Lead Isotope Analysis of Copper Alloy Objects from the Iron Age Baba Jilan Archaeological Site, Luristan, Western Iran

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Abstract

This paper presents the lead isotopic composition of 15 tin bronze objects recovered at Baba Jilan, an Iron Age cemetery located in northern Luristan. The results are compared with a compilation of data available for ore sources, metal objects and slags from across Iran. These data show that the objects have a lead isotopic composition that is compatible with multiple copper sources in Iran, primarily from the Urumieh-Dokhtar and the Sanandaj-Sirjan, where ancient mining activities have been identified, and that were suggested as possible sources for the production of metals recovered at other sites on the Iranian Plateau.

Keywords: tin bronzes, Luristan, Iron Age, lead isotopes, sourcing

Introduction

Despite its central role in early metallurgy and the development of tin bronzes, the Iranian Plateau remains understudied in terms of identifying ore sources used for metal production. The earliest examples of tin bronze objects date to the Early Bronze Age (dated from the end of the 4th millennium BC to early 3rd millennium BC) in western Iran (Fleming et al. 2005; Thornton 2009; Helwing 2013), where it developed during the Bronze Age (ca. 3000-1500 BC) (Pigott 2004; Thornton 2009) and expanded during the Iron Age (1500-550 BC) (Moorey 1969, 1982; Haerinck 1988; Muscarella 1990; Overlaet 2005). The proliferation of tin bronze objects in the Iranian Plateau during the Iron Age shows the importance of this alloy for the manufacture of commonplace and ritual objects during this time, especially in western, northern and north-western Iran, and also in the Middle and Neo Elamite civilization in the south-western Iranian Plateau (Moorey 1969; Fleming et al. 2006; Mortazavi et al. 2011; Oudbashi and Davami 2014; Oudbashi and Hessari 2017; Oudbashi et al. 2019). The Luristan bronzes are among the most famous bronze collections from western Iran and include a large collection of funerary and religious objects from Iron Age cemeteries and sanctuaries from the Luristan region (central Zagros). It is however important to note that among the thousands of bronze objects held in museum collections that are attributed to Luristan, only a small percentage has been scientifically excavated and recorded (Calmeyer 1969; Moorey 1969; Muscarella 1990; Overlaet 2004, 2006; Pigott 2004).

Although numerous bronze objects dating to the Iron Age have been found in the Luristan region during the past 100 years, there is no evidence for tin bronze metallurgy in the region, and the question regarding the origin of these objects and the ore sources used for their production remains open.

Iran is rich in copper ore sources, with the richest metallic deposits found in the Urumieh-Dokhtar, the Sanandaj-Sirjan, Central Iran and the Lut Block (Ghorbani 2013). Iranian copper sources may have been used in ancient times (probably prehistoric period) for the extensive metallurgical industry observed in the Iranian Plateau (Momenzadeh 2004; Pigott 2004; Thornton 2009). Despite the lack of a detailed chronology of mine use, archaeological and geological surveys conducted on the Iranian Plateau have demonstrated the extensive mining of the region's ores during ancient times and more than 400 copper deposits are known in Iran with 79 of them bearing traces of ancient mining activities (Nezafati et al. 2008).

At present, the origin of tin resources in the area remains more enigmatic, and although sources of tin exist in Afghanistan, or Anatolia (see Berger et al. 2019 for a recent review of tin sources), the sources used for the production in Luristan have not been identified yet. In this context, the discovery and study of a copper-tin mine at Deh Hosein (eastern Luristan, western Iran) was one of prime importance and it was proposed that the mine was used for the production of bronze alloy from the Bronze Age to the Iron Age in the eastern Mediterranean region and western Iran (Momenzadeh et al. 2002; Momenzadeh 2004; Nezafati et al. 2006, 2009).

Over the last decades, lead isotopic analysis of ancient copper-based objects has led to significant progress in our knowledge of metal provenance and circulation (e.g., Gale 1997; Stos-Gale et al. 1997, 2009; Ling et al, 2014; Pernicka 2014; Pollard and Bray 2014, Artioli et al. 2020 and references therein). Despite the success of the approach, only a few studies have used lead isotopes to identify the ores used for the production of copper and bronzes in ancient Iran. Analyses were conducted at Arisman, Tappeh Sialk, and Shahr-i Sokhta (Hauptmann et al. 2003; Nezafati et al. 2008; Pernicka et al. 2011), but sources used to produce copper-base objects during the Bronze Age and Iron Age of the Iranian Plateau are poorly documented (Nezafati 2006; Cuénod et al. 2015) with only one major study on copper-base Bronze Age objects from Pusht-i Kuh (Luristan) (Begemann et al. 2008).

Due to its role as a centre for the emergence of early tin bronze metallurgy during the Bronze Age, and because of the extensive bronze findings in the region in comparison with others on the Iranian Plateau during the Iron Age, as well as the discovery of Deh Hosein as a possible tin source in antiquity in western Iran, Luristan reveals the importance of the central Zagros region in the metallurgy of copper alloys and the existence of possible ancient tin resources in the Luristan or neighbouring regions.

This paper will evaluate objects found at Baba Jilan, an Iron Age cemetery located in northern Luristan, to determine if their lead isotopic composition is compatible with that of copper sources in Iran, and in doing so contribute to our understanding of possible ore sources used for the production of bronzes in a region central to the development of metallurgy. To this end, we collected and compiled lead isotopic data available in the literature for Iranian copper ores sources with well documented provenance, as well as copper objects and slags recovered at multiples sites dating from different periods.

Materials and Site

The bronze objects analysed in this study were recovered from the Baba Jilan, an Iron Age cemetery in Pish-i Kuh of Luristan, which is located in northern Luristan (Figure 1). The cemetery is located about 30 km west of the city of Nurabad in the northern part of the modern Lorestan province and is situated on the southern side of the Sar Kashti mountain in the Delfan region (Hasanpur et al. 2015). The site was first surveyed in 2006 after damage from looting activities was reported in 2005. Subsequently, the site was excavated in 2007–2008 in two individual seasons. Based on the excavations, 11 tombs were discovered, including six cist graves as well as five jar burials. Archaeological materials discovered during excavations included potteries and a diversity of objects made of metal, bone, stone, blue frit and shell (Hasanpur et al. 2015; Oudbashi and Hasanpour 2016, 2018). Radiocarbon dates obtained on three human bone samples revealed that the cemetery was used during the Iron Age around 700-800 BC (Hasanpur et al. 2015; Oudbashi and Hasanpour, 2018). It can therefore be concluded that the Baba Jilan represents a typical graveyard of the Iron Age II period containing bronze objects related to Luristan Bronzes (Hasanpur et al. 2015).

The metallic objects analysed in this study include 15 tin bronze objects selected from a small collection of objects that were previously analysed by ICP-MS and SEM-EDS for the identification of their chemical composition and microstructure (Oudbashi and Hasanpour 2016, 2018). While the objective of the present paper is not to revisit the conclusions previously obtained, a summary of the main findings resulting from the bulk chemistry analysis and the metallography is reported here for the purpose of their integration with the isotopic results. The 15 objects analysed for lead isotopic composition include eight vessels, one vessel's spout, three buttons, two pins, and one spring bead (Table 1). The quantitative ICP-MS and semi-quantitative SEM-EDS analysis of these 15 samples demonstrated that the objects were made of binary copper-tin alloy with variable tin concentration, between 5.26 and 11.16 %, meaning that all of them were low tin bronzes based on the classification used in archaeometallurgy (low tin corresponds to a level below 15%). The vessels showed, in general, slightly higher tin levels but no correlation was found between the type of object and the tin content (Oudbashi and Hasanpour 2018). These analyses also determined that lead and arsenic concentrations were low, ranging between 0.01 and 0.44 %, and between 0.04 and 0.51 %, respectively, which indicates that the two elements should be considered as impurities from the ore rather than as an addition for alloying purpose. In general, all the other elements analysed, including Ag, Co, Fe, Ni, Sb and Zn, are present as minor/trace amounts in these objects. The metallographic analysis revealed that the objects analysed here were produced through several cycles of cold working by hammering and heat working by annealing. The findings published in Oudbashi and Hasanpour (2016, 2018) are in agreement with previous results that showed that the addition of lead for the casting of alloy was not a technique used in Luristan metallurgy, and the Iron Age tin bronzes found in Iran tend to present a variable tin content (Oudbashi 2019a, 2019b). The comparison of the Baba Jilan objects with other Iron Age objects confirmed the pattern of no correlation between the tin content and any specific type of object as also observed in further Luristan bronzes (Oudbashi 2019a, 2019b). Oudbashi and Hasanpour (2016, 2018) suggested that the Iron Age tin bronzes from Luristan were produced using an uncontrolled alloying process such as co-smelting, cementation or the application of copper-tin bearing complex ores.

In co-smelting, the copper (sulphidic or sulphidic/oxidic copper) and tin ores (cassiterite, SnO_2) are smelted directly together to make bronze, while cementation corresponds to the process of adding cassiterite to metallic copper in crucible in a reducing atmosphere to make tin bronze (Pigott 2004; Rovira et al. 2009; Erb-Satullo et al. 2015; Oudbashi et al. 2016). Of course other alloying methods are noted in literature including melting metallic copper and tin together, recycling or using copper-tin bearing complex ores (Coghlan 1975; Pigott 2004; Oudbashi et al. 2016; Oudbashi and Hessari 2017).

Based on the cuneiform texts, the ancient Mesopotamian bronze production recipes are known to have a Sn/Cu ratio of 1:6, 1:8 and 1:9 (16.6, 12.5 and 11.11 percent of tin respectively) for different types of bronze objects (Potts 1997; Joannes 1997; Helwing 2009). Nevertheless, further evidence suggests that tin content could not be easily controlled (Moorey 1994). According to previous archaeometallurgical works on the Luristan bronzes, it can be suggested that they were produced with an uncontrolled process that caused the bronze objects to present variable tin content (e. g. Oudbashi and Davami 2014; Oudbashi and Hasanpour 2018). The results of comparative studies on the composition of tin bronze objects (for example thin sheet vessels) revealed that there is no correlation between the tin concentration and object typology in the Luristan bronzes (e.g. see Oudbashi 2019a, 2019b). These evidences are in contrast with the ancient texts about tin bronze production in the ancient Near East. Furthermore, the colour of low-tin bronze alloy with less than about 15 percent of tin leading to formation of alpha bronzes (theoretically), is golden hue in comparison with pure copper that is salmon-red in colour. So, the final product of all tin bronze production methods, does not have obvious difference in colour in the prehistoric period because most ancient bronze alloys have less than 15 percent of tin (Meeks 1993a,1993b; Scott 2010). Some limited number of tin bronze objects with higher amount of tin were observed in the prehistoric period of Iran (Fleming et al. 2006) but tin content is less than 20 percent in these objects that caused this partially high-tin objects to present an appearance similar to that of other low-tin objects. Of course, it should be mentioned that the bronze objects with higher that 15 percent of tin are rare in the Iron Age Luristan (Oudbashi 2019a, 2019b).

The recycling or re-melting the broken or useless tin bronze objects to make metallic objects. The results of analytical and experimental studies on the recycling process shows that this process leads to produce tin bronze ingots with low amount of tin due to its preferential oxidation in respect to copper (Figueiredo et al. 2010; Valério et al. 2010). This metallurgical process could also have been used to produce Luristan bronzes, as is observed in the prehistoric time around the world (Karageorghis and Kassianidou 1999; Bray and Pollard 2012; Vernet et al. 2019; Charalambous and Webb 2020).

It should be mentioned that tin ingots were traded in the ancient time in the Near East and Anatolia (Pulak 2000) but that there is so far no evidence for the use or trade of metallic tin in Iranian Plateau during the Prehistoric period. So it is reasonable to suggest the trade cassiterite for application of cementation or co-smelting of copper and tin ores to produce bronze alloy in the Bronze Age and Iron Age of Iran (Oudbashi and Hessari 2017). On the other hand, the evidences from the recent discovered ancient copper-tin mine of Deh Hosein in the east of Luristan, shows probable use of tin-bearing coper ores to obtain tin bronze, dated back to second and first millennium BC (Nezafati 2006).

These evidences show that the production of the Luristan bronzes has been performed by using one (or more) uncontrolled alloying method because, as mentioned above, tin concentration is strongly variable in the composition of the Luristan bronzes and because there is no evidence of relationship between typology and composition of the objects.

Finally, two hypotheses can be derived from a variable tin bronze composition:

- The ancient metalworkers could not produce bronze alloy with distinct composition due to the use of uncontrolled alloying processes to make bronze. The processes explained above may not lead to make bronze with specific proportions of alloy constituents.
- The Luristan bronzes are religious/ritual objects that are deposited in graves or temples and there was no importance to make them with bronze alloys with specific compositions.

Methods and Analytical Conditions

A fragment of each object (25-70 mg) was cut with a plier or using a diamond disk mounted on a micro-drill. Each fragment was cleaned using sand paper to remove the corrosion layer and then cleaned in mQ water (sonication for 30 minutes), rinsed with mQ water and finally dried at room temperature. The samples were digested in polypropylene tubes using a mixture of 14N Optima Grade HNO_3 and 12N Optima Grade HCl (1-1 ml respectively) and were heated at 85°C in a warming unit for approximately 12 hours. The following steps were realized in a clean room that provided the clean environment and low background required for isotope analyses.

An aliquot of the solutions was taken to obtain approximately 2000 ng of Pb. These aliquots were evaporated at 90°C in PFA vials on a hot plate, the dry residue was re-dissolved in 6N HCl and then evaporated at 90°C. The dry residue was re-dissolved in 0.5N

HBr. The lead extraction and the sample preparation for lead isotope analysis were conducted as described in Renson et al. (2016).

Lead isotope analyses were conducted using a Nu Plasma II (Nu Instruments) multi-collector - inductively coupled plasma - mass spectrometer (MC-ICP-MS) in operation at the Missouri University Research Reactor (MURR). The instrument was optimized to obtain a minimum of 100 mV for mass 204. The samples and SRM981 standard were spiked using a Tl solution to obtain identical Pb and Tl concentrations in the samples and in the standard (approximately 200 ng g^{-1} in Pb and 50 ng g^{-1} in Tl). The SRM981 was measured after every two samples. The lead isotopes values of the standard and the samples were corrected for mass fractionation using the NIST value for the $^{205}Tl/^{203}Tl$ natural ratio (2.38714) and for Hg isobaric interference at mass 204 using a 0.229883 value for the $^{204}Hg/^{202}Hg$ natural ratio. The mean values for the SRM981 were 36.685 ± 0.007 (2SD), 15.488 ± 0.002 (2SD) and 16.935 ± 0.002 (2SD) for $^{208}Pb/^{204}Pb$, $^{207}Pb/^{204}Pb$ and $^{206}Pb/^{204}Pb$, respectively (n=17). The values measured for the samples were corrected by standard-sample bracketing method (White et al. 2000; Weis et al. 2006) using the values published by Galer and Abouchami (1998) to eliminate instrumental drift.

As mentioned above, the lead concentration is below 0.5 % in all objects from Baba Jilan (Oudbashi and Hasanpour 2018). Moreover, in most cases, tin sources (cassiterite, stannite or ingots) present very low lead concentrations in comparison with copper sources (Begemann et al. 1999). The lead present in these objects is therefore considered to be associated with the copper sources and the lead isotopic composition representative for that of the copper ores.

Results

The fifteen objects from Baba Jilan have a lead isotopic composition that varies between 38.622 and 39.341, 15.626 and 15.732, 18.540 and 18.972, 2.05938 and 2.08670 and between 0.82775 and 0.84541 for $^{208}Pb/^{204}Pb$, $^{207}Pb/^{204}Pb$, $^{206}Pb/^{204}Pb$, $^{208}Pb/^{206}Pb$ and $^{207}Pb/^{206}Pb$ ratios, respectively (Table 1 and Figure 2). The lead isotopic signature of the 15 objects does not display systematic relation with the concentration of any element, including that of tin. The three pin samples (BJ-07, BJ-08, and BJ-17) have higher

$^{208}Pb/^{206}Pb$ values for a same $^{207}Pb/^{206}Pb$, and higher $^{208}Pb/^{204}Pb$ for similar $^{207}Pb/^{204}Pb$ and $^{206}Pb/^{204}Pb$ ratios values, in comparison with the rest of the objects. No other systematic relation appears between the type of object and the isotopic signature, however, a few pairs of samples display an almost identical isotopic composition using all five ratios, and for two of these pairs, the elemental composition is very homogeneous. The relative standard variation is used below to illustrate the range of variation of the elemental chemistry observed between these few specific couple of objects in comparison with that observed within the 15 objects (Table 2) (data provided based on Oudbashi and Hasanpour 2018). Two vessel samples (BJ-14, and BJ-19), have the almost exact same lead isotopic composition and similar elemental signature, with RSD between 0 and 17% for the 10 elements (Table 2). The two pins (BJ-03, and BJ-06) have the almost exact lead isotopic and elemental composition, with RSD between 0 and 4.3% for the 10 elements (Table 2). Two other samples, a vessel (BJ-02) and a vessel's spout (BJ-01), have similar isotopic composition but are characterized by larger differences in their elemental composition, with RSD between 3 and 88% for the 10 elements (Table 2). These results likely indicate that some of the objects were made of specific ore sources using a similar production processes. Considered all together, the isotopic variation within the 15 samples is rather large and likely results from the use of multiple sources for the production of these objects. This is examined below through a comparison between the Baba Jilan objects with around 125 data for Iranian copper ore sources and associated mineralizations (Figure 3), and then with data for 68 slags, prills, and objects found at multiple sites in Iran (Figure 4). References for the Iranian copper ore sources and for the objects, prills and slags (Chegini et al. 2000; Hauptmann et al. 2003; Nezafati 2006; Begemann et al. 2008; Nezafati et al. 2008; Shafiei 2010; Pernicka et al. 2011; Nezafati and Stöllner 2017; Isotrace Oxalid) are detailed in Table 3. Part of these isotopic data are for ore deposits where no ancient mining activities have been documented so far. These comparisons are conducted by looking at the assemblage of 15 objects, and then by looking at the different couple or single objects. The comparisons are also conducted based on all five isotopic ratios: $^{208}Pb/^{204}Pb$, $^{207}Pb/^{204}Pb$, $^{206}Pb/^{204}Pb$, $^{208}Pb/^{206}Pb$, and $^{207}Pb/^{206}Pb$ (Figures 2, 3 and 4).

The data used for comparison are derived from multiple studies conducted using different instruments, with various analytical conditions, and that do not necessarily provide the different corrections or do not report the error ranges that apply to the data. This can cause offsets between data from different studies and difficulties when comparing these data (see Baron et al. 2013), which limits our ability to identify the best isotopic compatibility. Despite this limitation, we used the values of the five isotopic ratios to identify potential ore sources based on the compatibility of the ratios. It is also important to note that data currently available for the ore deposits used hereafter for comparison are not necessarily representative for the entire range of their isotopic variability. The deposits are also presented in relation with the geologic region they belong to, but, similarly, these deposits do not necessarily represent the entire range of isotopic variability for the geological zone they belong to.

Data available for copper deposits from the Lut Block at Chehel Kureh and Qaleh Zari are not compatible with Baba Jilan objects. The number of data available for ores from the Lut Block is very limited, and the incompatibility observed here is valid only for these few samples and cannot be interpreted any further at this point. We are considering successively the association with different geological zones and with specific deposits.

The isotopic composition of objects from Baba Jilan differs from the ores from Central Iran (Figure 3) and none of the ore deposits from the Anarak region have an isotopic composition that can explain that of the Baba Jilan objects. BJ-01 and BJ-02, however, have a signature close to that of Bagh Ghorogh (Baghorogh) and Chah Gorbeh based on the $^{208}Pb/^{206}Pb$ vs $^{207}Pb/^{206}Pb$ ratios but have higher $^{207}Pb/^{204}Pb$ ratio values for a same $^{208}Pb/^{204}Pb$ and $^{206}Pb/^{204}Pb$ ratios.

Except for one sample (BJ-07), the objects from Baba Jilan display a lead isotopic composition that largely overlaps with that of the ores from the Urumieh-Dokhtar Zone (values reported here are for deposits from the Karkas, including Veshnaveh, from the Kerman Cenozoic volcanic arc, and from the north-western part of the Urumieh-Dokhtar), and with that of ores from the Astaneh-Sarband (values reported here are for deposits from Nezam Abad, Astaneh, and Deh Hosein) in the Sanandaj-Sirjan Zone (Figure 3).

BJ-01 and BJ-02, a vessel spout and a vessel that present very similar isotopic signature but different elemental chemistry, show some compatibility with the deposits of Deh Hosein (Astaneh-Sarband) based on the 206 normalized ratios, $^{208}Pb/^{204}Pb$, and $^{206}Pb/^{204}Pb$, but exhibits discrepancy with these sources using the $^{207}Pb/^{204}Pb$ ratio. These two objects also partly overlap with sources in the Karkas (Urumieh-Dokhtar) but no consistent match with a specific source can be observed. BJ-03 and BJ-06, the two pins that present very similar elemental and isotopic compositions, have a signature compatible with the field of the deposits of Astaneh (Astaneh-Sarband) and sources in the Karkas, including Veshnaveh (Urumieh-Dokhtar). The sample BJ-012, a vessel, have an isotopic signature close to that of the pins and shares the similarity with the same ore sources. BJ-13, a vessel, has an isotopic signature that closely corresponds with that of the ores from Veshnaveh and other sources in the Karkas (Figure 3). BJ-15, another vessel, has a composition compatible with that of sources in the Karkas, and with ores from Nezam Abad in the Astaneh-Sarband. Sample BJ-20 mainly overlaps with sources in the Kerman, the Karkas and Nezam Abad. Sample BJ-09, a bead, overlaps with the field of the Karkas sources. BJ-14 and BJ-19, two vessels with a similar isotopic and elemental signature, have a lead isotopic signature that differs from Veshnaveh, and the Karkas more largely, but overlaps with the field of the Nezam Abad sources in the Astaneh-Sarband. Sample BJ-04, a vessel, share a similar association with the Nezam Abad deposits. Samples BJ-07, BJ-08 and BJ-17, the three button samples do not show any consistent match with any source. BJ-07 has a signature that does not correspond with any of the sources presented here. Sample BJ-17 overlaps with the signature of the Nezam Abad deposit based on the ratios normalized to mass 204 but differs based on the ratios normalized to 206. Object BJ-08 is close to sources in the Kerman based on the $^{208}Pb/^{206}Pb$, $^{207}Pb/^{206}Pb$, $^{208}Pb/^{204}Pb$ and $^{207}Pb/^{204}Pb$ ratio values, but differs from that source based on the $^{206}Pb/^{204}Pb$ value.

In their study of the lead isotopic composition of 69 objects excavated at Pusht-i Kuh, and from the collection of Luristan objects from the Louvre Museum, Begemann et al. (2008) identified three main groups. Based on ratios normalized to 204 and 206, seven objects from Baba Jilan (BJ-01, BJ-02, BJ-03, BJ-06, BJ-08, BJ-12 and BJ-15) have a composition that is compatible with that of the group described in Begemann et al. (2008) as presenting the lowest $^{208}Pb/^{206}Pb$ and $^{207}Pb/^{206}Pb$ ratios (Figure 4). Two additional objects (BJ-13 and BJ-20) overlap with that isotopic field based on most ratios but are different based on $^{207}Pb/^{204}Pb$ value. The authors associate some of the objects from this isotopic group with sources at Veshnaveh and some with the mining district of Anarak in the central Iranian Plateau. Among these seven objects, four (BJ-01, BJ-02, BJ-03, BJ-06, BJ-08 and BJ-12) have a signature that is also compatible or close with samples from Arisman, although some differ based on the $^{207}Pb/^{204}Pb$ value (Figure 4). Two objects (BJ-01 and BJ-02) are also compatible with material from Tappeh Sialk (Figure 4). Three objects (BJ-13, BJ-15, and BJ-20) are compatible with material from Qoli Darvish and Tappeh Sarm (Figure 4). Qoli Darvish is a site located in central Iran that provided an outstanding stratigraphic sequence 4th to 2nd millennium BC (the Late Chalcolithic to the Late Bronze Age) and Tappeh Sarm is an Iron Age II/III cemetery contemporaneous with the Luristan bronzes (Nezafati and Stöllner 2017). Object BJ-17 has a composition that falls between that of two samples from Pusht-i Kuh identified as singletons and unclearly sourced by Begemann et al. (2008). Tentatively, the authors related these two objects to ore sources in Eastern and Central Anatolia, but acknowledged that these are not the most likely sources due to their arsenic content. Four objects (BJ-04, BJ-07, BJ-14 and BJ-19) present no clear compatibility with any material used here for comparison (Figure 4).

Because the concentrations of some trace elements can be used to further decipher between sources (i.e., Pernicka 2014; Pollard and Bray 2014), a comparison was performed between trace element content of the 15 objects from Baba Jilan (Oudbashi and Hasanpour 2018) with that of the ores and metallic objects that are used for lead isotopic comparison. It is important to mention that, while trace element concentration data are available for most of the sources from the Astaneh-Sarband used for the isotopic comparison, only a very limited number of samples from the Urumieh-Dokthar can be used to that end. Indeed, most of the studies used here for isotopic comparison, either did not report elemental data at all, or did not do it consistently for the samples and/or trace elements examined here. The slags and prills were not included as their elemental content differ from that of the objects. The comparison attempted below is therefore mainly meaningful for the sources from the Astaneh-Sarband and the objects from other sites. The scatter plot presenting Sb versus As shows that the Baba Jilan samples overlap with objects from Tappeh Sarm and some of the ore samples from Deh Hosein deposits (Figure 5a). It is worth noting that the objects analysed from the Bronze Age of Pusht-i Kuh (Luristan) Begemann et al. 2008) show no relationship with the Baba Jilan objects while they have weak correlation with some samples from Deh Hosein, Astaneh and Nezam Abad ore deposits (Astaneh-Sarband area). The scatter plot presenting Ni versus Ag shows that some Baba Jilan samples overlap with Pusht-i Kuh objects and one sample from Tappeh Sialk as well as with a few ore samples from Deh Hosein and Nezam Abad deposits (Figure 5b). Based on these two plots, no systematic correlation appears between the 15 samples from Baba Jilan and any of the groups of objects from other sites or ore deposits. The comparison based on a few trace elements conducted here however shows that objects from Baba Jilan have a composition that is in part compatible with that of ore sources in the Astaneh-Sarband, as well as that of some objects from Pusht-i Kuh, Tappeh Sarm, Qoli Darvish and Tappeh Sialk. These observations concur with the results of the lead isotopic analysis.

Discussion

Most of the objects analysed here (14/15) exhibit a lead isotopic composition that is compatible with multiple ore sources located in north-western Iran in the Urumieh-Dokhtar and the Sanandaj-Sirjan regions. Two of the objects (BJ-01 and BJ-02) are also possibly compatible with sources in Central Iran. Both the Urumieh-Dokhtar and the Sanandaj-Sirjan are among the richest Iranian lithotectonic domains for metallic deposits (Momenzadeh 2004). One sample (BJ-07) exhibits poor compatibility with any of these sources and displays higher 208 ratios, which suggests a source with a different U/Th ratio. The lead isotopic composition of 11 objects may have resulted from the use of multiple sources from the Astaneh-Sarband area with some of the samples corresponding to ores from the Nezam Abad deposit, some from the Astaneh deposit, some from Deh Hosein, and/or could be the result of a mixing of ores from all of these deposits. Three samples (BJ-09, BJ-13 and BJ-20) have a signature that is close to the field of the Astaneh-Sarband but have slightly lower $^{207}Pb/^{204}Pb$. The Deh Hosein deposits alone have an isotopic composition that only overlaps with two samples from Baba Jilan, but other objects have a composition compatible with deposits in the vicinity of Deh Hosein (approx. 15 km radius) at Astaneh and Nezam Abad. The pattern displayed by most of the objects (11/15) may also be the result of a mixing between these three ore sources. Based on this evidence, it can be proposed that the ores deposits from the Astaneh-Sarband area represent possible sources for the production of 11 objects from Baba Jilan. However, at this time it is impossible to know if objects were made from these ores separately of if they were made from a variable mixing of ores from multiple deposits. The hypothesis that the Astaneh-Sarband area could be the source for some of the objects found at the Iron Age cemetery in Luristan would support the findings of Nezafati (2006) who suggested that deposits from the Deh Hosein area were used for the production of Luristan Bronzes. The investigations conducted so far at Deh Hosein and the surrounding area in the Astaneh-Sarband revealed mining activities dating to 1775-1522 BC, and likely earlier (Nezafati et al. 2006). The lead isotopic composition of a broad diversity of mineralizations from Deh Hosein was found to display a narrow range of values (Nezafati 2006). A comparison between the lead isotopic signature of the Deh Hosein ore deposits with that of objects recovered at multiple sites in Luristan, Mesopotamia, Southern Persian Gulf, and Western Turkey, showed an overlap in signature between the Deh Hosein ores and number of these objects, which led to suggest that ores from Deh Hosein could have been used for the production of these objects and that Deh Hosein could have been a major supplier of the tin used across a wide area from western Turkey to southern Persian Gulf, and may have already been known and exploited as early as the 3rd millennium BC (Nezafati 2006; Nezafati et al. 2006). Because of the unique presence of tin in combination to copper so far attested in the proximity of Luristan and Mesopotamia, the study of these deposits represent an important discovery and should be included for comparison in sourcing studies of tin bronzes in the region. However, to this day, no evidence for smelting or other metal production activity has been identified at Deh Hosein or in the surroundings and the possible association between the objects found at Baba Jilan and these sources has therefore to be considered with caution.

Some of the Baba Jilan objects also clearly overlap with the ore sources of Veshnaveh. The mine of Veshnaveh, located in the Urumieh-Dokhtar, represents a type of mineralization that provides high-grade ore rich in copper that was likely attractive to miners in ancient times (Momenzadeh 2004; Nezafati and Stöllner 2017). Veshnaveh ores were exploited as early as the beginning of the 3rd millennium and up to the late 2nd millennium BC (Chegini et al. 2000; Nezafati and Stöllner 2017). Veshnaveh ores were suggested as a possible source for metal objects found at Tappeh Sarm, an Iron Age cemetery near Kahak, and for slags from Qoli Darvish, a Late Chalcolithic to Early Bronze Age site located south of Qom (Nezafati and Stöllner 2017). The authors also concluded, based on the chemical composition of the metals, that they were produced using a mixing of multiple ore sources. In their study of metallurgy at Arisman, a Chalcolithic site that provides some of the earliest evidence for smelting, Pernicka et al. (2011) suggested that a mixing of ores from Veshnaveh with ores from the Anarak region could explain the isotopic composition of slags and copper material found at the site. Some of the copper objects from Arisman have a lead isotopic composition similar to that of Veshnaveh ores but the authors do not favour these ores as the source because of their elemental chemistry, including low arsenic content (Pernicka et al. 2011). It is worth noting that the process of speiss production to provide arsenic and alloying of arsenic and copper has been attested in Arisman explaining that high level of arsenic in the objects from Arisman is due to deliberate alloying process leading to make arsenical bronze objects (Rehren et al. 2012). Veshnaveh was also suggested as one of the possible copper sources used for the production of some Bronze Age Luristan metals found at Pusht-i Kuh (Begemann et al. 2008).

In this study, we identified more Luristan Bronzes with a lead isotopic composition compatible with that of two objects identified as singletons, which were not clearly assigned by Begemann et al. (2008). For these objects, the present study suggests possible sources in the Astaneh-Sarband area and in the Urumieh-Dokhtar.

Even if the isotopic compatibility between an object and an ore source does not unequivocally indicate that this specific ore was used to produce that specific artefact, the present results show that several sources located in the Sanandaj-Sirjan and the Urumieh-Dokhtar, between 150 and 300 km east of Luristan, could be the source of raw material used for the production of objects found at Baba Jilan. Some of the possible sources, the Astaneh-Sarband area and the Veshnaveh deposits, were exploited during earlier periods for metals and were suggested as sources for metal production at multiple sites on the

Iranian Plateau. The results presented here show that there is not a single ore source that can explain the full range of isotopic variation presented by the Baba Jilan objects. The raw materials used to manufacture these objects are therefore likely derived from multiple sources, which has been previously suggested (Nezafati and Stöllner 2017). Whether the objects resulted from the use of various sources, and/or from a mixing of different sources, is difficult to determine at this point. It is also impossible to know whether or not these objects were produced at different workshops.

These results confirm those obtained on some Luristan objects from the Bronze Age (Begemann et al. 2008). They also support findings by Nezafati (2006), which suggest that the Deh Hosein area could have been an important source for raw material used for the production of tin bronze in Luristan.

The vast amount of Luristan Bronzes recovered from both archaeological excavations and illegal activities demonstrates large-scale bronze production on the Iranian Plateau during the Iron Age. Because these objects are from cemeteries and sanctuaries, and no evidence of Iron Age settlement has been found in the Luristan region, some researchers have attributed these objects to nomad populations who lived in western Iran (Overlaet 2004, 2005). These groups are thought to have moved from other parts of the Iranian Plateau to the highland of Luristan and placed the objects in the cemeteries and sanctuaries located in what could have been holy regions (Overlaet 2004). Moreover, no evidence for tin bronze metallurgy dating to the Iron Age has been found in Luristan, which suggests that tin bronzes were produced outside the region. Survey of the region to locate possible sites or workshops containing evidence for archaeometallurgical activities related to Iron Age bronze production is still needed, however, the geographical features of Luristan, with high mountains and deep valleys, makes it difficult to identify archaeometallurgical sites used in the prehistoric period. The results of the present study show that different ore deposits may have been used to produce tin bronze objects found at the Baba Jilan site. This may also be the result of using different ore deposits or the production of tin bronze in one (or more) archaeometallurgical workshops in Luristan. The results confirm that the tin bronze metallurgy during the Iron Age of Luristan was a complex process in which different ore deposits might have been used to produce bronze objects in Luristan and neighbouring regions.

From an archaeological viewpoint, the Luristan bronzes show a specific artistic style, but the analytical data shows that they could have been produced in different workshops and/or by using different ore resources. Accordingly, it is possible that they have been produced in different regions of the Iranian Plateau (or neighbour regions) by using their local and accessible ores and have been imported to the Luristan region or that the smelted raw materials (probably tin bronze ingots) or copper (and tin) ores have been obtained elsewhere outside Luristan and then have been imported to Luristan to produce objects in the specific style in the metalworking workshops. These results show the necessity to develop multi-analytical studies to more accurately address questions related to the production of Luristan bronzes. The application of the isotopic tool in future analyses of objects and sources should contribute to the identification of possible sources used for the production of metal in one of the central regions for its earliest development.

Conclusion

Most of the 15 samples recently excavated at Baba Jilan, and analysed here, exhibit a lead isotopic composition compatible with that of ore sources in Iran. Most of these sources are located in the Urumieh-Dokhtar zone and the Sanandaj-Sirjan, which constitute the richest areas for copper sources in Iran, and also possibly in Central Iran. These results support evidence from other sites that suggested the use of ores from these mineralization zones as the sources for the production of objects found at other sites. The objects from Baba Jilan show compatibilities with different sources and were likely made from multiple ore sources. Whether these objects originated from different production centres, or from one production centre using ores from multiple deposits, is unknown at this point.

The advent of tin bronze metallurgy in the Luristan region is an important event in the archaeometallurgy of the Near East. More field studies, as well as analytical and isotopic investigations, are necessary to reveal different aspects of this ancient technology from metallurgical and geoarchaeological points of view. Nevertheless, the results of this study revealed connections between the Luristan highland and the central Iranian Plateau during the Iron Age for the production of tin bronze alloy.

Acknowledgements

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Figure Captions

Figure 1. Site location, geological zones, and location of some ore deposits mentioned in the text (modified after Nezafati 2006).

Figure 2. Three-isotope plots for the objects from Baba Jilan. a. $^{207}Pb/^{204}Pb$ vs $^{206}Pb/^{204}Pb$ three-isotope plot, b. $^{208}Pb/^{206}Pb$ vs $^{207}Pb/^{206}Pb$ three-isotope plot.

Figure 3. Three-isotope plots comparing tin bronze objects from Baba Jilan and ore sources from the main geological zones in Iran. Lead isotope data for the ore sources from the main geological zones in Iran, with details for the sources from the Sanandaj-Sirjan and the Urumieh-Dokhtar zones. Isotopic data are reported from the references provided in Table 3.

Figure 4. Three-isotope plots comparing tin bronze objects from Baba Jilan and objects, prills and slags from multiple sites in Iran. Lead isotope data for samples from the different sites are reported from the references provided in Table 3.

Figure 5. Biplot comparing trace element content in the Baba Jilan objects with that of ore sources and objects used in the lead isotopic comparative analysis. a. Sb vs As, b. Ni vs Ag. Values are reported in % using a logarithmic scale. Values reported for ore sources and objects from other sites are from Nezafati 2006, Pernicka et al. 2011, Begemann et al. 2008, Nezafati et al. 2008, and Nezafati and Stöllner 2017.

Figure-1

Click here to access/download;Figure;Figure-1.jpg

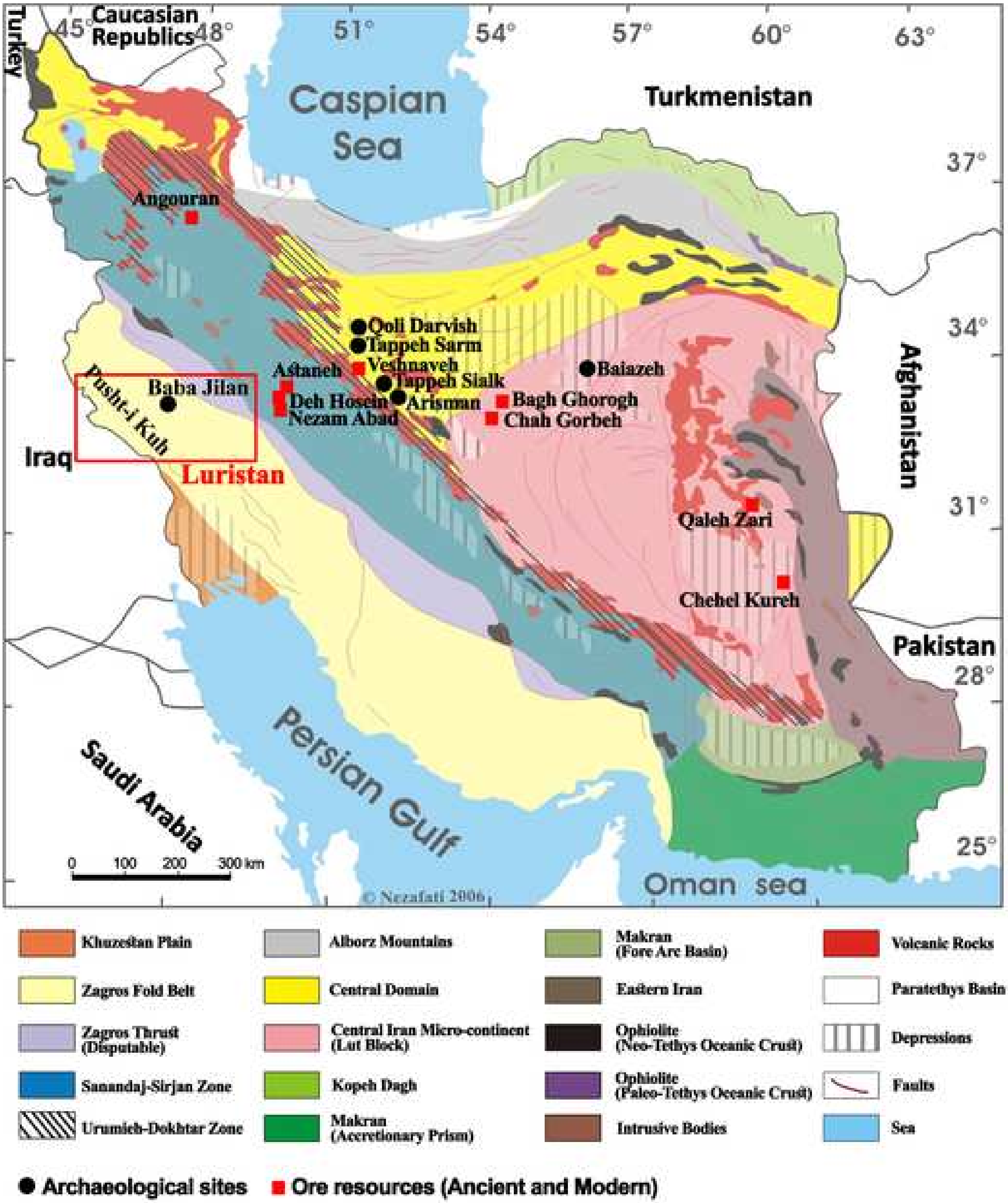

Figure-2

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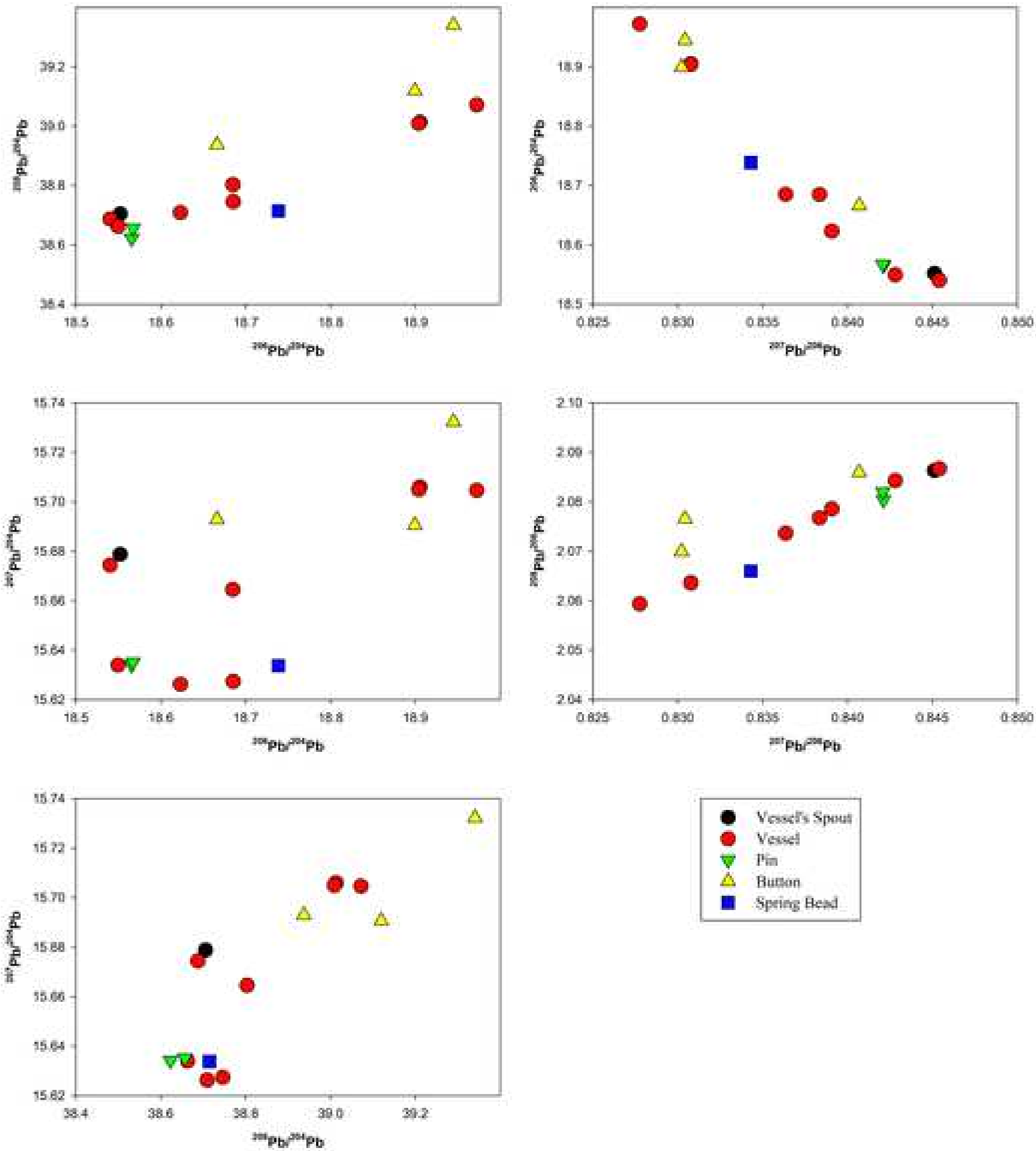
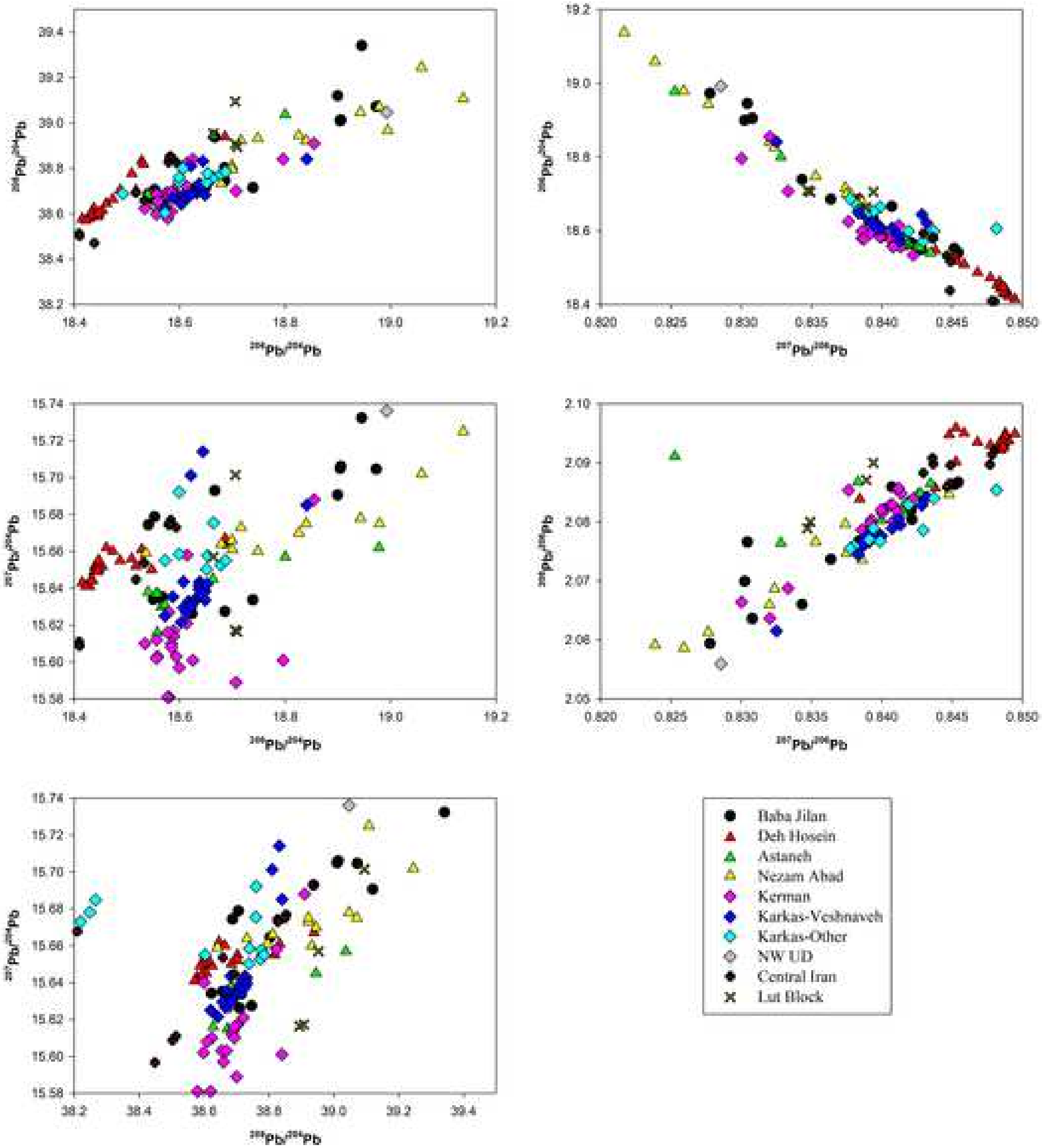




$^{208}Pb/^{204}Pb$
$^{206}Pb/^{204}Pb$
$^{207}Pb/^{206}Pb$
$^{207}Pb/^{204}Pb$
$^{208}Pb/^{206}Pb$
Baba Jilan
Deh Hosein
Astaneh
Nezam Abad
Kerman
Karkas-Veshnaveh
Karkas-Other
NW UD
Central Iran
Lut Block

Figure-4

Click here to access/download;Figure;Figure-4.jpg

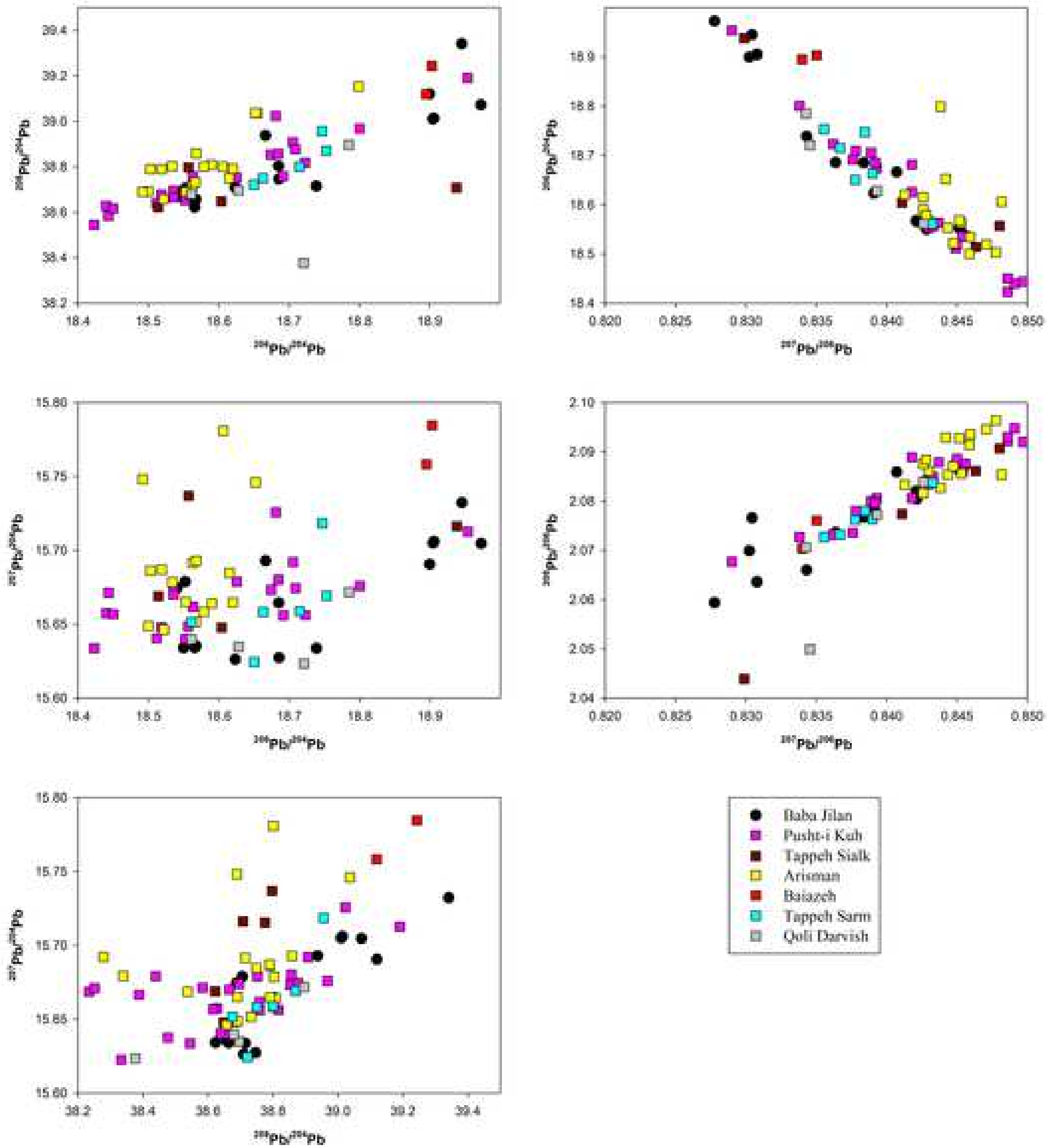
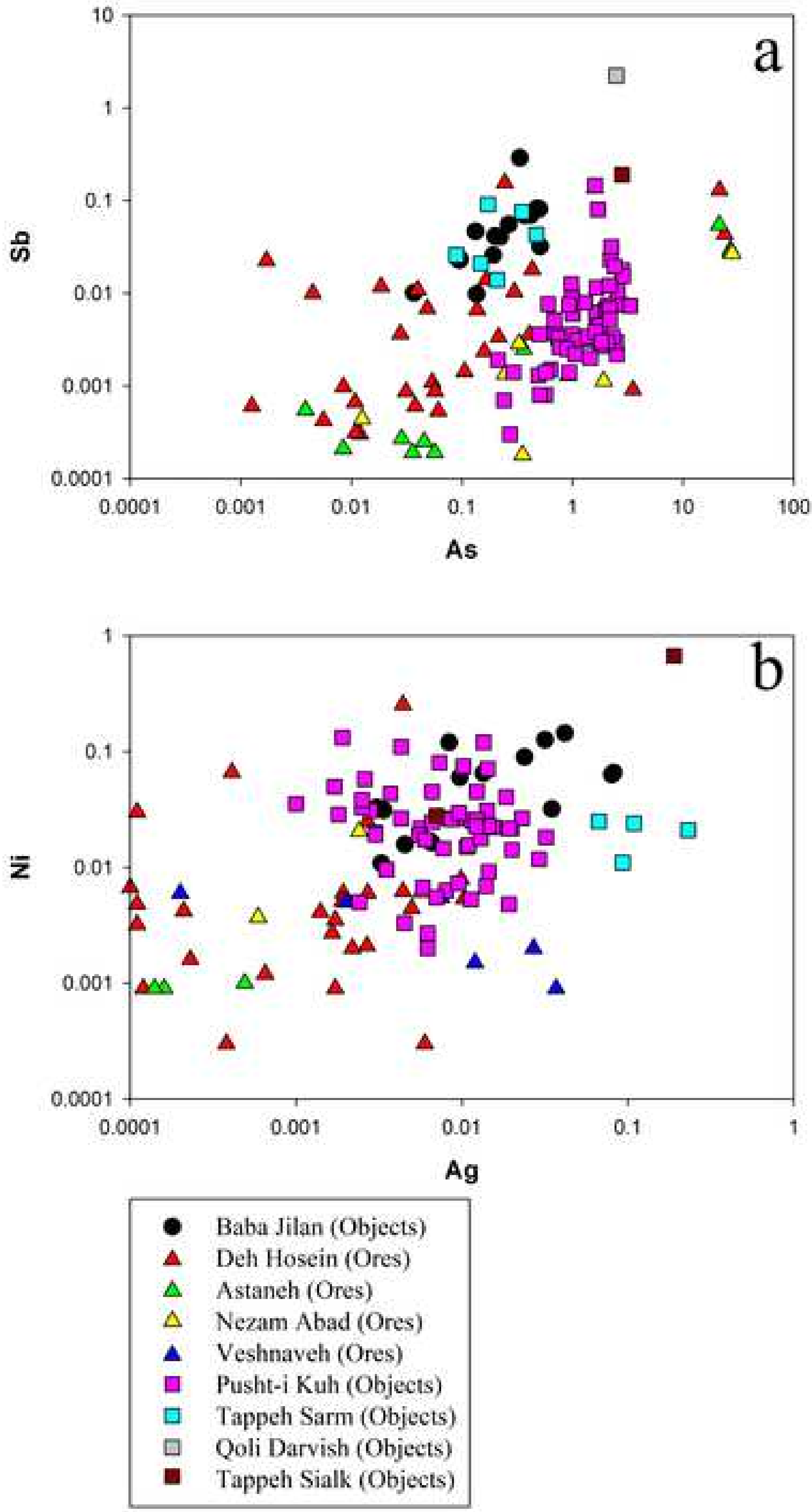


Figure-5



a
Sb
As
10
1
0.1
0.01
0.001
0.0001
0.0001
0.001
0.01
0.1
1
10
100
b
Ni
Ag
1
0.1
0.01
0.001
0.0001
0.0001
0.001
0.01
0.1
1
Baba Jilan (Objects)
Deh Hosein (Ores)
Astaneh (Ores)
Nezam Abad (Ores)
Veshnaveh (Ores)
Pusht-i Kuh (Objects)
Tappeh Sarm (Objects)
Qoli Darvish (Objects)
Tappeh Sialk (Objects)

Table-1

Table 1. Type of objects, sample ID and lead isotopic ratios for the 15 objects from Baba Jilan. Object typology and ID from Oudbashi and Hasanpour (2016, 2018).

Sample	Object	$^{208}Pb/^{204}Pb$	2se	$^{207}Pb/^{204}Pb$	2se	$^{206}Pb/^{204}Pb$	2se	$^{208}Pb/^{206}Pb$	2se	$^{207}Pb/^{206}Pb$	2se
BJ-01	**Vessel's Spout**	38.7051	0.0022	15.6788	0.0007	18.5521	0.0008	2.08631	0.00004	0.84513	0.00001
BJ-02	**Vessel**	38.6872	0.0020	15.6744	0.0006	18.5402	0.0007	2.08670	0.00003	0.84541	0.00001
BJ-03	**Pin**	38.6224	0.0026	15.6342	0.0010	18.5657	0.0009	2.08039	0.00003	0.84212	0.00001
BJ-04	**Vessel**	39.0719	0.0021	15.7046	0.0007	18.9724	0.0008	2.05938	0.00005	0.82775	0.00001
BJ-06	**Pin**	38.6567	0.0035	15.6352	0.0006	18.5673	0.0007	2.08202	0.00013	0.84208	0.00001
BJ-07	**Button**	39.3405	0.0033	15.7323	0.0010	18.9449	0.0010	2.07658	0.00006	0.83042	0.00001
BJ-08	**Button**	38.9372	0.0028	15.6929	0.0010	18.6662	0.0010	2.08589	0.00004	0.84069	0.00001
BJ-09	**Spring Bead**	38.7145	0.0022	15.6338	0.0008	18.7385	0.0008	2.06600	0.00003	0.83430	0.00001
BJ-12	**Vessel**	38.6628	0.0021	15.6340	0.0008	18.5497	0.0009	2.08429	0.00003	0.84282	0.00001
BJ-13	**Vessel**	38.7091	0.0025	15.6262	0.0009	18.6231	0.0009	2.07857	0.00004	0.83908	0.00001
BJ-14	**Vessel**	39.0133	0.0029	15.7060	0.0010	18.9054	0.0010	2.06363	0.00003	0.83077	0.00001
BJ-15	**Vessel**	38.8030	0.0021	15.6646	0.0006	18.6849	0.0008	2.07674	0.00003	0.83836	0.00001
BJ-17	**Button**	39.1195	0.0020	15.6906	0.0007	18.8993	0.0008	2.06990	0.00003	0.83022	0.00001
BJ-19	**Vessel**	39.0094	0.0023	15.7049	0.0008	18.9041	0.0009	2.06357	0.00003	0.83077	0.00001
BJ-20	**Vessel**	38.7459	0.0021	15.6274	0.0007	18.6853	0.0009	2.07365	0.00003	0.83635	0.00001

Table-2

Table 2. Presenting elemental concentration averages, standard deviations, relative standard deviations for all 15 objects and within selected objects having similar isotopic composition. Elemental data used for calculations are reported in Oudbashi and Hasanpour 2018. Values in wt%.

	Ag	As	Co	Cu	Fe	Ni	Pb	Sb	Sn	Zn
All 15 samples										
Average	0.03	0.27	0.02	89.36	0.20	0.06	0.15	0.06	9.12	0.09
Std dev	0.03	0.16	0.01	1.68	0.22	0.04	0.14	0.07	1.82	0.02
RSD	**89**	**60**	**52**	**2**	**111**	**72**	**96**	**113**	**20**	**17**
BJ-01 and BJ-02										
Average	0.03	0.18	0.02	89.69	0.08	0.08	0.19	0.04	8.73	0.09
Std dev	-	0.11	0.01	2.90	0.03	0.07	0.16	0.03	3.36	0.01
RSD	**-**	**63**	**47**	**3**	**35**	**88**	**82**	**71**	**38**	**8**
BJ-14 and BJ-19										
Average	0.08	0.21	0.01	88.93	0.08	0.07	0.25	0.04	9.74	0.08
Std dev	0.00	0.01	0.00	0.57	0.01	0.01	0.04	0.00	0.50	0.00
RSD	**0**	**7**	**0**	**1**	**9**	**11**	**17**	**0**	**5**	**0**
BJ-03 and BJ-06										
Average	0.01	0.49	0.03	87.94	0.17	0.02	0.02	0.08	10.34	0.09
Std dev	0.00	0.01	0.00	0.18	0.01	0.00	0.00	0.00	0.04	0.00
RSD	**0.0**	**3**	**0.0**	**0.2**	**4.3**	**0.0**	**0.0**	**0.0**	**0.4**	**0.0**
BJ-03, BJ-06 and BJ-12										
Average	0.02	0.34	0.03	87.66	0.32	0.02	0.06	0.06	10.61	0.11
Std dev	0.01	0.26	0.01	0.49	0.27	0.01	0.08	0.04	0.47	0.03
RSD	**69**	**76**	**22**	**1**	**84**	**25**	**119**	**71**	**4**	**27**

Table 3. References for lead isotopic data used for comparison: main geological zones in Iran and ore deposits/locations, and sites and type of material/composition.

Geological zone	**Ore/deposit/location**	**References**
Sanandaj-Sirjan zone	Deh Hosein, Astaneh, Nezam Abad	Nezafati 2006
Urumieh-Dokhtar zone	Kerman Cenozoic magmatic arc, Karkas, Veshnaveh, Angouran	Chegini et al. 2000; Shafiei 2010; Pernicka et al. 2011; Nezafati and Stöllner 2017; Isotrace Oxford
Central Iran Zone	Anarak Area	Pernicka et al. 2011
Lut block	Chehel Kureh, Qaleh Zari	Hauptmann et al. 2003
Site/location	**Material and/or composition**	**References**
Baiazeh	Slag	Isotrace Oxford
Arisman	Slag, Cu prill	Chegini et al. 2000; Pernicka et al. 2011
Pusht-i Kuh (Luristan)	Objects: Cu, CuPbSn, CuPb, CuSn, Sn, CuAs, CuPbAs,	Begemann et al. 2008
Tappeh Sialk	Objects, slag	Chegini et al. 2000 ; Nezafati et al. 2008
Tappeh Sarm	Metals	Nezafati and Stöllner 2017
Qoli Darvish	Metal, slag furnace lining, smelting slag, Cu-rich slag	Nezafati and Stöllner 2017