Geologic provenience analysis of agate and carnelian beads using laser ablation-inductively coupled plasma-mass spectrometry (LA-ICP-MS): A case study from Iron Age Cambodia and Thailand

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Abstract:

Agate and carnelian beads, imported from South Asia, were widely exchanged in Southeast Asia during the Iron Age period (500 BCE – 500 CE). Recent studies have identified changes in bead types and manufacturing methods over time, as well as evidence for possible local production. In order to understand the broader implications of these developments, geochemical analysis using laser ablation-inductively coupled plasma-mass spectrometry (LA-ICP-MS) was undertaken on 73 beads from 10 Iron Age sites in Cambodia and Thailand and 64 geologic samples from four sites in India, Iran, and Thailand. The results show that many of the beads were produced from raw material derived from the Deccan Traps, India and that there is not yet strong evidence for bead production using a Southeast Asian source. Secondly, we find that there is not yet clear evidence for a change in the different geologic sources used to produce beads over time. This study adds to the growing body of literature highlighting the utility of LA-ICP-MS in differentiating and assigning provenience to agate/carnelian and other silicates.

Keywords

Agate/Carnelian
LA-ICP-MS
Provenience analysis
South Asia
Southeast Asia
Beads

1. Introduction

Contact with South Asia was deeply influential to the socio-political development of many early polities in Southeast Asia, beginning around the fourth or fifth centuries BCE (Higham, 2014). One of the earliest indicators of this contact is stone and glass beads, believed to have been produced in South Asia, and widely traded throughout Southeast Asia. In the past, most scholars assumed that all beads of this kind were imports from India, and were proxies for South Asian influence in the region (e.g. Francis, 1996; Glover, 1990). More recently, the discovery of unfinished beads, unusual styles of beads, and possible manufacturing debris at Southeast Asian sites has led many to argue that at least some of these ornaments were manufactured locally, perhaps using regionally available raw materials (Bellina, 2014; Francis, 2002, Theunissen et al., 2000).

At the same time, recent archaeological work has highlighted how exchange with South Asia transformed over the Iron Age, with intensifying interaction during the early first millennium CE (Bellina and Glover, 2004). Studies of glass beads in Southeast Asia have identified a regional shift in the recipes used to produce glass artifacts during the first few centuries CE (Carter, 2010; Lankton and Dussubieux, 2013). Examinations of stone beads, and especially agate and carnelian beads, have also identified morphological and manufacturing changes over time (Bellina, 2014; Carter, 2013, Carter, 2015). Generally, these shifts in stone and glass bead types reflect the circulation of increasing quantities of mass-produced goods (Carter, 2015). Identifying the geologic sources used to produce beads would aid in understanding this shift, the interaction networks at play during this period, and their effects on emerging socio-political complexity.

In the current study we use laser ablation-inductively coupled plasma-mass spectrometry (LA-ICP-MS) to characterize 73 agate/carnelian artifacts from 10 sites in Cambodia and Thailand as well as 64 samples from geologic sources in three regions (India, Iran, and Thailand). In doing so, we aim to address the following questions:

- 1) Can we identify potential raw material sources used to produce the agate and carnelian beads using elemental analysis?
- 2) Is there evidence that a central Thai source was used to produce beads, as has been hypothesized elsewhere in Southeast Asia by Theunissen et al. (2000)?
- 3) Can we detect changes in the raw material source used to produce agate/carnelian beads over time? Are these related to morphological and manufacturing differences identified in earlier and later period beads?

Answers to these questions have implications for understanding resource exploitation networks, the organization of stone bead production, the involvement of elites in the exchange and production of beads, and interaction networks within Southeast Asia and between South and Southeast Asia.

2. Background

2.1 Agate and carnelian

Agate and carnelian are generally considered to be varieties of chalcedony, which like jasper, flint and chert, is a type of microcrystalline quartz (Bauer, 1969; Butler, 1995; Pabian et al., 2006). Agate contains mostly silica (>97 %) and frequently forms in igneous rock, but can also appear in sedimentary rocks (Leudtke, 1992). Chalcedony can come in a variety of colors, however, agate frequently refers to a translucent type of banded chalcedony, with brown, black, and white colors, with carnelian referring to stones that range from yellow-orange to red, red-

orange, or brown-red (see Butler, 1995). The stone colors of both agate and carnelian are frequently enhanced by bead makers through heat-treatment and other techniques (Francis, 2002; Kenoyer, 2003). Mineral and trace element impurities aid in differentiating geologic sources and assigning provenience to artifacts (Roll et al., 2005). Stones collected for beadmaking are usually those that have already eroded from their primary context host rocks and ended up in a secondary context, such as a spread of loose agate nodules in a riverbed (Law, 2011; see also Francis 1982 for a discussion of mining of agate nodules from the Narmada River floodplain).

A principle objective of provenience analysis is to measure geochemical variability within a raw material source, with the hope that the variability will be greater between sources than within a single source (Weigand et al., 1977). However, because secondary context agate deposits may potentially contain materials that formed across extremely wide areas and in very different geologic episodes and/or environments, samples from such sources can be expected to have a high degree of geochemical variability (Luedtke, 1992: 51). Additionally, as many agate/carnelian raw material sources are frequently collected from secondary locations, compositional analysis will not necessarily inform us about the place where these materials were collected, but instead about the place of origin for the agate/carnelian.

2.2 Previous studies of agate and carnelian beads and artifacts

LA-ICP-MS has been used extensively by many researchers wishing to measure the quantity of trace elements within chalcedony and chert (e.g. Baldwin et al., 2011; Götze et al., 2009; Möckel et al., 2009; and Schmidt et al. 2012). Recent archaeological studies have also shown the effectiveness of using LA-ICP-MS in analyzing quartzite (Pitblado et al., 2013) and chert (Andreeva et al., 2014; Roll et al., 2005; Speer, 2014a, 2014b) artifacts. A study by Insoll and others (2004; see also Fraser et al., 2005) used UV-LA-ICP-MS to examine carnelian trade between India and Africa, and identified a possible geologic stone source for some artifacts from western Africa. The study included 13 artifacts from sites in western Africa, which were compared to 13 geologic samples from the secondary Ratanpur deposit in Gujarat, India. Although the authors found some overlap between the two groups, there were also samples that were compositionally distinct from the Ratanpur source.

Other researchers have analyzed agate and carnelian artifacts using other techniques. Murillo-Barroso and others (2015) recently characterized three quartz objects, including a carnelian bead, using X-Ray Diffraction (XRD) and SEM-EDS analysis, although they did not compare their results to any geologic sources. Theunissen and others (2000) used the non-destructive Proton Induced X-Ray Emission/Proton Induced Gamma Emission (PIXE/PIGME) technique to explore the possibility of local production of agate and carnelian beads in Southeast Asia. Their study analyzed beads from the Thai Iron Age sites of Noen U-Loke (n=9) and Ban Don Ta Phet (n=9) and compared this with archaeological material found at different bead production sites located in India and Sri Lanka, as well as two samples from the Ban Khao Mogul agate source in Lopburi province, central Thailand. They found that there was a compositional similarity between the two geologic source samples from Ban Khao Mogul and the carnelian and agate artifacts at Ban Don Ta Phet and Noen U-Loke, raising the possibility of local manufacture of beads using this raw material source (Theunissen et al. 2000:98).

This study rightfully questions the Indian origin for all agate and carnelian beads in Southeast Asia, however it was limited in that nearly all of the objects analyzed in their study were artifacts rather than geologic source samples. Additionally, only two samples from the Ban Khao Mogul source were analyzed. Although there is not a set number of samples required to

characterize a source, we argue that two samples cannot capture the geochemical variability that exists in agate/carnelian deposits. The current study expands on this dataset by including a larger number of samples from the Ban Khao Mogul source.

Another recent study by Law and others (2013) used instrumental neutron activation analysis (INAA) to analyze a large set of samples from four sources, Ratanpur and Mardak Bet in Gujarat, India, the Ban Khao Mogul source in Thailand, and Shahr-i-Sokhta, Iran, as well as a set of agate/carnelian beads from Afghanistan. This study proved to be highly effective at characterizing agate/carnelian sources and convincingly assigning artifacts to sources (Law et al. 2013; Law 2011). However, INAA is a destructive technique in most cases, and thus not ideal for analyzing complete beads on a scale large enough to address questions regarding regional exchange patterns and the potential for local craft production amongst sites in Southeast Asia. In the current study we analyze materials from these same sources using the less destructive technique of LA-ICP-MS (Speakman and Neff, 2005) and find as good if not better statistical separation between the sources as in the previous INAA study.

Recently another study has analyzed a variety of silica-based samples using multiple techniques, including LA-ICP-MS, scanning electron microscopy (SEM), micro-Raman spectroscopy, and X-ray diffraction, to measure major, minor, and trace elements (Gliozzo et al., 2014). This study was able to characterize a variety of silica-based artifacts, which were then compared to a database of published geologic source sample data. Although an exact source for the artifacts was not identified, the authors felt confident in ruling out a possible Indian source for their archaeological materials (Gliozzo et al. 2014). The authors compared their results to a dataset of published material, although they observed several problems in the quality of published data, including the inability to rely on major elements due to the multiple techniques used, the reporting of average concentrations over single measurements, and the unreliability of results for specific elements which made multivariate statistics difficult (Gliozzo et al., 2014). Furthermore, they specifically report the lack of comparative data for carnelian. In the current study, we greatly expand on the agate/carnelian database and were able to accurately measure major, minor, and trace elements using LA-ICP-MS alone.

3. Materials

3.1 The geologic sources

Geologic samples came from four sources in three regions (Figure 1). Samples from these same sources had already been characterized previously using instrumental neutron activation analysis (INAA) at the University of Missouri Research Reactor (MURR) (Law, 2011; Law et al., 2013). In this earlier study, good to excellent statistical separation was achieved between the four individual agate/carnelian sources comprising the geologic dataset. We expected similar or better results using the minimally destructive and less costly technique of LA-ICP-MS (Speakman and Neff, 2005).

Two of the four geologic sources characterized in this study are located in the Indian state of Gujarat (Figure 1). Agate/carnelian sources in this region have been exploited for 4000 years or more (Allchin 1979; Kenoyer et al. 1994; Roux 2000). The Gujarati city of Khambhat (Cambay) has been a major bead production center since at least the 16th century AD (Arkell 1936). We suspected that this area was a major supplier of beads to Southeast Asia during the Iron Age period as well. The agate and carnelian sources of this region are part of the large igneous formation known as the Deccan Traps (Figure 1) (Mahoney 1988). The Deccan Traps are not a homogeneous formation, but rather consist of a series of flood basalt layers. Agates and

other minerals were formed in vesicles within these basalts.

Figure 1. Map showing the location of the Deccan Traps, Gujarat province, and the agate/carnelian sources in India and Iran analyzed in this study.

Samples derived from the Deccan Traps were collected by Randall Law from deposits at Ratanpur (n=15) and Mardak Bet (n=15) (Figure 1). The Ratanpur source (henceforth RTP) is located within a Miocene conglomerate known as the Babaguru Formation. This secondary context agate/carnelian deposit potentially contains stone from an extensive geographic area brought to the site through fluvial processes (Gadekar 1977; Law 2011). The Mardak Bet (henceforth MB) source is located on a small island in the seasonally flooded salt flats known as the Little Rann of Kutch. Samples were collected from two beds of loose agates that had eroded from a decomposed outcrop of basalt. A large number of flakes with a heavy patina were observed on the surface, suggesting exploitation of these raw materials into antiquity (Law, 2011:277). Samples from both beds were treated as a single source due to their geographic proximity to one another.

Also included in the geologic dataset are nodule fragments recovered during Italian excavations at the proto-historic site of Shahr-i-Sokhta, Iran (henceforth SIS) (Figure 1). These samples were treated as a proxy source located in eastern Iran, as agate/carnelian pebbles “may be collected along the dried out beds and ancient branches” of the Helmand River delta (Tosi, 1969: 374). Even though it was not expected that raw material from SIS were used to make

beads found in Southeast Asia, the samples from this source were analyzed in order to verify that LA-ICP-MS could differentiate this source from others in the dataset.

Agate/carnelian from Southeast Asia is represented by the deposit at Ban Khao Mogul (henceforth BKM), in Lopburi Province, central Thailand (Figure 2). Several scholars have discussed this site as a possible raw material source location for beadmakers in central or northeast Thailand (Bellina 2007; Glover 1990; Theunissen et al. 2000). The deposit consists of small nodules of agate and other microcrystalline materials eroding from a small limestone outcrop. As recently as 15 years ago, several villagers were quarrying material from this source to produce polished stones and cabochons (Nigel Chang, personal communication, 2007). However, the age of the quarry is not known and during Carter's visit in 2007 only one villager was still practicing this craft.

It should be noted that this is not the only potential raw material source in Southeast Asia, as other sites with agate/carnelian have been reported elsewhere in Thailand, Vietnam, Indonesia, and Myanmar (see discussion in Carter, 2013: 233-238). Beads made from petrified wood, in which a mineral such as quartz has replaced organic remains, have been produced for hundreds of years in Myanmar (see Moore and Myint, 1993). Decorative pieces of agate are currently exported from Indonesia and agate has also reportedly been used in jewelry in Vietnam (see Carter, 2013: 233 for further discussion). However, to the best of our knowledge, none of these sources have clear evidence for having been exploited archaeologically. Several geologic sources from Southeast Asia were analyzed by Carter (2013) in her dissertation and will be discussed in future publications.

Figure 2: Map of Iron Age sites considered in this study and the Ban Khao Mogul (BKM) agate/carnelian source in central Thailand.

3.2 The agate and carnelian beads

The agate and carnelian beads (n=73) came from 10 Iron Age sites in Cambodia and Thailand (Figure 2 and Table 1) (see Carter, 2013 for further discussion). An earlier morphological study of these beads has identified a difference in the manufacturing techniques and overall bead quality between these sites. In one group of sites, agate and stone beads were generally of a higher quality, with more complex shapes and made with skilled and time-consuming manufacturing techniques (Carter, 2013, 2015). Glass beads and ornaments made from potash glass were also frequently found (Carter, 2010, 2013, 2015). Sites with these types of beads, which we will refer to as Group 1 sites, generally dated to the early Iron Age period (late centuries BCE) and included several mortuary sites in southeast Cambodia (Figure 2): Village 10.8, Prohear, and Bit Meas. A single bead was analyzed from Krek 52/62, a residential circular earthwork site located near Village 10.8. Also included in Group 1 was the bead manufacturing site of Khao Sam Kaeo in peninsular Thailand. Recent archaeological work at Khao Sam Kaeo shows strong evidence for the presence of Indian craftsmen who lived and worked at the site, producing stone and glass ornaments (Bellina, 2014; Lankton et al., 2008). Beads from this site likely came from workshop contexts and included several unfinished beads.

Khao Sam Kaeo is not the only site in Southeast Asia with evidence for bead manufacture, although it is the best studied. Other sites, including those in Myanmar, central and peninsular Thailand, Malaysia, and southern Vietnam, have been suggested as possible bead manufacturing locations. However, evaluating these claims is problematic due to extensive looting. Waste material can travel with finished beads from workshops, so the presence of unfinished beads at a site is not necessarily a strong indicator that manufacturing was undertaken in that location. Further work is needed to identify multiple stages of bead production within a single site, as seen at Khao Sam Kaeo, before scholars can be confident in identifying a bead manufacturing location (for further discussion see Carter, 2013: 157-163).

The second group of sites, which we will call Group 2, had different types of stone and glass beads. In contrast to the Group 1 sites, the Group 2 sites contained beads in simpler shapes and made using less complex and less skilled manufacturing techniques, including some techniques that indicate the mass production of beads (Carter, 2013, 2015). Instead of potash glass beads, the majority of glass beads at these sites were made from high alumina mineral soda glass (Carter, 2010, 2013, 2015). Group 2 sites generally dated to the later Iron Age (early-mid first millennium CE) and included Angkor Borei, Phnom Borei and Phum Snay in Cambodia (Figure 2). Lastly, two sites spanned both the early and late Iron Age period: Ban Non Wat in northeast Thailand and Promptin Tai in central Thailand. Both sites contained burials that date from approximately 500 BCE – 500 CE and included glass and stone beads typical of both Group 1 and Group 2 sites (Carter, 2013, 2015).

The shifting glass compositions seen at Group 1 and Group 2 sites is part of a regional change that was happening across Southeast Asia during the early first millennium CE (Lankton and Dussubieux, 2013) and appears to reflect intensifying trade and interaction with South Asia (Bellina and Glover, 2004). The presence of lower quality agate/carnelian beads also signals a change in the organization of production towards mass-produced beads, which raises several questions. Evidence from Khao Sam Kaeo suggests the local manufacture of beads during the early Iron Age, including high quality beads (Bellina 2014). Were these beads made using a local

raw material source or using raw materials imported from India? The use of a local raw material would suggest that elites at Khao Sam Kaeo were exercising control over nearly every stage of manufacture, from raw material acquisition to the production of the finished products. We also wondered if the shift towards mass production of beads during the later Iron Age period resulted in a change in the raw material source used to produce the beads. This could be due to a new workshop producing these objects and exploiting a different raw material source. Furthermore, Bellina (2007) has suggested Southeast Asian workshops may have been making lower quality beads, perhaps using a local source, while higher quality beads continued to be imported from India. Geochemical analysis is one way to begin addressing these questions.

4. Methods

4.1 LA-ICP-MS

Analyses were performed at the Elemental Analysis Facility (EAF) at the Field Museum in Chicago. The equipment used includes an Analytik Jena quadrupole ICP-MS connected to a New Wave UP213 laser unit. The parameters of the ICP-MS are optimized to ensure a stable signal with a maximum intensity over the full range of masses of the elements and to minimize oxides and double ionized species formation (XO^+/X^+ and $X^{++}/X^+ < 1$ to 2 %). For that purpose the argon flows, the RF power, the torch position, the lenses, the mirror and the detector voltages are adjusted using an auto-optimization procedure.

For better sensitivity, helium is used as a gas carrier in the laser. The choice of the parameters of the laser ablation not only will have an effect on the sensitivity of the method and the reproducibility of the measurements but also on the damage to the sample. We used the single point analysis mode and 70 % of the laser energy (0.2 mJ) with a pulse frequency of 15 Hz. As for some other potentially heterogeneous materials, such as ceramics (Dussubieux et al., 2007), agate and carnelian samples were ablated 10 times with a laser diameter of 100 microns. For comparison, for glass, a homogeneous material, only 4 ablations are performed with a laser diameter of 55 microns (Dussubieux et al., 2009). This insures that a relatively large volume of representative material is sampled. The locations for sampling were selected from the portion of the sample that was clearly visible within the chamber. However, for samples that were visibly heterogeneous, such as banded agates, locations were selected to reflect the range of variation within the sample (e.g. light and dark bands). The sample is pre-ablated for 20s to avoid surface contamination as much as possible. The average of the 10 measurements of 58 elements, corrected from the blank, was considered for the calculation of the compositions.

To improve reproducibility of measurements, the use of an internal standard is required to correct possible instrumental drifts or changes in the ablation efficiency. The element chosen as internal standard has to be present in relatively high concentration so its measurement is as accurate as possible. In order to obtain absolute concentrations for the analyzed elements, the concentration of the internal standard has to be known. The isotope Si29 was used for internal standardization. Concentrations for major elements, including silica, are calculated assuming that the sum of their concentrations in weight percent in glass is equal to 100 % (Gratuze, 1999).

Fully quantitative analyses were obtained by using external standards. To prevent matrix effects, the composition of standards has to be as close as possible to that of the samples. In the absence of standard reference materials (SRM) with a high silica content matrix doped with a large range of trace elements, several synthetic silicate glasses were selected. SRM 610 and SRM 612 are manufactured by the National Institute for Standards and Technology. They are soda-lime-silica glass doped with trace elements in the range of 500 ppm (SRM 610) and 50 ppm

(SRM 612). Certified values are available for a very limited number of elements. Concentrations from Pearce et al. (1997) were used for the other elements. Two other standards were manufactured by Corning. Glass B and D are glasses that match compositions of ancient glass (Brill, 1999, vol. 2, p. 544). The detection limits were calculated as 3 times the standard deviation obtained from the measurement of 10 blanks, ranging from less than 1 ppb to ~1 ppm for copper. In order to start with a dataset that has a normal distribution, the base-10 logarithm of the concentration of all elements in ppm were considered. Following Speer (2014a, 2014b) elements that were below the limits of detection (LOD) were replaced with a zero for the statistical analyses. Appendix A presents the compositional results for the geologic sources and agate/carnelian artifacts. Major elements are listed in weight percent and the minor and trace elements are presented in ppm.

4.2 Determining elements to use in statistical analyses

Previous work has emphasized the importance of trace elements in identifying and distinguishing between geologic sources of silicate material (e.g. Gliozzo et al. 2014; Roll et al. 2005; Speer 2014a, 2014b). However, we did not feel that all the trace elements could be relied upon due to the low concentrations of certain elements within our samples that were close or below the LOD of for the instrument. Table 2 lists the elements measured in LA-ICP-MS and number of samples that were at or below the LOD for each element. Some elements, such as Sc, Sr, and Zr were measured in all samples, while others, such as Cr and W, were at the LOD in the majority of samples. In order to best characterize and differentiate the source samples from one another we needed to rely on elements that were measured in nearly all of samples. For this reason, we used elements that were measured above the LOD in 90% or more of the 64 total geologic source samples in our statistical analyses. These 16 elements are listed in bold in Table 2.

In addition to these elements, exploratory bi-plots identified several additional elements that were determined to be diagnostic for specific sources, despite being measured above the LOD in fewer than ninety percent of the geologic samples. Potassium (K) was present in measurable quantities in all but 2 of the BKM samples and sodium (Na) was present in all but one of the SIS samples, while (U) and antimony (Sb) were fully measured in the SIS and BKM samples. These four elements were useful for distinguishing these sources from the RTP and MB sources. The concentration of titanium (Ti) also varied between the four sources and assisted in differentiating the sources from one another. These five elements were joined with those in bold in Table 2, so that the elements used for all statistical analyses were: Al, B, Ba, Ca, Ce, Fe, K, La, Mg, Na, Ni, Pb, Rb, Sb, Sc, Si, Ti, U, Y, and Zr. Table 3 lists the average concentrations and standard deviations for the elements used in the statistical analyses for each of the four geologic sources.

4.3 Statistical analysis

Principal Components Analysis (PCA) was the primary statistical method undertaken to identify compositional differences between the sources and between the sources and artifacts. However, Canonical Discriminant Analysis (CDA) was used as an additional technique as a means of maximizing the difference between sources and assigning possible provenience to artifacts. This method has been used successfully in other studies assigning provenience to agate/carnelian artifacts (Law et al., 2013) and chert artifacts (Speer, 2014a, 2014b). In CDA, linear combinations of variables called discriminant functions are generated that produce a

maximum degree of separation (discrimination) between various defined groups of cases, which in this instance are the individual sets of samples collected from different geologic sources (see Baxter 1994).

Discrimination success is evaluated by a cross-validation technique in which each case is left out of its group in turn and compared to the dataset as an ungrouped case. This cross-validation technique allows for a more accurate picture of the CDA results (Kovarovic et al., 2011). A percentage is generated based on the number of cases that were correctly assigned to the groups to which they actually belong. The next step involves adding the agate/carnelian artifacts as ungrouped cases, which are then assigned to the group whose center (or centroid) in multidimensional space they are nearest, producing a Mahalanobis distance value. The data are displayed on a bivariate plot using the first and second discriminant functions.

It is important to note that ungrouped cases do not always conclusively belong to their assigned groups. CDA assumes that all possible groups are present and will always assign an ungrouped case to a group. In reality, the group assignments could change as additional geologic sources are added to the dataset. As the current study represents only a fraction of potential stone sources, we should consider the group assignments to merely reflect predicted group membership (PGM). CDA also produces two PGM assignments, reflecting the group with the second smallest Mahalanobis value. Unknown cases can easily be misclassified due to similarities between the different defined groups (geologic sources) or because the case itself is an outlier of a group. For this reason, it is important to consider both the first and second PGMs when evaluating the CDA. Both statistical methods were performed in IBM SPSS 20.0. The casewise statistics and standardized canonical discriminant function coefficients are listed in Appendix B.

5. Results

5.1 The geologic sources

Figure 3 displays a 3-D scatterplot of the first three principal components of a PCA of the geologic sources, accounting for 69% of the variation. There is fairly good separation between the sources from three different regions. From the PCA and the average concentrations listed in Table 3, it is clear that there are several elements that distinguish the sources from one another. Generally, the BKM source has higher concentrations of several elements, including Ba, Ce, La, Rb, Y and Zr, but comparatively low levels of B. Conversely, the SIS source is notable for high concentrations of B, Pb, and U and lower levels of other elements. The Deccan Traps sources also have lower concentrations of many elements, but higher quantities of Ni and Sc than the other two sources. Figure 4 displays several biplots showing the varying concentrations of B, Sb, and Sc in the geologic sources samples. The large standard deviations seen in several of these elements also testify to the wide range of compositional variation seen within these geologic sources (Table 3).

There is considerable overlap between the two Deccan Traps sources: MB and RTP. This is perhaps unsurprising as the two sources come from the same geological formation, and Ratanpur is a secondary source. Due to the heterogeneous nature of this formation, it may be difficult to characterize the different Deccan Traps agate sources. However, the compositional differences between the three regional sources (Thailand, India, Iran) are promising for future studies that consider geologic sources from different areas.

The compositional differences between the sources are emphasized in CDA. Figure 5

shows a bi-plot of a CDA demonstrating the distinct compositional groups between the three regions, with 86% of the cross-validated cases correctly classifying. As seen in the PCA, much of the overlap is between the RTP and MB sources, with 3 RTP and 2 MB samples misclassifying with one another (Appendix B). Additionally, 2 SIS samples misclassified as belonging to the BKM source, one BKM sample misclassified with the MB source, and one MB source misclassified with the SIS source (Appendix B).

Figure 3. A 3-D scatterplot of the agate/carnelian geologic sources plotted by their first, second, and third components.

Figure 4. Bi-plots (top row, bottom row-left) and a three-dimensional scatterplot (bottom row- right) using the elements B, Sc, and Sb showing the compositional differences between the four geologic sources. Ellipses serve to demarcate the geologic sources and are not statistically significant. Element concentrations are in PPM and have been logged.

Figure 5. A bi-plot of the agate/carnelian geologic sources plotted by their first two functions.

5.2 The agate/carnelian artifacts

The agate and carnelian artifacts were added to the first PCA with the four geologic sources and Figure 6 shows the geologic sources and artifacts plotted by the three principal components, making up 55% of the total variation. From this PCA it is clear that nearly all of the artifacts are compositionally similar to and plot with or near the Deccan Traps sources. This is reaffirmed with the CDA, which assigned only one of the artifacts to the BKM source (Figure 7 and Table 1). Four additional artifacts had a second PGM to the BKM source (Table 2 and Appendix B).

The bead with primary PGM to the BKM source was from the site of Angkor Borei (AKC03036) and consisted of a rough spherical carnelian bead from a burial context. Of the 4 other beads with secondary PGM to the BKM source an additional carnelian rough spherical bead was from Angkor Borei (AKC03042), two beads were from Ban Non Wat, including a rounded square agate pendant (AKC02063) and a spherical carnelian bead (AKC02067), and a short carnelian bicone from the site of Village 10.8 (AKC0344). The beads do not exhibit any obvious characteristics which might indicate they were all produced in the same workshop, nor are they unusually distinct from the other beads in this study. One possible exception might be the bead from Village 10.8, which has a low-luster polish that is unusual for the beads in this study.

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448 **Figure 6. Top-A 3-D scatterplot of the agate/carnelian geologic sources and the**
 449 **agate/carnelian artifacts plotted by their first, second, and third components. Bottom- The**
 450 **MB and RTP sources and agate/carnelian artifacts plotted by their first three components,**
 451 **the SIS and BKM samples have been removed for clarity.**
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Figure 7. A bi-plot of the agate/carnelian geologic sources with the agate/carnelian artifacts plotted by their first two discriminant functions. The five labeled artifacts are those that have either a primary or secondary PGM assignment to the BKM source.

6. Discussion and Conclusion

We can now return to the questions posted in Section 1. First, we asked if we could identify potential raw material sources used to produce the agate and carnelian beads. The results of this preliminary study indicate that most, if not all, the agate and carnelian beads were made from raw materials from the Deccan Traps in northwest India. However, some artifacts appear to vary somewhat from the Deccan Traps sources measured, and may derive from another untested source. The inclusion of additional geologic sources will assist with addressing this issue in the future and continuing to analyze samples from the dozens if not hundreds of potential sources in

South and Southeast Asia is ongoing. Nevertheless, it is clear that the agate/carnelian beads are compositionally distinct from the SIS and BKM geologic sources and did not derive from these sources.

This brings us to our second question, regarding the possibility of the local production of beads using the BKM source. None of the agate/carnelian artifacts in this study plot with this source (Fig. 6 and 7). While a small number of artifacts had either a primary or secondary predicted group membership to the BKM source, these beads also did not plot near the BKM group, indicating a degree of geochemical difference. CDA will always assign unknown samples to one of the known groups, therefore while the beads could be outliers to the BKM source they may have more likely been made from an as of yet untested geologic source.

Despite an earlier study by Theunissen et al. (2000) who found that beads from two sites in northeast and central Thailand might have been produced using the BKM source, the beads in this study from central (Promtin Tai) and northeast (Ban Non Wat) Thailand were largely made from Indian raw materials. If Theunissen's results are correct, then the locally made beads found at the two sites in his study may not have been widely circulated.

We cannot rule out that beads may have been produced locally using imported raw materials from India. This would be consistent with evidence for the production of glass bangles and beads at the site, which were believed to have been made from imported glass (Lankton et al., 2008; Lankton and Dussubieux, 2013). All of the beads from Khao Sam Kaeo, including the unfinished beads were assigned to the Deccan Traps sources in the CDA, and plot with or near the Deccan Traps geologic sources in the PCA. Data from studies of the beads and workshop materials at Khao Sam Kaeo suggest that these craftsmen may have been working under the patronage of local Southeast Asian elites and making beads in a local style (Bellina 2014). However, results from the current study suggest that the craftsmen working at the site may have brought raw material with them for manufacture into beads in Southeast Asia.

Lastly, there is not clear evidence for changes in the raw material source used to produce agate/carnelian beads over time. Despite clear morphological differences between earlier and later period agate/carnelian beads, both groups of beads appear to derive from the Deccan Traps sources. This is complicated by the fact that the two Deccan Traps sources appear compositionally analogous to one another making it difficult to clearly see changes in the use of sources within this formation over time. We can cautiously suggest that results from CDA may point toward the use of different sources within the Deccan Traps over time. Roughly 70% of the beads from the Group 1 sites and the early Iron Age period burials at Ban Non Wat were assigned a primary PGM to the RTP source, while approximately 60% of the beads from the Group 2 sites and later burials at Ban Non Wat were assigned primary PGM to the MB source. This could be an indication of shifting raw material resources within the Deccan Traps over time, but analyses of additional sources in the Deccan Traps is needed before we can consider this in more depth. This work does reinforce the importance of this geologic formation for the production of beads for thousands of years, from the prehistoric Indus Valley civilization through the medieval period.

The results of this study reaffirm previous work (Andreeva et al., 2014; Gliozzo et al., 2014; Insoll et al., 2004; Pitblado et al., 2013; Roll et al., 2005; Speer, 2014a, Speer, 2014b), which demonstrated that LA-ICP-MS is an effective tool for characterizing and differentiating agate/carnelian and other silicate materials. Ongoing work analyzing additional geologic sources promises to elucidate resource exploitation networks and the exchange of finished products in

the ancient world. As briefly demonstrated here, this has implications for understanding the interaction networks and socio-political changes happening in many ancient communities.

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Site Name and Dates	Database ID	Agate/ Carnelian	CDA PGM 1	CDA PGM 2
Angkor Borei, Cambodia 200 BCE - 200 CE	AKC03035	Carnelian	Ratanpur	Mardak Bet
	AKC03036	Carnelian	Ban Khao Mogul	Mardak Bet
	AKC03037	Carnelian	Ratanpur	Mardak Bet
	AKC03038	Carnelian	Mardak Bet	Ratanpur
	AKC03039	Carnelian	Ratanpur	Mardak Bet
	AKC03040	Carnelian	Mardak Bet	Ratanpur
	AKC03041	Carnelian	Ratanpur	Mardak Bet
	AKC03042	Carnelian	Mardak Bet	Ban Khao Mogul
	AKC03043	Carnelian	Mardak Bet	Ratanpur
	AKC03044	Carnelian	Mardak Bet	Ratanpur
	AKC03045	Agate	Mardak Bet	Ratanpur
	AKC03046	Quartz	Mardak Bet	Ratanpur
Ban Non Wat 420 BCE - 600 CE	AKC02060	Agate	Mardak Bet	Ratanpur
	AKC02061	Carnelian	Mardak Bet	Ratanpur
	AKC02062	Carnelian	Ratanpur	Mardak Bet
	AKC02063	Agate	Mardak Bet	Ban Khao Mogul
	AKC02064	Agate	Ratanpur	Mardak Bet
	AKC02065	Agate	Mardak Bet	Mardak Bet
	AKC02066	Agate	Ratanpur	Mardak Bet
	AKC02067	Carnelian	Mardak Bet	Ban Khao Mogul
	AKC02068	Carnelian	Mardak Bet	Ratanpur
	AKC02069	Agate	Mardak Bet	Ratanpur
	AKC02070	Agate	Mardak Bet	Ratanpur
	AKC02071	Agate	Mardak Bet	Ratanpur
Bit Meas, Cambodia, Contemp. with Prohear	AKC00730	Agate	Ratanpur	Mardak Bet
	AKC00732	Carnelian	Mardak Bet	Ratanpur
Khao Sam Kaeo, Thailand 4th -2nd	AKC03500	Carnelian	Ratanpur	Mardak Bet
	AKC03501	Agate	Ratanpur	Mardak Bet
	AKC03502	Agate	Ratanpur	Mardak Bet
	AKC03503	Agate	Ratanpur	Mardak Bet

centuries BCE	AKC03504	Agate	Ratanpur	Mardak Bet
	AKC03505	Agate	Ratanpur	Mardak Bet
	AKC03506	Agate	Ratanpur	Mardak Bet
	AKC03507	Agate	Mardak Bet	Ratanpur
	AKC03508	Agate	Ratanpur	Mardak Bet
	AKC03509	Agate	Ratanpur	Mardak Bet
	AKC03510	Carnelian	Mardak Bet	Ratanpur
	AKC03511	Carnelian	Ratanpur	Mardak Bet
	AKC03512	Agate	Ratanpur	Mardak Bet
	AKC03513	Agate	Ratanpur	Mardak Bet
	AKC03514	Carnelian	Mardak Bet	Ratanpur
	AKC03515	Carnelian	Ratanpur	Mardak Bet
	AKC03516	Agate	Mardak Bet	Ratanpur
	AKC03517	Agate	Ratanpur	Mardak Bet
Krek 52/62, Cambodia Late first millennium BCE	AKC00657	Carnelian	Ratanpur	Mardak Bet
Phnom Borei 200 BCE - 0 BCE/CE	AKC01950	Carnelian	Mardak Bet	Ratanpur
	AKC01951	Carnelian	Mardak Bet	Ratanpur
Phum Snay, Cambodia 350 BCE - 200 CE	AKC00003	Agate	Mardak Bet	Ratanpur
	AKC00016	Carnelian	Ratanpur	Mardak Bet
	AKC00020	Carnelian	Mardak Bet	Ratanpur
	AKC00025	Carnelian	Mardak Bet	Ratanpur
	AKC00026	Carnelian	Mardak Bet	Ratanpur
	AKC00035	Carnelian	Mardak Bet	Ratanpur
	AKC00044	Carnelian	Ratanpur	Mardak Bet
	AKC00053	Carnelian	Ratanpur	Mardak Bet
Prohear, Cambodia 200 BCE - 100CE	AKC00643	Carnelian	Ratanpur	Mardak Bet
	AKC00644	Agate	Ratanpur	Mardak Bet
	AKC00646	Agate	Mardak Bet	Ratanpur

Promptin Tai, Thailand 500 BCE - 500 CE	AKC00922	Carnelian	Mardak Bet	Ratanpur
	AKC00923	Agate	Ratanpur	Mardak Bet
	AKC00932	Carnelian	Ratanpur	Mardak Bet
	AKC00980	Carnelian	Mardak Bet	Ratanpur
	AKC01051	Carnelian	Mardak Bet	Ratanpur
	AKC01108	Agate	Mardak Bet	Ratanpur
	AKC01111	Agate	Mardak Bet	Ratanpur
Village 10.8, Cambodia 400 BCE - 50 CE	AKC00303	Carnelian	Ratanpur	Mardak Bet
	AKC00308	Agate	Ratanpur	Mardak Bet
	AKC00344	Carnelian	Ratanpur	Ban Khao Mogul
	AKC00348	Carnelian	Ratanpur	Mardak Bet
	AKC00364	Carnelian	Ratanpur	Mardak Bet
	AKC00433	Carnelian	Ratanpur	Mardak Bet
	AKC00437	Carnelian	Ratanpur	Mardak Bet

Table 1: Agate/carnelian artifacts analyzed in this study and their primary (CDA PGM 1) and secondary (CDA PGM2) predicted group memberships from Canonical Discriminant Analysis.

	Ratanpur 15 samples	Mardak Bet 15 samples	Shahr-i-Sokhta 17 samples	Ban Khao Mogul 17 samples	<i>Total</i>
SiO₂	0	0	0	0	0
Na₂O	14	10	1	12	37
MgO	1	0	0	0	1
Al₂O₃	0	0	0	0	0
P ₂ O ₅	15	15	17	13	60
K ₂ O	8	12	12	2	34
CaO	0	1	0	2	3
MnO	6	3	6	7	22
Fe₂O₃	1	0	2	2	5
CuO	10	7	4	10	31
SnO₂	2	2	1	3	8
PbO	1	0	0	0	1
Li	5	9	11	3	28
Be	5	11	1	0	17
B	0	0	0	1	1
P	15	15	14	13	57
Sc	0	0	0	0	0
Ti	2	1	3	2	8
V	0	0	2	4	6
Cr	13	14	14	13	54
Ni	3	0	0	0	3
Co	7	8	6	8	29
Zn	7	5	0	1	13
Rb	1	0	1	0	2
Sr	0	0	0	0	0
Zr	0	0	0	0	0
Nb	6	9	1	0	16
Ag	11	9	5	10	35
Sb	1	6	0	0	7
Cs	6	6	3	3	18
Ba	0	0	1	0	1
La	0	0	3	2	5
Ce	0	0	0	0	0
Pr	2	2	3	4	11
Ta	9	11	11	8	39
Au	3	5	0	0	8
Y	0	0	1	0	1
Bi	4	6	5	10	25
U	7	2	0	0	9

W	11	12	6	10	39
Mo	9	6	3	3	21
Nd	2	3	5	7	17
Sm	7	9	11	10	37
Eu	5	9	10	8	32
Gd	6	10	11	10	37
Tb	7	8	8	9	32
Dy	7	10	7	10	34
Ho	4	3	6	9	22
Er	8	8	5	10	31
Tm	9	9	5	9	32
Yb	9	10	1	11	31
Lu	10	10	2	9	31
Hf	9	12	8	7	36
Th	6	6	5	6	23

Table 2: Elements measured in LA-ICP-MS and the number of samples from each geologic source in which the sample was below the limits of detection (LOD) for that element. The elements in bold are those that were used in statistical analyses.

	Ratanpur	Mardak Bet	Shahr-i-Sokhta	Ban Khao Mogul
SiO ₂	997,681 ± 1026	997,561 ± 1148	995,025 ± 9574	988,199 ± 16045
Na ₂ O	<LOD	780 ± 280*	917 ± 1230*	511 ± 426*
MgO	36 ± 29*	128 ± 78	270 ± 538	1018 ± 2605
Al ₂ O ₃	538 ± 330	584 ± 439	1791 ± 5287	6864 ± 9855
K ₂ O	131 ± 91*	236 ± 108*	559 ± 551*	1943 ± 3623*
CaO	1269 ± 699	1061 ± 422*	1069 ± 1730	714 ± 632*
Fe ₂ O ₃	425 ± 258*	399 ± 403	863 ± 949*	1515 ± 2399*
B	14.53 ± 9.45	16.58 ± 12.03	58.93 ± 26.85	4.78 ± 4.19
Ba	2.30 ± 2.96	15.08 ± 38.79	15.76 ± 40.31	47.27 ± 52.64
Ce	0.08 ± 0.08	0.17 ± 0.19	0.45 ± 1.26	4.52 ± 10.16
La	0.04 ± 0.04	0.06 ± 0.06	0.24 ± 0.73	2.24 ± 4.84
Ni	3.42 ± 2.19*	2.43 ± 1.84	0.90 ± 1.02	0.72 ± 0.73
Pb	1.75 ± 3.15*	1.60 ± 1.74	3.47 ± 4.74	1.66 ± 1.99
Rb	0.28 ± 0.20*	0.38 ± 0.35	0.56 ± 0.47*	3.59 ± 5.56
Sb	0.43 ± 0.31*	0.25 ± 0.33*	1.89 ± 0.89	3.29 ± 3.31
Sc	1.97 ± 0.42	3.19 ± 2.05	0.97 ± 0.24	1.31 ± 0.59
Sr	0.99 ± 0.64	1.79 ± 2.16	16.62 ± 34.12	12.62 ± 16.72
Ti	6.09 ± 8.54*	7.87 ± 15.51*	37.54 ± 128.26*	161.98 ± 419.05*
U	0.01 ± 0.03*	0.14 ± 0.12*	8.21 ± 9.93	0.97 ± 1.78
Y	0.15 ± 0.21	0.16 ± 0.22	0.66 ± 1.87*	5.01 ± 11.33
Zr	0.27 ± 0.50	0.36 ± 0.41	6.66 ± 21.55	9.35 ± 15.73

Table 3: Average concentrations and standard deviations of the elements used in statistical analyses in ppm for each of the four geologic sources. *Refers to elements that had one or more samples with measurements at the LOD.