CHAPTER 12

LEAD, SILVER AND COPPER ACQUISITION NETWORKS

CHAPTER INTRODUCTION: METALS

Of the many rock and mineral varieties found at Indus Civilization sites, archaeologists have by far expended the most time and effort in the study of artifacts made of metals, in particular copper (for recent overviews see Agrawal 2000; Agrawal and Kharakwal 2003; Biwas 1996; Chakrabarti and Lahiri 1996; Kenoyer and Miller 1999; Shrivastava 2006). Despite all of the attention, attempts to correlate metal artifacts with their potential sources have been few and limited in nature. Sana Ullah (1940) compared the bulk chemical compositions of copper ores from deposits in Afghanistan and India to copper artifacts from Harappa and suggested that, based on the presence of nickel and arsenic, Harappan copper was probably derived from sources in Rajasthan. Much later Hegde and Ericson (1985) made seven Pb isotope assays on samples of chalcopyrite and galena from deposits in Rajasthan. Although their work was an important first-step for provenience research of this kind, they did not follow up with analyses of additional sources samples or, more importantly, of artifacts that could have been correlated with potential ore sources.

In an effort to advance this line of research another step forward, in this chapter, I compare metal artifacts from Harappa and eight other prehistoric sites to various metal ore deposits in Pakistan, India, Oman and Iran using *lead isotope analysis*. Geologists and archaeologists working in these areas have already isotopically assayed many of the lead, lead-silver and copper sources that would have been accessible to the ancient peoples of these regions. Here, previously published lead isotope data are compiled and presented along with the results of nearly 150 new assays of geologic source samples conducted specifically for this study. Together, these provide reasonably representative geologic databases, which I use to make provisional geologic provenience determinations for 86 metal artifacts or archaeological ore minerals composed of or containing lead. I have determined that, among other things, residents of Harappa acquired lead resources from deposits in at least three regions: Jammu and Kashmir, southern Balochistan and one other source area that has yet to be identified.

I begin this chapter by reviewing some of the forms in which the element lead (denoted by the abbreviation "Pb" - from the Latin "plumbum") is found in both nature and in archaeological materials. A discussion of Pb isotopes and the technique used in this study to sample and measure them follows. Next, an explanation of how isotopic data are evaluated and displayed is provided. At that point, the Pb isotope database for lead deposits is presented in a region-by-region overview of isotopically assayed ore occurrences. After the potential lead sources have been evaluated and readied for comparison, the results of Pb isotope assays made for all of the lead ores, finished artifacts, slags and residues from Harappa are plotted in relation to them and provisional geologic provenience determinations are made. Assays of lead and/or silver artifacts from the sites of Mohenjo-daro, Allahdino, Nagwada, Gola Dhoro, Mehrgarh, Nausharo, Mundigak and Shahr-i-Sokhta are also evaluated in relation to the lead database. Finally, seven archaeological copper ore fragments from Harappa are compared to a second Pb isotope

database put together from published data and new analyses of copper ores and slags collected previously unassayed copper deposits. In the conclusion of this chapter, I briefly discuss how these results inform our understanding of the lead, silver and copper acquisition networks in which Indus Civilization peoples, especially those at Harappa, were involved.

LEAD, LEAD ARTIFACTS AND ARTIFACTS CONTAINING OR DERIVED FROM LEAD

Lead, like most other metal elements (gold excepted), is relatively uncommon in its native state and instead is usually found mineralized in sulphide deposits (Rapp 2002: 140). Within those deposits its most common form is the mineral galena – lead sulphide (PbS) (Deer *et al.* 1992: 604). Galena frequently co-occurs with sphalerite (zinc sulphide) and also sometimes contains significant amounts of copper, silver and antimony (ibid.). Cerussite (PbCO₃), anglesite (PbSO₄) and massicot (PbO) are all common alteration products of galena and are typically found along the weathered margins (gossan) of lead deposits that are adjacent to calcareous rocks (Guilbert and Park 1986: 811).

Archaeologically, lead has been found in several forms at Indus Valley and Balochistan Tradition sites. Finished items made of this metal include small vessels and utensils, hooks, cones or "plumb bobs," rods, rings and ingots (Francfort 1989: 147-148; Kenoyer and Miller 1999: 119; Mackey 1938: 453; Lal 1985: 656; Ratnagar 2004: 197). Raw, unmodified lead ores of various kinds have been found at several sites as well as lead slags and non-descript lumps (Hargreaves 1929: 33; Mackay 1943: 188; Marshall 1931c: 30). Lead residues that may be the remains of cosmetics or pigments are also known (Sana Ullah 1931: 691).

Even though it is not readily apparent, the

element lead is often present in objects made predominantly of other metals. For instance, it occurs as a natural impurity in copper ores such as chalcopyrite (Deer et al. 1992: 595) and copper objects made from such ores usually retain trace amounts of lead. In addition, lead is often deliberately added as an alloy during manufacture of copper alloy items (Agrawal 1971). Metals extracted from lead ore might also be expected to retain a trace amount of the element. The term argentiferous describes metals ores (lead or copper) with a high silver content. Silver artifacts have been found at many Indus Civilization sites and it has often been suggested that much of that metal may have been derived from the smelting of argentiferous lead ores (Asthana 1993: 276; Ratnagar 2004: 193; Pascoe 1931: 675; Sana Ullah in Mackay 1938: 599).

Determining the sources of lead, whether it occurs as a finished item, a raw ore, residue or trace component of an artifact made of another metal like silver or copper, can provide important insights into long-distance acquisition networks in which the residents of Harappa were involved. As is demonstrated below, it is possible to determine the geologic provenience of lead artifacts (and perhaps copper and silver ones too) using lead isotope analysis (Agrawal 2000: 30; Ratnagar 2004: 195).

LEAD ISOTOPE ANALYSIS

The element lead (Pb) has four isotopes (²⁰⁸Pb, ²⁰⁷Pb, ²⁰⁶Pb, ²⁰⁴Pb) that vary in absolute amounts depending on its geologic age and the conditions in which it mineralizes (Guilbert and Park 1986: 286-90). Thus, Pb isotope analysis has long been employed in the geosciences to determine the age and evolutionary environment of ore deposits (Doe and Zartman 1979). This analytical technique has also been effectively applied to the study of archaeological metals over the past 30 years (Lambert 1997: 188-191).

The main reason for this is that Pb isotopes do not undergo physiochemical fractionation when an ore is smelted or when the extracted metal is fashioned (and re-fashioned) into finished products (Budd *et al.* 1995: 127). Thus, an artifact containing lead from a *single* deposit will retain the original isotopic composition of that deposit. Although it is a problematic possibility that alloyed and/or recycled metal artifacts may contain lead derived from multiple sources, Pb isotope data are, nonetheless, extremely useful for archaeological studies attempting artifact-to-ore source correlation.

Measuring the abundance of the four isotopes of lead within a geologic or archaeological sample is done with a mass spectrometer. Until fairly recently, the majority of Pb isotope analyses were conducted using a thermal ionization mass spectrometer (TIMS) - an instrument which still today produces the most accurate and precise measurements (Pomies et al. 1998). All of the previously published isotope values for geologic samples presented in Appendix 12.1 were made using a thermal ionization mass spectrometer (TIMS). Measuring Pb isotopes using an inductively coupled plasma mass spectrometer (ICP-MS) is, however, becoming increasingly more common (Pollard and Heron 1996). Although the analytical error factor for ICP-MS isotope measurements is somewhat larger than for TIMS, data derived using both techniques can be, and have been, used together effectively (Attanasio et al. 2001; Ingo et al. 1997).

EDTA SAMPLING OF LEAD AND SILVER ARTIFACTS AND ANALYSIS USING ICP-MS

Most of the new Pb isotope assays of geologic and archaeological lead and silver samples made for this study were conducted using the ICP-MS at the LARCH. A handful of analyses were made using a Neptune multi-collector ICP-MS at the Keck Isotope Laboratory, UC-Santa Cruz. In order to get lead from samples into a liquid solution for analysis, a new technique (Figure 12.1) was used that employs ethylenediaminetetraacetic acid (EDTA) a hexadentate chelating agent that forms coordinate bonds with lead atoms (Law and Burton 2008; Law and Burton 2006a; Reslewic and Burton 2002). A non-toxic sampling solution is prepared that consists of ultra-pure water and 0.05% dissolved EDTA (Figure 12.1 A). A lead artifact that has previously been cleaned in purified water is placed into a disposable plastic sampling tray and approximately 50 ml of the sampling solution is poured into the tray, immersing the artifact (an inscribed lead bar from Harappa [H2000/2174-321] is pictured in Figure 12.1 B). The sample remains in the solution for two minutes while the tray is lightly agitated at 20 second intervals. This short immersion time is sufficient to extract lead from a sample in concentrations from 100 ppb to as much as 100 ppm - orders of magnitude more than minimally required for ICP-MS analysis. After two minutes, the now lead-enriched solution is poured into a sample vial for return to the lab (Figure 12.1 C). The artifact is rinsed in ultra-pure water, allowed to dry and then returned to its place of storage or display. Although this sampling method is technically destructive (Pb atoms form 1:1 bonds with the EDTA and are removed from a sample), the short immersion time in the sampling solution does not result in any macroscopic alteration of lead or silver artifacts whatsoever.

Lead-enriched solutions sampled from archaeological and geologic materials (sites and locations to be discussed) were returned to Madison and analyzed on the ICP-MS in groups of five. For accurate counting statistics, 107 counts per second (approx. 10 ppb) were required against a background of approximately 104 counts per second. An EDTA-sampled solution of the NIST Common Lead Isotopic Standard (SRM 981) was assayed before and after each group run. Based on repeated measurements (n=44) of this standard, the overall *precision* (Figure 12.1 D) – an expression of analytical error based on the relative standard **Figure 12.1** A technique for non-destructive Pb isotope sampling and analysis of lead and silver artifacts using ethylenediaminetetraacetic acid (EDTA) and ICP-MS.

A. A non-toxic solution of purified water and 0.05% EDTA is taken to the site or museum at which a lead or silver artifact is stored.



B. The artifact is placed in a small tray; the EDTA solution is poured over it and is then left to sit for two minutes.



C. The now lead-enriched solution is put into a sample vial and the artifact is washed, dried and returned to the collection.



D. Solutions are analyzed with an ICP-MS.
The analytical precision (below) of the
instrument was determined by repeated
measurements of NIST Lead Isotopic
Standard SRM 981.

	208/207	207/206	207/204
NIST Standard	2.3696	0.9150	15.4970
mean of 44 measurements	2.3724	0.9157	15.5544
SD of mean	0.004384	0.00175	0.13352
Precision (%RSD)	0.19%	0.19%	0.86%

E. The **accuracy** of this technique was gauged (right) by analyzing ore samples from deposits that had already been well characterized using TIMS. EDTA/ICP-MS derived isotope values for samples from the Zawar and Rajpura-Dariba/Rampura-Aguchalead bodies closely matched previously published (Deb et al. 1989) TIMS data.



deviation (%RSD) of all of those measurements, was calculated to be under 0.2% for ratios constructed (discussed below) using the isotopes ²⁰⁸Pb, ²⁰⁷Pb and ²⁰⁶Pb. It was significantly higher (0.86%) for ratios using ²⁰⁴Pb. Although considerably less precise than TIMS, which typically produces measurements with a precision of 0.005 to 0.01 %RSD (Heumann *et al.* 1998), the ICP-MS has, nonetheless, provided values (for all ratios except those using ²⁰⁴Pb) for which it is possible to be confident of to at least three significant digits. Furthermore, the frequently assayed NIST standard has been used to adjust (correct) measured isotopic values on the samples to more accurately reflect what their true values likely are. This was done by taking the average of the values measured for the two bracketing standards (one run before and one after five samples), calculating how that average varied from the set NIST standard and then adjusting the measured values on the samples by the difference.

In order to gauge the accuracy of this technique and the corrections applied to the data it provided, one galena sample each from two isotopically wellcharacterized lead occurrences in southern Rajasthan – Zawar and Rajpura-Dariba/Rampura-Agucha, was analyzed using the EDTA/ICP-MS method. The data were compared to the published TIMS produced values from those two ore fields (Deb *et al.* 1989). In both cases, the corrected EDTA/ICP-MS isotopic values for the geologic test samples closely matched those of the highly accurate TIMS method (Figure 12.1 E).

Although not as precise as TIMS, there are many advantages to using a combination of EDTA sampling and ICP-MS for the analysis of lead artifacts. Sampling using this technique is, for all practical purposes, non-destructive. No material need be chipped off or scraped from an artifact for digestion in acid. Although EDTA sampling is "destructive" in the sense that some Pb ions are physically removed from the surface of the sample, this only takes place at an atomic level. Macroscopic alteration of artifacts is in no way evident. Ease, speed and cost of sample preparation and analysis are other advantages. Preparing lead samples for TIMS is an extended and laborious process that results in such analyses being quite expensive and time consuming. The EDTA/ ICP-MS method is considerably less difficult and comparatively less expensive. No hazardous acids or purification columns need be used. The simple and short sample preparation process and the ease of ICP-MS (as compared to TIMS) help keep the

cost of isotopic analysis using this method reasonably low. A final advantage is that artifacts sampled for Pb isotopes in this way need not be removed from location at which they are stored. A small kit containing purified water (for artifact cleaning before and after sampling), the non-toxic EDTA solution, disposable containers (for artifact immersion in the solution) and vials (for holding the sampled solution) can be taken directly to a site being excavated or a museum collection and lead artifacts can be quickly and easily sampled.

Presenting and plotting PB isotope data

Lead isotope data are normally presented as ratios of the absolute amount of one isotope against the absolute amount of another - for example ²⁰⁸Pb/²⁰⁴Pb. Although only three *distinct* ratios can exist (as there are only four isotopes of lead), a dozen variations are possible depending on how one chooses to construct them. Those chosen to be presented often vary from publication to publication. Three ratios (²⁰⁸Pb/²⁰⁷Pb, ²⁰⁷Pb/²⁰⁶Pb and ²⁰⁷Pb/²⁰⁴Pb) are reported in the various appendices for this chapter. If future users of this data wish to employ combinations not presented here it is possible to easily extrapolate them from the ratios provided. For example, the ratio ²⁰⁶Pb/²⁰⁴Pb may be generated simply by dividing the ratios ²⁰⁷Pb/²⁰⁴Pb by ²⁰⁷Pb/²⁰⁶Pb. In the course of compiling the databases in this chapter, extrapolation was used when published isotope ratios were different from the three selected to be presented here. *Caution* is advised, however, when constructing any ratio using ²⁰⁴Pb from a sample analyzed at the LARCH, as measurements made for that isotope are significantly less precise than those made for ²⁰⁸Pb, ²⁰⁷Pb and ²⁰⁶Pb.

Pb isotope data are graphically represented on a bivariate plot of the values for two ratios. Since three distinct ratios exist, two such plots are required to fully display all measured data (Weeks 2004: 131). However, only one plot is used in this chapter (with 208 Pb/ 207 Pb for the *y* axis and 207 Pb/ 206 Pb for the *x*

axis) because of, once again, the imprecision of the ²⁰⁴Pb values for samples analyzed at the LARCH. Although omitting the third dimension by dropping ²⁰⁴Pb does, admittedly, create some limitations, it does not necessarily impede one's ability to discriminate between lead sources and/or to make provenience determinations. Klein and others (2004) were able to differentiate Roman lead and copper sources without using ²⁰⁴Pb data. In fact, when Sangster and others recently (2000) compared Pb isotope data from 151 lead deposits around the world, they found that:

Statistical analyses suggested that while ²⁰⁴Pb is critical for identifying a small proportion of environmental Pb sources, about 86% of the source discrimination power is due to the ²⁰⁶Pb, ²⁰⁷Pb, and ²⁰⁸Pb isotopes. Thus, the requisite analytical precision, rather than a lack of ²⁰⁴Pb data, is the most critical issue with respect to unequivocal identification of Pb sources in most cases (Sangster *et al.* 2000: 115).

For this study, very good analytical precision overall measuring ²⁰⁶Pb, ²⁰⁷Pb and ²⁰⁸Pb was achieved using the EDTA/ICP-MS technique. I will show that convincing statements regarding the probable geologic proveniences of metal artifacts from Harappa and other sites can be made based a single plot generated using those three Pb isotopes alone.

LEAD AND SILVER

Sir Edwin Pascoe, who was Director of the Geological Survey of India during the 1920s, noted that "with the exception of iron, there is perhaps no metal whose ores appear to have been worked to so large an extent in India as those of lead" and added that much of the emphasis in the exploitation of this mineral was directed toward the "extraction of silver" (Pascoe 1931: 676). Yet instead of focusing on sources in northwestern South Asia, he mentioned locations stretching from Burma to Tunisia where these metals are found (ibid.: 675-677). Although some of the places he listed are adjacent to the Indus Basin and would have been, more or less, accessible to Harappan consumers, most of the regions he states "might be looked upon as possible sources" (ibid.: 675) of lead and silver are located in South India, Afghanistan, Iran, Burma and even Armenia!

It is somewhat mystifying as to why Sir Edwin largely ignored (and presumably felt that Harappans would have ignored) the deposits of lead and leadsilver that practically ring the Indus Basin (Figure 12.2). In the intervening years some scholars have recognized the importance of these indigenous sources (Agrawal 2000; Chakrabarti and Lahiri 1996) and, in some areas, have even conducted detailed studies of ancient mining and production (Craddock et al. 1989). Others, however, still repeat (almost verbatim) what Pascoe wrote 75 years ago (Asthana 1993; Biwas 1996). Although I do not ignore the possibility that metals like silver were imported from sources far outside of the Greater Indus region (Ratnagar 2004: 199), here I emphasize the isotopic characterization and comparison of lead and silver deposits in closer areas where they were more "easily procurable" (Chakrabarti 1990: 143).

The Pb isotope database of potential Harappan lead and silver sources

The Pb isotope database for lead deposits assembled here (Appendix 12.1) consists of 232 analyses made on lead ore samples from 58 individual localities throughout India, Pakistan and Oman (Figure 12.2 and 12.3). One hundred thirty-three of those isotopic determinations were drawn the geologic literature. Ninety-nine new Pb isotope analyses were conducted at the LARCH and the Keck Isotope Laboratory, UC-Santa Cruz on samples collected from previously uncharacterized (or undercharacterized) lead and lead-silver deposits in Gujarat,



Rajasthan, Uttaranchal, Himachal Pradesh, Jammu and Kashmir, Balochistan and Oman. Not included in the primary database are seven assays of lead slags (and a few ores) from the argentiferous galena deposit at Nakhlak, in central Iran (Appendix 12.6), which are considered only in examinations of lead and silver artifacts from sites in Iran (Shahr-i-Sokhta) and Afghanistan (Mundigak).

In the following sub-sections, potential lead and lead-silver sources are discussed on a region-byregion basis, as are important deposits that have yet to be characterized and, importantly, the ancient cultures and settlements located in those regions that Harappans would have almost certainly been directly or indirectly interacting with in order to obtain this mineral resource.

Lead deposits in Balochistan

In Pakistan's Balochistan Province, lead occurs

at multiple locations within the Las Bela, Khuzdar and Chagai districts (Ahmad 1975: 63-65; Shams 1995b: 246-47). During the latter half of the third millennium BC, both Indus Civilization settlements and those belonging to the local Kulli culture, which possessed a great many traits that are considered to be typically "Harappan," could be found in the area of southeastern Balochistan encompassed by the former two districts. Although it remains to be determined if it is appropriate to characterize Kulli society as "the highland form of the Indus Civilization" (Possehl 1986: 61), it seems evident that there was a high degree of interaction taking place at this time between those dwelling in this region and the Indus Valley. The lead sources of southern Balochistan, therefore, would have likely been among the most accessible to Indus Civilization consumers. Somewhat less accessible, but still important, would have been the deposits in the Chagai district, which are located along what would



Figure 12.4 Map of the central Kanrach Valley, Las Bela District, southern Balochistan and lead deposits sampled for this study.



Figure 12.5 View looking north from the Duddar lead deposit, Kanrach Valley, Balochistan.



Figure 12.6 Sampling a galena seam at Bamph Nala, Kanrach Valley, Balochistan.

have been one of the major trade and communication routes between the Indus Valley and southwestern Afghanistan and eastern Iran.

Lead mineralization can be found at several places in the Las Bela district (Ahsan and Bhutta 1991; Heron and Crookshank 1954: 93) but the largest and most economically viable deposits occur in the central part of the north-to-south running Kanrach Valley (Figure 12.4). Isotopic analyses of four galena samples from the Duddar and Kharrari areas (N 26° 05' 33", E 65° 50' 16") had been undertaken prior to this study (Bhutta 1992; Siddiqui 1994, Bhutta and Qureshi 1997). Twelve additional assays were performed at the LARCH on samples collected in May 2001 from the Kharrari North area (Figure 12.5) and a third location in nearby Bamph Nala (Figure 12.6). It is especially important to note that all of these locations are found within one to two kilometers of the prehistoric site of Bakkar Buthi, which contains both Kulli and "purely Harappan" occupation levels (Franke-Vogt *et al.* 2000: 196). A.H. Kidwai (in Heron and Crookshank 1954: 93) reported "weathered ore, slags of lead and copper, [and] furnace clay" at a location noted at "ruins" (near Thana Kanrach) around 20 km to the north of the site. Although no such items were found during the limited excavations at Bakkar Buthi (Ute Franke-Vogt *personal communication* 2003), it is by far the nearest of all Harappan settlements to a source of lead.

Lead in the Khuzdar district (formerly part of the Kalat district) occurs at several places in the vicinity of Khuzdar town including Gunga, Shekran and Surmai (Ahmad 1975: 64; Jankovic 1986; Shams 1995b: 246; Siddiqui and Sharp 1993). Shams noted (1995b: 246) that the ore at Gunga (N 27° 44' 30", E 66° 32') was highly argentiferous, containing up to 3000 ppm silver (recall that 10 ppm is considered viably argentiferous - Craddock 1995: 211). Three isotopic analyses of galena samples from that deposit were performed by Siddiqui (1994). Two additional samples from this location were supplied by Drs. Mehrab Khan and Khalid Mahmood of the University of Balochistan - Quetta and analyzed at the LARCH. Numerous old mines and slag fields have been noted throughout this part of the Khuzdar district (Hassan 1989; Heron and Crookshank 1954: 92; Siddiqui and Sharp 1993). All are located within 25 km of the ancient mounds at Sohr Damb (Nal), where galena, cerussite, lead slags and silver artifacts have been recovered (Hargreaves 1929: 33; Yule 1995). Importantly for this study, a "Kulli-Harappan" has recently been documented at that site (Franke-Vogt 2005).

Although rich and extensive polymetallic sulphide deposits are found in the northwestern part of the Chagai District (Ahmad 1975; Shams 1995b: 243-247), showings of galena tends to be minor in nature (Kazmi and Jan 1997: 449). Nevertheless, Pb isotope assays were made at the LARCH on a total of 12 galena samples from deposits at Koh-i-Sultan (\approx N 27° 07', E 62° 47') and Rekodiq (\approx N 29° 10', E 62° 14'), which were supplied by Drs. Mehrab Khan and Khalid Mahmood of the University of Balochistan – Quetta.

Lead deposits in the NWFP

Lead ore in Pakistan's North-West Frontier Province (hereafter NWFP) is found primarily in the Chitral, Allai Kohistan, Swat and Hazara districts (Ahmad 1969: 39-40; Shams 1995b: 245-46). There are several reasons to believe that Harappans may possibly have obtained this metal from deposits in these districts. Numerous Early Harappan period (Kot Dijian) sites can be found in or directly adjacent to the southern parts of the province (Allchin 1984). Also, the most direct route to Shortughaï, the Indus Civilization outpost in northern Afghanistan (Francfort 1984b), would have been through the NWFP. Lastly, but certainly not least, I have already show that two other important varieties of stone steatite (Chapter 7) and vesuvianite-grossular garnet (Chapter 9), seem to have been acquired from sources located in this region.

Lead mineralization, sometimes co-occurring with copper and antimony, has been noted at nearly a dozen locations in the southern part of the Chitral district (Ahmad 1969: 39-40). Tahirkheli and others (1997) performed Pb isotope analysis on three galena samples from a zone of copper mineralization in the Shi Shi Valley area near Drosh (\approx N 35° 39', E 71° 54'). Farther to the southeast, five Pb isotope determinations were made by Shah and others (1992) for the Lahor and Pazang lead deposits in the Besham area (\approx N 34° 56', E 72° 52'), Allai Kohistan.

Among the lead sources in the NWFP that have not yet been isotopically assayed are those near Ushu (N 35° 34', E 72° 43') in the Swat district (Tahirkheli 1959) and several zones of galena mineralization around the Abbottabad area (\approx N 34° 11', E 73° 03'), Hazara district (Shams 1963; Ahmad 1969: 40). It will be especially critical to acquire material from these sources for future analysis. Early Harappan period interaction with the Swat region is suggested by finds of Kot Dijian-like ceramics at Ghalegay



Figure 12.7 Select sites and lead deposits in Jammu and Kashmir.

Cave (Stacul 1987: 29-49). The Kot Dijian sites of Sarai Khola, Hathial and Jhang (Khan 1983; Halim 1972) lie around 50 km or so south of the Hazara district galena sources, which themselves are situated within a few kilometers of the steatite deposits shown (Chapter 7) to have quite probably been the major source of that material for residents of Harappa.

Lead deposits in Jammu and Kashmir

In the Jammu and Kashmir region, lead mineralization occurs in the districts of Doda, Riasi, Baramulla, Anantnag and Muzaffarabad (Ahmed 1997; Srivastava 1977; Varadan 1977: 52-54). The geologist C.S. Middlemiss noted (1929: 2) that the "lead-silver deposits of the State [*appear to*] have been worked in a primitive way during ancient times." Importantly, some of these deposits are located (Figure 12.7) in the vicinity of Indus Civilization settlements and/or along the natural routes through which Northern Neolithic and Kot Dijian/"Late" Kot Dijian phase peoples interacted with one another.

Sporadic occurrences of lead-zinc are found within the Great Limestone formation of the Riasi district (Nayak and Sharma 1991). Four galena samples from abandoned workings near Sersendu village (N 33° 06' 53", E 74° 54' 00") were isotopically assayed by Raha and others (1978). Additional assays were made at the LARCH on 11 samples from this same area. I collected six of those from an old mine at Kheri Kot (on the ridge above Sersendu) in June of 2003 (Figure 12.8). Five more were provided by Dr. Rajesh Sharma of the Wadia Institute of Himalayan Geology who collected them from the nearby Darabi area. The Riasi deposits are among the closest lead sources to Harappa and are located less than 30 km north of Manda, where Joshi and Bala (1982) identified both Early Harappan and Harappan occupational phases.

In the Baramulla district, lead can be found at various places in the Buniyar area along Hapatkhai Nala (≈ N 34° 06', E 74° 12'), about a kilometer south



Figure 12.8Top - View from Kheri Kot looking north, above Sersendu Village and the Chenab River.Bottom images - Visiting one of the old lead mines at Kheri Kot and sampling galena.

the Jhelum River (Middlemiss 1929; Raina 1977). The Jhelum River is an important route connecting the Kashmir Valley to the Potwar Plateau and upper Indus Basin. The cache of carnelian beads in a Kot Dijian style pots recovered at the Northern Neolithic site of Burzahom (Pande 2000) provides clear evidence for interaction and exchange between the ancient peoples of those regions. Lead from the Buniyar area, which sometimes contains minor amounts of sphalerite and chalcopyrite in addition to galena (Sharma and Sachan 1998), could have been an important trade good for Northern Neolithic Phase Kashmiris, who were settled as near as 20 km to the northeast at Kanispur (Mani 2000). Pb isotope determinations were made at the LARCH on seven galena samples from the Buniyar occurrences provided by Dr. Rajesh Sharma of the Wadia Institute of Himalayan Geology at Dehra Dun.

Several lead sources in Jammu and Kashmir have yet to be isotopically characterized. At Treri (N 34° 26' 48", E 73° 43' 10") in the Muzaffarabad district, galena is found with zinc and copper (Ahmad *et al.* 1992; Ahmed 1997). Nodules of galena can be found at Hapatnar (N 33° 50', E 75° 21') and Shumahal (N 33° 50', E 75° 17') in the Anantnag district (Varadan 1977: 52). In the Doda district, galena and cerussite are found near Chichhe village as are old workings for those minerals (Srivastava 1977).

Lead deposits in Himachal Pradesh and Uttaranchal

D.P. Agrawal argued (1999) that the central Himalayas were an important, but largely overlooked, region with regard to the study of South Asian archaeometallurgy. Indeed, the states of Uttaranchal and Himachal Pradesh are exceptionally rich in both base and noble metals. Geologists have noted the existence of scores of disused mine shafts, pits, slag heaps and waste dumps that indicate the varied deposits of the region were widely exploited in former times (Sharma 2002). Ropar, Bara and Kotla Nihang Khan are all proto-historic settlements found within 10 km of the point near which the Sutlej River leaves the foothills of the lesser Himalayas (Sharma 1982). Harappan period remains were also unearthed at Chandigarh, not far from where the Ghaggar River meets the plains (Joshi 1990: 15). Indus Civilization peoples dwelling at any of these sites would have been well positioned (Figure 12.9) to access the rich leadsilver deposits of the central Himalayas.



Figure 12.9 Select Himachal Pradesh lead deposits and Punjab archaeological sites.



Figure 12.10 Sampling trip to the old silver mine at Uchich, Parvati Valley, Kullu District, Himachal Pradesh.



Figure 12.11 Old working and lead sample taken at Amba Kala, Himachal Pradesh.

In Himachal Pradesh, lead deposits occur in the Kinnaur, Kullu, Simla and Sirmaur districts (Chowdhury and Sehgal 1962; Geological Survey of India 1989a: 32-33; Srikkantia and Bhargava 1998: 335-36). Prior to this study, no Pb isotope determinations had been made for any of the deposits in the state, many of which are highly argentiferous. The lead mines in the Kullu Valley and others adjoining it have long been famous as sources of silver (Calvert 1873). Five Pb isotope determinations were made on samples collected (Figure 12.10) in June of 2004 from the old silver mine at Uchich (N 32° 00' 41", E 77° 22' 27") near Manikaran in the Parvati Valley, Kullu (formerly Kangra) district (Sehgal 1964). Samples were also collected at that time from the galena mines at Panuh (N 30° 58' 12", E 77° 01' 15") and Tal (N 31° 09' 57", E 76° 57' 18") in the Solan district (Chattopadhyay and Verma 1992; Kumar 1992). Ten Pb isotope analyses for the Panuh deposit and five for the Tal deposit were conducted at the LARCH. Galena at Amba Kala (N 30° 38' 29", E 77° 27' 24") in the Sirmaur district is reported to contain up to 250 ppm silver (Geological

Survey of India 1989a: 49). Five Pb assays were made on samples collected from that deposit (Figure 12.11) in May of 2003.

Although lead mineralization occurs across Uttaranchal, the richest deposits are found in the eastern portion of the state in the Kumaun region (Valdiya 1980: 263). Many of these deposits are highly argentiferous, such as the one near Birgana village, Garwal District (Shrivastava and Kapoor 1981). Sarkar and others (2000) assayed three galena samples from the lead-copper deposits found in the vicinity of Bageshwar (\approx N 29° 48', E 79° 46') (Banerjee 1977; Srivastava and Gaur 1979). Dr. Rajesh Sharma of the Wadia Institute of Himalayan Geology, Dehra Dun supplied galena samples from two additional Kumaun region deposits for Pb isotope analysis at the LARCH. Three samples were from the polymetallic sulphide deposit at Askot (≈ N 29° 46', E 80° 20') in the Pithoragarh district near the border with Nepal (Acharya 1988; Ghose 1976). Two samples came from an area of galena mineralization at Khansue, Okhalkanda subdivision, Nainital district.



Figure 12.12 Isotopically assayed lead sources and select Harappan and Ahar-Banas sites in the southern Rajasthan and northern Gujarat regions.

Lead deposits in Rajasthan and Gujarat

The base metal deposits of Rajasthan are extensive and numerous. There is evidence they were exploited beginning at least as far back as the Early Historic period of South Asia. Lead mineralization can be found in the Udaipur, Rajsamand, Bhilwara, Sirohi, Ajmer, Sawai Madhopur, Chittorgarh, Pali, Banswara and Bharatpur districts (Geological Survey of India 1994; Indian Bureau of Mines 1995: 29-38). In this section, only those deposits that have been isotopically characterized are discussed.

The Khetri copper belt is the dominant zone of sulphide mineralization in northern Rajasthan (Geological Survey of India 1994: 2-34) and was almost certainly a major source of that metal during the Harappan period (Agrawala 1984). Sites belonging to the Ganeshwar-Jodhpura culture complex are found across this region. Stylistic analyses of copper arrowheads suggest that there were contacts between peoples of that phase and Harappans (Rizvi 2007: 152). However, is unlikely that much, if any, of the raw lead used by Indus Civilization peoples was derived from northern Rajasthan sulphide deposits. The only location where galena mineralization can be found in the Khetri copper belt is on its southern margins at Saladipura (N 35° 34', E 72° 43') in the Sirohi district. There it occurs in trace amounts together with sphalerite and pyrite (Das Gupta 1970). Two Pb isotope analyses of galena from this location were conducted by Deb and others (1989).

Although the zone of mineralization is largely obscured under the alluvium, "extensive old workings for galena" are still evident at Chauth-ka-Bawara (N 26° 02' 59", E 76° 11' 05") in the Sawai Madhopur district (Geological Survey of India 1994: 135) of eastern Rajasthan. Five galena samples collected from



Figure 12.13 Old lead mine shaft and smelting slags at Rajpura-Dariba, Udaipur District, southern Rajasthan.

this deposit in June of 2004 were analyzed at the LARCH.

others (2001).

The largest concentration of lead deposits in this database is situated in the southern Rajasthan/ northern Gujarat region. A supplementary map (Figure 12.12) is provided for the discussion of potential sources in this region. Individual locations in the Ambaji-Sendra sulphide belt are identified as well as important Harappan and Ahar-Banas complex sites.

Lead occurs at numerous locations within the Ajmer and Pali districts of central Rajasthan (Geological Survey of India 1994: 53-62). Old workings are found (N 26° 29' 08", E 74° 39' 10") on the outskirts of Ajmer city at Lohakhan (Nath 1972). One sample collected at this location in January of 2004 was analyzed at the LARCH. Abandoned mine shafts penetrating as deep as 40 m can be seen at Punagarh Hill (N 25° 48', E 73° 26') in the Pali district (Gupta 1977; Sahai *et al.* 1997: 180). Two galena samples from this location were analyzed by Deb and

The lead deposits of the Udaipur, Rajsamand and Bhilwara districts of Rajasthan are the richest in India and the most isotopically well characterized. Twenty-one Pb isotope determinations have been made (Balasubramanyan and Chandy 1976; Deb et al. 1989) on galena samples from the lead-zinc deposit at Zawar (N 24° 20', E 73° 41') in the Udaipur district. Evidence suggests that mining and beneficiation of ores from this deposit began around 2000 years ago (Freestone et al. 1985; Craddock et al. 1989). Collectively there have been 31 isotopic determination made (Balasubramanyan and Chandy 1976; Deb et al. 1989; LARCH 2004) on galena samples from the Rajpura-Dariba lead belt (≈ N 24° 38', E 74° 18') in the Rajsamand district and the Rampura-Agucha deposit (N 25° 49', E 74° 43') in the Bhilwara district. Occurring in the same geologic terrain, these two zones of polymetallic sulphide mineralization are isotopically indistinguishable and so are treated as a single source in this study. The lead from both the



Figure 12.14 Sampling galena at Khera Mawal, Banaskantha District, northern Gujarat.



Figure 12.15 Top images - Modern lead-zinc-copper mine at Ambaji, Banaskantha District, northern Gujarat and a galena speckled sample taken at that location. Bottom images - Smelting slags near Ambaji at Kumbariya.

Rajpura-Dariba and Rampura-Agucha deposits is richly argentiferous (Mukherjee *et al.* 1991; Lahiri and Ravindranath 1988) and there is abundant evidence (Figure 12.13) for mining and smelting at those locations extending as far back as the midsecond millennium BC (Willies 1989). If it is could be shown that Indus Civilization peoples might have obtained some of their lead and/or silver from these sources then it would provide compelling evidence of long-distance interaction with peoples belonging to early phases of the Ahar-Banas culture of southern Rajasthan (Chakrabarti 1968; Fairservis 1975: 335-340; Shinde *et al.* 2005).

In the Sirohi district near the border with Gujarat, pockets of galena are found at Khera Mawal (N 24° 24' 37", E 72° 42, 46") intermittently along a 500m zone (Geological Survey of India 1994: 68). Deb and others (2001) produced a single isotope determination for a galena sample from this location. Data for two additional samples was produced at the Keck Isotope Laboratory, UC-Santa Cruz after the deposit was visited in January of 2004 specifically for this project (Figure 12.14).

Beginning on the northern border of Gujarat at Ambaji and extending approximately 240 km into southern Rajasthan near Sendra is a geologic terrain that contains many areas of massive sulphide mineralization (Deb *et al.* 2001). Old workings, mine dumps and slag fields (Figure 12.15) are founds at numerous locations across this zone (Murty and Shekar 1975: 32; Sahai et al. 1997: 182; Geological Survey of India 1994: 34; personal observations). Sixteen Pb isotope determinations in total have been made (Deb et al. 1989; Deb et al. 2001; LARCH) on ore samples from following locations within the Ambaji-Sendra sulphide belt: Ambaji (N 24° 20' 43", E 72° 50' 48"), Amli-Mal (N 24° 21' 53", E 72° 48' 22"), Kumbariya (N 24° 18' 36", E 72° 53' 48"), Danva (N 24° 46' 10", E 73° 02' 10"); Deri (N 24° 20' 43", E 72° 50' 48"), Basantgarh (N 24° 43' 30", E 73° 01' 15") and Birantiya Khurd (N 26° 07', E 74° 08'). The Indus Civilization peoples at sites like in north Gujarat like Dholavira or Nagwada might have accessed these deposits directly or via interaction with huntergatherer groups of the North Gujarat Plain (Possehl 1980: 73).

Lead mineralization is much more minor in nature elsewhere in Gujarat. The sporadic occurrences that do exist, however, could conceivably have been exploited by the Harappan or related cultures of



Figure 12.16 Sampling the lead occurrence at Khandia, Vadodara District, eastern Gujarat.

the region (Ajithprasad 2002; Possehl 1980, 1992; Sonawane 2002). A small deposit of galena (Figure 12.16) is located near Khandia (N 22° 19' 26" E 73° 33' 39") in the eastern part of the Vadodara district (Shah *et al.* 1985; Yellur 1969). Five isotopic analyses of samples collected from this deposit in May of 2003 were performed at the LARCH. In the Junagadh district of southern Saurashtra, galena together with specks of chalcopyrite (Shekar and Mukul 1969) is found in the Gir Forest near Banejnes (N 21° 03' 15", E 70° 53, 48"). Five isotopic analyses of samples collected from this deposit in May of 2003 were performed at the LARCH.

Lead occurs with pyrite in the Amba Dongar fluorite deposit of southeastern Gujarat (Simonetti and Bell 1995). A single sample of galena from this deposit was assayed by Venkatasubramanian and others (1982).

Lead deposits elsewhere in South Asia

In this section, I briefly review Pb isotope analyses of galena from deposits in South Asia that are either minor in nature or not encompassed by the current study area – the Greater Indus region. Although many of these deposits were probably not utilized by Harappans, in the interest in producing a comprehensive database, these data are included in the appendix and plotted on the accompanying charts.

Deb and others (2001) made a single Pb isotope determination on galena from the polymetallic ore deposit at Tosham Hill (N 28° 52' 32", E 75° 54' 41"), district Bhiwani, Haryana. This deposit has been proposed as a possible source of tin for the Indus Civilization (Kochhar *et al.* 1999). However, only trace amounts of galena occur with the other metals found there (Awasthi *et al.* 1981). Although Tosham Hill cannot be ruled out as a source of Harappan lead, it is unlikely that much, if any, galena was extracted from this location.

In total, 15 Pb isotope analyses have been made

on galena samples from deposits in eastern India. In the state of Bihar, three isotope determinations exist for the lead deposit at Amjhor (Balasubrahmanyan and Chandy 1976) and five for those in the Hesatu-Pindura sulphide belt (Singh *et al.* 2001). Three samples from Gorubathan, West Bengal and four from Rangpo, Sikkim have also been analyzed (Sarkar *et al.* 2000). All of these deposits are over 800 km away from the easternmost Harappan site. It seems highly unlikely Harappans would have exploited these sources when much closer ones were at hand.

Lead minerals occur in many parts of southern India (not pictured on Figure 12.3). Venkatasubramanian and others (1982) conducted isotopic analyses on individual galena samples from various deposits in the states of Andhra Pradesh, Karnataka and Tamil Nadu. For her provenience study of the Early Historic Period South Indian bronze icons, Srinivasan analyzed (1999) nine samples from the lead occurrence at Agnigundala, Andhra Pradesh.

Lead deposits in Oman and Iran

One area outside of the Greater Indus region to which Indus Civilization peoples travelled is the eastern part of the Arabian peninsula. Harappan artifacts have been recovered at a number of coastal and inland sites located present-day Oman and the United Arab Emirates (Cleuziou 1984, 1992; Cleuziou and Vogt 1985). Part of the impetus for Indus peoples to make such a distant journey may have been the acquisition of metals, in particular copper, from the region's rich sulphide deposits (Weisgerber 1984). However, although a fair amount of Pb isotope data exists for eastern Arabia, most of the analyses made to date have been conducted on pyrite, chalcopyrite or various volcanic rocks (Calvez and Lescuyer 1991; Chen and Pallister 1981; Prange 1999; Stos-Gale et al. 1997). I have located only three published Pb-Pb determinations made on lead minerals from Oman - a single sample each from Wadi Mayh (≈ N 23° 25'



Figure 12.17 Sampling the old lead mine at Wadi Nujum, Oman.

53", E 58° 33'), Qumayrah (N 23° 55' 57", E 56° 11' 42") and a section of the Semail ophiolite near Ibra (N 22° 41' 25", E 58° 32' 04"). Insofar as I have been able to determine, each of these occurrences are only trace showings of galena and not significant lead sources. The only viable deposit in all of Oman seems to the one at Wadi Nujum (N 23° 23' 43", E 58° 10' 44"), where an old mine (Coleman and Bailey 1981: 93-95) and Bronze Age stone mining tools (Weisgerber 1997: 198, Figure 206) have been documented. I collected galena at this location (Figure 12.17) in February 2009. Four samples were analyzed at the Keck Isotope Laboratory, UC-Santa Cruz.

The metalliferous ore deposits of Iran are numerous and widespread but nowhere are they richer or more varied than in the Central Persian Desert (Wertime 1968). "At Anarak-Nakhlak nearly all the minerals of the desert come together in a remarkable and evident juxtaposition; thus this region was one of the earliest homes of metallurgy anywhere in the world" (ibid.: 927). The argentiferous lead deposit at Nakhlak (N 33° 33' 50", E 53° 50' 40"), which have been assayed at silver 715 grams per ton, appears to have been worked since ancient times (Holzer and Ghasemipour 1973). Stos-Gale (2001) published the results of Pb isotope analyses on slags and ores collected from this deposit, the values of which have been extrapolated for use in the present study (Appendix 12.6).

Afghanistan

Referring back for a moment to Figure 12.2, it is plain to see that Afghanistan is a conspicuous "hole" in this dataset that, although not comprehensive, is otherwise reasonably representative of the different regions immediately surrounding the Indus Basin where Harappans might potentially have acquired lead or silver (if extracted from lead). There are five major lead deposits and nearly 100 more documented occurrences and showings of that metal throughout Afghanistan (ESCAP 1995: 30). One of the more important sources in terms of this study is the Asad Qala (Kalai-Assad) deposit (Berthoud et al. 1977; Jarrige and Tosi 1981), which is around ten kilometers north of the Helmand Tradition site of Mundigak (Casal 1961). Isotopic assays have been made for a series of lead and silver artifacts from Mundigak and are presented below. Until geologic materials from Asad Qala can be analyzed, those artifacts may serve

as proxy samples for that deposit (of course, I am making the assumption that the metal was locally procured).

Plotting and evaluating the Pb isotope data for South Asian lead deposits

When the 232 Pb isotope determinations comprising the lead deposits database are placed on a bivariate plot of the ratios ²⁰⁸Pb/²⁰⁷Pb and ²⁰⁷Pb/²⁰⁶Pb, the majority (211) of samples cluster into a loose linear pattern (Figure 12.18). Twenty-one samples (from Banejnes, Uchich, Besham and six deposits in South India) are isotopically distant outliers (Figure 12.19) to the main body of lead deposits. Although there is a degree of overlap between many of the individual deposits and ore fields, by and large, most major lead sources are isotopically distinct from one another - especially a when considered at a regional scale. For example, most of the lead deposits of Rajasthan cluster in the lower right area of Figure 12.18. The Ambaji-Sendra belt, which extends from Gujarat into south central Rajasthan, falls toward the middle of the plot. A few remaining deposits in Rajasthan trend toward the upper left. All of the lead deposits in Balochistan, however, plot squarely in the far upper left of the figure. Although samples from sources in the Himalayas and the NWFP fall at several different places across the trend, they mostly plot apart (although there are degrees of overlap in places) from the other deposits. The complex geologic histories and processes that affect the isotopic composition lead occurrences in these regions and cause them to plot as they do is not the subject matter of this book. What is important with regard to this study is that lead ores deriving from the various regions are, to a very large degree, isotopically distinct from one another. Therefore, when a question is posed such as - Did the metal for these lead or silver artifacts come from sources in Rajasthan, Balochistan, the Himalayas or elsewhere? - then providing an answer may very well be possible with reference to this Pb











Figure 12.20 Isotope "fields" of the "main body" of South Asian lead ore deposits.



Figure 12.21 Harappa site plan with numbered trenches and areas where lead ores, objects, slags or residues have been recovered. Dateable artifacts = (period). Others are surface/disturbed. Multiple artifacts = x number.

isotope database. I address these very questions in the upcoming sections of this chapter.

To aid in interpretations of the archaeological Pb isotope data, I have created a new version (Figure 12.20) of the bivariate plot in which the different groups of lead ore occurrences (or "fields") that comprise the main body of geologic samples have been outlined and labeled. The shapes demarcating ore "fields" were drawn by hand and are *not* statistical confidence intervals. They are meant only to be visual guides (it is easier to see individual points plotted against solid shapes versus numerous data points). Individual data points are still visible as lightly superimposed geometric shapes. Crossed bars representing the overall range of analytical error (standard deviation of the mean of repeated measurements of the NIST lead isotope standard) are present on each plot.

Determining the probable geologic proveniences of lead artifacts from Harappa

Thirty-four "lead" artifacts, in total, have been recovered during HARP excavations and Pb isotope data have been produced for all of them using the EDTA/ICP-MS method. Nineteen are raw lead ores (Appendix 12.2) while the remaining ones are finished items, lead slags, melted lumps and residues (Appendix 12.3). Examples have been found on every mound and in the cemetery area (Figure 12.21). One ore fragment was found in Ravi Phase (Period 1) levels, two finished items were found in Period 3B burial contexts and a total of 10 artifacts (including ores, residues, slags and finished items) come from



Figure 12.22 Three varieties of lead ore found at Harappa and pigments derived from them.



Figure 12.23 Raw lead ore fragments from Harappa plotted against South Asian lead ore fields.

Period 3C levels. The remaining 22 lead artifacts are from surface or disturbed contexts.

Lead ores

When performing isotopic or elemental analysis of archaeological metals that have been altered in some way (smelted, forged, cast, alloyed, etc.) there is always the possibility that material from two or more geologic sources may be present in a single artifact. This can potentially lead to the misassignment of an artifact's provenience due to the obscuring of the chemical and/or isotopic characteristics used to match it to a geologic source. When the artifacts under examination are unadulterated metal ores, however, the potential mixing of sources is not an issue. For this reason, raw ore, when it is present at a site, is undoubtedly the best form of archaeological metal to study when attempting artifact-to-source correlation. Lead isotope analysis has been employed successfully in the past to help identify the probable sources of lead ore (galena) fragments excavated at sites in Egypt (Hassan and Hassan 1981) and North America (Farquhar and Fletcher 1984).

As discussed in Chapter 4, XRD analysis has shown that three distinct minerals (Figure 12.22) are represented among the 19 lead ore artifacts at Harappa: massicot (n=5), cerussite-anglesite (n=3)and galena (n=11). Besides being important ores for the metal lead (and, if argentiferous, silver), each are common mineral pigments/cosmetics and go by names like "plumbic ochre" (massicot), "lead white" (cerussite) and "surma" or "kohl" (galena). I discuss this aspect further in the section below where I examine lead residues found in small ceramic and faience bottles, which are possibly cosmetics. Lead minerals are also frequently employed in traditional medicines in South Asia (Murthy 1983) and the Arabian Gulf region (Worthing and Sutherland 1996). It is quite likely that when lead was intentionally brought to Harappa in raw form, it was for one of these uses.

When Pb isotope data for the 19 lead ore fragments from Harappa (Appendix 12.2) are placed on the bivariate plot of the main body of geologic samples (Figure 12.23), they more or less cluster in three areas. Nine artifacts fall within the fields of the Jammu and Kashmir deposits. Although a few of those are isotopically more analogous to either the Riasi samples or the Buniyar samples, most cluster in or near the area where the two sources overlap. Five of the ores plot with samples from deposits in Kanrach Valley and Khuzdar areas of southern Balochistan - sources that have largely indistinguishable Pb isotope characteristics. Here is an instance where having useable ²⁰⁴Pb measurements (and/or even more accurate ²⁰⁸Pb, ²⁰⁷Pb and ²⁰⁶Pb data) might help to resolve these two sources. On the other hand, it might not. Although the two areas of lead mineralization lie around 180 km apart, both belong to a zone in Jurassic sediments along the western margin of the Indian Plate known as the "Las Bela-Khuzdar metallogenic province" (Jankovic 1986: 1). For now, the two sources have been grouped into one "field" that represents the region of southern Balochistan. The source association of the remaining five ore fragments is more ambiguous. Although two are near geologic samples from the Amba Kala deposit in Himachal Pradesh, all five fall in a blank gap on the lead deposits plot. However, because the artifacts are unadulterated lead minerals, we can be certain that a lead source (or sources) physically exists somewhere with these precise isotopic characteristics. It just has not yet been located, sampled and included in the database.

Interestingly, the nine ores that appear to be from the Jammu and Kashmir sources are all galena fragments. The other two galena artifacts that plot in the "ambiguous" area. The only three examples of cerussite-anglesite from Harappa all plot closely together with the samples from southern Balochistan sources. It is not presently possible to isotopically differentiate lead occurrences in the Khuzdar area from those in the Kanrach Valley and the mineral could have come from deposits in either area. Cerussite has been reported to occur in "small quantities" in the Las Bela district (Gazetteer of Las Bela 1907: 119) and is found in the oxidation zone of the deposit at Gunga (Jankovic 1986: 4) in the Khuzdar district. The "chief ore" at Shekran is cerussite (Heron and Crookshank 1954: 93) and fragments the mineral have been recovered during both previous (Hargreaves 1929: 33) and current (Ute Franke-Vogt personal communication 2005) excavations at the prehistoric site of Sohr Damb/ Nal, which is around 15 to 25 km west of the Khuzdar deposits.

Two of the fragments of massicot – a mineral that likewise forms in the oxidation zones of lead deposits, also appear to come from southern Balochistan sources. The three other examples of that mineral plot with galena fragments in the "ambiguous" region.

"Finished" lead artifacts

Six "finished" lead artifacts have been recovered at Harappa (Figure 12.24). These include a rivet or plug used to repair a shell ladle (Dales and Kenoyer 1989a: Fig. 58), several small rectangular "bars" one of which (H2000/2174-321) appears to be inscribed, a corroded tubular "rod" fragment and a tiny squarish lead "piece." All of them appear to be composed of entirely of lead metal. As this study now shifts from raw ores to artifacts that have been manipulated in various ways, it is worth emphasizing again that such artifacts, be they "finished" items, slags or residues, might potentially possess Pb isotope characteristics that reflect the mixing of lead from multiple geologic sources. When an artifact is composed of metal from only two sources, its measured isotopic values will plot somewhere along a mixing line between them (Klein et al. 2004: 470). If the end members of a mixing line are well known and no other deposits are believed to exist that have isotopic values intermediate to them, then a case can be made that

artifacts plotting along that line contain metal from both sources (ibid.). However, if other geologic deposits exist between the end members of a potential mixing line, then those same artifacts could be misassigned to them or, alternately, if an intermediate deposit is unknown then an artifact derived from it could be misinterpreted as being composed of a mix of materials from the two end member sources. The situation is further complicated when materials from more than two deposits are used to fashion an artifact or when lead derived from different types of metals are mixed (for instance if copper containing trace lead is alloyed to silver extracted from a lead deposit). In short, when examining any type of artifact other than a raw ore, the possibility *always* exists that measured isotopic values may be a reflection of something other than the actual isotopic characteristics of single geologic source. This is a fact that must be kept in mind when interpreting data of this kind, most especially assays of "finished" artifacts.

When the Pb isotope determinations (Appendix 12.3) for the six "finished" artifacts are placed on the bivariate plot (Figure 12.25), they are distributed among some of the same areas where the lead ores previously clustered. The lead composing two of the bars (including the inscribed one) appears to have been derived from sources in southern Balochistan (although one bar plots at the point where the isotopic values of that source region begin to overlap with values for samples from the Panuh deposit of Himachal Pradesh). The lead used to make the ladle plug and the rod falls into the same general "ambiguous" area that several of the lead ores previously fell. Also plotting in this same general area but directly adjacent to the single data point for galena from the Wadi Mayh area of Oman is the remaining bar and the small lead "piece." These, if real, are interesting and important source associations. However, caution in the interpretation of these data is advised as Wadi Mayh is represented by only one sample and the isotopic characteristics of these



Figure 12.24 "Finished" lead artifacts from Harappa.



Figure 12.25 "Finished" lead artifacts from Harappa plotted against South Asian lead ore fields.



Figure 12.26 Lead slags and melted lead lumps from Harappa.



Figure 12.27 Lead slags and melted lead lumps from Harappa plotted against South Asian lead ore fields.

artifacts could be the result of source mixing.

Lead slags and lumps

Isotopic assays were made of six metallurgical craft indicators (Figure 12.26) from Harappa composed of or containing significant amounts of lead. Heather Miller conducted (1994a, 1994b, 1999) a series of surface and subsurface surveys of the site in an effort to locate areas where different types of craft activities were conducted. It was on the southern slope of Mound E that she encountered the heaviest concentration of metallurgical craft indicators (metal or metalliferous scraps, slags, prills, crucible and kiln fragments) (Miller 1999: 414-419). Most of these were related to copper production but a few of the slags quite clearly had a significant lead component (i.e. yellow oxidized patches and/or visible lead prills). A yellow patch on one slag sample $(H_{93/3804-5})$ that Miller had analyzed using XRD and XRF (X-ray fluorescence) was shown to contain, among other things, lead bromide and massicot (Heather Miller personal communication 2003). A few non-descript melted lumps of lead have also been recovered during surveys and excavations at Harappa.

When the Pb isotope determinations (Appendix 12.3) for the lead-rich metallurgical slags and melted lead lumps are placed on the bivariate plot (Figure 12.27), they again cluster in the same three general areas as the ores and finished artifacts did. One of the lumps (H93/6006-1) and a frothy slag with lead prills (H93/3892-69) fall closely together on the upper left margin of the southern Balochistan cluster, while another slag (H93/3804-5) plots on the right margin of that cluster. A third slag (H93/3563-23) falls directly adjacent to the Wadi Mayh sample and a fourth (H93/3511-37) lie in the "ambiguous area," away any of the sources in the database. The other lead lump (H2001/11701-6) is isotopically analogous to deposits in the Jammu and Kashmir region.

The possibility (even the probability) that these artifacts contain lead from multiple geologic sources

should be kept closely in mind when interpreting this data. Copper oxidation (identified by XRD as atacamite [Cu2Cl(OH)3] - Heather Miller personal communication 2003) is, in addition to the previously mentioned patch of lead oxidation, quite clearly evident on slag H93/3804-5 (Figure 12.16 far right). Although most of the lead measured on this sample would have been extracted from the lead-rich portion of the artifact, it is still unknown whether or not that was representative of more than one geologic source or how any trace amounts of lead in the copper-rich portion may have affected the results. Some of these artifacts, therefore, may fall upon a yet to be defined mixing line. For instance, a mixture of lead from a source in southern Balochistan with that from one in the Jammu and Kashmir region would probably have isotopic values much like those exhibited by slag H93/3511-37. The possibility that this occurred is evaluated again later in this chapter when isotopic data for all lead artifacts are examined together.

Lead residues

In this section, I examine the compositions and probable geologic proveniences of lead residues found in small bottles and/or in other forms at Harappa. Lead minerals were used in many parts of the ancient world to produce pigments for both painting and self-adornment (Nriagu 1983; Rapp 2002: 210-211). Archaeological lead residues found at Indus sites are very likely the remains of substances that Harappans used as cosmetics. "Surma" (or "kohl" as it is known in Arabia and across the Near East) is a silvery-black eye salve/cosmetic that is ostensibly made using the mineral antimony and, thus, the thin, elongated objects found at Indus sites that were probably cosmetic applicators are often dubbed "antimony rods. " In reality, this cosmetic/salve usually contains the lead mineral galena (see my analysis of modern surma in Appendix 12.7 as well as studies by al-Hazzaa and Krahn 1995; Hardy et al. 1998; Parry and Eaton 1991; and Vaishnav 2001). Surma/kohl, other lead-



Figure 12.28 A small faience bottle (H98/8158-26) with patches of lead residue adhering to its interior (see inset detail).

based cosmetics (such as "lead white" made from powdered cerussite) and the vessels designed to hold them ("kohl" pots, jars or tubes made of a wide range of materials) are very well known from ancient Egypt and the Near East (Lucas and Harris 1999; Ungár *et al.* 2002; Walter *et al.* 1999). Although it is commonly assumed that the many miniature ceramic, faience, stone or metal vessels found at Harappa and Mohenjo-daro held, among others things, similar kinds of cosmetics, documentation of these substances has been limited. A small alabaster pot at Mohenjodaro was found to contain galena (Mackay 1938: 665) and a white substance in a small beaker at Harappa was identified as cerussite (Vats 1940: 312).

Numerous tiny vessels and vessel fragments have been recovered during HARP excavations. The contents of two contained residues that were sampled for this study. The first (Figure 12.28) is from a small faience bottle with a very constricted opening $(H_{98}/8_{158-26})$. The base of the bottle was broken and patches of a thin yellow residue could be seen on its interior. The second is from a small bottle fragment (H87/539-80) that still had a thick yellow and white residue adhering to its interior (Figure 12.29). XRD analysis of that residue indicated that it was composed of cerussite and another phase that, although I have not yet been able to conclusively identify it, has a major peak that is reminiscent of a copper mineral of some kind. Although this residue is likely the remains of a cerussite-based cosmetic like "lead white", it is not impossible that the bottle may have originally held a galena-based surma that has since oxidized as cerussite - a phenomenon documented in a study of kohl from Roman-era Palestine (Grüner 2002: 80).

I also had the opportunity to examine small vessels from past excavations at Harappa that were



Figure 12.29 A small ceramic bottle fragment (H87/539-80) with a thick layer of residue in its interior that was identified by XRD as mainly being composed of cerussite.

stored in the reserve collection of the site museum. Most contained no visible evidence of any residue that may have provided clues as to what they once held. However, the rims or interiors of six small bottles (Figure 12.30) were darkened as if they had contained a black substance such as surma. Each bottle was sampled using the EDTA technique to see if enough (or if any) lead was present on it for isotopic analysis. After the solutions were returned to Madison and assayed, it was determined that only one had contained any lead at all – a small jar from Vats' excavations (1940) identified by the number *3906* (Figure 12.30 far right). If the other vessels had once held a black cosmetic, it was probably made from organic substances such as "lamp black mixed with fat" as Ernest Mackay had once proposed (1938: 665), rather than a lead mineral.

One final, particularly interesting residue from the HARP excavations was originally thought to be a metal "rod" (Figure 12.31 – artifact H88/197-1). It was recovered in the grave of an adult woman in a disturbed (in antiquity) cemetery area burial



Figure 12.30 Six tiny ceramic vessels from Harappa Museum's Reserve Collection. Only the one on the far right (# 3906) was found to have once contained a lead substance.



Figure 12.31 Artifact H88/197-1, which was originally described as a lead "rod" but is now believed to be a solidified lead residue.

dating to Period 3B (Figure 12.32 A). The "rod", which is somewhat fragile and not as heavy as would normally be expected of an item made of metal, was excavated already in a fragmentary condition (Figure 12.32 B). The material that it is composed of was initially classified by Dales and Kenoyer (1989: 91) as "lead/orpiment" because of its bright orange-yellow (very orpiment-like) oxidized exterior. When I first examined this artifact in December of 2003, I noticed that a small amount of powder had accumulated in the bag holding it. This was collected and analyzed using XRD. The resulting pattern (Figure 12.32 C) indicated that the "rod" (or at least the powder coming off of it) was composed primarily of *wulfenite*



Figure 12.32 Excavation and analysis of artifact H88/197-1. [A] The artifact was found within Feature 194a – a Period 3B burial. [B] Excavation of the "rod" in two pieces. [C] XRD revealed that it was composed of wulfenite (lead molybdate) and graphite. [D] BSE image of the "rod" in section. [E] Detail of the artifacts non-metallic, heterogeneous matrix with galena crystals imbedded within it.

(peaks identified with a W on the XRD scan) and *graphite* (identified with a Gr). Wulfenite – lead molybdate (PbMoO4), is another mineral found in the oxidation zones of lead deposits and sometimes has an orange-yellow appearance (Read 1979: 466).

Graphite is, of course, pure native carbon. It crossed my mind that the carbon in the artifact, although identified as graphite, may actually be charcoal, "bone black", or "lamp black" – carbonaceous substances used as black cosmetic pigments in South Asia (O.P.



Figure 12.33 [A] Wooden "kohl tube" from Gurob, Egypt, Image used courtesy of the Petrie Museum of Egyptian Archaeology, University College London. [B] Reconstruction of a wooden "surma tube" with a applicator set into the powered lead-based cosmetic held within it. [C] Reconstruction of the oxidized remains of the surma after the tube and applicator. [D] Photograph and BSE scan section of artifact H88/197-1 for comparison.



Figure 12.34 Lead residues from Harappa plotted against South Asian lead ore fields.

Agrawal 1999: 193-194). However, carbons such as these are amorphous and would not have registered peaks on the XRD scan.

A small portion of one of the broken ends of the "rod" was removed for further study using EMPA. The BSE image of this piece in section (Figure 12.32 D) revealed that it is not composed of solid metal (hence the artifact's light weight) but rather is a composite of several granular substances of varying consistencies that have been mixed together. EDS scans indicated that one of these substances was lead sulphide (galena). Cubic crystals of galena can be clearly seen in the artifact's matrix (Figure 12.32 E).

I believe that this artifact, rather than being the remnants of a rather fragile "rod", may instead be the solidified residue of surma powder that was once held in the tube-shaped interior of a container made of a perishable material like shisham wood or bamboo. Cylindrical "kohl tubes" made of wood or reed are well known from ancient Egypt. The one shown in Figure 12.33 A was excavated by Flinders Petrie at New Kingdom site of Gurob (Thomas 1981: 63 [UC 7891]). There is a groove running down the lengths of the Harappan "rod"/residue fragments (visible in section on the upper left of the BSE scan on Figure 12.32 D) that may be the place where a thin cosmetic applicator (probably also perishable) was set into the powder. Over time, the wooden tube and applicator (Figure 12.33 B) disintegrated leaving only the lead-based powder, which consolidated but cracked as it oxidized, in the tubular rod-like shape of the container's interior with a small groove running down its length (Figure 12.33 C). I believe this reconstruction best accounts for the features exhibited by this artifact (Figure 12.33 D).

The Pb isotope compositions of the four residues analyzed (Appendix 12.3) indicate (Figure 12.34) that the lead in each of them was probably acquired from sources in the Jammu and Kashmir region. It should be noted, however, that although the "rod"/ residue (H88/197-1) that contained wulfenite would

seem to have come from one of the Buniyar sources, this mineral has not as yet been reported in that area or, for that matter, anywhere in Jammu or Kashmir. Even so, wulfenite can form due to lead-molybdenum impurities in sphalerite (Guilbert and Parks 1986: 809) - a zinc mineral which is found throughout the zone of sulphide mineralization at Buniyar (Sharma and Sachan 1998). Moreover, mineralization there is thought to perhaps be epigenetically related to intrusive granitoid plutons in the vicinity (Kaul 1981), which could have easily contributed the molybdenum impurities that oxidized with lead as wulfenite $(PbMoO_{4})$. Also significant with regard to the "rod"/ residue, is the fact that graphite associated with igneous rocks is abundant in the vicinity of Buniyar (Mehta 1957) as well as nearby the unassayed lead deposits of the Doda district (Gupta and Guha 1988).

Although more geologic research remains to be done in order to fully understand the nature of lead deposits in Jammu and Kashmir, it is of interest to note here that the southernmost occurrences in the Buniyar area (those nearest to the igneous intrusives) are found around the village of "Surmawali" (Raina 1977), which is presumably so named (as many villages in the vicinity of lead deposits seem to be, e.g. *Surmai* village in the Khuzdar district of Balochistan) because the raw material for surma is obtained at this location.

ISOTOPIC ASSAYS OF LEAD AND SILVER ARTIFACTS FROM EIGHT OTHER PREHISTORIC SITES

In this section, I present the results of a series of Pb isotope analyses made on various kinds of lead artifacts from the prehistoric sites of Shahr-i-Sokhta, Mundigak, Mehrgarh, Gola Dhoro, Nausharo and Mohenjo-daro (Appendix 12.4). Isotopic assays of silver artifacts from Mundigak, Nagwada, Gola Dhoro, Mohenjo-daro and Allahdino (Appendix 12.5) have been made and are also evaluated in relation to the lead deposits database. Although these eight ancient settlements are not the focus of this study,



Figure 12.35 Lead and silver artifacts from the site of Mundigak, Afghanistan.

incorporating data on artifacts from them into it, 1) allows the lead acquisition networks that residents of Harappa were involved in to be viewed from a perspective that is more holistic, 2) provides an indication (by proxy) of what the isotopic character of lead deposits in certain regions (southern Afghanistan and eastern Iran) not represented in the database *may* be like and 3) permits a material type (silver) to be examined that was widely used by Indus Civilization peoples, but for which examples from Harappa are not available for analysis.

The sites and artifacts

Lead artifacts from six prehistoric sites were sampled and analyzed using the EDTA/ICP-MS technique. Data for and descriptions of individual



Figure 12.36 [A] Galena fragment and [B] copper surma bottle from Mohenjo-Daro.



Figure 12.37 A flattened lead lump (BSR-5930) and a silver ring from (BSR-7053) from the site of Gola Dhoro, Gujarat.



artifacts can be found in Appendix 12.4. All site locations are noted on figures 12.2 and 12.3.

Dr. Massimo Vidale graciously provided access to two galena samples from the Helmand Civilization urban center of Shahr-i-Sokhta in the Seistan Basin of eastern Iran (Tosi 1982). As the majority of metallurgical finds from this site date to the midthird millennium BC (ca. 2700 to 2500 BC) levels there (M. Tosi cited in Hauptmann 2003: 198), it can probably be assumed that these archaeological ores, which are part of a larger collection of mostly copper artifacts and manufacturing debris presently under examination from that site (Giardino *et al. in preparation*), were deposited at a time roughly ranging from late Period 2 to early Period 3A at Harappa. Although few in number, these artifacts can provide a general idea of what the isotopic character of lead deposits in eastern Iran *may* be like, assuming, of course, they were obtained from that region.

Pieces of galena along with sheet-like lead fragments and a ring made of that metal (Figure 12.35 – MGK 1 to 12, & 14) were recovered in Period III and IV levels (ca. Harappa Period 2 to mid-Period 3) at the Helmand Civilization settlement of Mundigak (Casal 1961) in southern Afghanistan. These artifacts are now in the collections of the Centre de Recherches Archéologiques Indus-Balochistan, Asie Centrale et Orientale at the Musée Guimet, Paris, which is presently headed by Dr. Jean-François Jarrige. I was kindly allowed to extract lead from these artifacts, which were quite possible derived from the source just 10 km from Mundigak at Asad Qala, (Jarrige and Tosi 1981).

Also in the collections at Musée Guimet are a

number of lead artifacts from the ancient settlements of Mehrgarh and nearby Nausharo, which are located at the foot of one of the major passes connecting the Indus Valley to the central Balochistan highlands and beyond to the Helmand Basin. Dr. Jean-François Jarrige, who directed excavations at both sites, generously permitted lead to be extracted from these artifacts (not pictured) for analysis. The Mehrgarh artifacts are galena fragments from the site's Balochistan Tradition Neolithic levels (Period IIB – ca. 5000 BC). The artifact from Nausharo is a lead ring from the initial Indus Civilization levels (roughly equivalent to Period 3A at Harappa) at that site. The nearest lead sources are in the Khuzdar region, around 200 km to the southwest.

Two lead artifacts from the Indus Civilization city of Mohenjo-daro in northern Sindh were assayed. The first is a galena fragment (Figure 12.36 A) recovered during the surface surveys by the German-Italian mission to that site (Jansen and Urban 1984), which is now stored at the Department of Archaeology and Museums' Excavation Branch collection in Karachi. The other sample was extracted from a "surma" residue in small copper vessel (Figure 12.36 B) on display in the cases of the Mohenjo-daro site museum. Although the vessel is presumably from one of the early excavations at that site, no accession numbers or other identifying information could be found for it. The closest lead deposits to that site are those in the Khuzdar region – around 170 km to the west-northwest. It is recognized that, because the sample was taken from a copper vessel, the Pb isotope.

A flattened lead lump (Figure 12.37 – artifact BSR-5930) from the Indus site of Gola Dhoro (Bhan *et al.* 2004) in northern Saurashtra, Gujarat was sampled with the kind permission and assistance of two of that site's excavators, Dr. P. Ajithprasad and Dr. Ambika Patel of Department of Archaeology at Maharaja Sayajirao University, Gujarat. The artifact belongs to Phase-II, which is the "Classical/Urban" Harappan Period at the site.

The Pb isotope characteristics of silver artifacts from five sites were determined using ICP-MS. Most were sampled using the EDTA lead extraction technique. Immersion time in the sampling solution was tripled to six minutes because, presumably, only a trace amount of lead was present in the metal. Although lead was indeed extracted from silver items in amounts considerably lower than for those composed of lead, concentrations were still sufficient for isotopic analysis. Lead from one artifact (from Nagwada), which was in Madison undergoing other types of analysis, was brought into solution by dissolving precisely 0.02 grams of metal from it in ultra-pure nitric acid. Isotopic data for and descriptions of the silver artifacts discussed below can be found in Appendix 12.5. Site locations are noted on Figure 12.2.

A small silver ring (Figure 12.23 A – MGK 13) from Mundigak Period IV was sampled for Pb isotopes at the same time as the lead artifacts from that site (discussed above).

A detailed analysis of a silver ring fragment (Figure 12.38 A) from the site of Nagwada (Hegde et al. 1988) in the Rupen estuary of northern Gujarat was begun in collaboration with one of the excavators of that ancient settlement, Dr. Kuldeep Bhan of the Department of Archaeology at Maharaja Sayajirao University, Gujarat. The artifact is heavily corroded. BSE imaging (Figure 12.38 C) and EDS scans revealed that the silver (Ag) metal of the ring's exterior has undergone alteration to silver bromide (AgBr), encompassing some sand grains (SiO2) from the sediment it was buried in as it did so. The interior of the artifact (Figure 12.38 D), however, is unaltered. Five WDS scans (Figure 12.38 B) were conducted across the center of this unaffected portion and then averaged in order to quantify the absolute amounts of silver, copper (Cu), lead and gold (Au) in the metal. The silver ring was found to be very pure (\approx 98.5 %); even more so than those silver artifacts from Mohenjo-daro and Lothal that have been analyzed



Figure 12.39 Silver artifacts from Mohenjo-Daro sampled for this study.

(summarized in Kenoyer and Miller Table 5.3). Copper (\approx 1.5 %) makes up most of the ring's nonsilver content. Although it is likely that copper was deliberately alloyed with the silver during production of the ring, it is not impossible that it was retained from the extraction of silver from copper or lead ores. Although very little lead was present in the metal (\approx 0.2 %), enough could be brought into solution (as described above) for isotopic analysis using the ICP-MS.

This tiny ring fragment is of special interest because it was from recovered from Nagwada's earliest occupational phase, which immediately precedes the Harappan Period at the site (Hegde et al. 1988: 62). Certain ceramics forms are found in this level and in equivalent contexts at several other sites in general vicinity (Dholavira, Sukotada, Moti Pipli - Figure 12.7), which exhibit close similarities to those used by the Early Harappan Amri-Nal and/or Kot Dijian peoples of southern Balochistan and Sindh (Possehl 1999: 603-609). These finds probably reflect the initial expansion of those cultures into the northern Gujarat area (Possehl 2002b: 40-41). If the silver used by Early Harappan peoples was extracted from lead ore as some have proposed (discussed in Chapter 4), then it will be of great interest to know whether or not the lead in the Nagwada ring is isotopically more analogous to the closest deposits of argentiferous lead in southern Rajasthan (specifically, the Zawar deposit

and the Rampura-Agucha/Rajpura-Dariba belt – beginning around 225 km to the northeast of the site) or to the next nearest sources in the Khuzdar region (around 700 km away), which just so happens to be the northern part of the Amri-Nal culture area.

A small metal ring (Figure 12.37 – artifact BSR-7053) from Gola Dhoro was sampled at the same time as the flattened lead lump from the same site described above. It belongs to Phase-II, which is the "Classical/Urban" Harappan Period at the site. I have included it here with the silver artifacts because it was largely covered with a dark gray layer that is reminiscent of oxidized silver. However, the ring clearly has a high lead content (it may even be mostly lead). There are yellow patches on its surface that resemble lead oxidation and the EDTA solution in which it was washed was, unlike most silver artifacts that I have sampled, fully saturated with lead.

Lead was non-destructively extracted from five silver items excavated at Mohenjo-daro (Figure 12.39), which are currently on display at the National Museum in Karachi. The silver bangle sampled (NM-1) was one of two from a jewelry hoard (Marshall 1931b: Plate CXLVIII *in hoard*, Plate CLXIV b *restored*) found in a "Late Period" (roughly equivalent to Harappa Period 3C) room in the DK area (Mackay 1931a: 250, 1931c: 529). The other silver items sampled – three tiny beads (NM-2, 3 & 4) on a strand of 18 and a button or nose stud (NM-5), are from the same



Figure 12.40The Allahdino jewelry hoard - top image.The 10 silver ornaments from the hoard sampled for this study - bottom image.



Figure 12.41 Lead and silver artifacts from eight sites plotted against South Asian lead ore fields.

excavations but are not specifically identified in the site report.

Prof. Walter Fairservis discovered a jewelry hoard (Figure 12.40 *top image*) at the small Harappan site of Allahdino, in southern Sindh (1993: 109). Ten silver items from that hoard (Figure 12.40 *bottom image*) were sampled at the Department of Archaeology and Museum's head office in Karachi.

Results

When the results of the Pb isotope analyses (appendices 12.4 and 12.5) of the lead and/or silver artifacts from the eight sites discussed above are placed on the bivariate plot of South Asian lead ore sources (Figure 12.41), all cluster in the upper left corner of the plot in two of the same general areas that many of the lead artifacts from Harappa previously fell. None appear to be even remotely related to sources in Jammu or Kashmir or to the many sources in Rajasthan and Gujarat (Amba Dongar excepted). Because all of the artifacts cluster in one section of the plot and a great many of them overlap, I have created two separate figures – one for lead (Figure 12.42 A) and one for silver artifacts (Figure 12.42 B), which focuses closely in on the area around where they fall. On these plots I have also placed the Pb data that was extrapolated (Appendix 12.6) from Stos-Gale's published (2001) isotope assays of galena samples



 Figure 12.42
 [A] Lead artifacts from six sites plotted against select lead ore fields. [B] Silver artifacts from five sites plotted against select lead ore fields.

from the well-known argentiferous lead deposit at Nakhlak in central Iran.

- Lead artifacts

On Figure 12.42 A, twenty-six lead artifacts from six sites have been plotted in relation to select ore sources in South Asia, Oman and Iran. Before I begin to discuss the possible geologic provenience of those artifacts, first note that the error bars, when viewed from this closer perspective, are naturally much larger. Whereas on the previous bivariate plots the three areas where artifacts tended to fall/cluster were separated by distances that were far greater than the analytical uncertainty of the results, here many of the individual artifacts and groups of artifacts that I discuss are situated, relative to one another, within or near the range of possible error. Nevertheless, many artifacts cluster in distinct groups (rather than spread widely and randomly) that appear to be genuine and meaningful.

Starting with the Mohenjo-daro lead artifacts, we see that the single galena fragment from that site plots on the margins of the southern Balochistan source area. The cluster of Nakhlak (Iran) source samples is within the range of analytical error. However, given the location of the site, it is far more probable that the archaeological ore originally came from the former region. The lead extracted from the "surma" bottle falls in the "ambiguous" area where several ores, artifacts and slags from Harappa have previously plotted. Still, it would seem that the lead-based substance that was once in that bottle came from a different source area than that in the substances once held in similar bottles at Harappa or the galena fragment just discussed. It is, of course, possible that lead from multiple sources are present in the residue and what is we are seeing is an isotopic value plotting along a mixing line.

The Nausharo lead ring and most of the Mehrgarh galenas plot in an extended linear group, the majority of which fall within the area defined by the southern Balochistan sources. A few of the galena fragments, however, continue along the linear trend into areas where the Nakhlak and the Chagai Hills samples lie. It is difficult to judge whether or not the Mehrgarh galenas represent ores from a single source or multiple ones, although the linear manner in which they group indicates it may be the former. Perhaps when additional geologic samples from deposits in western Balochistan (Chagai region) are analyzed they will begin to isotopically overlap with the southern Balochistan deposits or vice versa. Although I doubt any of the archaeological ores actually came from the central Iran source (given that it is nearly 1400 km away and there were many other deposits that were closer), it is quite conceivable that Chagai Hills galena found its way to the Bolan Pass region.

The Mundigak lead artifacts plot in interesting ways. Of the ten galena fragments, seven (all of the unlabeled Mundigak data points on the figure) fall into a loose linear group (which may or may not represent a single source) that partly overlaps with the southern Balochistan deposits and then extends into an area where none of the geologic samples in the lead database lie. The three remaining Mundigak galenas (labeled by numbers 4 through 6) plot squarely within the southern Balochistan source area, with the Mehrgarh-Nausharo samples just discussed. Admittedly, given the degree of analytical uncertainty at this level, all of these lead ore artifacts could in reality be isotopically more similar (or dissimilar) than is evident here. However, I believe there are probably at least two sources represented among the Mundigak ores. Refer back to Figure 12.35 for a moment. The white weathered exteriors (probably cerussite) of artifacts MGK-04, 05 and 06 set them apart from other galenas, which have yellowish exteriors. These three ore fragments, which were excavated together, could actually be from a southern Balochistan source while the group of seven might be from the Asad Qala mine nearby Mundigak (samples from that deposit will eventually need to be

analyzed in order to confirm this). The lead ring and sheet fragments (noted on the figure) plot differently still. Isotopically, the ring falls on the margin of the area defined by the Chagai Hills source samples and lies adjacent to a few of the Mehrgarh galenas. The lead sheet fragments fall well away from the other Mundigak artifacts – one is adjacent to two Oman data points while the other plots nearby the single datapoint for the galena occurrence at Amba Dongar, Gujarat. The sheets have a white oxidized exterior like MGK-04 through 06 and could be a mixture lead from the same ore source and one or more others that are presently unknown. It is unlikely that they are actually from Oman or Gujarat, however.

Next we consider the two galena fragments from Shahr-i-Sokhta in eastern Iran. One of them plots in the area defined by the southern Balochistan source samples. However, it also groups closely with the cluster of Mundigak ores that may have been acquired from a deposit in the vicinity of that site. The second fragment falls in an area where data points for galena samples from both the Chagai region and Oman lie. It is entirely possible that the two ores could have actually have come from any of these areas. Close ties between Shahr-i-Sokhta and Mundigak clearly existed during the Integration Era of the Helmand Tradition (Shaffer 1992); the lead deposits in the Chagai region of western Balochistan are the closest (\approx 200 km) sources of that metal to the site; graves with Nal culture jars in the cemetery area at Shahr-i-Sokhta (Piperno 1979: 125) suggest connections with southern Balochistan; and there is good evidence that long-distance trade relationships extended as far as the Gulf region and Oman (Ratnagar 2004: 66-67). Unfortunately, the lead deposits in the most pertinent region, Iran, are either poorly characterized or (mostly) not characterized at all. Making a stronger statement about the provenience of these artifacts is, therefore, not possible at this time and, as I will discuss, it may not be possible using Pb isotope data alone and/or using an ICP-MS to make isotopic

measurements.

Lastly, the flattened lead lump from Gola Dhoro appears to be composed of metal derived from southern Balochistan. It seems then that Harappans dwelling here had access to the same extensive raw material network through lead from that region was being acquired by Indus Civilization peoples all the way from Haryana in the north down to Gujarat in the south.

- Silver artifacts

Now we move on to the 19 silver objects from five sites. The data for these are plotted on Figure 12.42 B in relation to select lead sources in South Asia, Oman and Iran.

The ring from Mundigak plots with the three samples from the Shi Shi Valley lead occurrence in Chitral, NWFP. I believe it unlikely, however, that the silver for this item was extracted from that particular lead source as it somewhat "off the beaten path" and is not reported to be argentiferous (Tahirkheli et al. 1997). Certainly more accessible to residents of Mundigak would have been the silverbearing lead deposits in the Ghorband and Panjshir valleys of north-central Afghanistan (Collins 1894; Pascoe 1931: 675). Those sources are located along the major trade and communication routes crossing the Hindu Kush of northern Afghanistan (Howland 1940) and, although unassayed, occur in the same general geologic terrain as the Chitral deposits. Of course, it is also possible that the ring contains lead from multiple sources and that its evident isotopic characteristics are a reflection of that rather and an individual deposit.

The data point for the Nagwada silver ring fragment falls within the area encompassed by the southern Balochistan lead deposits. Although it is possible that the isotopic characteristics of this artifact actually reflect the mixing of lead from multiple sources, it is very unlikely that one of those was the argentiferous deposit at Rajpura-Dariba in

southern Rajasthan, which is situated at the opposite end of the bivariate plot of the main ore sources (refer back to Figure 12.41). The only deposits anywhere in Rajasthan or Gujarat with isotopic characteristics that remotely resemble those of the lead in the ring is the small occurrence at Khera Mawal and the showing at Amba Dongar. However, even from the closer perspective used for the plot of silver items (Figure 12.42 B), the artifact and the data points for that source are separated by a distance greater than the range of possible analytical error. A geologic provenience of southern Balochistan for this silver ring from the initial occupation of Nagwada is very much consistent with other evidence for the movement of Early Harappan peoples from that region and Sindh into northern Gujarat during the early third millennium.

The five silver items sampled from Mohenjo-daro plot in a loose group around the somewhat ill-defined area where so many lead artifacts from Harappa have also fallen. The bangle is directly adjacent the single data point for the Wadi Mayh (Oman) lead occurrence. The three beads group nearby. The button/nose stud plots slightly farther away but still in a manner that seems to suggest that it and the others ornaments are very possibly composed of metal containing lead (either natural or added) from the same source or sources. The Wadi Mah occurrences is not, as far as I have been able to determine, known to be argentiferous and, therefore, the probable geologic source of the silver used to make these items remains "ambiguous."

Eight of the ten silver ornaments from the Allahdino hoard plot within the area defined by geologic samples from the southern Balochistan lead deposits. Of the two remaining pieces, one – AD-9 (number 9 on the figure), falls with the five Mohenjo-daro silver artifacts in the "ambiguous" area. The other – AD-6, could simply be an outlier of the southern Balochistan deposits (AD-5 lies nearby, within that defined area). On the other hand, AD-6 (and perhaps AD-5) might be on an isotopic mixing line between one of those deposits and the ambiguous "source" where AD-9 and the Mohenjo-daro items fall. Or it may be from silver extracted from a different source area, one that is not represented in the database. Whichever the case actually is, the isotopic characteristics of the lead in most of the Allahdino samples are analogous to the sources of that metal closest to the site itself. Still, it is important to note that two examples (AD-1 & AD-8) fall near (within the range of analytic uncertainty) the Nakhlak argentiferous lead deposit of central Iran and one (AD-5) lies similarly near to the Amba Kala deposit (also argentiferous) of Himachal Pradesh.

Lastly, the silver/lead ring from Gola Dhoro falls squarely among the cluster of three data points representing lead occurrences in Oman at Ibra, Qumayrah and Wadi Nujum. It also plots within the range of error for lead deposits in the Chagai Hills of far western Balochistan. However, if we assume for the time being that the ring is made from metal derived from only one deposit, then it seems more likely the source was probably in Oman rather than the Chagai Hills given the links between the Indus region and eastern Arabia during the Harappan Period.

INTERPRETATION OF THE PB ISOTOPE DATA FOR LEAD AND SILVER ARTIFACTS FROM ALL SITES

Pb isotope assays have been made on 79 lead and/or silver artifacts from Harappa and eight other prehistoric sites in the Indus Valley and Helmand regions. When the results for all are placed on the bivariate plot of lead ore deposits (Figure 12.43) the data points representing them cluster in three main areas. Based on these patterns of clustering, the "probable geologic provenience" of each lead artifact from Harappa is stated in the final columns of appendices 12.2 and 12.3.

Those artifacts that cluster in the Pb isotope fields defined by the geologic samples from the





Jammu and Kashmir region (Buniyar and Riasi area deposits) are, at present, the most secure in terms of their geologic provenience associations. With the exception of a slight degree of overlap with the Ambaji-Sendra belt (which, in actuality, is due to just two data points that are distant outliers of the main group of Ambaji-Sendra belt assays), there are no lead sources in the database with isotopic characteristics that even remotely resemble this one. Of course, this may eventually change as more sources are analyzed. Until that time, however, I believe it can be stated with a high degree of confidence that the lead in these particular artifacts probably came from sources in "Jammu and Kashmir."

Conversely, the artifacts with the least secure geologic provenience associations are those that cluster in what I have repeatedly called the "ambiguous" area around the Wadi Mayh and Amba Kala deposits. Although many fall next (or are within the range of analytic error) to the data points for those three deposits, I am reluctant to make even a provisional provenience association for any of them because the "isotopic space" in this area so poorly defined in relation to known geologic sources. The characteristics that cause most of these artifacts to plot in this space *could* be due to the mixing of lead from two or more sources. Even if that is so, however, we may be certain that at least one geologic deposit with the same general isotopic characteristics as the artifacts in this cluster also exists and was exploited, simply because several of those artifacts are raw ores of lead (galena and massicot fragments from Harappa) that have not been subject to source mixing. In fact, there is probably more than one geologic deposit represented in this cluster and I have placed dashed ellipses on the figure around the areas where I think they could be. These ellipses are purely speculative and only further characterization of the geologic deposits around them (as well as of new ones) will allow us to determine if the artifacts that they group genuinely belong together. So although

some of these lead and silver items may possibly come from sources in Oman or Himachal Pradesh, for the Harappa artifacts I can only state that their geologic proveniences are "unclear at this time"

Artifacts in the third cluster are the most difficult to interpret. They fall mainly in the southern Balochistan "field" defined by the Khuzdar and Kanrach Valley lead deposits. Some, however, extend beyond that field and plot on or near data points belonging to samples from deposits in Iran, Oman and the Chagai region, as well as into areas of the bivariate plot that are not currently defined by any geologic sources. Those that do this belong to either the Helmand Civilization sites of Mundigak and Shahr-i-Sokhta, where residents may have been exploiting some of the lead sources in the regions around their settlements, or to Mehrgarh, which may have been acquiring lead through long-distance trade networks extending to some of those very same source regions. It seems then almost certain that multiple geologic deposits dispersed across a wide geographic area are represented among the lead and silver artifacts making up this cluster.

The "fields" that are located in the section of the bivariate plot where artifacts in the third cluster fall are, at the present time, fairly distinct, i.e., there is minimal overlap between them. However, as additional ore samples from these and other sources are incorporated into the database there is reason to expect that they will begin to overlap with one another considerably more. That reason is because the geologic age (the principal factor influencing isotopic composition) of most of the metallogenic zones in which lead (and copper) is found in Balochistan, eastern Iran, southern Afghanistan and even Oman are broadly similar (Bazin and Hübner 1969; Samani 1998; Shams 1995b; Wolfart and Wittekindt 1980). One of the purposes for including data from the Nakhlak deposit of central Iran was to illustrate just how isotopically alike lead deposits from those different regions can be. An

artifact with measured Pb isotope values causing it to plot with the Nakhlak data points would be still be within the range of analytic error to sources in Oman, southern Balochistan and the Chagai Hills region (Figure 12.42 A shows this best). Admittedly, Pb isotope measurements made using an ICP-MS do leave something to be desired (namely, extremely high precision and useable ²⁰⁴Pb values) when attempting to assign artifacts to sources within the limited isotopic space that this cluster encompasses. TIMSgenerated data would undoubtedly improve source resolution in this part of the bivariate plot by lowering analytic uncertainty and providing a third dimension for comparison. However, as the database of assayed deposits in the Indo-Iranian highlands and Oman increases, no amount of precision will enable one to differentiate overlapping areas that are isotopically identical. This could limit the ability to use Pb isotope data alone to identify the provenience metal artifacts from sites located within those regions.

The situation is entirely different for Harappa and sites in the Indus Valley because, as previously discussed, they are surrounded by regions in which the isotopic characteristics of the metallogenic zones within them are, on the whole, extremely different from one another. The precision provided by the ICP-MS is more than sufficient to differentiate artifacts coming from one regional zone or another. So as for the probable provenience of the artifacts from Harappa and other Indus sites that are among the third cluster under discussion, all fall into the field defined by the Khuzdar/Kanrach Valley data points. Although this field may eventually be obscured by data from other deposits, for now we can state, with a good degree of confidence, that the lead for those artifacts was acquired from nearest isotopically analogous sources - those in "southern Balochistan."

The same may be stated of the Nagwada ring and most of the silver artifacts from the Allahdino hoard. Although it is certainly possible the metal for those ornaments was extracted from isotopically analogous lead deposits in a different area (perhaps coming to the Indus region through trade with Mesopotamia as Ratnagar has suggested [2004: 199]), I believe that, with these new findings, the evidence that southern Balochistan was a silver source for Indus Civilization peoples is now fairly good. At least one of the Khuzdar occurrences (Gunga) is known to be highly argentiferous (Shams 1995b: 246) and sediment surveys of nalas (streams) coming off of the Mor Range in the lower Kanrach Valley area detected elevated levels of silver suggesting that the existence of sources in this region are quite good (Naseem 2002). Old lead mines and smelting areas are well-documented around Khuzdar area (Hassan 1989; Siddiqui and Sharp 1993) and the only silver artifacts reported in this region are from Sohr Damb/ Nal (Hargreaves 1929), which is in vicinity of Gunga and these old workings. The Early Harappan Nal and Indus Civilization era Kulli occupations at that site and in the southern Balochistan highlands provide the cultural links through which silver could have made its way to Allahdino in lower Sindh and Nagwada in northern Gujarat. Short of discovering archaeological remains that document the cupellation of silver from lead deposits in this area, the evidence (isotopic, artifactual and circumstantial) that at least some Harappan silver likely came from southern Balochistan sources cannot get too much better.

ONGOING STUDIES OF LEAD AND SILVER ARTIFACTS AND SOURCES

Studies of lead and silver artifacts and their potential geologic sources continue. Analyses of such artifacts from the Indus cities of Dholavira and Rakhigarhi, as well as from the Bronze Age burial site of HD-10 (Salvatori 2001) in the Ra's al-Hadd region of Oman, were in progress as this book was being prepared for publication. These new Pb isotope assays are being conducted using either MC-ICP-MS or TIMS, which will provide useable ²⁰⁴Pb data and even more accurate ²⁰⁸Pb, ²⁰⁷Pb and ²⁰⁶Pb values. Eventually, all of the artifacts and geologic samples ran on the old ICP-MS at the LARCH will be reanalyzed using these instruments. Although this work is ongoing, a few brief, preliminary observations can be made here.

Evidence continues to mount suggesting that southern Balochistan was a major source area for lead and silver during this period. Metal or raw ore derived from occurrences in that region, which was being used by Indus Civilization peoples at Harappa, Mohenjo-Daro, Allahdino, Nagwada and Gola Dhoro, was also being acquired by residents of both Rakhigarhi in Haryana and Dholavira in Gujarat. The deposits of southern Balochistan, specifically those in the Kanrach Valley, may have also been the source of four lead beads found in a cairn burial at HD-10 in Oman.

New source areas are also coming to light. Although residents of Dholavira acquired most of their lead from occurrences in southern Balochistan, a minor amount appears to have been derived from the Ambaji deposits of northeastern Gujarat.

COPPER

The focus of this chapter now shifts to copper, which was undoubtedly a material of enormous importance not only for Indus Civilization peoples but also for their contemporaries in many parts of Asia and Europe. Only through a concerted research program that combines isotopic and compositional analyses with stylistic and contextual studies (a la the recent work by Lloyd Weeks [2004] on copper alloy artifacts from Bronze Age sites in the United Arab Emirates) will it be possible to begin to fully understand the exploitation, production, trade, use and re-use of this metal at Harappa and elsewhere in ancient South Asia. My intention here is not that ambitious. In this section, I use Pb isotope analysis to compare the few examples of raw copper ore that have been recovered at Harappa to a small database of isotopic assays made of geologic samples and copper production debris (slags) representing sources in five broad regions from which Indus Civilization peoples may have acquired this important metal. Although this study is small in size, it is the first direct analytic comparison of copper artifacts from Harappa to geologic materials from potential sources since Sana Ullah's early work (1940).

COPPER ORE AT HARAPPA

Only seven copper minerals have been identified from among the more than 2600 copper or copperalloy artifacts recovered at Harappa. In addition, the only vitrified craft indicators related to this metal are things like crucible and kiln-wall fragments that are made of clay-based materials (Miller 1999: 414). The scarcity of ores and the absence of siliceous slags is indicative of production activities that involved melting and working metal that had already been processed in some way rather than the smelting of raw copper ore in order to extract metal (Miller 1994b: 505). This is not at all surprising. It certainly would have made little sense to transport large amounts of raw ore over 400 km to the site (the minimum distance to deposits of any significance) when it could have been reduced to easily manageable quantities of metal at or closer to its source. The seven ores (Appendix 12.9) - four small fragments of chalcocite and three of malachite (Figure 12.44); all come from the south slope of Mound E, which is where the heaviest concentration of copper production debris was found (Miller 1999: 414-415). All are from surface or disturbed contexts except for a single malachite fragment from Period 3C levels.

THE PB ISOTOPE DATABASE FOR COPPER ORE SOURCES

Kenoyer and Miller suggested (1999: 115) that Harappan metalsmiths might have acquired copper from deposits in four main source regions: the broad region west of the Indus Valley that includes the







Figure 12.45 Assayed copper ore deposits and archaeological sites featured in this section.

combined areas of Balochistan and Afghanistan; Oman; the Aravalli mountain range of Rajasthan and northern Gujarat; and, perhaps, Iran. I basically agree with this but would add a fifth region – the Himalayas and related highland areas of northern India and Pakistan, as a potential source area, especially since it now appears that some of the lead found at Harappa was acquired from deposits in Jammu and Kashmir.

Prior to this study, Pb isotope assays of copper

ores had only been made for deposits located in Oman, Iran and the Aravallis. The database (Appendix 12.8) presented here includes isotope values extrapolated from those previously published data along with new analyses of copper ores and/or slags from seven locations in Balochistan, Waziristan, the Himalayas and the Aravallis. Below, I focus mainly on the assayed the deposits that are featured in the database (Figure 12.45). For more comprehensive overviews that detail individual occurrences and the many old workings found in these regions see the following: for Pakistan and India (Ahmad 1969; Chakrabarti and Lahiri 1996; Geological Survey of India 1994; Kazmi and Jan 1997; Nandan *et al.* 1991); Afghanistan (ESCAP 1995: Peters *et al.* 2007); Iran (Bazin and Hübner 1969); and for Oman (Weeks 2004).

The Aravallis

The Aravalli Range of Rajasthan and northern Gujarat is considered by many to have likely been one





Figure 12.46 Top image - Looking toward the Khetri mines from Singhana with the small "mountains" of slag in the foreground. Bottom images - Fifteen meter high layered slag heap at Singhana.

of the main sources (some believe *the* main source) for the copper used by Indus Civilization peoples (Agrawal 2000; Agrawala 1984; Allchin and Allchin 1982; Asthana 1993; Chakrabarti and Lahiri 1996; Dhavalikar 1997; Kenoyer and Miller 1999; Pascoe 1931; Sana Ullah 1940). Nineteen major zones of base metal mineralization occur intermittently along its approximately 600 km length and old workings have been noted in every one of them (Geological Survey of India 1994). The richest deposits are found in the northern part of the range in the zone known as the Khetri "Copper Belt." Although it was not possible to obtain ore samples from deposits within this zone, copper smelting slags from two locations were provided by Kishore Raghubans, then a PhD student at M.S. University, Baroda who did his dissertation research on the Ganeshwar-Jodhpura culture complex, which was an indigenous, non-urbanized Chalcolithic society inhabiting the northern Aravalli region during the third millennium BC (Agrawala and Kumar 1982). The first set of slags was acquired at Singhana (near Khetri), where the numerous small hills (heaps) composed entirely of smelting debris provide an indication of the intensity and duration of the copper extraction/production activities that have taken place in this region (Figure 12.46). The second set comes from a smelting area located less than one kilometer from the site of Ganeshwar, where more than 5000 of copper artifacts were recovered from third millennium BC levels (Hooja and Kumar 1997: 328). From here and from other Ganeshwar-Jodhpura culture settlements along the Khetri copper belt, Harappan consumers would have been living only from 100 to 150 km away in the southern Haryana.

In 1985, Hegde and Ericson conducted Pb isotope assays of single chalcopyrite samples from five deposits in the Aravallis. The values produced for two of those samples (from Khetri and Kho Dariba) were not even remotely comparable to other Pb isotope determinations made for massive sulfide deposits in the Aravallis or, for that matter, to any typical Pb-Pb isochron (Dickin 2004: 149-151). Until the accuracy of those particular cases can be confirmed they will not be used. Hegde and Ericson's published values from the other three samples do, however, appear to be accurate. The datapoint for a chalcopyrite sample from the Ambaji deposit (discussed below) is consistent with both the previously published TIMS data (Deb *et al.* 1989) and the analyses that I have made on samples from that occurrence. Details on the two remaining assayed deposits (Kankaria and Piplawas) are scarce but they are reported to be located in the central portion of the Aravalli Range.

The Ambaji polymetallic (Pb-Zn-Cu) sulfide deposit is located in the southernmost portion of the Aravalli Range. Old pits and slag fields extending for several kilometers can be found in this area (recall Figure 12.15), which lay only 150 km to the northeast of the Harappan site of Nagwada. Literary evidence and ¹⁴C dates from beams in deep (480 m) disused mine shafts suggest that ore has been extracted here since at least the first century BC (Nandan et al. 1981: 58). Previous isotopic characterizations of this deposit were made using galena samples (Deb et al. 1989). Although the isotope values were expected more or less identical, new assays of ten chalcopyrite samples collected from the Gujarat Mineral Development Corporation's open pit mine at Ambaji were made for this database.

The Himalayas

Geologists conducting fieldwork in areas of the Himalayas ranging from Kashmir to Uttaranchal have noted hundreds of old mine shafts, open pits and/or slag heaps related to the extraction and production of copper (Dass *et al.* 1964; Middlemiss 1929; Nandan *et al.* 1981; Sharma 2002). Dr. D.P. Agrawal correctly observed (1999) that this broad region has largely been overlooked in past studies of ancient metallurgy and he singled out the extensive polymetallic sulphide deposit at Askot in eastern Uttaranchal (previously discussed in the relation to the lead ore database) as an important potential source for ancient consumers of copper in the northern part of the Subcontinent. The seven assays from this location in the copper database were made on samples provided by Dr. Rajesh Sharma of the Wadia Institute of Himalayan Geology, Dehra Dun.

Chalcopyrite (reported as pyrite – Director - Punjab Haryana and Himachal Pradesh Circle -Geological Survey of India 1971: 188) occurring in a granite pegmatite was collected near Chargaon (Figure 12.47) in the Kinnaur District, Himachal Pradesh (N 31° 30' 55", E 78° 07' 10"). Although this mineral appeared to be a minor accessory ore in what is reported as an old lead-silver mine (neither of those metals were located, however), five assays were included in the database because they are the only other copper minerals from a Himalayan deposit, besides those from Askot, that I currently had in my possession. There are <u>many</u> other potential copper sources across this region remaining to be characterized.

Sources west of the Indus Valley

Porphyry copper deposits of broadly similar geologic ages are found across the western Balochistan - eastern Afghanistan regions (Afghanistan Geological Survey 2006; Chmyriov et al. 1973; ESCAP 1995; Ludington et al. 2007; Sillitoe 1978). One of the largest occurs in Cretaceous to Oligocene era island arc volcanic rocks in Balochistan's Chagai district (Ahmad 1975: 40-46; Shams 1995b). The extensive slag fields found in that region (Vredenburg 1901: 292) and directly to the north in the Gardan Reg area of southwestern Afghanistan (Dales 1971; Dales and Flam 1969) bear testament to the intensive exploitation of these deposits during the early to midthird millennium BC (Kenoyer and Miller 1999). Seven ore samples from the Saindak occurrence (Wolfe 1974) were provided by Abdul Razique and Razzaq Abdul-Manan of Tethyan Copper Ltd.

Copper occurrences are also found in the ophiolite sequences that were emplaced during the Cretaceous to Lower Eocene eras along the western



Figure 12.47 View of mine and detail of copper oxidation at Chargaon, Kinnaur District, Himachal Pradesh.



Figure 12.48 Shin Kai copper prospect North Waziristan and detail of the copper oxidation/mineralization found there.



Figure 12.49 Copper deposits and towns (Harappan and modern) in relation to the Las Bela ophiolite of southern Balochistan.

margins (the "Indus suture zone") of the Indus Basin at various points from the NWFP to Balochistan (Asrarullah *et al.* 1979). Several prospects are located within the Waziristan ophiolite (Badshah 1985), around 60 km up the Tochi River from where Neolithic and Early Harappan settlements of the Bannu Basin lie (Khan *et al.* 1988). In June of 2001, chalcopyrite and malachite was collected for this study from the Shin Kai prospect (Figure 12.48) near Mohammad Khel (Badshah *et al.* 1997) (\approx N 32° 55', E 69° 52'). Several minor deposits of the same nature are also found in the Zhob ophiolite (Heron and Crookshank 1954: 58-59) some 150 km to the southsouthwest.

One potential source of copper from which samples have not yet been acquired for this study but is still very much worth mentioning here is the Las Bela ophiolite of southern Balochistan. "Massive copper sulphide prospects" are found between 15 to 30 km northeast of Uthal town at Saap and Ann Dhoros (Ahsan and Quraishi 1997: 44-45). Although these are said to have "no economic value" (ibid.) by today's standards, they are quite likely the very same places "in the hills between Liari and Bela" where 19th and early 20th reports indicated copper could be found in "large quantities" (Gazetteer of Las Bela 1907: 118). Del'Hoste (1844) related a colorful account by two Hindu traders who said that they had easily smelted a good quantity of the metal from this source before they were run out of the area under threats from the local tribal chief of being "burned alive" if they returned.

Although they are trifling in relation to copper deposits in most of the other source areas discussed in this section, the Las Bela ophiolite occurrences are important because there were permanent Harappan settlements located in the vicinity (Figure 12.49). Bakkar Buthi lies just 20 km to northeast across the Mor Range and Balakot is situated around 50 km due south. All of the other potential sources are in regions external to the "core area" of Indus Civilization sites. Although it is unfortunate that I am not able to include samples from these deposits in the copper database, it still may be possible to examine them by proxy. Although the lead deposits in the Kanrach Valley are hosted in Jurassic sedimentary rocks, the mineralization is thought to be associated with the Las Bela ophiolite zone (Zaigham and Mallick 2000: 487). The isotopic character of the copper deposits might, therefore, be similar to those of the Kanrach Valley galena samples.

Iran

The copper deposits of Iran are numerous, widespread and have evidently been exploited since antiquity (Bazin and Hübner 1969: 4-6; Berthoud *et al.* 1976; Wertime 1968). For Indus Civilization consumers, however, they would have been rather remote – the nearest are more than 1,100 km west of Harappa as the crow flies. Hauptmann and others recently (2003) conducted a Pb isotope analysis on samples from two copper deposits in eastern Iran (Chechel Kureh and Qaleh Zari) and set of copper ores, slags and artifacts from Shahr-i-Sokhta. The isotopic values for the archaeological ores (which have been included in this database along with those for the two geologic deposits) most closely matched those of the samples from the Qaleh Zari source, 300 km northwest of the site.

Oman

The exploitation and production of copper by the ancient peoples of the eastern Arabian Peninsula region (Figure 12.50) has been the subject of much study (Berthoud and Cleuziou 1983; Craddock *et al.* 2003; Prange 1999; Weeks 2004; Weisgerber 1984). Some feel that metal from deposits in Oman made its way to Harappan consumers (Kenoyer and Miller 1999) while others are doubtful that the region was an important source (Agrawala 2000; Chakrabarti 1998). The existence of trade links between the Indus Valley



Figure 12.50 Isotopically assayed massive sulphide deposits and sites having Harappan materials in Oman.

and this region are not in doubt, however. Harappan artifacts have been found at coastal settlements like Ras al Jinz (Cleuziou and Tosi 1994) and at inland oasis sites like Hili (Cleuziou and Vogt 1985).

Most copper in Oman occurs in massive sulphides deposits within ophiolites that are located either on the mainland of the eastern Arabian Peninsula (the Semail ophiolite) or on Masirah Island, just off of the country's southeastern coast (Weeks 2004: 12-14). Non-ophiolitic deposits exist but are minor in nature (ibid.). Pb isotope analyses of massive sulphide ores (usually chalcopyrite or pyrite) from eleven deposits in this region have been conducted by Chen and Pallister (1981) and Calvez and Lescuyer (1991). The assayed deposits include one non-ophiolitic source (al Ajal) and ten occurrences within the Semail ophiolite (location names are in the second column of Appendix 12.8 and on Figure 12.50).

Analysis and results

In addition to the seven copper ore fragments from Harappa, Pb isotope assays were made for 49 ores and/or slags from deposits in each of the potential source areas discussed above. Ore samples (archaeological and geologic) were brought into solution by dissolving 0.02 grams of fresh material from them in ultra-pure nitric acid. Slag samples (15 each from Singhana and the smelting area near Ganeshwar) were crushed into a coarse powder and then examined under low magnification for any minute copper prills that could be used for analysis. When no metal could be found or the sample weight could not be reached, the siliceous part of the slag was used. Many examples had patches with a distinct greenish cast, which were preferred for analysis because the coloring was presumably due to copper content. Although the sample weight for some the slags was doubled to 0.04 grams in an effort to get more lead into solution, in exactly half of those analyzed there was still too little of it for the ICP-MS to make accurate measurements. In the end, Pb

isotope data was obtained for 11 of the 15 Ganeshwar slags but only for four of the slags from Singhana.

The ores and slags analyzed at the LARCH, together with the previously published Pb isotope values, provide 85 datapoints representing copper deposits in the five potential source areas being considered here. When their ratios are placed on a bivariate plot (²⁰⁸Pb/²⁰⁷Pb to ²⁰⁷Pb/²⁰⁶Pb) along with those of the seven archaeological copper ores from Harappa, they spread widely, clustering in three areas (Figure 12.51 A). The chalcopyrite samples from Chargaon mine in Himachal Pradesh cluster in the upper left portion of the plot while those from the polymetallic sulphide deposit at Askot, Uttaranchal fall at the other extreme in the bottom right corner. Plotting along with the latter group are the datapoints for the assays that Hegde and Ericson (1985) conducted on samples from the Kankaria and Piplawas copper deposits of central Rajasthan. All of the remaining source samples, along with the ores from Harappa, fall in a cluster of largely overlapping groups in the center of the plot. The isotopic characteristics of the artifacts are clearly much different from either of the two Himalayan sources or the samples from central Rajasthan. This does not mean, however, that the Harappan ore fragments could not have come from other copper occurrences in those regions - just not these particular deposits or, most probably, not from any other deposits that are geologically related to them.

For Figure 12.51 B, the area of the main cluster in which the Harappan artifacts fall has been enlarged. Starting from the bottom left of the plot; the datapoints for the Ambaji mine in northern Gujarat cluster in a fairly well-defined group and are consistent with the single assay that Hegde and Ericson (1985) conducted for that deposit (noted on the plot with "H&E"). Although two Singhana slag samples representing the Khetri copper belt of northern Rajasthan fall nearby, most from that zone (including the Ganeshwar slags) cluster higher to the



left, in the middle of the plot. From that point begins the area where most of the datapoints for the various Oman sulphide assays fall. Three of the Ganeshwar slags plot there as well. The Oman assays terminate in a tight group (it is difficult to see on the plot but there are nine data points there) exactly at the point where the linear clusters of the Shin Kai (Waziristan) and Saindak (Chagai) datapoints begin, overlapping with several of them. The latter two groups extend from there to the point where the Iranian ore samples (geologic and archaeological) begin, with two of the Saindak datapoints overlapping with the Shahr-i-Sokhta artifacts and Qaleh Zari deposit samples.

The datapoints for five of the seven copper ores from Harappa fall upon (or directly adjacent to) the small area where the Oman, Chagai and Waziristan samples overlap that. This is most definitely a situation where having high precision isotope ratios and useable ²⁰⁴Pb values might be of great benefit in helping to differentiate sources and resolve the provenience of artifacts. Or it might not. The Waziristan and Semail ophiolites both formed in the mid-to-late Cretaceous era (Gnos et al. 1998; Weeks 2004: 9) as did some of the older intrusive volcanic rocks that host the Saindak deposit (Shams 1995b: 243-244). Therefore, these massive sulphide occurrences (and others, like the occurrences in the Zhob and Las Bela ophiolites, of roughly equivalent age) and porphyry copper deposits may ultimately be indistinguishable from one another using Pb isotope data alone. Of the remaining two Harappan ores; one plots with two of the Singhana slags (but within the range of error to the Oman cluster) and another falls parallel to but apart from where some of the Oman samples and Ganeshwar slags group. The former could be from the Khetri copper belt but I would feel more confident in such an assessment if 1) the samples representing that source were ores rather than slags and 2) the analytical precision were better than it is.

It is possible to state that the Harappan ores probably do not come from those deposits in the database representing eastern Iran, southern Rajasthan or the Himalayas. Five of them could be from sources in Oman or from geologically similar deposits located in regions to the west of the Indus Valley like Waziristan or western Balochistan. Of the remaining two, the provenience of one is unclear (perhaps it comes from Western sources also) while the other *may* have come from the Khetri copper belt of northern Rajasthan.

A BRIEF NOTE ON FURTHER AND ONGOING studies of Harappan copper

Recently, twelve copper alloy artifacts from Harappa were assayed using TIMS and compared to the copper database presented in this section (Hoffman and Miller 2009). The results largely mirrored those for the seven copper ore fragments from Harappa. That is to say, the Pb isotope characteristics of the artifacts suggested that all or most were made from metal derived from sources to the west of the Indus region or Oman rather than those in the Rajasthan.

Pb isotope studies of copper ores and alloy objects from the Indus city of Dholavira are just beginning. However, the initial data indicate that both metal and raw ore from the Ambaji deposits of northern Gujarat were being acquired by site residents. Some copper from a source either west of the Indus region or in Oman also seems to have been used at the city. However, if the trend of the preliminary results continues then the primary source of copper for residents of Dholavira may well have been to the east at Ambaji.

COMPARISON OF THE LEAD AND COPPER DATASETS

Many of the metal ore sources featured in this chapter are dominated by either lead mineralization or copper mineralization – e.g., the Kanrach Valley



Figure 12.52 [A] Pb isotope data for copper ores and artifacts superimposed on the "fields" of the main body of lead deposits. [B] Select copper ore data & lead "fields" plotted with copper and select lead artifacts.

(lead) and Khetri (copper) deposits. Other sources, like Ambaji and Askot, are rich in both metals. Rather than trying to evaluate all metal artifacts in relation to sources with very different and complex geologies, a decision was made early on in this study to treat lead and copper separately, the idea being, as it were, to compare apples-to-apples and orangesto-oranges. So, two databases were assembled that contained Pb isotope assays made one just one or the other type of material rather than including all analyses made for a metallogenic zone or deposit. For instance, Calvez and Lescuyer (1991) produced Pb isotope data for galenas, copper-bearing massive sulphides, gossans, pelagic sediments and volcanic rocks from locations in northern Oman. The galena assay values from that study went into the lead ore database and the sulphides into the copper database - the rest were excluded. However, now that the main comparative studies are complete, it may prove informative to briefly evaluate the databases in relation to one another and to compare certain artifacts to both of the databases combined.

In Figure 12.52 A, I have superimposed the Pb isotope data for the main cluster copper sources and artifacts (Figure 12.51 B) directly on the bivariate plot of the main body of lead ore fields. All of the copper data falls in the upper left quarter of the plot. This area is enlarged for Figure 12.52 B, where I have also added the data points and dashed ellipses for those lead artifacts that clustered in the "ambiguous" area of the lead deposits database.

The first thing that is evident from these plots is the way in which the Pb isotope assays of copper and lead samples from the same metallogenic zones, not at all unexpectedly, tend to correlate well with one another. The copper samples from the Saindak deposit (white circles with black borders) in the western Chagai District of Balochistan plot either on or directly adjacent to the isotope field (dark green) defined by galena fragments from related porphyritic deposits (Rekodiq and Koh-i-Sultan) located nearby. Copper and lead samples from the Ambaji mine similarly correlate with one another. In future versions of these databases, assays of both copper and lead minerals from these sources and others like them can probably be safely used together. It remains to be determined whether or not the same will be true for lead and copper deposits found within a common metalliferous zone or just located in the same regions. However, because the two databases are far from being complete, it is worth the effort to briefly consider where certain lead and copper artifacts plot in relation to geologic samples in the opposite databases (i.e. lead-to-copper and copper-to-lead). This might, at least provisionally, help to fill some of the existing isotopic "gaps" or "ambiguous areas."

Note that on Figure 12.52 B the five copper ores from Harappa that cluster closest together, all plot within the isotope field for the southern Balochistan lead deposits (and a sixth falls adjacent to it). Most of the galena samples defining that field are from occurrences in the Kanrach Valley, which (as discussed above) may be geologically (and isotopically) related to the nearby Las Bela ophiolite and the copper-bearing sulphide deposits within it. Even if those copper and lead deposits are shown to be isotopically analogous it would not rule out the other possible sources (Oman, Waziristan, Chagai) for the archaeological ores. It would, however, establish the southern Balochistan region as an isotopically analogous source area in which both copper deposits and Indus Civilization settlements can be found.

Consider also the various lead artifacts from Harappa and silver artifacts from Mohenjo-daro (noted on Figure 12.52 B as white circles with red borders) that cluster in the "ambiguous" area on the bivariate plot of lead sources. Many of the slags representing the Khetri copper belt also plot in that area. Although this is an interesting association, it must be examined realistically. Lead mineralization is exceedingly rare in this part of northern Rajasthan. One of the few occurrences that have been reported - trace galena in a deep coring at Saladipura (Deb *et al.* 1989), is isotopically very unlike the slags and the artifacts. Silver is extracted today from argentiferous copper ores in the Khetri belt (Rao *et al.* 1997). However, there is no evidence that it was exploited there during earlier eras, nor have any silver artifacts reported been reported from Ganeshwar-Jodhpura sites in the region. Still, although the chance that the metal for these lead and silver artifacts actually came from northern Rajasthan is very slim, it is important to keep an open mind to the possibility.

A final point to note is how, collectively, copper samples from sources in Balochistan, Oman and Iran form a linear swath of datapoints that plot across the isotope fields defined by lead samples from those same regions (and southern Afghanistan too if the Mundigak ores are used as proxy source samples). This illustrates the point I tried to make at the conclusion of the section on lead and silver - as more samples from more deposits across these regions are incorporated the database, there is almost certainly to be a significant degree of isotopic overlap between them. Perhaps with high precision TIMS measurements the sources in this part of the plot can be differentiated from one another but that is by no means certain. For metal artifacts from Indus Valley sites having Pb isotope characteristics that cause them to plot in this area it may, ultimately, only be possible to state that they probably came from sources to the west of the Indus Valley or from Oman.

CHAPTER CONCLUSION

The Pb isotope measurements produced using the ICP-MS at the LARCH are, admittedly, not as accurate as those obtainable using TIMS or MC-ICP-MS. However, they are accurate enough to differentiate lead and copper ores sources as isotopically diverse as those found in and around the Greater Indus region. Moreover, because of the ease of sample preparation and non-destructive nature of the EDTA sampling technique, many artifacts were assayed that would otherwise be unavailable for analysis. In addition, the comparatively low cost of analysis means that large numbers of artifacts and, importantly, comparative geologic samples could be assayed.

Much work remains to be done to create comprehensive Pb isotope databases with which to evaluate the potential lead and copper geologic provenience of artifacts from Harappa and other archaeological sites in the region. With regard to sources that may have been utilized by Indus Civilization peoples, there are many deposits in the study regions that remain uncharacterized. There is currently no isotopic data for any metalliferous ore bodies in Afghanistan. The need to assay the lead deposits of Haraza, NWFP and copper deposits in Jammu and Kashmir is particularly acute as I have provided evidence in this chapter and elsewhere in this book that Harappans were probably acquiring several rock and mineral varieties from these regions to the north of the site. One area, which I have not previously discussed in this chapter, where both lead and copper mineralization have been reported (Heron and Crookshank 1954 95-96; Siddiqui and Shams 1988: 3), is eastern portion of the Salt Range, just 230 km north of Harappa. This is, in fact, the source area *nearest* to the site for either of those metals! The copper mineralization is said to be trifling but the galena occurrences were apparently popular sources of surma for the local population (Punjab Government 1907: 204; Wynne 1878: 300-301). Obtaining and analyzing samples from this region is of the highest priority and their absence in the databases is another reason why the geologic provenience determinations made in this chapter should always be considered provisional.

In addition to expanding the databases to incorporate new sources, it will be equally important to produce many more assays on the deposits already in it. Currently, the average number of analyses per source is around five. It is generally held that at least 20 analyses of a lead ore field need to be conducted in order to adequately define its isotopic boundaries (Pollard and Heron 1996: 328). Still others have argued that having a bare minimum of 40 analyses is more realistic, especially when the isotope data for an ore field are not normally distributed (Baxter et al. 2000). By those standards some sources in this database, such as the Rajpura-Daribia/Rampura-Agucha lead ore field, which has 31 data points, are fairly to very well characterized. Nearly all other sources in it are dramatically under-characterized, with several even being represented by only a single data point. Even at this early stage, however, a great deal new has been learned with regard to the lead, silver and copper acquisition networks in which the residents of Harappa, as well as some of their predecessors and contemporaries at sites in the Greater Indus region and beyond, were involved.

We now know that residents of Harappa were acquiring raw lead ore (galena) from the Jammu and Kashmir region at the time of the site's foundation (Ravi Phase) and were still doing so during the latter part of the urban phase (periods 3B and 3C). They apparently used some of the lead from this source to make surma. Residue in a surma bottle from Mohenjo-daro suggests residents of that site used lead from a different (unknown) source to make this cosmetic. During Period 3C, Harappans also appear to have been acquiring raw lead from at least two (possibly more) other regions. Cerussiteanglesite and massicot ores from sources in southern Balochistan (the deposits near Sohr Damb/Nal in the Khuzdar district are the strongest candidates) were probably used as mineral pigments or cosmetics. Some of the lead used by Indus peoples at Mohenjodaro, Dholavira, Rakhigarhi, and Gola Dhoro, as well as the early residents of Mehrgarh, also came from occurrences in that region. Harappans acquired lead from at least one or more additional sources, the

locations of which are not known at this time. These unknown sources do not appear to be the same ones that residents of the Helmand Civilization sites of Mundigak and Shahr-i-Sokhta probably exploited in southern Afghanistan and east Iran. Harappans melted lead from all three regions (Jammu and Kashmir, southern Balochistan and the unknown source or sources) to fashion various kinds of items.

It appears that the silver used by Indus Civilization peoples was being extracted from at least three, possibly more, different argentiferous lead ore sources. The Early Harappan settlers of Nagwada may have brought this metal with them from deposits in southern Balochistan (probably around Khuzdar) as they moved into northern Gujarat from regions to the northwest. The silversmiths who created the artifacts in the Allahdino hoard seem to have used some metal from deposits in that region as well. They also appear to have used metal from the same source(s) as those who made the silver artifacts analyzed from Mohenjodaro. The location of that source is presently unknown, however. The silver/lead used to make a ring from the site of Gola Dhoro may have originated in eastern Arabia. Further studies of potential sources in that region need to be conducted in order to confirm this, however.

Finally, we now see that residents of Harappa did not acquire a great deal of raw copper ore (malachite or chalcocite). Only seven examples have been recovered at the site to date. Most of these (5 or 6 of the 7) were probably derived from sources to the west of the Indus Valley or, perhaps, Oman. One ore fragment may have come from deposits in the northern part of Rajasthan's Aravalli Range. Recent research (Hoffman and Miller 2009) suggests that finished copper alloy objects, which constitute a major part of Harappa's rock and mineral assemblage, might follow a pattern similar to that of the ores. This situation might be very different at the southernmost Indus city of Dholavira, however. Preliminary results indicate that site residents were acquiring much of their copper (as well as some lead) from deposits occurring in northern Gujarat near Ambaji.

In the next chapter, I take the provenience determinations made for all of the lead artifacts and

copper ores from Harappa and, together with the determinations made for each of the other materials examined in the preceding seven chapters, address the three lines of inquiry outlined in Chapter 1.