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High precision isotopic analyses of lead ores from New Mexico by MC-ICP-MS: implications for tracing the production and exchange of Pueblo IV glaze-decorated pottery

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ABSTRACT

Between ca. 1275 and 1700 CE, Pueblo groups in the northern Southwest United States produced and exchanged ceramic bowls decorated with lead-based glaze paints. Previous studies of these glaze-decorated bowls have used lead isotopic analysis by ICP-MS to identify the sources of lead used by Pueblo potters, and investigate how social or economic factors may have influenced resource use among different Pueblo communities (e.g. Habicht-Mauche et al., 2000, 2002; Huntley et al., 2007; Huntley, 2008). However, interpretations of much of this isotopic data have remained provisional because of overlap among the isotopic ratios of potential sources and because the isotopic composition of many glaze paints do not clearly match any known source. Here, we use multi-collector ICP-MS to re-measure the lead isotopic composition of 48 samples of lead sulfide (galena) and lead carbonate (cerussite) from sources in New Mexico that were potentially utilized by Pueblo potters, including mines within the Cerrillos Hills, Magdalena, Hansonburg, and Joyita Hills mining districts. These results define the isotopic composition of lead ores from these districts with greater precision and accuracy than achieved in previous studies and better distinguish among these mining districts in lead isotope space. Most significantly, we find that galena mineralization within the Cerrillos Hills only has a modest degree of isotopic variation, with $^{206}Pb/^{204}Pb$ ratios from 18.508 to 18.753, $^{207}Pb/^{204}Pb$ ratios from 15.580 to 15.607, and $^{208}Pb/^{204}Pb$ ratios from 38.388 to 38.560. These ranges are far narrower than previously reported, and should supersede previously published values for this district. In total, we conclude that isotopic measurements of both ores and glaze paints made by MC-ICP-MS will provide new information about the provenance of lead in glaze paints and allow for more detailed interpretations about resource procurement and exchange in the Pueblo world.

1. Introduction

Glaze-decorated ceramics were both produced and widely exchanged by Pueblo groups in the northern Southwest United States (U.S.) between ca. 1275 and 1700 CE. Lead (Pb), an effective flux which lowers the melting points of silica and alumina, is a major component of many glaze decorations, often present at concentrations of at least 35–50% by weight (Habicht-Mauche et al., 2000; Huntley et al., 2007; Huntley, 2008).

Since 2000, lead isotopic measurements of glazes have been employed to infer the sources of lead ore used by Pueblo potters in the Galisteo Basin (Habicht-Mauche et al., 2000, 2002; Nelson and Habicht-Mauche, 2006), Salinas area (Huntley et al., 2007) and Zuni region (Huntley, 2008) of New Mexico (Fig. 1). Determining the sources of raw materials used by Pueblo potters provides information about the interactions between communities that produced glaze wares across eastern Arizona and New Mexico, and the social and economic patterns that influenced resource use. Isotopic comparisons of glaze paints and ores indicate that certain mining districts, such as those located in the Cerrillos Hills and Magdalena Mountains, may have been particularly important to Pueblo potters (Fig. 1, Habicht-Mauche et al., 2000, 2002; Huntley et al., 2007; Huntley, 2008). However, interpretations of lead procurement remain provisional because published isotopic ratios of glaze samples often do not precisely match the previously published

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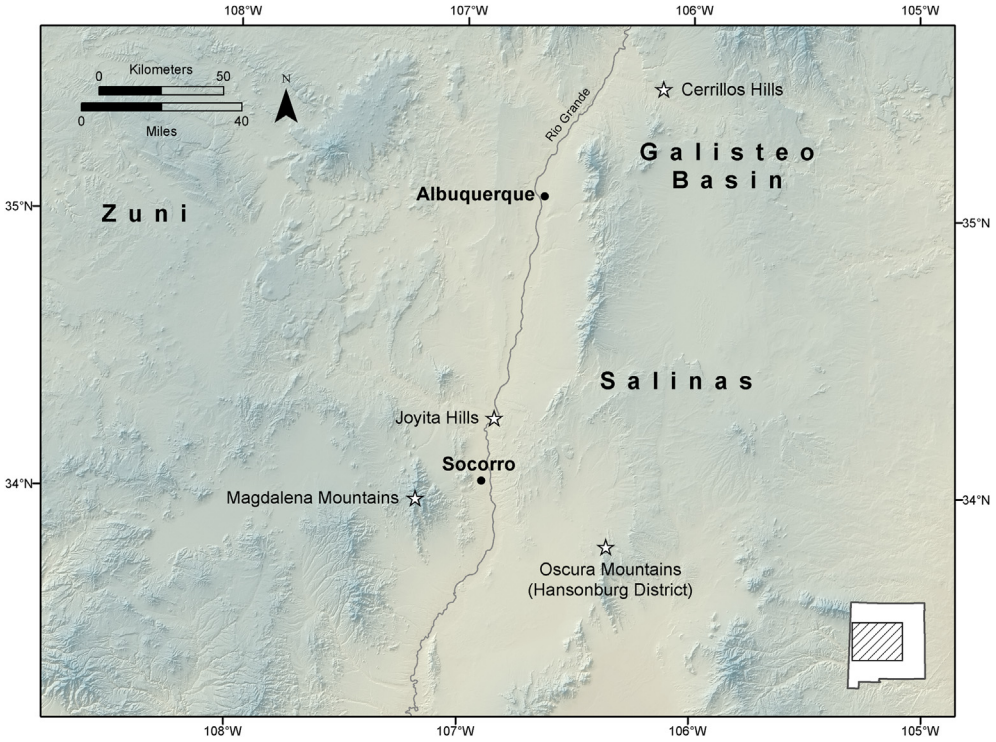

Fig. 1. Location of ore deposits sampled for this study and regions of glaze ware production discussed.

ratios of any of the potential ore sources, or cannot be clearly assigned to a single source (Huntley et al., 2007; Huntley, 2008). These uncertainties have been attributed to (a) the prehistoric use of ore sources which have not yet been characterized with lead isotopes, (b) the mixing of ores from different sources to make glazes, and (c) isotopic overlap among the ore sources.

The advent of multi-collector inductively coupled mass spectrometry (MC-ICP-MS) in the late 1990s now makes it possible to routinely generate high precision measurements of lead isotopic ratios. However, this technique has never been applied to measure the isotopic composition of either lead ore from the potential sources used by Pueblo potters, or lead-rich glazes found on Pueblo IV ceramics. Our goal in this paper is to demonstrate that high-precision lead isotopic measurements can provide much tighter clusters of isotopic ratios for lead sources than were achieved by earlier techniques.

In the present study, we use MC-ICP-MS to re-measure the lead isotopic composition of galena (PbS) and cerussite ($PbCO_3$) from the Cerrillos Hills, Hansonburg, Joyita Hills, and Magdalena districts in New Mexico (Fig. 1). All these areas are considered to be possible sources of lead ores used by potters in the Galisteo, Salinas, and Zuni Pueblos (Habicht-Mauche et al., 2000, 2002; Huntley et al., 2007; Huntley, 2008). Among these areas, we place particular emphasis on characterizing ores from the Cerrillos Hills because of the district's established importance as a site of ancient lead and turquoise mining in the Southwest U.S. (Schroeder, 1979; Warren and Weber, 1979; Warren and Mathien, 1985; Mathien, 1998; Bice et al., 2003).

We compare our results using MC-ICP-MS to isotopic measurements that have been made on the same ore deposits using single-collector ICP-MS (Habicht-Mauche et al., 2000, 2002; Huntley et al., 2007) and thermal ionization mass spectrometry (TIMS) (Ewing, 1979; Habicht-Mauche et al., 2002; Stacey and Hedlund, 1983). These new measurements, which are made on the same samples used by Habicht-Mauche et al. (2000), Habicht-Mauche et al. (2002), and Huntley et al. (2007), allow us to (a) narrow the range of lead isotopic ratios associated with these ore deposits, (b) re-visit questions of isotopic overlap among sources and (c) consider how regional geologic controls influence the isotopic signature of lead ores across these regions of New Mexico and thus impact studies of lead provenance. The results indicate that high-precision lead isotopic measurements are likely to both simplify and revise interpretations about the source of lead used to create glaze-decorated ceramics, and thus advance our knowledge of interaction and exchange among Pueblo communities in the Pueblo IV and protohistoric Southwest.

2. Advantages of MC-ICP-MS

Isotopic ratios of lead in archaeological materials were first measured by TIMS (Brill and Wampler, 1967; Gale and Stos-Gale, 1982), but the technique requires laborious and expensive sample preparation and offers relatively low sample throughput. Single collector ICP-MS was developed in the 1980s and 1990s (Vanhaecke et al., 2009) and presented a quicker and lower cost alternative for isotopic determinations. Previous studies of lead provenance in southwestern glaze paints have relied upon the determination of isotope ratios of both glazes and ores by solution analyses via single collector magnetic sector high-resolution (HR)-ICP-MS.

While single-collector ICP-MS has advantages over conventional TIMS measurements in terms of both the cost and efficiency of lead isotope analysis, the precision of the analyses is worse (Vanhaecke et al., 2009). Habicht-Mauche et al. (2002) demonstrate the limitations of the HR-ICP-MS dataset on ores from New Mexico by comparing the lead isotopic ratios of three ore samples (all from the Mina del Tiro mine, in the Cerrillos Hills) measured using three different techniques: acid dissolution HR-ICP-MS, laser ablation (LA)-HR-ICP-MS, and TIMS. Of these analyses, the measurements made by TIMS have the least scatter and are the most accurate representation of the true isotopic signature of ore from the Mina del