THE 'TIN PROBLEM' IN THE PREHISTORIC NEAR EAST – FURTHER INSIGHTS

FROM A STUDY OF CHEMICAL DATASETS ON COPPER ALLOYS FROM IRAN

AND MESOPOTAMIA.

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Abstract

A great number of ancient bronzes have been recovered in the Near-East, but the source of

the tin used for their production remains elusive. This paper proposes new insights into the

'tin problem' by using a 'big data' approach. Chemical analyses of over 5000 Bronze Age

and Iron Age copper alloys from Iran and neighbouring regions have been assembled. By

interrogating this dataset within a suitable framework we can now show that different

systems of exchange existed, where tin was added at different stages in the process, and we

highlight the likely importance of tin sources in western Iran during the Iron Age.

Keywords

Tin, bronze, Iran, Mesopotamia, metal trade

I. Introduction

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Whilst the appearance of tin bronze is considered to be a major technological innovation in the ancient world, the scarcity of tin finds and the relative rarity of known tin deposits have resulted in much scholarly speculation on what has been branded "the tin problem": the difficulty in locating the source of the tin used to make bronze in the ancient Near-East¹. Expeditions in the 1970s by Cleuziou and Berthoud revealed the presence of several tin deposits in Afghanistan², compatible with textual evidence from Bronze Age Mesopotamia indicating that tin came from the east³. The relative proximity in Afghanistan of tin with sources of lapis lazuli and gold, two materials also highly sought after by the Mesopotamian elite, subsequently lead to a general acceptance of Afghanistan as one of the most plausible sources for Near-Eastern tin⁴. However the discovery of a new potential tin source in western Iran⁵ and the publication of new datasets in the last decade have started to challenge this view⁶.

In order to address this problem, we have taken a 'big data' approach, assembling all the available datasets of chemical compositions of Bronze and Iron Age Iranian and Mesopotamian copper-base objects into a single database. With a total of 5524 objects of (reasonably) known provenance for which the tin-content has been measured, it is possible to study the chemical dataset as a body of evidence in its own right rather than as a mere support or illustration for textual, geological or archaeological evidence. Indeed, geological and mining data on the one hand and archaeological and textual evidence on the other undeniably hold crucial information for the study of ancient metal, the former notably on the provenance of the metal, the scale and technology of the extraction, the latter including information on trade arrangements as well as on the social context of metal production, use and deposition. However we argue that the metal in itself, through its chemistry, lead isotopy and microstructure, holds an additional (though obviously related) set of information that has often been overlooked in the past: it can help us identify geographical and chronological

patterns of metal occurrence and gives us indications on metalworking technology, metal movement, mixing and recycling. In this paper we choose to focus on this set of evidence and more precisely on the chemical data as we have not yet been able to assemble a big enough set of lead isotope or metallographic data. Further work will nevertheless be needed to reconcile the findings presented here with the other types of evidence discussed above. In this paper we merely propose an introduction to this method and seek to demonstrate how it can help start to provide new information on the main questions surrounding the use of tin in Mesopotamia and Iran:

- 1) How did the tin travel? Was it in ingot form, as cassiterite (tin oxide), or pre-alloyed as tin-bronze?
- 2) Where did the tin come from?
- 3) Why did tin-bronze come to replace un-alloyed copper and arsenical copper?

II. Method

II.1 Available analytical data

The data collected for this project are taken from published excavation reports, private and museum collections catalogues and scientific publications, listed in Table 1. For Iran, the chemical compositions of 2143 different artefacts, from 38 different publications, were included in the database. Data from a number of neighbouring countries was also added in order to include possible source areas for copper and tin, and major trading networks. For these regions, which include modern Iraq, Oman, United Arab Emirates, Pakistan, Afghanistan and Uzbekistan, the chemistry of an additional 3381 artefacts from a further 16 studies was collected.

The publications vary in the number of objects analysed, the region and time-period they focus on, and the analytical techniques used (Table 1). As a result, the dataset is very fragmented and it is crucial to define the best possible geographical and chronological framework to overcome this and to assess the extent to which the results can be used in a single dataset. This was rendered difficult by problems in the chronology of some Iranian sites, and objects of uncertain provenance. The objects known as the "Luristan bronzes" were particularly problematic: the term refers to ornate bronze objects of a type that started to appear on the art market in the 1920s. These are often looted items for which it is impossible to ascertain an exact provenance. Although the chemical data for these bronzes were added to the database, we have chosen here to run the analysis without any of the unprovenanced objects. Only Luristan bronze recovered on excavations were therefore taken into account in the preparation of the graphs and tables below. For all provenanced objects, the dating proposed in the original publications has been used, as a detailed reassessment of site chronologies is now largely impossible.

	Number of objects	Analytical technique		
Iranian sites				
Halm 1935	4	nm	Tepe Giyan, Western Iran	
Riesch and Horton 1937	11	nm	Tepe Hissar, Gorgan	
Desch 1938	7 (0)	nm	Unprovenanced, Luristan	
Halm 1939	1	nm	Tepe Sialk, Central Iran	
Burton Brown 1951	21	OES	Geoy Tepe, Azerbaijan	
Birmingham 1963	18 (0)	OES	Unprovenanced, Luristan	
Dono 1965	6	nm	Ghalekuti, Amlash	
Moorey 1971	127 (0)	OES	Unprovenanced, North-Western Iran	
Muscarella 1974	4 (0)	OES	Various Iranian sites	
Vatandoost-Haghighi 1977	140 (137)	AAS	Various Iranian sites	
Berthoud and Françaix 1980	34	OES + SSMS	Various Iranian sites	
Heskel 1981	37	OES	Various Iranian sites	
Tylecote and McKerrell 1986	4	XRF	Tepe Yahya, South-East	
Malfoy and Menu 1987	436	OES	Susa, Khuzistan	
Muscarella 1988	99 (30)	ICP-AES	Various Iranian sites	
Pigott 1989	2	PIXE	Hasanlu, Azerbaijan	
Hopp et al. 1992	85 (3)	nm	Various Iranian sites	
Gunter and Jett 1992	3 (0)	EDXRF	Unprovenanced, Iran	
Riederer 1992	44 (1)	nm	Unprovenanced, North-Western Iran	
Mahboubian 1997	119 (0)	EMP	Various Iranian sites	

Hakemi 1997	5	XRF?	Shahdad, South-East	
Vatandoust 1999	30 (24)	ICP-AES + SEM- EDS	Various Iranian sites	
Thornton et al. 2002	102	ICP-MS + EMP	Tepe Yahya, South-East	
Pigott et al. 2003	30	PIXE	Tal-e Malyan, Fars	
Pigott et al. 2003	11	PIXE	Tal-eMalyan, Fars	
Krause 2003	77 (55)	OES	Hasanlu, Azerbaijan; Susa, Khuzistan; Luristan	
Hauptmann et al. 2003 (republication of Helmig 1986)	21	AAS	Shahr-i Sokhta, South-East	
Fleming et al. 2005	16	PIXE	Various cemeteries, Luristan	
Fleming et al. 2006	41	PIXE	War Kabud, Luristan	
Nezafati 2006	29 (3)	EDXRF	Unprovenanced, Luristan	
Begemann et al. 2008	102 (54)	ICP-OES + NAA	Pusht-i Kuh area + unprovenanced, Luristan	
Meier 2008	19	EDXRF	Shahdad, South-East	
Nezafati et al. 2008	4	EDXRF	Tepe Sialk, Central Iran	
Thornton 2009 (re-publication)	229	OES EMP AAS PIXE	Tepe Hissar, Gorgan	
Frame 2010	60	WDS	Godin Tepe, Western Iran	
Vatandoust et al. 2011	28	EDXRF	Arisman, Central Iran	
Fleming et al. 2011	113	PIXE	Hasanlu, Luristan	
Oudbashi et al. 2012	20	SEM-EDS	Sangtarashan, Luristan	
Total Iranian sites	2139 (1562)			

Neighbouring regions				
Caley 1972	2	nm	m Darra-i Kur, Bactria	
Moorey 1985 (re-publication)	127	nm + AAS + XRF	Various sites, Mesopotamia	
Craddock 1985	30	AAS	Ras al-Khaimah, Oman Peninsula	
Hauptmann et al. 1988	43	ICP-AES + AAS	Various sites, Oman Peninsula	
Weeks 1997	118	SEM-EDS	Tell Abraq, Oman Peninsula	
Kenoyer and Miller 1999 (republication)	53	nm	Various sites, Indus Valley	
Weeks 2000	20	PIXE	Sharm tomb, Oman Peninsula	
Prange 2001	207	ICP-AES	Various sites, Oman Peninsula	
Prange and Hauptmann 2001	86	ICP-AES	Selme hoard, Oman Peninsula	
Weeks 2003	83	PIXE	Various sites, Oman Peninsula	
Krause 2003	158	OES	Tepe Gawra, Assyria; Barbar, Bahrain; Ur, Mesopotamia	
Craddock et al. 2003	20	AAS Ra's al Hadd, Oman Peninsula		
Hauptmann and Pernicka 2004	2068	XRF + NAA Various sites, Mesopotamia		
Kaniuth 2006	193	nm	Dzarkutan and Sapalli Tepe, Bactria	
Ponting 2013	112	ICP-OES	Nimrud, Assyria	
Goy et al. 2013	61	ICP-AES	Masafi Area, Oman Peninsula	
Total neighbouring regions	3381			
Total all regions	5524 (4943)			

Table 1. Publications containing chemical datasets for copper-based artefacts from Iran and neighbouring regions. The numbers in brackets are the numbers of objects of known provenance. nm = analytical technique not mentioned.

II.2 Geographical framework

The 81 sites for which compositional analyses were collected have been grouped into 14 regions as shown in Figure 1.

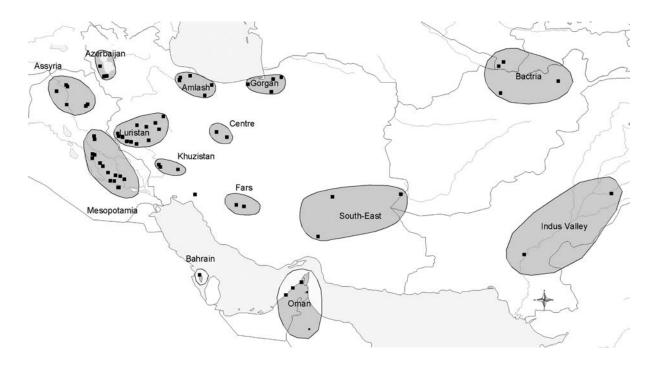


Figure 1. Map showing the regional divide and the main sites used in this study

The dearth of data for the Gorgan sites meant that both Gorgan and Tepe Hissar had to be grouped under one region, labelled here as Gorgan. This grouping is not fully deprived of archaeological sense as highland and lowland settlements are thought to have been in contact on a local scale. Thornton in particular noted that Tepe Hissar was closely connected to the sites of the Gorgan plain despite the Elburz Mountain range between them⁷. It should also be noted that the region labelled 'Oman' here represents the Oman Peninsula, grouping both modern Oman and the United Arab Emirates.

II.3 Chronological framework

Two chronological scales were used for the analysis. The first is a simple division into thirds of a millennium (e.g., early, mid and late 2nd millennium). Each object was attributed to one of these time periods based on the dating provided in the original publication. For objects at the interface of two time periods, the earlier one was arbitrarily chosen for the sake of consistency. The time ranges given in the original publications only rarely spanned less than 300 years, meaning that defining a tighter chronology would have been unwise. A division into thirds of a millennium seems to give a coherent chronology, but is far from perfect. Indeed, there are a number of objects that have been attributed to a time period which could actually have been deposited up to two thirds of a millennium earlier or a full millennium later. Because of these chronological problems, and in order to have more data points for each region, a second, less detailed, scale was used. Period 1 and Period 2 were therefore defined as follows: Period 1, from the early 3rd to mid-2nd millennium inclusive, is what can very roughly be called the Bronze Age, and Period 2, from the late 2nd to the mid-1st, approximately corresponds to the Iron Age. A comparatively small amount of data was available for the 2nd millennium BC, which limits the number of objects that might have been misattributed to either of these two blocks. Unfortunately, not all regions are well represented in each time period and the results presented below systematically filter out regions with less than five objects for a given time period.

II.4 Working with legacy data

Inevitably, the 54 studies used here, conducted over 80 years, used a wide range of experimental methods, reflecting the broader evolution of archaeometry. The precision, accuracy and detection limits of the measurements will vary from one technique to another. Measurements obtained by a single technique can also be affected by differences in the

calibration, by the lack of homogeneity within an object, and by different degrees of corrosion in each object. How then is it possible to directly compare the data from all of these studies, especially when the exact experimental procedures are very rarely published? To minimise these problems, three principles were applied:

Firstly, it was decided not to focus on the composition of individual artefacts, but rather look only at large groups of artefacts. This limits potential problems in the precision (poor reproducibility under fixed conditions) of a technique. Indeed, the variations in precision will tend to average out over a large sample size. Similarly, variations due to the possible inhomogeneity of the objects will also tend to average out.

Secondly, the simple question of the presence or absence of an element was used in many cases. This enabled the use of data that can only be considered as semi-quantitative (or even qualitative), whether as a result of the analytical technique used, or because of other problems such as inhomogeneity or corrosion. The latter was notably observed in objects from Tell Abraq where the very high tin contents were interpreted as the result of a preferential corrosion of the copper due to the acidic soil, and therefore only considered as qualitative information on the presence of tin bronze at the site⁸. The question of presence or absence can also help overcome variations in accuracy (how close a measurement is to the 'true' value) between different techniques. Assessing the accuracy of each publication was rendered impossible because, in a great majority of cases, no information on standards was published. Given that most studies are limited to a particular region and time-period, there is a risk, when comparing these groups of objects, of picking up variations related to offsets between techniques instead of true variations between the different assemblages. It must be noted that in the few cases where groups of artefacts were analysed by more than one technique⁹, the techniques showed a general agreement. While there were some outliers where the two measurements were drastically different (again emphasizing the importance of not

commenting on isolated values), these cases were rare and for tin the different techniques agreed on the question of presence or absence (as defined below) in 100% of cases.

A potential issue with the simple question of presence or absence is the arbitrary nature of the choice of cut-off value, and the sensitivity of this boundary, which brings us to another muchdiscussed question surrounding bronze: at what tin percentage can an object be defined as a tin-bronze? A tin bronze, it could be argued, should be an object in which tin has been deliberately added as opposed to being present accidentally 10. Traditionally scholars have used 1 or 2% as a cut-off between copper and bronze. To choose a limit, one can indeed consider how much tin is likely to appear as a trace in a finished object given the composition of copper ores. Cleuziou and Berthoud¹¹ chose 0.5%, arguing that copper ore from that area is rarely associated with significant amount of tin. Another approach would be to calculate the amount of tin necessary to show a significant modification to the hardness or colour of the objects, but this is potentially blurred by re-melting, mixing and recycling. Should objects obtained by mixing tin-bronze with copper qualify as "deliberate" tin bronzes? An arbitrary limit of 2% is chosen here to distinguish what we call "tin-less copper" from "tin-bronze". Since objects with between 0.5 and 2% tin represent 7% of the total dataset, which is not insignificant, we did however test the sensitivity of the analysis to the choice of cut-off, as discussed below.

Finally, the third principle applied here was to prefer elemental distribution graphs (see for example Figures 2 and 3) to mean values or medians whenever possible. Not only are graphs less likely to lead to overstating the case of small numerical differences that can derive from technique-related discrepancies, but they also tell us more about the metallurgy of the area in question. As shown below, the shape of a distribution of the tin content and in particular whether it is skewed or presents several modes can give information on metallurgical practices.

III. How did the tin travel? At what stage was it added to copper?

Before we address the thorny question of the origin of the tin, we show how the chemical dataset can shed some light on the form in which tin was transported from the mining area to the consumers, and at what stage it was added to copper. Several possibilities can be envisaged, including as tin ore, as tin ingot, or as pre-alloyed metal. It can be exchanged and transported directly in ore form, as cassiterite or stannite. This possibility was suggested by Charles, in a study aimed at considering the reasons behind the scarcity of tin finds ¹². It can also be made into tin ingots near the mining area and traded as such. In both cases, the tin is added to the copper at a late stage of its trade, and the process of alloying, and therefore the quantity of tin added, are controlled by the consuming society. But tin can also be traded in alloyed form: in bronze ingots or in finished objects, either used as such, or altered. In these cases, at least part of the tin in the finished objects in the consumer region has been added to copper at an early stage and is not entirely controlled by the metal-smiths of this region. It is also possible to envisage a trade in master alloys of bronze, with a very high tin content.

The question of how tin travelled remains largely unanswered. Here we show how using a simple classification of the type of copper in circulation can help to elucidate patterns of tin flow. Following the method proposed by Bray¹³, the dataset was divided into copper metal groups based solely on the presence or absence of arsenic, antimony, silver and nickel in the objects. Here we focus only on two of these groups: one named 'As Only' with elevated amounts of arsenic (>=0.1%), but low antimony, silver or nickel (<0.1%), and one, named 'AsSbNi', with elevated amounts of arsenic, antimony and nickel (>=0.1%), but little to no silver (<0.1%). When looking at the percentage of objects represented by these groups in each region for "Period 1" (roughly Bronze Age), 'As Only' metal occurs most prominently

along the Caspian Sea, in south-eastern Iran and in Bactria where it represents about a third or more of the assemblages, whilst it is less common in the west and in the regions bordering the Persian Gulf (see Table 2). 'AsSbNi' on the other hand represents about a third of the assemblage in Assyria, but is almost entirely absent in eastern Iran and quite rare along the Persian Gulf. From this it can be postulated that the former is more likely to have come from one or several sources on the Iranian Plateau or further to the north or east, while the latter is more likely to originate somewhere in the west or north west or, potentially, Central Asia. It is important to note that these metal groups are not meant to equate to metal from a single copper source but are simply designed as a tool to start teasing out various potential provenances for copper. These copper sources are not the subject of this paper and are not discussed, but one can refer to Pigott¹⁴ for a review of potential copper sources in greater Iran and Vatandoust¹⁵ for a comprehensive map of Iranian copper deposits.

	% As Only	% AsSbNi	Number of
			objects analysed
Azerbaijan	29	6	17
Amlash	32	0	25
Gorgan	33	1	138
Luristan	16	5	103
Khuzistan	6	22	220
Fars	0	0	10
South-East	40	1	129
Centre	i.d.	i.d.	1
Indus Valley	8	0	13
Bactria	35	24	17
Oman	13	13	142
Bahrain	7	4	75
Mesopotamia	14	17	1814
Assyria	8	32	223

Table 2. Percentage of the assemblage represented by 'As Only' and 'AsSbNi' metal for "Period 1" in the regions defined in Figure 1. i.d. = insufficient data.

The distribution of the tin content for bronze objects (>2% Sn) of these two metal groups in Mesopotamia is shown in Figure 2. Two very different profiles can be seen: for the 'As Only'

group, the tin content presents an almost symmetrical unimodal distribution with a mode around 8-10 %. For the 'AsSbNi' group, the tin contents show a very different pattern: the distribution is skewed towards much lower values (2 to 5% Sn). The 'As Only' distribution is likely to indicate that tin has been added at a late stage in the life of these objects, with little subsequent mixing, re-melting or secondary alloying occurring between this alloying event and the deposition of the object, as these processes would result in a wider range of tin compositions. The addition of tin at a late stage in the life of this copper could mean either that Mesopotamia imported copper and tin separately and alloyed the metal once there, or that they imported a fairly standard bronze and did not alter it after import. If the 'As Only' type copper comes from the eastern Iranian Plateau, then the second possibility seems rather unlikely, as eastern Iran appears to use very little tin in this period (see below). If on the contrary it comes from further east, Bactria or Central Asia for example, where this copper type is also common and where tin mines have been discovered, then the import of prealloyed bronze is a definite possibility. However, the existence of Mesopotamian texts with precise recipes for the alloying of copper and tin¹⁶, the documented trade of tin reaching Mari from Elam in ingot form in the first half of the 2nd millennium BC¹⁷ and the absence of recovered bronze ingots until a later period (see below), might support a separate trade in copper and tin.

Moreover, although uncommon, metallic tin objects and ingots are not unknown in the Mediterranean and the Near East¹⁸. The most striking examples are the tin ingots discovered in shipwrecks in the eastern Mediterranean representing a total of over a tonne of tin and dated to the second half of the 2nd millennium BC¹⁹. As for objects, a tin bracelet was found in Thermi IV and a ring and a flask were recovered in Egypt and dated to the 14th century BC²⁰. Closer to our area, five bracelets and two rings from the 19th century BC were found in Tell ed-Der²¹. In Kültepe, where tin was a common trading item between Assyrians and

Anatolians in the early 2nd millennium, three flat circular objects might have been tin ingots, but this identification has not been verified by chemical analysis²². Finally, Weeks reports a tin ring from Tell Abraq in Oman dated to the late 3rd millennium BC²³. The scarcity to this day of tin-ingot finds in Mesopotamia and Iran, especially in the 3rd millennium might reflect the fact that tin was too valuable to be left behind, or perhaps that tin was mostly traded as cassiterite, which has perhaps not been systematically identified or reported in excavation reports.

Ingots of copper alloy occur with a higher frequency in the Near-East and some have been subjected to chemical analysis. Of the 70 analysed ingots found in Susa, Tepe Hissar, Shahr-i Sokhta, Oman and Bahrain, most of which are bun-shaped, and dating to the mid or late 3rd millennium BC, none contain tin in more than trace amounts. The earliest evidence of bronze stock in the region comprises of three bars found in Tal-e Malyan from the late 3rd or early 2nd millennium BC²⁴. The presence of bronze at the site in this period can potentially be explained by the fact that as an Elamite capital it was closely linked with Susa, which is known to have traded tin in the early 2nd millennium BC²⁵. Bronze ingots, bar-, plate- and bun-shaped are found only later on in Hasanlu (ca. 800 BC)²⁶. However, even at the beginning of the 1st millennium BC, tin-less copper ingots are still common as attested by the 27 ingots analysed from Masafi (U.A.E) and Nimrud (Assyria) of which all but one have been found to have less than 2% tin²⁷.

This evidence therefore points towards a trade of unalloyed copper and tin separately in the 3rd millennium, and possibly the 2nd millennium BC if the evidence from the eastern Mediterranean shipwrecks is representative of the trade further east.

The Mesopotamian recipes, mentioned above, indicate ratios of copper to tin of six to one from the pre-Sargonite period to the Neo-Babylonian Period which translates to about 14.3%

tin. A ratio of seven to one (12.5% Sn) is also known from the Ur III period²⁸. This percentage is higher than the maximum of the distribution in Figure 2. Interestingly, 24th century BC recipes from Ebla in western Syria indicate a wide range of tin contents from 0.83 to 21.55% Sn, but the very low and very high values are rare and the vast majority of objects are produced with between 11 and 17% tin²⁹, which is still high compared to the measured tin contents seen in Figure 2. However, these recipes specify the initial amount of tin added into the mix, not the tin in the finished object and thus do not take into account oxidative loss. It is also conceivable that the recipes in fact indicate an amount of cassiterite, mistranslated as tin, which could explain the lower measured tin contents in the artefacts. Indeed, after much discussion on the meaning of the Sumerian word AN-NA (akkadian anāku) and specifically on whether it should be translated as tin, lead or arsenic³⁰, it is now generally accepted that the term refers to tin. However there has been little debate on whether it could in fact in some cases designate the tin ore cassiterite, which would indicate that tinbronze was produced by a cementation process rather than by the addition of solid tin. While Muhly did reflect on the question he offered no definite answer: in 1973 he pointed out that the equation of AN-NA to the Hittite *dankui-, derived from an adjective meaning dark, may point to the use of the dark tinstone that is cassiterite rather than to the grey-whitish tinmetal³¹. In 1985 however, he argued that the production of tin-bronze by cementation was very unlikely given how difficult it would have been to control³². An interesting direction for further research would therefore be a reassessment of the production processes of tin-bronze in order to explain the discrepancies between recipes and analytical results and clarify the meaning of the AN-NA and anāku.

The very different profile for the tin content of the bronzes of the AsNiSb group (Figure 2), for which the copper is more likely to have come from the west or north-west or from Central Asia, probably indicates that much has happened to the copper between a primary alloying

event and deposition. Indeed, given the known Mesopotamian recipes, it is difficult to imagine that such a low tin content would have been purposefully added to the copper. We can imagine that this copper was not the subject of an organised trade but reached Mesopotamia as copper and pre-alloyed bronze objects perhaps as down-the-line trade or in events of booty-taking³³ and was subsequently treated as scrap material and re-cast to suit the Mesopotamain taste. Another possibility to explain this type of distribution might be the smelting of mixed copper tin stannite ore known to have been present at Mushiston for example³⁴ and that would have resulted in the production of bronze of uncontrolled tin content. Though some work has been done recently on the use of this type of ores in the Balkans³⁵, the range and profile for the tin content to be expected by the smelting of mixed stannite ores is still unclear and merits more attention.

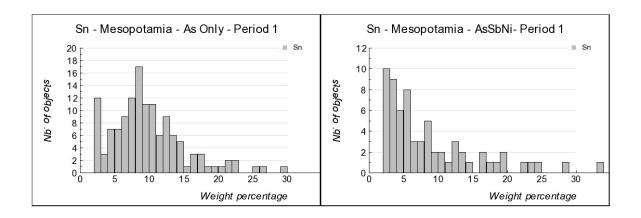


Figure 2. Tin distribution in bronze objects (>2% Sn) of the 'As Only' and 'AsSbNi' groups for Period 1 (early 3rd to mid-2nd millennium inclusive) in Mesopotamia.

IV. The provenance of tin

The question of the provenance of the tin used in ancient bronzes has long been, and remains today the subject of much academic research. Promising advances are for example currently being made in the field of tin isotopy³⁶. However much work remains to be done on

characterising the variability of isotope ratios within and between tin sources, and on the suitability of this method for objects in which tin from different sources might have contributed to the bronze. It is hoped that this approach will soon add a useful dataset that can be folded into the broader tin discussion. At present however, there remains no clear understanding of the provenance of the tin for Near-Eastern bronzes. It is not the purpose of this paper to review once again the geological, textual and archaeological evidence of potential tin sources for the Near-East, since very comprehensive recent reviews have been written by Stech and Pigott, Weeks and Helwing and Pigott³⁷. Instead, after a very brief summary of the potential sources, we attempt to show how a large dataset of compositional analysis can add to this picture.

Wertime and colleagues found cassiterite mines in Egypt in close proximity to 22nd century BC inscriptions³⁸, while Cleuziou and Berthoud found tin deposits in Afghanistan³⁹. Since then, more substantial evidence for tin mining was recovered: the site of Kestel/Göltepe in the Taurus Mountains of southern Turkey was suggested as a viable source of cassiterite ore in the 3rd millennium BC by Yener and Laughlin⁴⁰. Recently, another 3rd millennium tin source was identified in the Kayseri Province, also in Anatolia, not far from Kültepe, which interestingly is known to have bought tin from Assyria in the early 2nd millennium BC⁴¹. Tin mines have also been found in the region of the Zeravshan valley (modern Uzbekistan and Tajikistan) where the sites of Karnab, Lapas, Changali and Mushiston provide evidence of the mining of tin mainly during the Andronovo-Tazabag' jab period in the first half of the 2nd millennium BC, but which possibly started as early as the second half of the 3rd millennium BC at Mushiston and extended to the late 2nd or early 1st millennium BC at Karnab⁴². The production at these sites is thought to have exceeded local needs. Finally the recently discovered tin and copper mine of Deh Hosein on the eastern border of Luristan in western

Iran exploited from the mid-2nd millennium to the early 1st, if not earlier, has been presented as an important new clue in the provenance of tin by Nezafati and colleagues⁴³.

Here a study of the tin content in "Period 2" (late 2nd to the mid-1st mill. BC) objects is given as an example of how interrogating a large dataset of analyses can contribute to the debate about the source of tin. Figure 3 shows the tin distribution profiles for the regions for which more than 20 objects have been analysed for "Period 2". In the three regions of western Iran (Azerbaijan, Amlash and Luristan), tin-less copper is very rare and the distribution of tin content is quite similar: a main peak at 8-10% Sn and a secondary one around 4% Sn which can provisionally be interpreted as the result of mixing of copper and bronze. In Assyria, the tin distribution is very similar to western Iran, but with a higher proportion of tin-less copper objects (about 20%). The tin distribution profile in Mesopotamia is very different: tin-less copper represents about half of the assemblage (46%) and the tin bronzes appear to have either higher or lower tin contents with the 8-10% range being a minimum of the distribution instead of a maximum. This possibly indicates that tin was difficult to obtain in Mesopotamia in that period and that the tin bronzes that are present are mostly the result of mixing and heavy recycling. From this evidence, it is tempting to exclude a Persian Gulf trade of tin in this period, as Mesopotamia would have been likely to be at the receiving end of such a trade. If tin was one of the commodities traded through the Gulf by Harappa, which seems quite likely although there is at present no firm evidence, this lack of tin in Mesopotamia in Period 2 would be consistent with the fact that the Harappan civilisation collapsed in the beginning of the 2nd millennium BC, which resulted in an interruption of the Persian Gulf trade.

The Mesopotamian profile contrasts greatly with western Iran where, on the contrary, it would appear that tin was in constant supply and these distribution profiles can be interpreted as primary production profiles. Based on these observations, an attractive explanation for this would be the presence of one or several sources in western Iran or Anatolia. This would be

compatible with Muhly's interpretation based on the phrasing of ancient texts, stating that tin came overland from the Zagros⁴⁴. Of course the discovery of Deh Hosein has now come as a solid proof for the presence of tin in this region and the question that follows is whether this source, would have provided enough tin to support the western Iranian bronze industry. The evidence available at present reveals an undeniably important mining complex at Deh Hosein: more than 75 big open depressions, each bordered by waste dumps occur in an area of several square kilometres. In the vicinity of the depressions were found two small settlements or workshops presenting evidence of ore-processing. Moreover, a small program of lead isotope analysis has shown a good match between Deh Hosein ores and a set of 'Luristan' bronzes⁴⁵. This site therefore appears to have been of importance in the metallurgy of the region between the mid-2nd and the early 1st millennium BC, but it remains unclear how much tin (as opposed to copper or other minerals) it would have been able to produce. For example there is at present no firm evidence of beneficiation of the ore for its tin content. However this doesn't necessarily mean that tin wasn't recovered altogether, as we can envisage an indiscriminate smelting of copper and tin minerals and an export in an "uncontrolled" bronze, or a recovery of cassiterite in nugget form in streams as has also been reported to exist in the area⁴⁶. It seems possible, as suggested by Nezafati, that other such yet undiscovered sources are present in the Sanandaj-Sirjan zone which would help explain the distributions observed in Figure 3 and indeed, the Nezam Abad prospect about 12 km from Deh Hosein has already revealed tin and copper and some ancient workings⁴⁷. There have also been claims of tin near Tabriz in Iranian Azerbaijan⁴⁸, but the Geological Service of Iran was not able to verify its existence⁴⁹. Tin sources in western Iran could also have provided tin for Assyria. Indeed, one can easily envisage tin from the Zagros being traded along the Lower and Greater Zab Rivers to Assur in Assyria. Of course, another explanation for the tin patterns observed in Figure 3 could be that the tin present in western Iran in this period came

overland from further east, from Karnab for example, where the production might have continued into the late 2nd or early 1st millennium BC⁵⁰, but more data from the eastern part of our study area in this time period would be crucial in answering this question. If this was the case, the trade seemingly stopped before it reached Mesopotamia.

Perhaps is it also time to reopen the question about tin sources in the Caucasus, as the presence of tin in this region could help explain what seems to be a well-developed tinbronze industry in Iranian Azerbaijan in particular (Figure 3). While debates in the 1970s discussed the likelihood of this area being a provenance for the tin used in Mesopotamia and Iran⁵¹, it is surprisingly rarely cited in more recent reviews on the subject. Interestingly though, the Caucasus presents a comparatively high number of metallic tin or tin-lead objects⁵² and relatively early tin-bronzes (late 4th to early 3rd millennium BC at Talin and early to mid-3rd millennium BC at Velikent)⁵³. Evidence for workable tin deposits in the Caucasus remains however a subject of speculation: in his exhaustive review on mineral resources in the Caucasus Courcier explains that although the Caucasus is generally characterised by basic and ultra-basic rocks unfavourable to tin mineralisations, there exists two zones where tin is mineralised, but these haven't yet been the focus of any archaeological or geological research to assess the tin content of the minerals⁵⁴. In the light of the prevalence of tin-bronze in north-western Iran and of the evidence presented above, it seems that the presence of a workable tin source in the Caucasus remains a question worth consideration. The fact that no tin mines have been discovered to date may reflect the exploitation of relatively small sources in ancient times and questions the model of wide-scale tin trade versus the use of relatively small local deposits, at least in the Iron Age.

The tin profile in Oman is also intriguing. There, tin-less copper and tin-bronze are present in proportion of about 40% tin-less copper to 60% tin-bronze. The bronze objects fall in a strikingly narrow range of tin content. They are almost all bangles dated to c. 1100-600 BC

from the Selme hoard, which is thought to have been of indigenous origin⁵⁵. This indicates that, at least occasionally, tin was available in Oman in that period. The question of where this tin might have come from is an interesting one. There were clear contacts between Oman and Iran during the Iron Age as attested for example by the presence there of bridge-spouted vessels of Iranian style, of which some may have been direct imports⁵⁶. Could tin have been traded from western Iran to Oman? Or was tin still traded through the eastern part of the Gulf in the Iron Age? At this stage both possibilities are conceivable and data from the Indus Valley, Afghanistan and Bactria for this period would greatly help to bring light to this problem.

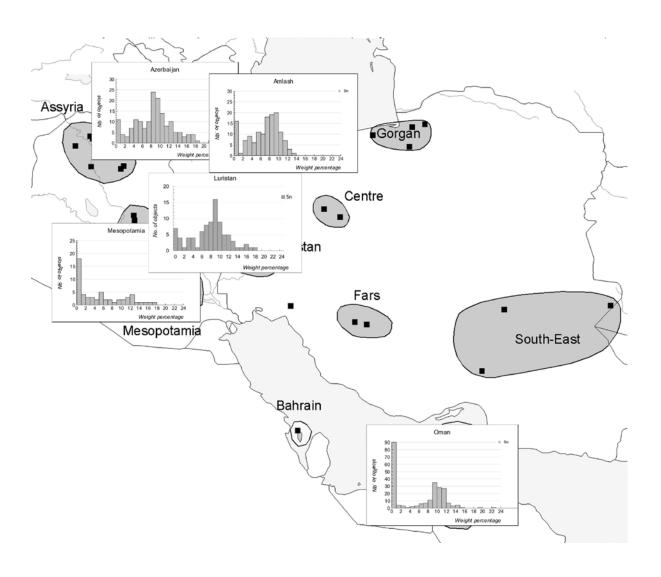


Figure 3. Distribution of the tin concentration in Azerbaijan, Amlash, Luristan, Mesopotamia and Oman between the late 2nd and mid-1st millennium BC.

V. Why did tin bronze replace un-alloyed copper and arsenical copper?

Figure 4 shows the evolution of the ubiquity of tin bronze in Iran as calculated using three different cut-off values for the presence/absence of tin. The general trend – tin-bronze gradually becoming more ubiquitous from the beginning of the 3rd millennium BC and reaching 90% of the assemblage in the Iron Age – does not critically depend on the choice of cut-off value.

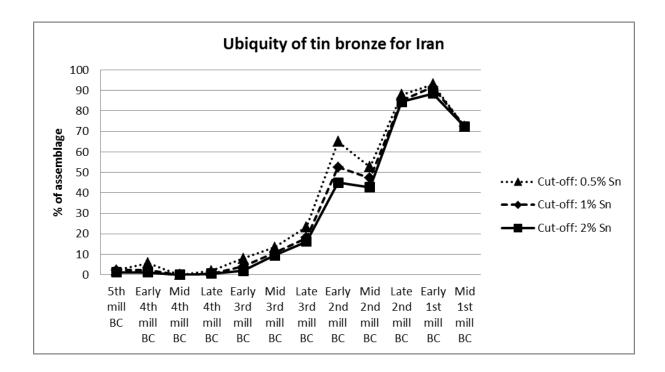


Figure 4. Evolution of the percentage of the assemblage represented by tin-bronze in Iran calculated using three different cut-off values for defining deliberate addition: 0.5%, 1%, and 2% tin. The apparent presence of tin-bronzes from the early 4th millennium is created by four objects out of 88. They came from the sites of Susa, Tepe Sialk and Tepe Hissar and had between 0.82 and 2.5% Sn. Two of these were analysed in the 1930s.

This highlights that the adoption of tin-bronze in Iran was on the whole not a sudden change. It took almost two millennia from the start of the 3rd millennium BC, and was used for almost all objects in the late 2nd millennium BC. Up until the early 2nd millennium, unalloyed copper and copper with variable amounts of arsenic prevailed. Interestingly, and counter-intuitively, bronze only really became the dominant copper alloy in the Iron Age, casting doubt on the idea that tin became a scarce commodity at the end of the 2nd millennium. It appears, therefore, that whatever the motivations to start using tin bronze, they were not sufficient to drive a rapid change in alloying traditions everywhere, or perhaps were challenged by difficulties in supplying enough tin. Figure 4 however only shows the general trend for the adoption of tin-bronze on a global scale (the whole of the modern territory of Iran) and more specific regional patterns are discussed in the following section.

V.1 Regional differences

A particular feature of the adoption of bronze in Iran, already well known⁵⁷ but highlighted by the present approach, is the difference between western Iran and eastern Iran. As can be seen on Figure 5, whilst regions of western Iran start using tin bronze in the first half of the 3rd millennium BC, both north-eastern Iran (Gorgan) and south-eastern Iran show no sign of any tin bronze in the 3rd millennium, and it remains relatively rare in the 2nd and 1st millennium BC in Tepe Yahya which is the only site of this region for which data was collected for this period. If we consider that Afghanistan or Bactria was a likely source of tin for regions further to the west, then the absence of tin is eastern Iran, which would have had to be crossed by overland trade routes, needs to be explained. This lack of tin is even more surprising given the presence in the east of stone vessels and semi-precious stones, such as lapis lazuli, initially thought to travel alongside tin⁵⁸. This has led to the idea of a maritime

"by-pass" of the Iranian Plateau and a Persian Gulf trade route controlled by central authorities in Mesopotamia or the Indus Valley⁵⁹. Another potential factor for the lack of tin bronzes in eastern Iran may be a "technological conservatism" at Tepe Hissar and Tepe Yahya in particular⁶⁰, where the use of arsenical copper was unchanged for most of the occupation of the sites. Reasons for such a conservative attitude to alloying have yet to be fully understood, but may show that the use of tin for alloying purposes was not universally considered necessary or desirable. It is also conceivable, as recently developed by Thornton and Giardino⁶¹, that the lack of tin in eastern Iran until the 2nd millennium BC reflects the fact that the exploitation of the Afghan sources may only have started around 2000 BC under the influence of people from the Bactria-Margiana Archaeological Complex of Central Asia, who are known to have migrated to eastern Iran at that time. Stöllner *et al.* on the other hand argue, based on Kaniuth's⁶² assessment that tin-bronze was uncommon in Bactria and Margiana before the early 2nd millennium BC, that the influence for tin use is more likely to have come from the Andronovo culture of central Asia, known to have mined tin from the late 3rd⁶³.

Although the graphs presented here do not at this stage provide a definitive answer to any of these questions, they allow us to go further than a qualitative description of when tin bronze first appeared or became "common" in a particular region (as for example in Thornton 2014, Table 23.1⁶⁴) and describe quantitatively its rate of adoption. They also highlight the areas for which there are gaps in our knowledge, for example here the scarcity of data for northwestern Iran before the Iron Age. It is hoped that such graphs can provide a more nuanced grounding to future discussions on the subject.

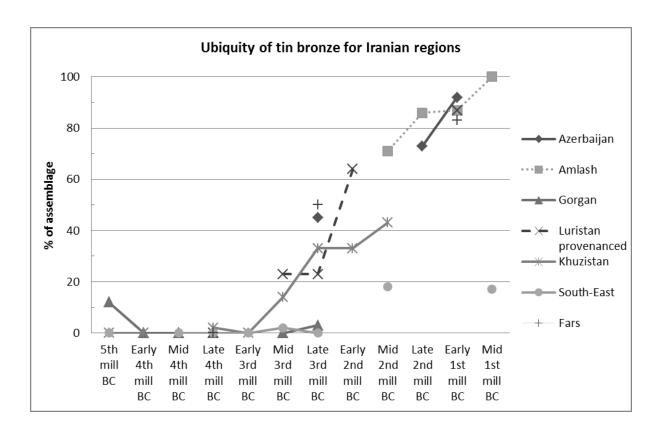


Figure 5. Chronological evolution of the percentage of the assemblage represented by tinbronze (>2% tin) for Iranian regions. Only periods for which more than five objects were analysed for a given region are represented. Not all regions show continuous distributions, since data are missing for some periods. The early 5th millennium signal in Gorgan is created by one pin (out of 8 objects) from Tepe Hissar with 2.12% Sn as analysed in the 1930s⁶⁵.

V.2 Analysis by object type

One of the sources for our understanding of the status of a certain material is the differential usage of that material for a variety of object types. It is often stated that tin-bronze was first used for ornaments, and only later for weapons and tools⁶⁶. This would indicate that tin-bronze was not used initially for its better mechanical properties, but was considered of value for other reasons, perhaps its rarity and novelty, or the colour change and lustre it brings to copper. The assembled dataset allows us to very simply test this statement: Figure 6 shows for Iran and Mesopotamia the evolution of the ratio of tin-bronze to tin-less copper in four different types of objects: weapons, tools, ornaments and vessels. For other regions, the

amount of data collected does not yet allow for a satisfactory chronological comparison of different object types.

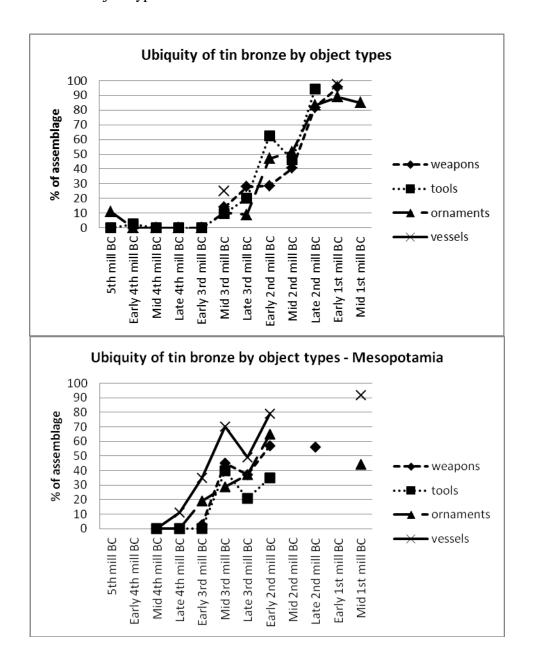


Figure 6. Chronological evolution of the percentage of the weapons, tools, ornaments and vessels made of tin-bronze (>2% tin) for Iran and Mesopotamia. Only periods for which more than five objects were analysed for a given object type are represented.

Bearing in mind the coarseness of the chronology used here, it now appears that in Iran ornaments and tools both started to be made of tin bronze at about the same time, in the mid-3rd millennium BC. In Mesopotamia, on the other hand, it would appear that vessels were

made of tin-bronze first, followed by ornaments and a few centuries later by tools and weapons. Tin bronze was also still used preferentially for vessels in the mid-1st millennium BC, when 92% of analysed vessels were made of tin bronze and has been noted by Müller-Karpe as the metal preferentially used to produce vessels at the Royal Cemetery of Ur in the mid to late 3rd millennium BC⁶⁷. It must have had properties that made it desirable for the production of vessels in the eyes of the Mesopotamians. This is particularly intriguing as it has been demonstrated that arsenical copper presents a greater ductility that tin bronze, which would have made it more suitable for the production of sheet-metal objects⁶⁸. An investigation into the mode of fabrication of these vessels could tell us more about the reasons behind this choice, but for now it appears possible that the Mesopotamians were trying to emulate the colour of gold for their vessels. The addition of tin certainly shifts the colour of the metal away from the dark red undertones of pure copper. Another potential explanation for the apparent earlier adoption of bronze for vessels could be a lag in the deposition of other kinds of objects (possibly due to differential recycling rates according to object type). This also requires further investigation, notably with a study of the context in with the vessels were recovered, compared to weapons and tools in particular. The actual amount of tin in the Mesopotamian vessels is not obviously different to that in tools, weapons or ornaments. Interestingly however vessels also seem to be treated slightly differently to other types of objects in the recipes recovered in Ebla, dated to the 24th century BC. These recipes, indicating the proportion of tin and copper for the manufacture of various objects in palace controlled workshop, show starting percentages of around 10% for vessels while most other types of objects are made with 12% Sn or more ⁶⁹. This can be explained by a technological constraint: at lower tin contents the majority of the tin can be absorbed in solid solution in the alpha phase dendrites that form upon casting, with less of the alpha-delta eutectoid being formed. As the eutectoid is hard and brittle, and therefore potentially

damaging to objects requiring heavy hammering, the ancient metal-workers might have tried to avoid its formation. However, the lower tin content found in the recipes for vessels could also denote a difference of status that has yet to be fully understood.

The role of tin-bronze as a prestige good charged with ideological and socio-political significance has convincingly been argued by Weeks for the region of the Persian Gulf⁷⁰. His discussion was based not only on its rarity and colour but also in the type of items it was used for, the contexts in which it was recovered and the distance and exchange mechanisms linked with its trade. The same arguments seem to apply in Mesopotamia as well: in addition to the preferential use for vessels clearly shown in Figure 6, its status has been demonstrated by Stech who studied its relationship to other grave goods and its presence in palatial contexts⁷¹. This research was based on the small number of analysed objects available in 1999 and a reassessment of similar questions would be of the greatest interest in light of the new data assembled here. It therefore seems that the prestige status of tin-bronze might have been the driving force behind its adoption in Mesopotamia and only later was it recognised as an alloy of superior mechanical properties well suited for the production of weapons and tools. Western Iran on the other hand possibly followed the trend set by its powerful neighbours and started using tin-bronze on a more global scale for all classes of objects at the beginning of the 3rd millennium BC, which is when Mesopotamia started using bronze for weapons and tools. The adoption of tin-bronze in western Iran might also be linked to the role it played in supplying tin for Mesopotamia whether it was as a source region (western Iran) or as a middleman in the trade $(Elam)^{72}$.

VI. Conclusion

This paper shows how assembling and analysing a database of over five thousand metal objects from Iran and the neighbouring areas can help identify and clarify trends that would otherwise have been lost, fragmented between 54 different publications. Of course an in depth understanding of the metal recovered at specific sites in relation to the rest of the material assemblage and the archaeological context there is crucial to develop an understanding of metal-working techniques, but it is our belief that interesting chronological and geographical patterns only emerge when a significant number of analyses are put together and looked at on a regional or national scale – a 'Big Data' approach.

For Greater Iran this has not only highlighted gaps in the available analytical data and raised a number of interesting research questions, but it has also provided us with an initial set of observations that might, when reconciled with textual and archaeological evidence, help "solve" the "tin problem". It has enabled us to formulate new hypotheses regarding three key questions in particular. The first one is how did the tin "travel", and when was it added to the copper. The distribution of tin content for metal likely to come from different places is very informative and we have argued that copper coming into Mesopotamia from the Iranian Plateau or further east was likely to have been mixed with tin at a late stage, probably by the Mesopotamians themselves. In contrast, copper coming from the west was probably treated differently and the tin in these objects may for example have come as already-alloyed objects from the west or may have been the result of copper from the west mixed with bronze from elsewhere.

The second question is the controversial assignment of a provenance to the tin. All that can be said with certainty at this stage is that the tin content of western and north-western Iranian objects in the Iron Age is compatible with the idea of a local source of tin, and that tin, at this time, is unlikely to have been extensively traded in the western part of the Persian Gulf. This may consolidate the claims of Nezafati and colleagues on the importance of the Deh Hosein

copper and tin mines⁷³. It however raises the question of how many of such occurrences would have been necessary to support the well-developed bronze traditions of the area. It certainly calls for more prospection work in the Sanandaj-Sirjan zone of the eastern Zagros, in western and north-western Iran in general and perhaps in the Caucasus to see if other ancient workings can be identified.

Finally, this approach has given us a better idea of the status of tin and tin-bronze throughout the Bronze Age and how it is likely to have been different in Mesopotamia, Western Iran and Eastern Iran. The "exotic" nature of tin and the golden colour that it would have conferred to the objects might have been what first attracted the Mesopotamians. In western Iran, aside from very early uses in Luristan, the idea of using tin-bronze may have come from the Mesopotamian neighbours. But while more and more objects were made of tin-bronze throughout the 3rd and 2nd millennia BC, it seems possible that the metal kept a valuable status well into the Iron Age: it is indeed only by the late 2nd millennium that almost all copper alloy work is bronze.

The hypotheses presented above are based on a number of patterns obtained by looking at the proportion of tin-bronze in the assemblage (Figures 4, 5 and 6) and at tin distribution profiles (Figures 2 and 3). In several cases we have proposed more than one scenario that could have led to the observed patterns (for example recycling, mixing or the use of mixed ores to explain the tin profile if the AsSbNi group in Figure 2) and tried to narrow them down to the most plausible hypothesis based on geological or archaeological considerations. Indeed the study of chemical datasets is made difficult by the problem of equifinality as different processes can lead to the same outcome in terms of chemical signature. As mentioned in the introduction, a more complete reconciliation of the presented patterns with other bodies of evidence is needed to convincingly discuss the processes that might have led to these. The bodies of evidence are of course textual, archaeological and geological, but research has

shown that an integrated study of big datasets of main alloying elements⁷⁴, minor elements composition⁷⁵ and lead isotopes data⁷⁶ can also help tease apart the mechanisms that might have led to specific patterns.

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