

Lead and strontium isotopes as tracers for Early Formative pottery exchange in ancient Mexico

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ARTICLE INFO

Keywords:

Lead isotopes
Strontium isotopes
Ceramics
Provenance
Mesoamerica
Early formative

ABSTRACT

The sourcing of ceramics contributes to resolve questions about the nature of interactions between ancient societies. One such question concerns the primacy of the Olmec center of San Lorenzo in the development of the first unified iconographic style and its role in the early evolution of Mesoamerican civilization. Applying a combination of the isotopic systems of lead and strontium to a selection of ceramics dating to Early Formative period (1500–1000 BCE), this research evaluates the isotopic approach as an independent tool to address previously debated questions. The results demonstrate the capacity of isotopic analyses to discriminate between production centers, identify imports, relate unassigned samples to their production center and ceramics to raw material, and produce insights into production strategies (ceramics used locally versus exports). It subsequently shows how this approach can help solving existing hypotheses relying on ceramic provenance in the Early Formative period in Ancient Mexico. The findings discussed in this study more largely demonstrate that the isotopic approach constitutes a powerful tool to source ceramics and should be considered where contradictory hypotheses exist on their provenance.

1. Introduction

Establishing the origins of exotic materials and identifying the trade networks through which the materials moved remain central to larger interpretations and debates in archaeology. One such question concerns the primacy of the Olmec center of San Lorenzo in the development of the first unified iconographic style and its role in the early evolution of Mesoamerican civilization. Artifact provenance studies provide hard evidence of material circulation patterns and therefore offer one basis for evaluating the nature of Olmec influence outside of the heartland. A study by Blomster et al. (2005) based on elemental chemistry of ceramic pastes and raw clays analyzed by neutron activation analysis (NAA) found that San Lorenzo exported substantial quantities of white kaolin pottery as well as vessels decorated with distinctive Olmec iconography but received no ceramic imports from elsewhere. This study both established that other regions imported Olmec pottery from San Lorenzo, in many cases making their own local emulations; it did not

attempt to provide an overarching model for the interaction and its impact on a pan-Mesoamerican basis, which varied and involved negotiations on many levels. This study, however, set off criticisms that questioned the choice of analytical methods (elemental chemistry vs. mineralogy), the relevance of chemical compositional groups to raw-material sources and geology, and the interpretation of samples left unassigned based on chemical characterization (Flannery et al., 2005; Stoltman et al., 2005; Sharer et al., 2006). These criticisms were rebutted on both methodological and interpretive levels (Neff et al., 2006a, 2006b). Projects that focused on petrography (Guevara, 2004; Cheetham et al., 2009) supported the patterns revealed by the NAA, and subsequent presentations on Gulf Olmec interaction have acknowledged the strength of the NAA results (Pool, 2007; Cyphers, 2012).

Here, we examine the circulation of Early Formative Olmec ceramics using a new approach based on the isotopic composition of lead and strontium, and determine how the combination of the two isotopic systems can further elucidate the origins of the materials that moved

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<https://doi.org/10.1016/j.jas.2020.105307>

Received 7 September 2020; Received in revised form 2 December 2020; Accepted 2 December 2020

Available online 17 December 2020

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across ancient Mesoamerica.

The abundances of the radiogenic isotopes of lead (^{208}Pb , ^{207}Pb , and ^{206}Pb) and strontium (^{87}Sr) vary according to the age, origin and evolution of rocks. This variation, commonly reported as ratios, makes them powerful tracers for sourcing materials used in the manufacture of a large range of artifacts. This has long been recognized and isotopic geochemistry has contributed to answering many crucial questions in archaeology through the identification of provenance and by helping to identify the movements of people and the objects they produced (e.g., Farquhar and Fletcher, 1980; Gale and Stos-Gale, 1982; Ericson, 1985; Degryse, 2012; Lugli et al., 2019; Artioli et al., 2020; Killick et al., 2020).

One of the major advantages of the isotopic approach is the degree to which the isotopic signature of materials is retained throughout the process of manufacturing ceramics. Because the transformations applied to the raw material occur without isotopic fractionation, the strontium and lead isotopic signatures of the objects align closely with that of the raw material(s), even if mixing occurs. The proof of concept for the application of the isotopic approach to ceramics was established through multiproxy studies of clay sources and ceramics from multiple contexts in the Mediterranean (Renson et al., 2011, 2013a, 2013b, 2014, 2016; Makarona et al., 2016; De Bonis et al., 2018; Kibaroglu et al., 2019). Like other approaches, isotopic geochemistry discriminates between ceramics manufactured in different regions. Additionally, however, it also relates the component of the ceramic body to their raw sources, and can help identify source mixings. This constitutes an advantage compared to the vast majority of studies based on elemental chemistry alone. The capacity of isotopes to identify a place of origin for ceramics through the identification of its raw material of course depends on the distribution of the raw material. As it is the case with other techniques, multiple sources across a region can also have a similar isotopic signature. A different isotopic composition however is a clear criteria to identify the presence of different sources, or mixings. Most provenance studies based on elemental chemistry are conducted by comparing ceramics to other ceramics. The signature of a ceramic production is the result of a combination of the raw material and the manufacturing process. Elemental and isotopic approaches are therefore by nature complementary information that improves our capacity to source ceramics.

Lead is present in minerals forming the Earth's crust where it comes as a replacement for K, Sr, Ba, Ca and Na, and mainly occurs in K-feldspars and micas. Through the alteration process that transforms these minerals into clay-minerals, lead tends to concentrate in the clay fraction (Wedepohl, 1974). Three of the lead isotopes, ^{206}Pb , ^{207}Pb , and ^{208}Pb , are the decay chain products of long-lived isotopes ^{238}U , ^{235}U , and ^{232}Th , respectively. Strontium comes in minerals as a replacement for Ca and less frequently K. In silicates, it is mainly present in plagioclases and K-feldspars. In sedimentary rocks, it is also abundant in carbonates. The isotope ^{87}Sr derives from the radioactive decay of ^{87}Rb . Because Rb and Sr have different chemical properties, they partition during igneous rock formation involving a variation of Sr isotopic composition of different minerals from a same rock. This, because of differential weathering rates of minerals can consequently induce a variation between the strontium isotopic composition of the residual clays and that of more resistant minerals derived from the same rock (e.g., Capo et al. 1998). In addition, Quaternary sediments often result from a broad diversity of erosion/deposition processes and can include material from a large diversity of rocks that can be unevenly distributed among the different grain size fractions. In ceramics, the lead isotopic composition will therefore be mainly representative for the clay fraction while strontium can be representative of both the clay fraction and the temper, with a possibility for the isotopic signature of Sr to differ between these two different components.

In this study, we apply the isotopic approach, combining Pb and Sr, to Early Formative Olmec-style ceramic sherds found at San Lorenzo in the Gulf Coast, Canton Corralito in southern Mexico, and Tlapacoya in the Basin of Mexico (Fig. 1) (Ortega-Gutierrez et al., 1992), all previously analyzed for their elemental signature by NAA (Blomster et al.,

2005; Cheetham, 2010). After decades of debates on Gulf Olmec interaction, informed primarily by stylistic and iconographic analyses, the NAA study contradicted the prevailing view of mutual exchange between different Early Formative centers across Mesoamerica. The large sample subjected to NAA, including raw clays and pottery sherds, focused both on sites traditionally implicated in Early Formative exchange (the Olmec center of San Lorenzo, sites in Chiapas, as well as the highlands sites of Tlapacoya and San José Mogote), as well as newly excavated sites in Oaxaca (Elatongo, Blomster, 2004) and Chiapas (Canton Corralito, Cheetham, 2010). The results were surprisingly unambiguous: San Lorenzo was the primary exporter of pottery, and despite the large sample analyzed from that Olmec center, not a single imported sherd was identified. Our objective in this project is to evaluate how isotopic geochemistry can contribute to solve crucial archaeological questions based on provenance research and improve our understanding of trade during the Early Formative period. This is accomplished by: 1) determining whether production centers and imports can be identified based on their isotopic composition; 2) evaluating if the isotopic approach is efficient in assigning samples that were unassigned based on their elemental chemistry; and 3) investigating how isotopic geochemistry can help differentiate between San Lorenzo ceramic types.

2. Material and context

A set of 40 Early Formative ceramic sherds from three sites was selected for lead and strontium isotope analysis. All ceramic fragments were previously analyzed by NAA (Cheetham, 2010) and sherds were selected to represent: 1) local productions from the three sites, 2) different types of ceramics produced at San Lorenzo, 3) samples produced at San Lorenzo but found at Tlapacoya and Canton Corralito, and 4) samples left unassigned using NAA. These different groups were identified as such based on the NAA study and were selected to confirm their assignment or clearly assign them using the isotopic approach. Ceramic wares examined here represent Calzadas Carved, Limon Carved-Incised, La Mina White (fine paste), Remolino White, El Tigre White, Mojonera Black, and figurines (Table 1).

The three sites are located in different geological and geographical contexts in Mexico (Fig. 1) (Ortega-Gutierrez et al., 1992): San Lorenzo in the Southern Gulf Coast, Tlapacoya in the Basin of Mexico, and Canton Corralito in Chiapas. The geology around San Lorenzo is dominated by terrigenous marine sedimentary rocks and volcano-sedimentary rocks of Tertiary age that include polymictic conglomerates, silica sand, sandstone, siltstone-claystone and rhyolitic tuffs, by Quaternary alluvial deposits, and by Quaternary sand, silt, clay sediments of lacustrine origin. The Tertiary-Quaternary Tuxtla volcanic field can be found about 30 km N of San Lorenzo, with the presence in its foothills of uplifted Miocene-Pliocene marl, sand and clay deposits, including kaolinite sources. Limestones and dolomites of Cretaceous age can be found 40–50 km S-SE of San Lorenzo. Igneous and metamorphic rocks from the Proterozoic and Paleozoic can be found 70–80 km from San Lorenzo in the S-SW direction. The site of Tlapacoya is located in the Basin of Mexico, in the Mexican Volcanic Belt, where the geology is mainly composed of volcanic Quaternary rocks of basaltic to andesitic composition and Quaternary sediments derived from their weathering. The site of Canton Corralito is located in the Pacific Coast Plain that developed between the Volcanic Highlands and the ocean. The volcanic highlands are composed of Tertiary lahar and pyroclastic deposits of andesitic composition. The sediments of the Plain are constituted of clays, silt, sand and gravel derived from the weathering of the rocks from the Volcanic Highlands.

3. Methods

Sherds were powdered in an agate mortar and dried overnight at 105 °C. Approximately 300 mg of powder were digested in 4 ml of 24 N

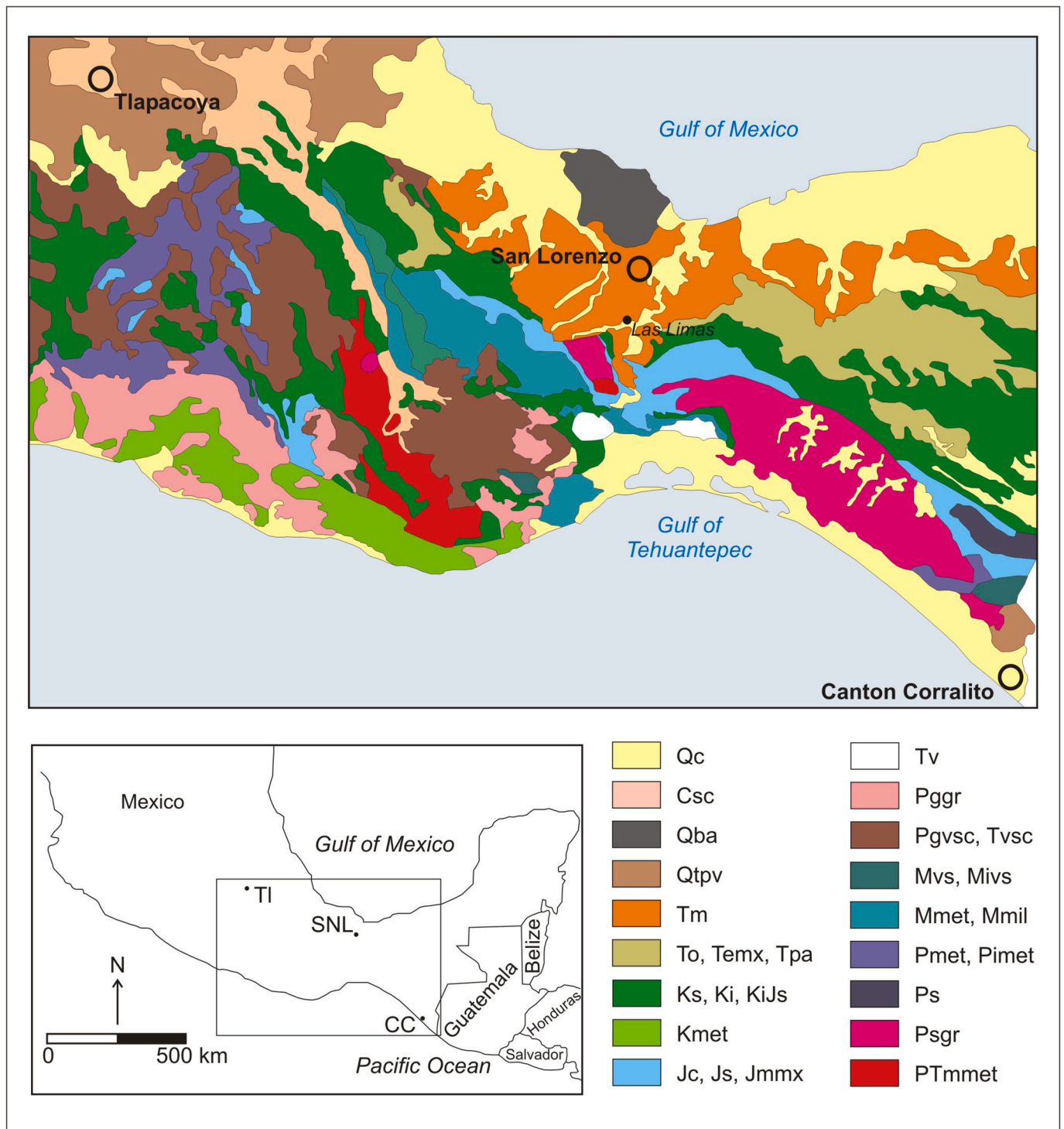


Fig. 1. Site locations and geological map of the area. Geological map simplified after (Ortega-Gutierrez et al., 1992). TI for Tlapacoya, SNL for San Lorenzo, CC for Canton Corralito. Chronostratigraphy and geological environments as reported elsewhere (Ortega-Gutierrez et al., 1992): Qc – Quaternary Sedimentary Continental, Csc – Quaternary and Neogene Sedimentary Continental, Qba – Quaternary Igneous Extrusive Volcanic Continental, Qtpv – Quaternary and Pliocene Igneous Extrusive Volcanic Continental, Tm – Miocene Sedimentary Marine, To – Oligocene Sedimentary Marine, Temx – Eocene Sedimentary Mixed, Tpa – Paleocene Sedimentary Marine, Ks – Upper Cretaceous Sedimentary Marine, Ki – Lower Cretaceous Sedimentary Marine, KiJs – Lower Cretaceous/Upper Jurassic Sedimentary Marine, Kmet – Cretaceous Metamorphic, Jc – Jurassic Sedimentary Continental, Js – Upper Jurassic Sedimentary Marine, Jmmx – Middle Jurassic Sedimentary Mixed, Tv – Tertiary Igneous Extrusive Volcanic Continental, Pggr – Paleogene Igneous Intrusive, Pgvsc – Paleogene Igneous Extrusive Volcano-sedimentary Continental, Tvsc – Tertiary Igneous Extrusive Volcano-sedimentary Continental, Mvs – Mesozoic Igneous Extrusive Volcano-sedimentary Marine, Mivs – Jurassic and Triassic Igneous Extrusive Volcano-sedimentary Marine, Mmet – Mesozoic Metamorphic, Mmil – Mesozoic Metamorphic, Pmet – Paleozoic Metamorphic, Pimet – Devonian to Cambrian Metamorphic, Ps – Permian and Carboniferous Sedimentary Marine, Psgr – Permian and Carboniferous Igneous Intrusive, PTmmet – Middle Proterozoic Metamorphic. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

Table 1

Isotopic results and information on samples. MURR sample ID, ceramic typology, chemical group assignment based on previous NAA results (Cheetham, 2010), Sr and Rb concentrations were previously measured by NAA (Cheetham, 2010), * for duplicate (i.e., entire procedure repeated for a same sample) and ** for replicate (i.e., analysis repeated for a same solution), se for standard error (instrument internal error), CC for Canton Corralito, SNL for San Lorenzo.

Sample ID	Site	Ceramic type	Chemical group	$^{208}\text{Pb}/^{204}\text{Pb}$	2se	$^{207}\text{Pb}/^{204}\text{Pb}$	2se	$^{206}\text{Pb}/^{204}\text{Pb}$	2se	$^{208}\text{Pb}/^{206}\text{Pb}$	2se	$^{207}\text{Pb}/^{206}\text{Pb}$	2se	$^{87}\text{Sr}/^{86}\text{Sr}$	2se	Sr ($\mu\text{g g}^{-1}$)	Rb ($\mu\text{g g}^{-1}$)
DCC012	Canton Corralito	Limon Carved-Incised	Canton Corralito local	38.5542	0.0013	15.6135	0.0005	18.7400	0.0005	2.05735	0.00003	0.83317	0.00001	0.70461	0.00002	428	32
DCC012 *	Canton Corralito			38.5622	0.0024	15.6150	0.0007	18.7430	0.0009	2.05746	0.00008	0.83312	0.00001	0.70460	0.00002		
DCC012 **	Canton Corralito													0.70458	0.00002		
DCC012 * **	Canton Corralito			38.5573	0.0018	15.6144	0.0006	18.7407	0.0006	2.05744	0.00004	0.83319	0.00001				
DCC014	Canton Corralito	Limon Carved-Incised	Canton Corralito local	38.5769	0.0027	15.6169	0.0006	18.7458	0.0007	2.05794	0.00009	0.83309	0.00001	0.70459	0.00002	485	41
DCC015	Canton Corralito	Limon Carved-Incised	Canton Corralito local	38.5521	0.0017	15.6134	0.0006	18.7385	0.0007	2.05741	0.00003	0.83324	0.00001	0.70469	0.00001	437	42
DCC204	Canton Corralito	Calzadas Carved	Canton Corralito local	38.5704	0.0059	15.6184	0.0015	18.7413	0.0012	2.05791	0.00017	0.83335	0.00002	0.70461	0.00001	359	35
DCC206	Canton Corralito	Calzadas Carved	Canton Corralito local	38.5393	0.0019	15.6100	0.0006	18.7329	0.0006	2.05733	0.00004	0.83330	0.00001	0.70466	0.00001	437	36
DCC207	Canton Corralito	Calzadas Carved	Canton Corralito local	38.5272	0.0023	15.6078	0.0008	18.7325	0.0009	2.05669	0.00003	0.83319	0.00001	0.70461	0.00002	385	28
DCC207 **	Canton Corralito													0.70462	0.00001		
DCC024	Canton Corralito	Limon Carved-Incised	Unassigned - likely CC	38.5443	0.0022	15.6128	0.0008	18.7315	0.0008	2.05766	0.00003	0.83348	0.00001	0.70468	0.00002	372	25
DCC071	Canton Corralito	Limon Carved-Incised	Unassigned - likely CC	38.5393	0.0016	15.6098	0.0006	18.7332	0.0007	2.05726	0.00004	0.83326	0.00001	0.70471	0.00001	423	44
DCC208	Canton Corralito	Calzadas Carved	Unassigned - likely CC	38.5014	0.0049	15.6025	0.0014	18.7167	0.0013	2.05716	0.00019	0.83363	0.00002	0.70440	0.00001	488	32
DCC214	Canton Corralito	Calzadas Carved	Unassigned - likely CC	38.5741	0.0021	15.6151	0.0008	18.7502	0.0009	2.05735	0.00005	0.83281	0.00001	0.70471	0.00002	479	40
DCC022	Canton Corralito	Limon Carved-Incised	San Lorenzo local (gray)	38.5327	0.0022	15.6388	0.0008	18.7213	0.0009	2.05826	0.00004	0.83535	0.00001	0.70761	0.00002	169	69
DCC022 **	Canton Corralito			38.5260	0.0029	15.6363	0.0010	18.7178	0.0010	2.05818	0.00003	0.83536	0.00001				
DCC146	Canton Corralito	Limon Carved-Incised	San Lorenzo local (gray)	38.6455	0.0030	15.6426	0.0013	18.8001	0.0012	2.05564	0.00003	0.83203	0.00001	0.70712	0.00001	169	101

(continued on next page)

Table 1 (continued)

Sample ID	Site	Ceramic type	Chemical group	$^{208}\text{Pb}/^{204}\text{Pb}$	2se	$^{207}\text{Pb}/^{204}\text{Pb}$	2se	$^{206}\text{Pb}/^{204}\text{Pb}$	2se	$^{208}\text{Pb}/^{206}\text{Pb}$	2se	$^{207}\text{Pb}/^{206}\text{Pb}$	2se	$^{87}\text{Sr}/^{86}\text{Sr}$	2se	Sr ($\mu\text{g g}^{-1}$)	Rb ($\mu\text{g g}^{-1}$)
DCC166	Canton Corralito	Limon Carved-Incised	San Lorenzo local (gray)	38.5644	0.0018	15.6405	0.0007	18.7173	0.0008	2.06035	0.00003	0.83562	0.00001	0.70762	0.00001	235	90
DCC482	Canton Corralito	Figurine	San Lorenzo white	38.5613	0.0024	15.6425	0.0010	18.7173	0.0012	2.06019	0.00003	0.83573	0.00001	0.70695	0.00002	267	60
DCC524	Canton Corralito	fine paste (La Mina White)	San Lorenzo white	38.7831	0.0023	15.6572	0.0008	18.8480	0.0009	2.05765	0.00004	0.83068	0.00001	0.70841	0.00001	127	59
DCC526	Canton Corralito	fine paste (La Mina White)	San Lorenzo white	38.6627	0.0032	15.6465	0.0012	18.8066	0.0013	2.05576	0.00007	0.83197	0.00001	0.71456	0.00001	0	72
DCC037	Canton Corralito	Limon Carved-Incised	Unassigned - likely SNL	38.6190	0.0022	15.6423	0.0008	18.7973	0.0008	2.05447	0.00005	0.83215	0.00001	0.70602	0.00001	352	57
DCC057	Canton Corralito	Limon Carved-Incised	Unassigned - likely SNL	38.6228	0.0027	15.6414	0.0008	18.7814	0.0007	2.05638	0.00010	0.83282	0.00002	0.70657	0.00001	165	73
DCC099	Canton Corralito	Limon Carved-Incised	Unassigned - likely SNL	38.6009	0.0025	15.6399	0.0009	18.7910	0.0011	2.05422	0.00004	0.83231	0.00001	0.70654	0.00002	375	81
DCC099 *	Canton Corralito			38.6059	0.0023	15.6403	0.0008	18.7944	0.0010	2.05416	0.00005	0.83219	0.00001	0.70652	0.00001	375	
DCC099 **	Canton Corralito													0.70652	0.00001		
DCC345	Canton Corralito	Calzadas Carved	Unassigned - likely SNL	38.6238	0.0021	15.6444	0.0009	18.7789	0.0009	2.05683	0.00003	0.83310	0.00002	0.70725	0.00001	154	57
DCC415	Canton Corralito	Calzadas Carved	Unassigned - likely SNL	38.6477	0.0026	15.6446	0.0009	18.7915	0.0010	2.05676	0.00004	0.83256	0.00001	0.70772	0.00001	106	83
DCC561	San Lorenzo	Limon Carved-Incised	San Lorenzo local (gray)	38.6442	0.0014	15.6413	0.0005	18.7921	0.0006	2.05644	0.00004	0.83234	0.00001	0.70842	0.00001	239	119
DCC562	San Lorenzo	Limon Carved-Incised	San Lorenzo local (gray)	38.6383	0.0018	15.6400	0.0006	18.7821	0.0006	2.05720	0.00004	0.83272	0.00001	0.70834	0.00002	179	102
DCC563	San Lorenzo	Limon Carved-Incised	San Lorenzo local (gray)	38.6400	0.0014	15.6437	0.0006	18.7681	0.0006	2.05882	0.00003	0.83352	0.00001	0.70884	0.00002	265	101
DCC564	San Lorenzo	Limon Carved-Incised	San Lorenzo local (gray)	38.6343	0.0015	15.6429	0.0005	18.7939	0.0005	2.05573	0.00003	0.83233	0.00001	0.70867	0.00001	215	106
DCC572	San Lorenzo	Calzadas Carved	San Lorenzo local (gray)	38.5828	0.0025	15.6431	0.0009	18.7489	0.0012	2.05796	0.00004	0.83436	0.00001	0.70944	0.00002	215	76
DCC573	San Lorenzo	Calzadas Carved	San Lorenzo local (gray)	38.6825	0.0027	15.6443	0.0010	18.8164	0.0012	2.05582	0.00004	0.83142	0.00001	0.70901	0.00001	150	121
DCC575	San Lorenzo	Calzadas Carved	San Lorenzo local (gray)	38.6134	0.0019	15.6415	0.0007	18.7648	0.0007	2.05778	0.00005	0.83356	0.00001	0.70831	0.00001	311	85
DCC578	San Lorenzo	Calzadas Carved	San Lorenzo local (gray)	38.6172	0.0019	15.6436	0.0006	18.7834	0.0007	2.05594	0.00004	0.83284	0.00001	0.70859	0.00002	179	98
DCC625	San Lorenzo	hollow limb; small leg	San Lorenzo white	38.6216	0.0016	15.6466	0.0005	18.7646	0.0005	2.05823	0.00004	0.83384	0.00001	0.70938	0.00001	358	80
DCC636	San Lorenzo	hollow head; Olmec style	San Lorenzo white	38.6029	0.0017	15.6442	0.0006	18.7938	0.0007	2.05402	0.00005	0.83242	0.00002	0.71129	0.00001	117	69

(continued on next page)

Table 1 (continued)

Sample ID	Site	Ceramic type	Chemical group	$^{208}\text{Pb}/^{204}\text{Pb}$	2se	$^{207}\text{Pb}/^{204}\text{Pb}$	2se	$^{206}\text{Pb}/^{204}\text{Pb}$	2se	$^{207}\text{Pb}/^{206}\text{Pb}$	2se	$^{87}\text{Sr}/^{86}\text{Sr}$	2se	Sr ($\mu\text{g g}^{-1}$)	Rb ($\mu\text{g g}^{-1}$)		
DCC639	San Lorenzo	solid head; Olmec style	San Lorenzo white	38.6010	0.0020	15.6441	0.0007	18.7715	0.0008	2.05638	0.00005	0.83340	0.00002	0.71492	0.00002	32	80
DCC585	San Lorenzo	Mojonera Black	Unassigned - likely SNL	38.7511	0.0019	15.6539	0.0007	18.7964	0.0008	2.06165	0.00004	0.83283	0.00001	0.71946	0.00002	0	121
DCC585**	San Lorenzo			38.7516	0.0018	15.6540	0.0009	18.7958	0.0008	2.06172	0.00004	0.83284	0.00001				
DCC591	San Lorenzo	La Mina White	Unassigned - likely SNL	38.6334	0.0019	15.6443	0.0007	18.7630	0.0008	2.05901	0.00004	0.83378	0.00001	0.70920	0.00001	266	83
DCC606	San Lorenzo	El Tigre White	Unassigned - likely SNL	38.5258	0.0017	15.6391	0.0006	18.7740	0.0006	2.05206	0.00004	0.83302	0.00001	0.70981	0.00001	186	47
DCC685	Tlapacoya	Calzadas white slip; incised	Tlapacoya	38.4318	0.0029	15.5929	0.0008	18.6665	0.0009	2.05888	0.00012	0.83535	0.00001	0.70434	0.00002	481	42
DCC687	Tlapacoya		Tlapacoya	38.4316	0.0016	15.5946	0.0006	18.6708	0.0006	2.05841	0.00003	0.83524	0.00001	0.70436	0.00001	587	38
DCC689	Tlapacoya	b/w; incised; banded-eye	Tlapacoya	38.4095	0.0015	15.5902	0.0005	18.6577	0.0006	2.05870	0.00004	0.83560	0.00001	0.70435	0.00002	574	49
DCC684	Tlapacoya	Calzadas God	San Lorenzo local (gray)	38.6559	0.0023	15.6433	0.0010	18.8064	0.0010	2.05544	0.00004	0.83184	0.00001	0.71046	0.00001	118	81
DCC699	Tlapacoya	Remolino White	San Lorenzo white	38.5912	0.0024	15.6458	0.0008	18.8102	0.0008	2.05167	0.00005	0.83178	0.00001	0.71259	0.00001	100	71

Trace Metal Grade HF and 1 ml of Optima Grade 14 N HNO₃ in microwave vessels (190 °C for 35 min) and then evaporated at 90 °C overnight. The dry residues were digested in 6 ml of 6 N Optima Grade HCl and transferred in Savillex® PFA vials. Vials were closed and warmed at 125 °C for 48 h. The solutions were split into two aliquots: one for Pb isotope analyses, the other for Sr isotope analyses. The solutions for both Pb and Sr analyses were evaporated at 90 °C. For lead extraction, samples were prepared using a protocol described elsewhere (Renson et al., 2016). For Sr isotope analyses, dry residues were digested in 2 ml of Optima Grade 7 N HNO₃ and evaporated. Strontium extraction was realized using a procedure adapted from De Muynck et al. (2009) using 2 ml Eichrom columns and 0.5 ml of Eichrom Sr-spec resin. Lead and strontium isotope analyses were conducted on a Nu Plasma II (Nu Instruments) Multi Collector - Inductively Coupled Plasma - Mass Spectrometer (MC-ICP-MS) in operation at the University of Missouri Research Reactor (MURR).

3.1. Lead isotope analysis

The instrument was optimized for ^{208}Pb maximum intensity (and minimum signal of 150 mV at mass 204). The samples and standard were spiked using a Tl solution to monitor and correct for mass fractionation. Sample solutions were prepared to measure Pb and Tl concentrations similar to those of the SRM 981 solution (approximately 200 ng g⁻¹ in Pb and 50 ng g⁻¹ in Tl). The SRM981 solution was measured several times at the beginning of each analytical session and after every two samples. Values were corrected for mass fractionation using a 2.38714 value for $^{205}\text{Tl}/^{203}\text{Tl}$ natural ratio (NIST value). A correction for mercury isobaric interference at mass 204 was also applied using a 0.229883 value for $^{204}\text{Hg}/^{202}\text{Hg}$ natural ratio. The mean values obtained for the SRM981 were 36.694 ± 0.013 (2SD), 15.491 ± 0.004 (2SD) and 16.938 ± 0.003 (2SD), for $^{208}\text{Pb}/^{204}\text{Pb}$, $^{207}\text{Pb}/^{204}\text{Pb}$ and $^{206}\text{Pb}/^{204}\text{Pb}$ ratios, respectively. These values are in good agreement with the NIST values and with the long-term values obtained at MURR on the Nu Plasma II. To correct for minimal shift observed during the runs, the sample values are corrected by standard-bracketing (White et al., 2000; Weis et al., 2006) using recommended values from Galer and Abouchami (1998). Values published by Galer and Abouchami (1998) for the SRM981 are 36.7219, 15.4963, 16.9405, 2.16771, and 0.91475 for $^{208}\text{Pb}/^{204}\text{Pb}$, $^{207}\text{Pb}/^{204}\text{Pb}$, $^{206}\text{Pb}/^{204}\text{Pb}$, $^{208}\text{Pb}/^{206}\text{Pb}$ and $^{207}\text{Pb}/^{206}\text{Pb}$ ratios, respectively. Three duplicates (i.e. entire procedure applied twice to the same sample) and three replicates (second analysis of the same solution) were measured to evaluate the reproducibility of measurements (Table 1).

3.2. Strontium isotope analysis

The instrument was optimized for ^{88}Sr maximum intensity (~7 V). The solutions were prepared to obtain the same strontium concentration in the sample and in the standard (150 ng g⁻¹). The SRM987 solution was measured several times at the beginning of each analytical session and after every two samples. The results presented here were acquired in two separate sessions. The values obtained for SRM987 during the first and the second session for the $^{87}\text{Sr}/^{86}\text{Sr}$ ratio were 0.71034 ± 0.00015 (2SD) and 0.71026 ± 0.00003 (2SD), respectively. Values were corrected for mass fractionation using the 0.1194 value for $^{86}\text{Sr}/^{88}\text{Sr}$ natural ratio. Isobaric interference at mass 87 was corrected using a 0.3857 value for the $^{87}\text{Rb}/^{85}\text{Rb}$ natural ratio. Isobaric interference of Kr at masses 86 and 84 were corrected by the iterative method. The sample results were corrected using the sample-standard bracketing method to eliminate any instrumental drift using the accepted value of 0.710248 for $^{87}\text{Sr}/^{86}\text{Sr}$ ratio (Thirlwall, 1991). Two duplicates and three replicates were measured (Table 1).

3.3. Statistical methods

A discriminant analysis was performed using the isotopic ratios of Pb and Sr to determine the ability of the isotopes to identify the sherds from each of the studied site. To this end, local sherds were assigned to a site (Canton Corralito, San Lorenzo gray, San Lorenzo white, and Tlapacoya) leaving the unassigned samples unclassified. This procedure determines which combination of isotopic ratios sustains the discrimination between the assigned classes (sites), calculates the percentage of correct classifications for each class (site), and estimates the probability of each unassigned sample to belong to each class (site). The analysis was performed using SPSS 24.0.

4. Results

For the presentation of the isotopic results, we use the ceramic typology and the site where the ceramics were found, as well as the chemical group identified by NAA. For samples attributed to San Lorenzo, two groups were identified by NAA: the San Lorenzo Gray that includes Calzadas Carved and Limon Carved-Incised, and the San Lorenzo White group that includes a broad diversity of types of ceramics and figurines. We have kept that distinction between the groups in our presentation of the results. The samples that were unassigned based on the NAA are labelled as “likely from San Lorenzo” or “likely from Canton Corralito”. It means that the probability of these samples to belong to these specific groups was lower than 2%. For lead isotopes, the ratio that

provides the best discrimination is the $^{207}\text{Pb}/^{204}\text{Pb}$, and the results and discussion are presenting the groups as displayed on the $^{207}\text{Pb}/^{204}\text{Pb}$ vs. $^{206}\text{Pb}/^{204}\text{Pb}$ three-isotope plot (Fig. 2a) and Table 1. Because the isotope ^{208}Pb derives from Th and can give further information of sources with a different initial U/Th value, the results obtained on the $^{208}\text{Pb}/^{204}\text{Pb}$ vs. $^{206}\text{Pb}/^{204}\text{Pb}$ are therefore briefly summarized where it helps further differentiating the samples, making reference to both Fig. 2b and Table 1. The strontium isotope results are displayed in the $^{87}\text{Sr}/^{86}\text{Sr}$ vs. $1/[\text{Sr}]$ plot (Fig. 3). The combination of all isotopic ratios is also further examined using a discriminant analysis (Fig. 4). Strontium and rubidium concentrations were previously obtained by NAA and are reported in Table 1. Because elemental chemistry was measured by NAA, no data are available for Pb concentrations. Note: the chemical group previously labelled as “Mazatan local” (Cheetham, 2010) is called “Canton Corralito” in the present manuscript.

Using the $^{207}\text{Pb}/^{204}\text{Pb}$ vs. $^{206}\text{Pb}/^{204}\text{Pb}$ three-isotope plot, the lead isotopic results show that the samples are distributed in three main groups, one sub-group and two singletons (Table 1 and Fig. 2a). A first

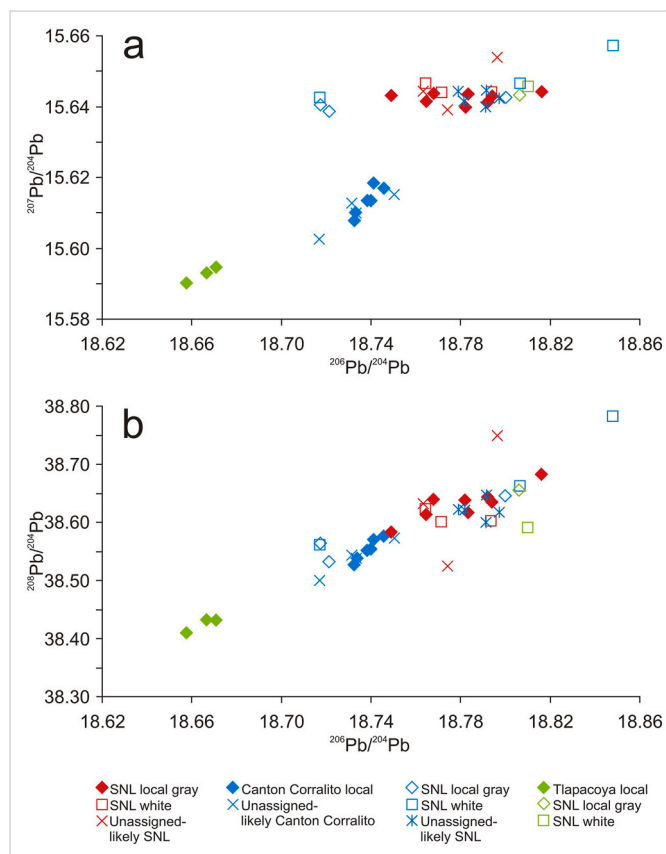


Fig. 2. Lead isotopic composition of all sherds. a. $^{207}\text{Pb}/^{204}\text{Pb}$ vs $^{206}\text{Pb}/^{204}\text{Pb}$ three-isotope plot, b. $^{208}\text{Pb}/^{204}\text{Pb}$ vs $^{206}\text{Pb}/^{204}\text{Pb}$ three-isotope plot. Colors are used to identify the sites where the samples were found. Red: samples found at San Lorenzo, blue: samples found at Canton Corralito, green: samples found at Tlapacoya. The different symbols represent the different chemical groups as identified previously by NAA. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

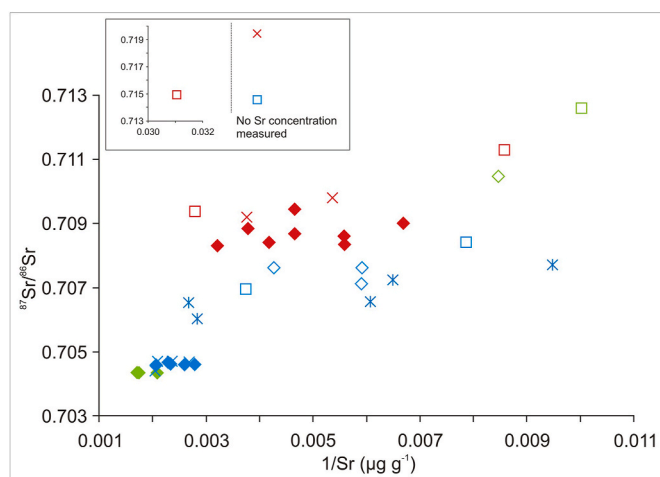


Fig. 3. $^{87}\text{Sr}/^{86}\text{Sr}$ isotopic ratio vs $1/[\text{Sr}] \mu\text{g g}^{-1}$ for all sherds. The $^{87}\text{Sr}/^{86}\text{Sr}$ isotopic ratios for two samples (DCC526 and DCC585) are reported outside the graph because no Sr concentration was measured by NAA for these samples. Color and symbol use is similar to Fig. 2. In the representation chosen here, with the Sr concentration reported as $1/[\text{Sr}] (\mu\text{g g}^{-1})$, a mixing between two end-members would be displayed as a straight line instead of a hyperbola. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

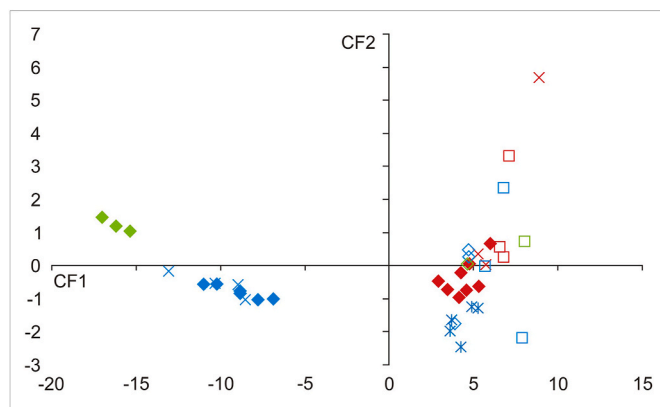


Fig. 4. Plot of the scores of the canonical functions (CF1 and CF2) showing the allocation of the sherds after the discriminant analysis based on the isotopic ratios. Color and symbol use is similar to Fig. 2. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

group is composed of ceramics found at Tlapacoya and identified as the local production based on elemental chemistry. A second group is composed of Calzadas Carved and Limon Carved-Incised ceramics found at Canton Corralito that were identified as local production or left unassigned based on elemental chemistry. A third group is composed of Calzadas Carved, Limon Carved-Incised, La Mina white, El Tigre, Remolino white and figurines found at San Lorenzo, Tlapacoya and Canton Corralito, and that were identified as San Lorenzo production or left unassigned based on the elemental chemistry. The sub-group which shares the $^{207}\text{Pb}/^{204}\text{Pb}$ ratio values with the third group is composed of two Limon Carved-Incised sherds and a figurine. These three samples have lower $^{206}\text{Pb}/^{204}\text{Pb}$ and $^{208}\text{Pb}/^{204}\text{Pb}$ values (Table 1 and Fig. 2a and b). All three were found at Canton Corralito and were identified as the result of San Lorenzo production based on elemental chemistry. The two singletons (Fig. 2a) are a Mojonea Black found at San Lorenzo that could not be assigned to any source based on elemental chemistry, and a La Mina white found at Canton Corralito and identified as a San Lorenzo product based on elemental chemistry. These two samples also differ from the main group based on their $^{208}\text{Pb}/^{204}\text{Pb}$ ratios, with higher values (Table 1 and Fig. 2b). In addition, two more singletons can be identified based on their lower $^{208}\text{Pb}/^{204}\text{Pb}$ only (Table 1 and Fig. 2b), and are a Remolino found at Tlapacoya assigned to the San Lorenzo White based on elemental chemistry, and an El Tigre found at San Lorenzo but unassigned based on elemental chemistry.

The strontium results show that 13 samples define a tight group with low isotopic ratio values and high strontium concentrations (Table 1 and Fig. 3). The 27 remaining samples show a large range of variation in both their isotopic composition and strontium concentrations (Table 1 and Fig. 3). The first group is composed of ceramics found at Tlapacoya and identified as the production from this site based on elemental chemistry, together with the Calzadas Carved and Limon Carved-Incised found at Canton Corralito, either identified as local products or left unassigned, based on elemental chemistry. The ceramics from Tlapacoya have lower isotopic values and higher strontium concentrations ($^{87}\text{Sr}/^{86}\text{Sr}$ range of 0.70434–0.70436, and $[\text{Sr}]$ of 481–587 $\mu\text{g g}^{-1}$) than those from Canton Corralito ($^{87}\text{Sr}/^{86}\text{Sr}$ range of 0.70440–0.70471, and $[\text{Sr}]$ of 359–481 $\mu\text{g g}^{-1}$). Among the Calzadas Carved and Limon Carved-Incised sherds identified as San Lorenzo production based on elemental chemistry, the samples found at the site have a relatively homogenous composition with $^{87}\text{Sr}/^{86}\text{Sr}$ (0.70800–0.70944) and strontium concentrations (150–311 $\mu\text{g g}^{-1}$). Those found at Canton Corralito, identified as San Lorenzo products or left unassigned based on elemental chemistry, have lower isotopic values (0.70603–0.70772) and display a larger range of concentrations (106–375 $\mu\text{g g}^{-1}$). The one Calzadas Carved found at Tlapacoya and associated with San Lorenzo production based on elemental chemistry has a higher isotopic ratio (0.71046) and lower Sr concentration (118 $\mu\text{g g}^{-1}$) than the samples found at San Lorenzo. The other types of ceramics (La Mina white, El Tigre, Remolino white, Mojonea black) and the figurines display larger ranges of isotopic composition (0.70695–0.71946) and concentrations (32–358 $\mu\text{g g}^{-1}$).

4.1. Discriminant analysis on the isotopic composition

The results described, so far, are based on a selection of isotopic ratios and strontium concentrations and suggest that the combination of Pb and Sr successfully identifies the production centers. A confirmatory discriminant analysis (DA) was performed using all isotope ratios and successfully assigned each sherd to its local site – the other samples were left unassigned. Further information on the DA is provided in Supplementary Material (Appendix A and B). Two canonical functions were found to be significant ($p < 0.05$) for the discrimination. The first function (CF1) is dominated by the $^{207}\text{Pb}/^{204}\text{Pb}$ values with shared contribution from $^{208}\text{Pb}/^{204}\text{Pb}$, $^{206}\text{Pb}/^{204}\text{Pb}$ and $^{87}\text{Sr}/^{86}\text{Sr}$ ratios (Appendix A, correlations). The second function (CF2), is only significantly associated to the $^{87}\text{Sr}/^{86}\text{Sr}$ ratios (Appendix A, correlations).

The CF1-CF2 scores plot shows a clear separation between types:

Tlapacoya and Canton Corralito sherds have negative (i.e. lower values for the isotope ratios) CF1 scores, while San Lorenzo sherds have positive (i.e. higher isotopic values) CF1 values (Fig. 4). San Lorenzo gray (chemical group composed of Calzadas Carved and Limon Carved-Incised) tends to show slightly lower CF1 scores than San Lorenzo white (chemical group composed of multiple ceramic types and figurines), the latter showing a larger heterogeneity in CF2 (i.e. $^{87}\text{Sr}/^{86}\text{Sr}$ ratios) values (four sherds plot close together, one shows a large negative score and two show large positive scores). CF2 mainly separates the Tlapacoya from the Canton Corralito sherds. Almost all previously assigned sherds were allocated to the group they belonged to. The only exception is a San Lorenzo gray (DCC572), which is isotopically closer to the San Lorenzo white group. The unassigned sherds were all allocated to the hypothetical group proposed based on their elemental composition. In the San Lorenzo group, most are isotopically closer to the San Lorenzo Gray, and only one (DCC585) is isotopically closer to the San Lorenzo white. This is the Mojonea Black that may represent a quite different source, given its large CF1 and CF2 positive scores (Appendix B). The reason for the allocation of the Mojonea Black into this type is the also relatively high scores for sherds DCC526 and DCC639. Overall, all samples showed a high probability (>0.8) of correct assignment to the site they were allocated based on NAA (Appendix B).

5. Discussion

The results obtained for the Calzadas Carved and Limon Carved-Incised are discussed separately from the isotopic results for the other types of ceramics and figurines. They are also discussed according to the previous groups identified based on elemental chemistry. In an attempt to further relate the ceramics to their possible clay sources, and therefore region of origin, the strontium signature of the ceramics is compared to relevant isotopic data for possible raw materials when available in the literature. Sufficient relevant data for lead isotopes are not available preventing a comparison at this time.

5.1. Calzadas Carved and Limon Carved-Incised - production centers, imports and unassigned samples

The lead isotopes clearly distinguish three groups amongst the Calzadas Carved and Limon Carved-Incised. The ceramics identified as locally produced at the three sites by NAA and recovered at their sites of origin present different lead isotopic compositions (Fig. 2a and Table 1). All of the samples found at Canton Corralito and Tlapacoya, and identified as imports from San Lorenzo by NAA (4/4), have a lead isotopic signature that corresponds with that of San Lorenzo production (Fig. 2a and Table 1). For these samples, the lead isotope ratios confirm the groups determined by elemental chemistry. These results show that lead isotopes allow for discrimination between different regional production centers, identification of imports, and the association of imported ceramics with their production center.

The strontium isotopic signature of the ceramics representing the three production sites demonstrates that Tlapacoya and Canton Corralito productions have very homogeneous compositions. Although they are similar to each other, they do not overlap and are clearly distinct from San Lorenzo production that presents higher $^{87}\text{Sr}/^{86}\text{Sr}$ ratio values (Fig. 3 and Table 1).

All of the Calzadas Carved and Limon Carved-Incised found at Canton Corralito that were left unassigned (9/9) but likely from Canton Corralito or San Lorenzo based on NAA, have lead isotopic compositions matching that of Canton Corralito and San Lorenzo, respectively (Fig. 2a and Table 1). Lead isotopes clearly allocate their origin to ceramics that could not be assigned based on their elemental composition. It also indicates that whatever process is responsible for an alteration of the elemental signature of these samples, it did not affect the lead isotopic composition of the ceramics. The combination of the isotopic and elemental analyses therefore constitutes a more robust approach to

sourcing ceramics in a Mesoamerican context.

The samples found at Canton Corralito and designated as unassigned-likely from Canton Corralito based on NAA (4/4) all have a strontium isotopic signature that is compatible with that of Canton Corralito (Fig. 3 and Table 1). For these samples, Sr isotopes also help in assigning the site of origin. The samples found at Canton Corralito and designated as unassigned-likely from San Lorenzo based on NAA (5/5), however, all present $^{87}\text{Sr}/^{86}\text{Sr}$ ratio values that are systematically lower than those of the San Lorenzo gray group found at San Lorenzo. This observation is addressed further below (Fig. 3 and Table 1).

Both, Tlapacoya and Canton Corralito are surrounded by a similar and homogeneous geology dominated by Cenozoic volcanic rocks and sediments derived from their alteration. When clays are derived from the weathering of isotopically homogeneous minerals, as it is the case for Sr in young rocks (Cenozoic) of andesitic composition, they will have the same signature as the parent rock. This means that there will be no difference in the Sr isotopic composition between the clay fraction and residual minerals.

The $^{87}\text{Sr}/^{86}\text{Sr}$ values displayed by the ceramics produced at Tlapacoya fall within the range of the strontium isotopic signature of the rocks from the Mexican Volcanic Belt, which varies between 0.703004 and 0.70505 (Cantagrel and Robin, 1978; Moorbath et al., 1978; Verma, 1983, 2000; McBirney et al., 1987; Nixon, 1988; Verma and Nelson, 1989; Verma et al., 1991; Wallace and Charnichael, 1994). More specifically, part of the basaltic scoria, hornblende-dacite and mixed lava from the Iztaccihuatl volcano, located 15–20 km E-SE of Tlapacoya, have the exact same composition as the ceramics from Tlapacoya (Nixon, 1988). Rocks from the Iztaccihuatl volcano are also rich in Ni and Cr, elements that are present in much higher concentrations in Tlapacoya ceramics.

The $^{87}\text{Sr}/^{86}\text{Sr}$ values displayed by the ceramics produced at Canton Corralito fall within the range of the strontium isotopic signature of the Volcanic Highlands and the Pacific Coast, which varies between 0.70380 and 0.70492 (Hodell et al., 2004).

The geology around San Lorenzo is dominated by a larger diversity of sedimentary rocks, volcano-sedimentary rocks, and sediments of multiple origins. The main types of rocks present in the vicinity of San Lorenzo are terrigenous marine sedimentary rocks and volcano-sedimentary rocks of Tertiary age that include polymictic conglomerates, silica sand, sandstone, siltstone-claystone and rhyolitic tuffs, Quaternary alluvial deposits, and Quaternary sand, silt, clay sediments of lacustrine origin. Less strontium isotopic data are available for these deposits but a range of $^{87}\text{Sr}/^{86}\text{Sr}$ values corresponding to 0.7088–0.7120 has been measured in the area of La Merced (Robles-Camacho, 2006; Schaaf et al., 2012). The Tertiary-Quaternary Tuxtla volcanic field, about 30 km N of San Lorenzo, presents lower $^{87}\text{Sr}/^{86}\text{Sr}$ values, between 0.703004 and 0.704202 (Nelson et al., 1995). Limestones and dolomites of Jurassic and Cretaceous age can be found 40–50 km S-SE of San Lorenzo. Older rocks from the Proterozoic and Paleozoic can be found 70–80 km from San Lorenzo in S-SW direction in the Guichicovi complex and the Mixtequita batholith. Based on the curve of the isotopic evolution of Sr in sea water, Jurassic-Cretaceous marine carbonates have $^{87}\text{Sr}/^{86}\text{Sr}$ values between 0.7068 and 0.7080 (e.g., McArthur et al., 2001). Part of the rocks from the Proterozoic metamorphic Guichicovi complex display highly radiogenic Sr signatures with values up to 0.803357 for $^{87}\text{Sr}/^{86}\text{Sr}$ (Weber and Köhler, 1999). The terrigenous fraction of the marine deposits formed during the Miocene has a crustal signature that could also range above the values measured for La Merced. The Quaternary sediments draining these different lithologies will therefore have a variable Sr signature along the drainages. Clay sources resulting from the alteration of these different formations are likely to have variable signatures as well. The strontium isotopic composition of the Calzadas Carved and Limon Carved-Incised produced at San Lorenzo and found at the site overlaps with that of values from the area of La Merced.

These comparisons show that materials produced and found at the

three sites have strontium signatures compatible with those of the raw materials locally available at their surroundings.

The $^{87}\text{Sr}/^{86}\text{Sr}$ ratio values of the samples excavated at Canton Corralito and identified as imports from San Lorenzo or unassigned based on elemental chemistry are, however, systematically lower than those found at San Lorenzo (Fig. 3 and Table 1). Two Limon Carved-Incised samples also show lower $^{208}\text{Pb}/^{204}\text{Pb}$ and $^{206}\text{Pb}/^{204}\text{Pb}$ values. The sample found at Tlapacoya identified as part of the San Lorenzo gray production also differs from the local San Lorenzo products, showing a higher $^{87}\text{Sr}/^{86}\text{Sr}$ ratio value. This is likely linked to the use of broader diversity of the raw materials around San Lorenzo or the Gulf Coast more largely. The region indeed displays multiple sedimentary and volcano-sedimentary deposits and is drained by diverse systems of rivers and streams that cross a diverse range of rocks of widely different ages and compositions that includes Proterozoic metamorphic rocks, Proterozoic-Paleozoic igneous rocks (granite-granodiorite), Jurassic and Cretaceous sedimentary rocks (limestone, dolomites, lutites, etc.). The carbonate fraction of marine rocks of Cretaceous-Jurassic age has a strontium isotopic composition that varies between 0.7068 and 0.7080 (e.g., McArthur et al., 2001). The Quaternary alluvium derived from weathering is therefore likely to present a somewhat variable mineralogy along the course of these drainages. The variability of the Sr isotopic composition could be related to the use of multiple clay sources or that of a type of clay naturally presenting a heterogeneous mineralogy. Another possibility is that different processes, such as grain-size selection or the addition of temper, could have been applied to the raw materials during the ceramic production process. Petrographic analyses conducted on San Lorenzo ceramics revealed that some of them contained volcanic glass, and that bentonites (clays derived from volcanic material) had been added to the ceramic clays (Guevara, 2004). Rocks of volcanic origin can be found 50–60 km N-NW of San Lorenzo along the Gulf Coast in the Tuxtla volcanic field, and these present low $^{87}\text{Sr}/^{86}\text{Sr}$ ratio values varying between 0.703004 and 0.704202 (Nelson et al., 1995). The use of raw materials naturally derived from this volcanic complex, or mixing of these materials with clays used for the San Lorenzo local production, would cause a shift of the $^{87}\text{Sr}/^{86}\text{Sr}$ ratio towards lower values. The rhyolitic tuffs present in the immediate vicinity (5–10 km) of San Lorenzo could have also been the source for the volcanic material found in the ceramics produced at the site. These hypotheses assume that the isotopic signature is controlled by the mineralogy.

Strontium isotope ratios of San Lorenzo-derived gray pottery found at Canton Corralito may imply a role for diagenetic modification of the Sr isotope signature. All eight Calzadas Carved and Limon Carved-Incised sherds found at Canton Corralito and identified as San Lorenzo imports based on elemental chemistry and/or lead isotopes have $^{87}\text{Sr}/^{86}\text{Sr}$ ratio values that are intermediate between local Canton Corralito and San Lorenzo (Fig. 3 and Table 1). Three of the Limon Carved-Incised have lower $^{87}\text{Sr}/^{86}\text{Sr}$ values together with higher strontium concentrations, when compared to the San Lorenzo Gray group found at San Lorenzo. These samples therefore display a pattern that is compatible with that of a mixing of strontium from two different sources, with one end-member representing the original composition of San Lorenzo Gray and the second representing the original signature of Canton Corralito production (i.e. as the proportion of Sr with the lower $^{87}\text{Sr}/^{86}\text{Sr}$ ratio increases the ratio decreases). In this model, we assume that the ceramics produced at Canton Corralito have the same strontium isotopic and elemental composition as the soils they are buried in. This is plausible considering the local geological context, the sediments developed on the Coastal Plain being derived from the weathering of the Cenozoic volcanic rock from the highlands and the groundwater percolating through local rocks and sediments also showing the local signature. In this interpretation, local strontium is hypothesized to have been added by re-precipitation in the pores of the ceramics during 3000+ years of burial at Canton Corralito. The other five samples (three Limon Carved-Incised and two Calzadas Carved), also have lower $^{87}\text{Sr}/^{86}\text{Sr}$ values than the San Lorenzo gray, but their strontium concentration is either similar

to or lower than that of the local San Lorenzo Gray group (in fact, their Sr/Rb ratios, 1.3–2.7, are within the range found for San Lorenzo gray ceramics, 1.2–3.6). Thus, if diagenesis is responsible for their lower isotopic signature, it would have to be through a more complex process of successive leaching and re-precipitation. The alternating wet and dry conditions at Canton Corralito are compatible with this hypothesis. (Two of the Calzadas Carved-Incised, as indicated above, also have different $^{208}\text{Pb}/^{204}\text{Pb}$ and $^{206}\text{Pb}/^{204}\text{Pb}$ values, which more likely points at different sources for these two samples). The hypothesis that diagenesis could have affected the strontium isotopic ratio of some of the ceramics deserves further analytical exploration, but such experimentation is beyond the scope of the present study. The data presented here, nonetheless, clearly highlight the importance of considering possible mobility issues when using geochemistry (elemental or isotopic) to identify the provenance of pottery, as is commonly the case for more porous material such as bones, for example. They also plead in favour of multiproxy approaches to reach strongly supported conclusions; in the case presented here, lead isotopes appear to constitute the proxy that is the most robust to post-burial impact on the ceramic chemical composition.

We are left with two distinct hypotheses about the San Lorenzo ceramic export industry. On one hand, the strontium isotopic ratios, and to a lesser extent both the $^{208}\text{Pb}/^{204}\text{Pb}$ and $^{206}\text{Pb}/^{204}\text{Pb}$ ratios, may show that local San Lorenzo products present a smaller range of isotopic variability than the San Lorenzo products exported to the other sites. Assuming these results accurately reflect compositional properties at the time of production, potters producing for export utilized different raw materials and/or processing practices compared to potters producing for local consumption; which has fascinating implications in terms of the nature of objects and symbols that were exported beyond San Lorenzo. Alternatively, diagenesis has altered the initial strontium content, which consequently affected the strontium isotopic composition. This second hypothesis calls for an evaluation of the possible impact of post burial processes on the isotopic signature of strontium when using this approach to source ceramics. This especially holds true in environments where very wet or alternating wet and dry conditions might be suspected to promote diagenetic replacements.

5.2. Other types and figurines

Five samples have a lead composition that corresponds with that of the Calzadas Carved and Limon Carved-Incised from San Lorenzo (San Lorenzo Gray chemical group) (Fig. 2 a, b and Table 1). Four of them belong to the San Lorenzo White chemical group and one was unassigned based on NAA results. For these five samples, the lead isotope signature appears again to support and complement previous results based on elemental chemistry. The $^{87}\text{Sr}/^{86}\text{Sr}$ ratio values, however, display larger variation with two samples presenting a signature compatible with that of San Lorenzo Gray group, and three presenting variable and higher $^{87}\text{Sr}/^{86}\text{Sr}$ ratio values (Fig. 3 and Table 1). One additional sample, a figurine that belongs to the San Lorenzo White chemical group, and to the sub-group identified based on the $^{207}\text{Pb}/^{204}\text{Pb}$ ratio, has lower values for $^{87}\text{Sr}/^{86}\text{Sr}$, $^{208}\text{Pb}/^{204}\text{Pb}$ and $^{206}\text{Pb}/^{204}\text{Pb}$ ratios. Finally, four of the samples have a different lead isotopic composition and are identified as singletons and discussed below. The elemental signature of this group has a larger range of variation than the San Lorenzo Gray group (Blomster et al., 2005), and the strontium isotopic results seem to display the same pattern. The majority of the samples from the San Lorenzo White group (7/10) have much lower Ca concentration, below $3000\text{ }\mu\text{g g}^{-1}$ (Cheetham, 2010), than the Calzadas Carved and Limon Carved-Incised (San Lorenzo Gray) samples. The variation observed in both signatures (elemental and Sr isotopes) of these objects is likely due to variation in the mineralogy of the raw materials used for their production. As mentioned above, the San Lorenzo White group includes multiple types of ceramic vessels and figurines, and a diversity of clays are available in the region around San

Lorenzo. The strontium signature of one figurine found at San Lorenzo, and one La Mina White found at San Lorenzo can be explained by the use of clay sources similar to those used for the production of San Lorenzo Gray. The low $^{87}\text{Sr}/^{86}\text{Sr}$ ratio value of a figurine found at Canton Corralito can be explained by the same hypothesis suggested for the San Lorenzo Gray group found at Canton Corralito, and discussed above that suggests the use of a different source for the material that was exported. A figurine found at San Lorenzo has a signature compatible with the general range of values for the La Merced area. The last two, a figurine found at San Lorenzo and a La Mina White found at Canton Corralito, have higher values, as shown in Fig. 3 insert, that are best explained by the use of clays derived from the Proterozoic and Paleozoic rocks, such as those present in the Guichicovi complex, about 70–80 km S-SW of San Lorenzo. The terrigenous fraction of Miocene rocks could also present relatively high $^{87}\text{Sr}/^{86}\text{Sr}$ values. It is worth noting that the kaolinite clay sources available in the region of San Lorenzo have been suggested as the source for the production of white ceramics at San Lorenzo (Coe and Diehl, 1980), and the kaolinite sources from Concepción formation were found to be high quality clays (Pool, 1990). Moreover, the clay sources derived from the uplifted Tertiary marine deposits present at the foothills of the Tuxtla volcanic field were used for the production of ceramics during the Classic period (Pool, 1990; Stoner et al., 2008). The clays from the Tuxtla region would therefore also deserve further isotopic investigation in view of identifying the source of some samples from the San Lorenzo White group. In the absence of data on these multiple sources, however, the two samples cannot be further assigned. Although their elemental and lead isotopic signature relate these two samples to San Lorenzo, the strontium isotopes indicate the use of different type of clays, or mixing of clays, whose origin could be located about 70–80 km S-SW from San Lorenzo. The Sr isotopic variation observed in this assemblage of objects may be related to the use of different sources for the production of the different objects, for some samples it could reside in the use of different tempers, and/or to use of different processing techniques, or to some combination of these effects. For a La Mina White and a figurine, the analysis of additional clays and ceramics representing these different types using the combination of analyses proposed here should help elucidate their provenance and circulation.

5.3. Singletons

Four singletons have a lead isotopic compositions that differ from the main San Lorenzo group, indicating that they were made from different clay sources (Fig. 2 a, b and Table 1). Two are unassigned samples but likely from San Lorenzo (a Mojonera Black and an El Tigre) and two are samples assigned to San Lorenzo White group (a La Mina White and a Remolino), based on NAA results. The Mojonera Black sample that was unassigned by NAA but proposed as likely being from San Lorenzo, has strontium and lead isotopic signatures that are incompatible with those of San Lorenzo products. This indicates that it was made using a different type of clay. The Mojonera black also has a unique elemental chemistry presenting high Fe, Sc, Rb and K levels but low Ca (no Sr was measured in this sample) that would be compatible with weathering of low Ca-granite, for example. The higher $^{87}\text{Sr}/^{86}\text{Sr}$ ratio value for the Mojonera Black is compatible with that of older rocks from the Proterozoic and Paleozoic, or their alteration products, which can be found 70–80 km S-SW from San Lorenzo. These sources are located about 30 km E-SE of Las Limas. The La Mina White has higher values for all three Pb ratios and was clearly made using a different clay source with more radiogenic values. The Remolino has a higher $^{87}\text{Sr}/^{86}\text{Sr}$ that is compatible with the signature of La Merced but has a lower $^{208}\text{Pb}/^{204}\text{Pb}$ value which could point to a source with a different U/Th ratio. Finally, the El Tigre sample is compatible with San Lorenzo production based on $^{87}\text{Sr}/^{86}\text{Sr}$, $^{207}\text{Pb}/^{204}\text{Pb}$ and $^{206}\text{Pb}/^{204}\text{Pb}$ ratios, but has lower for $^{208}\text{Pb}/^{204}\text{Pb}$ value which also indicates a different source. In the absence of Pb data for possible raw materials, no source can further be proposed for these last three samples.

Based on the results presented here, lead isotopes appear not only to discriminate between the three production centers, but also to assign many specimens that remained unassigned based on elemental chemistry. Indeed, virtually all of the previously unassigned specimens were matched to sources based on lead isotopes. Strontium isotopes also clearly assign the samples that were left unassigned and found at their site of production. Lead isotopic ratios are also more resistant to processes possibly affecting the clay during ceramic production than both strontium isotopes and elemental chemistry. The strontium isotopes, however, appear more useful for identifying the various raw material sources within the Gulf Coast area and/or processes applied to clay sources during the production of ceramics. But in addition, for part of the samples, the possible impact of diagenesis has to be examined. Comparison of ceramics with raw materials can provide further evidence useful in identifying the region of provenance. These results demonstrate that the combination of the two isotopic systems constitutes an excellent approach in the identification of ceramic provenance and circulation in the context of Early Formative Mexico and that the integration of the isotopic approach with elemental analyses will help resolve crucial questions about ceramic circulation patterns.

6. Conclusions and perspectives

The combination of the two isotopic systems (Pb and Sr) clearly enables the identification of the three production centers and supports previous results that were based on elemental chemistry. Samples that previously could not be assigned based on their elemental chemistry can be related to their production center using lead isotopic and for the most part using strontium isotopic ratios as well; this makes the combination of the elemental and isotopic approaches a powerful and more efficient tool for determining the provenance of ceramics than stand-alone approach. The isotopic signature of ceramics can also be used to establish a link with the raw material used for their production, which provides further insights into the provenance of the material. In the present case, some of the Calzadas Carved and the Limon Carved-Incised wares produced at San Lorenzo were made from clays presenting a variable chemistry/mineralogy. The strontium isotopes reflect that variability, while lead isotopes do not or are little affected. The robustness of lead isotopes in the face of different clay preparation processes was previously observed on fine Late Bronze Age pottery ware from Cyprus (Renson et al., 2013a). However, the strontium isotopic composition provides more insights into the possible sources used for the ceramic typologies and figurines belonging to the San Lorenzo White group than do lead isotopes, with the identification of possible sources located up to 70–80 km from the site. This chemical group was identified as presenting more elemental chemistry variability than the San Lorenzo gray group, composed of Calzadas Carved and Limon Carved-Incised. The combination of lead and strontium isotopes provides a powerful method for identifying and explaining variability within a group. In the case of the San Lorenzo White group, the use of isotopic analyses in combination with elemental chemistry will improve our capacity to identify the diverse sources used for their production, and will help clarify the circulation of these objects. This should hold true for the identification of any ceramic ware of unknown or debated origin in Mesoamerica, and the characterization of the isotopic signature of clay sources across the region should constitute a crucial contribution. The integration of the isotopic approach is a powerful tool to address current hypotheses for ceramic provenance in Mesoamerican contexts and the nature of inter-regional interaction.

Author contribution statement

V. R., H. N. and M. D. G. designed research, D. C. and J. P. B. provided material and context, V. R. performed isotopic analyses, V. R. and A. M. C. performed data analysis, V. R., H. N. and A. M. C. wrote the manuscript with contribution from all other co-authors.

Declaration of competing interest

The authors declare no conflict of interest.

Acknowledgments

We thank E. Peacock, J. Alvey and F. De Vleeschouwer for comments on previous versions of the manuscript. The acquisition of the Nu Plasma II MC-ICP-MS was funded by the National Science Foundation (grant #BCS-0922374). The Archaeometry Laboratory is supported by the National Science Foundation (grant #BCS-1912776). V. Renson was partly supported by a postdoctoral fellowship of the Belgian American Educational Foundation (BAEF). We thank the editor and three anonymous reviewers for their constructive comments.

Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.jas.2020.105307>.

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