Panamanian ceramics were mainly manufactured with Spanish Pb.

19.5 Final Remarks

Although we acknowledge the limitations of this work because of the relatively limited number of samples analyzed, it is important to highlight implications of this study for future research. This is one of the few studies to date that successfully combines Pb isotopic analysis and available chemical data of ceramic pastes, with archaeological and historical data to reconstruct the production chronology of one of the most important majolica production centers during colonial times. In addition, we have demonstrated that integrated studies, such as what we have described above, are critical for assessing the provenance and technology of colonial glazed ceramics in order to avoid erroneous conclusions about ceramic provenance and to provide additional insight into colonial technologies.

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