

RETHINKING THE BRONZE-IRON TRANSITION IN IRAN:  
COPPER AND IRON METALLURGY BEFORE THE  
ACHAEMENID PERIOD

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DPhil in Archaeological Sciences

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As the creeper that girdles the tree-trunk,  
the Law runneth forward and back;  
For the strength of the Pack is the Wolf,  
and the strength of the Wolf is the Pack.

*The Law of the Jungle*, Rudyard Kipling



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## Abstract

### RETHINKING THE BRONZE-IRON TRANSITION IN IRAN: COPPER AND IRON METALLURGY BEFORE THE ACHAEMENID PERIOD

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Iran, a country rich in mineral resources, has a long history of metal working. Copper objects first appeared in the 7th millennium BC and in the following millennia, copper became the material of choice for the production of many objects. Artefacts of iron began to appear in the mid 2nd millennium BC and by the mid 1st, iron had replaced bronze for most uses, but the reasons for this change remain unclear.

This thesis seeks to examine the transition from bronze to iron metallurgy from a new angle. By looking at changes in copper-based metallurgy between the Bronze Age and the Iron Age, it attempts to better understand the context in which iron metallurgy developed. To that end, the results of previously published chemical analyses of over 5000 copper-based objects from Iran and neighbouring regions and the lead isotope analyses of about 380 objects were assembled in a database.

The tin, arsenic, nickel, antimony and silver concentrations in particular are studied. The data is divided into 16 metal groups based on the absence or presence of the latter four elements. The study of the main groups allows us to describe interesting new patterns of metal movement and recycling. It appears that before the end of the Bronze Age, a number of copper sources and/or trade routes from both east and west declined, leading to a reliance on more local sources for copper and tin in the Iron Age. The practice of recycling from the 3rd millennium BC onward is also evidenced.

Overall, it seems that iron appeared within a thriving bronze industry, with a good access to metal resources and a developed understanding of the possibilities offered by copper (alloying, recycling, mixing...). Was it then the more 'permanent' nature of iron that attracted the ancient metal-workers and led to its advent?



## Contents

Acknowledgments .....	v
Abstract.....	vii
Contents.....	ix
Table of Figures.....	xiii
Chapter I. Introduction .....	1
1. Themes in archaeometallurgy: why study metals?.....	2
2. General concept of the DPhil.....	8
3. Geographical scope.....	11
a. Why Iran? .....	11
b. Broadening of the geographical scope.....	12
c. Landscape and resources .....	13
4. Historical scope .....	18
5. Summary of objectives and methods.....	26
Chapter II. Review of the development of metallurgy in Iran.....	29
1. The development of copper metallurgy.....	31
a. Sources of copper and mines in Iran.....	31
b. Early stages .....	40
c. Chalcolithic developments: the smelting and casting of copper.....	41
d. 4th and 3rd millennium BC: arsenical copper .....	43
e. The appearance of tin bronze.....	50
f. The problem of the origin of tin.....	52
g. Production processes.....	58
2. The appearance and development of ferrous metallurgy.....	60
a. Ore sources and mining .....	60
b. Chronological outline of the development of ferrous metallurgy.....	62
c. Smelting and forging: technological background.....	74
d. Metallurgical analysis of iron objects.....	80
3. Understanding the fundamental differences between bronze and iron metallurgy: a key to understanding the “bronze-iron transition” .....	90
4. Conclusion.....	94

Chapter III. The bronze data.....	95
1. Published data.....	95
a. A large and fragmented dataset .....	95
b. History of the publications.....	98
2. Integrating the data .....	126
a. The geographical and chronological framework .....	126
b. Integration of a large and varied dataset .....	136
3. Comparison with Chernykh’s study of Eurasian metallurgy.....	147
4. Conclusion.....	152
Chapter IV. The tin story .....	155
1. Introduction .....	155
a. Tin in the framework of this thesis .....	156
2. Diachronic analysis.....	157
a. Evolution of the ubiquity of tin bronzes .....	157
b. Evolution of the tin content .....	165
c. Artefact types .....	175
3. Definition of a “mid range” tin content.....	180
4. Synchronic analysis .....	189
a. Period 1 – Early 3rd to mid 2nd millennium BC.....	189
b. Period 2 – Late 2nd to mid 1st millennium BC .....	199
5. Discussion.....	201
a. Benefits of using a large dataset .....	201
b. Bronze Age/Iron Age transition.....	204
Chapter V. Overview of the copper composition and lead isotope analyses .....	211
1. Correction of the composition to take into account the deliberate addition of tin 211	
2. The arsenic content.....	214
a. Properties of arsenical copper .....	214
b. Chronological analysis.....	216
c. Inverse correlation between tin and arsenic and its contribution to the decrease in arsenic levels .....	219
d. Object types .....	226
e. Causes for the decrease in arsenic content.....	230
f. Regional specificities .....	231
3. The lead and zinc contents .....	234
a. Leaded copper.....	235
b. Brass.....	240
4. Nickel, antimony, silver and cobalt.....	245

a.	Nickel.....	245
b.	Antimony and silver.....	246
c.	Cobalt.....	248
5.	Lead isotope analysis.....	248
a.	Luristan .....	254
b.	Mesopotamia.....	256
c.	Oman.....	259
6.	Conclusion.....	264
Chapter VI. Metal flow and recycling.....		267
1.	Defining metal groups .....	269
2.	Type 1100: Arsenic Nickel.....	277
a.	Arsenic Nickel: geographical patterns .....	278
b.	Arsenic Nickel: chemistry and lead isotope ratios.....	281
c.	Relationship between the Arsenic Nickel group and the Nickel Only group	290
d.	Arsenic Nickel: A Tentative story .....	293
3.	Type 1111: Arsenic Nickel Antimony Silver.....	295
a.	Arsenic Nickel Antimony Silver: geographical patterns .....	296
b.	Arsenic Nickel Antimony Silver: chemistry and lead isotope ratios.....	298
c.	Arsenic Nickel Antimony Silver: A Tentative story .....	302
4.	Type: 1110: Arsenic Nickel Antimony .....	303
a.	Arsenic Nickel Antimony: geographical patterns.....	304
b.	Arsenic Nickel Antimony: chemistry and lead isotope ratios .....	306
c.	Arsenic Nickel Antimony: A Tentative story .....	310
5.	Type 1000: Arsenic Only metal.....	311
a.	Arsenic Only: geographical patterns.....	312
b.	Arsenic Only: chemistry and lead isotope ratios .....	316
c.	Relationship between the Arsenic Only group and the Clean Metal group...	319
d.	Arsenic Only: A Tentative story .....	321
6.	Other significant groups .....	323
7.	Conclusion.....	325
Chapter VII. Discussions and conclusion .....		329
1.	The appearance of iron in the context of bronze metallurgy.....	330
a.	Hypothesis of a response to economic pressure .....	330
b.	North-western influence on the appearance of iron.....	334
c.	Indigenous origin for Iranian iron-working.....	338
d.	Early understanding of the properties of iron – the question of craft specialization.....	339

e. The status and symbolism of early iron .....	343
2. Strengths and limitations of the synthesis of old datasets .....	344
a. The synthesis of old datasets: a promising approach.....	344
b. Limitations to this approach and further work.....	346
3. Conclusion: the study of large datasets as a basis of reflection on technological innovation .....	348
Bibliography .....	351
Appendix A: Chronology .....	377

## Table of Figures

Figure I-1. Lines of contact between Iran and the neighbouring regions. After Moorey (1994, p.xxi Fig.2 ) .....	17
Figure II-1: Modern copper ore bodies in Anatolia and on the Iranian Plateau. From Pigott 1999, Figure 4.6, p.83 .....	32
Figure II-2. Sources of copper, tin and gold likely to have supplied the Indus Valley civilisation (Figure 5.1 in Kenoyer & Miller, 1999, p.108) .....	37
Figure II-3: Location of the main sites mentioned in the text. ....	42
Figure II-4. Two of the arsenical copper daggers from the Louvre collection presenting respectively 4.5 and 4.1% As. (Pl. 14 in Begemann et al., 2008, p. 61) .....	49
Figure II-5. Map showing the possible trade routes used to bring tin into Mesopotamia and to redistribute it to the west (Fig 27.2 in Pigott, 2011, p.278).....	56
Figure II-6: Location of modern iron ore deposits in Anatolia, Iran and the Caucasus. (Drawing by Jon Snyder after Pigott 1981:Fig.2) (Pigott 1989, p.69) .....	61
Figure II-7: Iron finds in Iron Age I contexts (1450/1350 BC - 1100 BC) .....	65
Figure II-8. Bi-metallic lion pins recovered in Hasanlu (bronze body cast onto and iron shaft). (Fig. 13 in Pigott, 1989, p. 74).....	67
Figure II-9. Luristan short-sword from the Metropolitan Museum of Art. (Pl. 23, Fig. 1 in Leffert (1964, p.63)) .....	68
Figure II-10. Hilt of a Luristan sword cut for examination. (Fig. 4 in Bird & Hodges (1968, p.217)) .....	69
Figure II-11. Bi-metallic pins and bracelet attributed to Luristan (Ashmolean Museum n° 1931.33, 1951.277 and 1951.329).....	69
Figure II-12: Iron finds in Iron Age II contexts (1100-800 BC) .....	71
Figure II-13. Iron Age III sword (WK 66/467) and axe-head (WK 65/190) from the cemetery of War Kabud, Luristan. (Pl. 121 and Fig. 16 in Haerinck & Overlaet, 2004, p.49) .....	73
Figure II-14: Iron finds in Iron Age III contexts (800-600 BC).....	74
Figure II-15: Bi-conical bars possibly used for the trade of Iron. (Fig. 8 in Pleiner & Bjorkman 1974, p.295) .....	79
Figure III-1. Number of analyses entered in the database for each publication and analytical techniques used in these publications (Iranian sites) .....	97
Figure III-2. Number of objects entered in the database for each publication and analytical techniques used in these publications (Oman, Bactria, Indus Valley and Mesopotamia).....	98
Figure III-3. Map of the sites for which compositional data was collected .....	128
Figure III-4. Map showing the regional divide used in this thesis .....	129

Figure III-5. Time ranges given in the original publications to date the copper objects and time ranges to which they were attributed in the framework of this thesis .....	131
Figure III-6. Time ranges given in the original publications that overlap Period 1 (early 3rd to mid 2nd millennium BC) and Period 2 (late 2nd to mid 1st millennium BC). The periods in red are those to which 10 or more objects are associated. ....	133
Figure III-7. Number of objects entered in the database for each period (Iranian regions) .....	135
Figure III-8. Number of objects entered in the database for each period (neighbouring regions) .....	135
Figure III-9. Number of measurements entered in the database for each element.....	137
Figure III-10. Comparison of XRF and NAA tin measurements for the Mesopotamian sites .....	141
Figure III-11. Comparison of ICP-OES and AAS tin measurements for Selme.....	141
Figure III-12. Spread of the absolute difference in weight percentage of tin between two measures of one object using different techniques .....	141
Figure III-13. Comparison of Optical Emission Spectrometry and Mass Spectrometry lead measurements for Susa.....	142
Figure III-14. Comparison of OES and PIXE arsenic measurements for Hissar .....	143
Figure III-15. Comparison of OES and Mass Spectrometry arsenic measurements for Susa.....	143
Figure III-16. Comparison of XRF and NAA arsenic measurements for the Mesopotamian sites .....	143
Figure III-17. Spread of the absolute difference in weight percentage of nickel between two measurements of one object using different techniques .....	144
Figure III-18. Comparison of XRF and NAA antimony and silver measurements for the Mesopotamian sites .....	145
Figure III-19. Comparison of OES and Mass Spectrometry antimony and silver measurements for Susa .....	145
Figure III-20. Comparison of XRF and NAA tin measurements for the Mesopotamian sites. The red boxes represent the zones of the graph for which both techniques agree on the absence or presence of tin (with presence defined here as >0.2% tin).....	147
Figure IV-1. Distribution of the tin content for all of the objects of the database .....	158
Figure IV-2. Chronological evolution of the ubiquity of tin bronze in the copper-based metal assemblage .....	159
Figure IV-3. Chronological evolution of the ubiquity of tin bronze in the copper-based metal assemblage for Iranian regions .....	161
Figure IV-4. Number of objects in the database for each period for Iranian regions...	161
Figure IV-5. Chronological evolution of the ubiquity of tin bronze in the copper-based metal assemblage for neighbouring regions .....	163
Figure IV-6. Number of objects in the database for each period for neighbouring regions .....	163

Figure IV-7. Distribution of the tin content for objects from Oman from the late 3rd to mid 2nd millennium BC .....	166
Figure IV-8. Distribution of the tin content for objects from Oman from the late 2nd to the early 1st millennium BC .....	168
Figure IV-9. Evolution of the tin content in tin bronzes (>2% Sn) and of the ubiquity of tin bronzes in Luristan .....	168
Figure IV-10. Evolution of the tin content in tin bronzes (>2% Sn) and of the ubiquity of tin bronzes in Mesopotamia .....	169
Figure IV-11. Evolution of the tin content in tin bronzes (>2% Sn) and of the ubiquity of tin bronzes in Oman .....	169
Figure IV-12. Distribution of the tin content for Mesopotamian objects of the early 3rd millennium BC .....	171
Figure IV-13. Distribution of the tin content for Luristan objects of the mid 3rd millennium BC .....	171
Figure IV-14. Distribution of the tin content for Mesopotamian objects from the mid 3rd millennium BC to the mid 1st millennium BC .....	173
Figure IV-15. Distribution of the tin content for Luristan objects from the mid 3rd millennium BC to the early 1st millennium BC .....	174
Figure IV-16. Distribution of the tin content for objects from Azerbaijan from the early 1st millennium BC .....	175
Figure IV-17. Chronological evolution of the ubiquity of tin bronze in the copper-based metal assemblage for different object types in Iran (only periods for which more than 5 objects were analysed have been plotted on this graph).....	176
Figure IV-18. Chronological evolution of the ubiquity of tin bronze in the copper-based metal assemblage for different object types in Mesopotamia (only periods for which more than 5 objects were analysed have been plotted on this graph).....	177
Figure IV-19. Evolution of the tin content in tin bronzes (>2% Sn) for weapons, tools, ornaments and vessels in Iran .....	179
Figure IV-20. Evolution of the tin content in tin bronzes (>2% Sn) for weapons, tools, ornaments and vessels in Mesopotamia.....	180
Figure IV-21. Luristan spike butted axe-heads in the Ashmolean Museum. Top two: personal photography - accession n° 1951.158, 1951.160. Bottom two: the cutting edge was never ground down (Moorey, 1971) - accession n° 1965.188 and 1965.761.....	182
Figure IV-22. Tin content in different objects types in Iran between the early 3rd and the mid 2nd millennium BC .....	183
Figure IV-23. Tin content in different objects types in Iran between the late 2nd and mid 1st millennium BC.....	184
Figure IV-24. Percentage of tin in objects produced in Ebla as described in the tablets recovered there (after Archi, 1993) .....	185
Figure IV-25. Chronological evolution of the ubiquity of tin bronze with over 6% tin and tin bronze with between 2 and 6% tin in the copper-based metal assemblage.....	187

Figure IV-26. Chronological evolution of the ubiquity of tin bronze containing between 2 and 6% tin in the copper-based metal assemblage for Iranian regions. Only data points for regions where more than 5 objects were analysed are included in this graph.....	188
Figure IV-27. Chronological evolution of the ubiquity of tin bronze containing between 2 and 6% tin in the copper-based metal assemblage for neighbouring regions. Only data points for regions where more than 5 objects were analysed are included in this graph.....	188
Figure IV-28. Map representing the number of copper-based objects analysed for each region between the early 3rd and the mid 2nd millennium BC, the ubiquity of objects with more than 2% tin and the average tin concentration in the bronzes of these assemblages .....	191
Figure IV-29. Distribution of the tin concentration for each region between the early 3rd and the mid 2nd millennium BC. For each region a full histogram and a zoomed-in version are given. The first one allows to visualize the proportion of tin-less copper while the second one gives a better idea of the profile of the tin-content distribution.....	198
Figure IV-30. Map representing the number of copper-based objects analysed for each region between the late 2nd and mid 1st millennium BC, the ubiquity of objects with more than 2% tin and the average tin concentration in the bronzes of these assemblages .....	199
Figure IV-31. Distribution of the tin concentration for each region between the late 2nd and mid 1st millennium BC.....	201
Figure IV-32. Possible tin trade routes in Period 1 (early 3rd to mid 2nd millennium BC).....	206
Figure IV-33. Possible tin trade routes in Period 2 (late 2nd to mid 1st millennium BC) .....	207
Figure V-1. Variation of hardness with cold work for Cu-As alloys (Northover, 1989, p.113).....	215
Figure V-2. Evolution of the arsenic content in copper-based objects in Iran, Mesopotamia and Oman.....	218
Figure V-3. Evolution of the arsenic content and of the ubiquity of tin bronzes in the copper-based metal assemblage in Iran .....	219
Figure V-4. Distribution of the arsenic content in tin-less copper (<2% Sn), mid-range bronze (2-6% Sn) and high tin bronze (>6% Sn) in Iran (left) and Mesopotamia (right) .....	220
Figure V-5. Evolution of the arsenic content in Iran, Mesopotamia and Oman for tin-less copper objects (<2% Sn, left) and tin bronzes (>2% Sn, right).....	222
Figure V-6. Evolution of the arsenic content in tin-less copper objects (<2% Sn) and number of tin-less copper objects analysed for each period in Iran (top) and Mesopotamia (bottom).....	223
Figure V-7. Distribution of the arsenic content for tin-less copper objects (<2% Sn) in Period 1 (early 3rd to mid 2nd millennium BC, left) and Period 2 (late 2nd	

to mid 1st millennium BC, right) in Iran (top), Mesopotamia (middle) and Oman (bottom).....	224
Figure V-8. Distribution of the arsenic content for bronze objects (>2% Sn) in Period 1 (early 3rd to mid 2nd millennium BC, left) and Period 2 (late 2nd to mid 1st millennium BC, right) In Iran (top), Mesopotamia (middle) and Oman (bottom) .....	225
Figure V-9. Distribution of the arsenic content for weapons and tools in Period 1 (early 3rd to mid 2nd millennium BC, left) and Period 2 (late 2nd to mid 1st millennium BC, right).....	227
Figure V-10. Distribution of the arsenic content for ornaments and vessels in Period 1 (early 3rd to mid 2nd millennium BC, left) and Period 2 (late 2nd to mid 1st millennium BC, right).....	228
Figure V-11. Examples of different arsenic distribution profiles for tin-less copper objects (<2% Sn) in Period 1 .....	234
Figure V-12. Distribution of the lead content (top) and zinc content (bottom) for all of the objects of the database .....	235
Figure V-13. Chronological evolution of the ubiquity of leaded copper objects in the copper-based metal assemblage for Iranian regions.....	236
Figure V-14. Chronological evolution of the ubiquity of leaded copper objects in the copper-based metal assemblage for neighbouring regions.....	237
Figure V-15. Distribution of the lead content in copper-based objects for Khuzistan in the 4th millennium BC (left) and Oman for all periods (right) .....	238
Figure V-16. Chronological evolution of the ubiquity of leaded copper objects in the assemblage of tin-less copper objects (< 2% Sn, top) and of tin bronze objects (>=2% Sn, bottom) in Iranian regions.....	238
Figure V-17. Chronological evolution of the ubiquity of leaded copper objects in the assemblage of tin-less copper objects (< 2% Sn, top) and of tin bronze objects (>=2% Sn, bottom) in neighbouring regions.....	239
Figure V-18. Chronological evolution of the ubiquity of brass objects in the copper-based metal assemblage for Iranian regions .....	241
Figure V-19. Chronological evolution of the ubiquity of brass objects in the copper-based metal assemblage for neighbouring regions .....	241
Figure V-20. Maps showing the ubiquity of brass objects (>2% Zn) in Period 1 (early 3rd to mid 2nd millennium BC, top) and Period 2 (late 2nd to mid 1st millennium BC, bottom). For regions where brass represented more than 2% of the assemblage, the average percentage of zinc in the brass objects, as well as the type of the objects that were made of brass are is given. ....	243
Figure V-21. Zinc content against tin content for brass objects (>=2% Zn) in Bactria, south-eastern Iran and Mesopotamia).....	244
Figure V-22. Evolution of the nickel content in Iran, Mesopotamia and Oman over time. ....	246
Figure V-23. Evolution of the antimony content in Iran, Mesopotamia and Oman over time. ....	247

Figure V-24. Evolution of the silver content in Iran, Mesopotamia and Oman over time .....	247
Figure V-25. Distribution of the cobalt content in copper-based objects from Oman in Period 1 (early 3rd to mid 2nd millennium BC, left) and Period 2 (late 2nd to mid first millennium BC, right).....	248
Figure V-26. Lead isotope ratios for ores from Iranian mining areas .....	251
Figure V-27. Lead isotope ratios for ores from Oman, Afghanistan, Bactria, the Caucasus and the Indus Valley .....	252
Figure V-28. Overlap between the lead isotope abundance ratios of ores from various Near-Eastern regions. ....	253
Figure V-29. Lead isotope ratios for Iranian objects.....	254
Figure V-30. Lead isotope ratios for low nickel (<0.1% Ni), and high nickel (>0.1% Ni) objects (top), and low antimony (<0.1% Sb) and high antimony (>0.1% Sb) objects (bottom) from Luristan.....	256
Figure V-31. $^{207}\text{Pb}/^{206}\text{Pb}$ ratios for Mesopotamian tin-less copper (grey) and tin bronze (black) objects from different periods (from Begemann & Schmitt-Strecker, 2009, p.12, fig.5).....	257
Figure V-32. Lead isotope ratios for tin-less copper (<2% Sn), mid-range bronze (2-6% Sn) and high tin bronze (>6% Sn) objects (top) and low As (<0.25% As) middle As (0.25 to 1%) and high As (>1%) objects (bottom) in Mesopotamia.....	258
Figure V-33. Lead isotope ratios for artefacts from Oman in Period 1 and Period 2... 260	
Figure V-34. Plot of the arsenic content against the nickel content of tin-less copper objects (<2% Sn), mid-range bronzes (2-6% Sn) and high tin bronzes (>6% Sn) for Iran (top), Mesopotamia (middle) and Oman (bottom) in Period 1 (early 3rd to mid 2nd millennium BC) .....	262
Figure VI-1. Distribution of the nickel content of tin-less copper objects (<2% Sn) in Period 1 (early 3rd to mid 2nd millennium) in the South-East (left) and Khuzistan (right).....	271
Figure VI-2. Distribution of the silver content of tin-less copper objects (<2% Sn) in Period 1 (early 3rd to mid 2nd millennium) in Luristan (left) and bronze objects (>2% Sn) in Period 2 (late 2nd to mid 1st millennium BC) in Azerbaijan (right).....	272
Figure VI-3. Distribution of the antimony content of tin-less copper objects (<2% Sn) in Period 1 (early 3rd to mid 2nd millennium) in Luristan (left) and in Gorgan (right).....	272
Figure VI-4. Ubiquity of the 16 different copper groups .....	274
Figure VI-5. Number of objects falling in each copper category in Period 1 (top) and Period 2 (bottom).....	275
Figure VI-6. Chronological evolution of the percentage of the assemblage represented by each of the studied metal group .....	276
Figure VI-7. Percentage of the assemblage of each region represented by Group AsNi objects for Period 1 (top) and Period 2 (bottom). Percentages are only given for regions for which more than 10 objects were analysed. ....	280

Figure VI-8. Average levels of arsenic and nickel for tin-less copper objects (<2% Sn, left) and tin bronze objects (>=2% Sn, right) of the Arsenic Nickel group in various regions in Period 1 (early 3rd to mid 2nd millennium BC).....	283
Figure VI-9. Distribution of tin content of AsNi tin bronze objects (>2% Sn) in Mesopotamia, Assyria, Luristan and Khuzistan in Period 1 (early 3rd to mid 2nd millennium BC) .....	286
Figure VI-10. Lead isotope ratios for tin-less copper (<2% Sn) and bronze (>=2% Sn) objects of the AsNi group dating from Period 1 .....	287
Figure VI-11. Chronological evolution of the percentage of the assemblage represented by the AsNi, AsNiSbAg and AsNiSb groups in Mesopotamia, Assyria, Luristan and Khuzistan .....	288
Figure VI-12. Chronological evolution of the ubiquity of AsNi, AsNiSbAg and AsNiSb in south-eastern Iran and Oman.....	289
Figure VI-13. Chronological evolution of the ubiquity of AsNi and Nickel Only metal for all regions.....	290
Figure VI-14. Chronological evolution of the percentage of the assemblage represented by the AsNi and Nickel Only groups in Mesopotamia, Luristan, Khuzistan and Oman.....	291
Figure VI-15. Distribution of the tin content in objects of the AsNi type (left) and Ni Only type (right) in Luristan in Period 2 (late 2nd to mid 1st millennium BC).....	292
Figure VI-16. Tentative reconstruction of sources and movement of AsNi metal in Period 1 (early 3rd to mid 2nd millennium BC).....	294
Figure VI-17. Tentative reconstruction of sources and movement of AsNi metal in Period 2 (late 2nd to mid 1st millennium BC).....	295
Figure VI-18. Percentage of the assemblage of each region represented by Group AsNiSbAg objects for Period 1 (top) and Period 2 (bottom). Percentages are only given for regions for which more than 10 objects were analysed. ....	297
Figure VI-19. Average levels of arsenic, nickel, antimony and silver for tin-less copper objects (<2% Sn, left) and tin bronze objects (>=2% Sn, right) of the AsNiSbAg group in various regions in Period 1 (early 3rd to mid 2nd millennium BC) .....	298
Figure VI-20. Lead isotope ratios for tin-less copper (<2% Sn) and bronze (>=2% Sn) objects of the AsNiSbAg group dating from Period 1.....	300
Figure VI-21. Distribution of tin content of AsNiSbAg tin bronze objects (>2% Sn) in Mesopotamia, Assyria, Luristan and Khuzistan in Period 1 (early 3rd to mid 2nd millennium BC) .....	301
Figure VI-22. Tentative reconstruction of sources and movement of AsNiSbAg metal in Period 1 (early 3rd to mid 2nd millennium BC).....	303
Figure VI-23. Percentage of the assemblage of each region represented by AsNiSb objects for Period 1 (top) and Period 2 (bottom). Percentages are only given for regions for which more than 10 objects were analysed .....	305

Figure VI-24. Average levels of arsenic, nickel and antimony for tin-less copper objects (<2% Sn, left) and tin bronze objects (>=2% Sn, right) of the AsNiSb group in various regions in Period 1 (early 3rd to mid 2nd millennium BC).....	307
Figure VI-25. Lead isotope ratios for tin-less copper (<2% Sn) and bronze (>=2% Sn) objects of the AsNiSb group dating from Period 1.....	308
Figure VI-26. Lead isotope ratios for high Ni (>1.5% Ni) and mid-range nickel (0.1 to 1.5% Ni) tin-less copper (<2% Sn) objects of the AsNiSb group dating from Period 1 and ores from Oman and Anarak .....	308
Figure VI-27. Distribution of the tin content in AsNiSb bronze objects (>2% Sn) in Mesopotamia and Khuzistan in Period 1 (early 3rd to mid 2nd millennium BC).....	309
Figure VI-28. Tentative reconstruction of sources and movement of AsNiSb metal in Period 1 (early 3rd to mid 2nd millennium BC).....	311
Figure VI-29. Percentage of the assemblage of each region represented by As Only objects for Period 1 (top) and Period 2 (bottom). Percentages are only given for regions for which more than 10 objects were analysed .....	313
Figure VI-30. Chronological evolution of the percentage of the assemblage represented by the As Only and Clean Metal groups in the South-East, Gorgan, Luristan, Amlash and Mesopotamia .....	315
Figure VI-31. Average levels of arsenic for tin-less copper objects (<2% Sn, top left) and tin bronze objects (>=2% Sn, top right) of the As Only group in various regions in Period 1 (early 3rd to mid 2nd millennium BC) and of tin bronze objects in Period 2 (late 2nd to mid 1st millennium BC, bottom right) ....	316
Figure VI-32. Lead isotope ratios for tin-less copper (<2% Sn) and bronze (>=2% Sn) objects of the As Only group dating from Period 1 .....	318
Figure VI-33. Distribution of tin content for As Only tin bronze objects (>2% Sn) in Mesopotamia in Period 1 (early 3rd to mid 2nd millennium BC).....	319
Figure VI-34. Chronological evolution of the ubiquity of As Only and Clean Metal for all regions.....	320
Figure VI-35. Tentative reconstruction of sources and movement of As Only metal in Period 1 (early 3rd to mid 2nd millennium BC).....	322
Figure VI-36. Tentative reconstruction of sources and movement of As Only metal in Period 2 (late 2nd to mid 1st millennium BC).....	322
Figure VI-37. Tentative reconstruction of the movement of copper-based metal during Period 1 (early 3rd to mid 2nd millennium BC, top) and Period 2 (late 2nd to mid 1st millennium BC, bottom). The circles are not intended to represent the exact geographical location of a copper source, but the general area where metal of a certain type might have come from. ....	327

## Chapter I. Introduction

In the Near-Eastern section of Oxford's Ashmolean Museum, visitors come across a display of some of the ancient world's finest bronze-work: the famous Luristan bronzes.

Next to the display they can read:

*“‘Luristan Bronzes’ illustrate the artistic achievements and skills of metalworkers from around 1000 BC. Made using the ‘lost-wax’ technique, metalworkers were able to create highly complex decorative objects.”*

This display is far from unique: many other museums around the world exhibit in their galleries other examples of this remarkable art-work. But what do we actually know of these objects and of the people who made them? In the past 50 years, archaeometallurgists have been busy trying to answer this question, not just for Luristan but for the whole of Iran. Sites with remains of metal production have been excavated, collections of metal objects have been published and many objects have been subjected to scientific analysis. This work has resulted in a fairly good understanding of the main stages of metallurgical developments in different regions of Iran, most recently summarized in the excellent reviews by Pigott (1999b) and Thornton (2009a; 2009b). However it was felt that there was a need for a reassessment of the analytical dataset in itself. This thesis therefore aims to integrate all of the published compositional and lead isotope data for Iran as well as some from neighbouring regions. The main questions we seek to address by doing so are presented further in this chapter (I.2) but let us start with some introductory remarks on the study of archaeometallurgy.

## 1. Themes in archaeometallurgy: why study metals?

Traditionally, archaeometallurgical studies have mostly been focused around two main questions. The first one is that of the origin of metalworking i.e. ‘When and where did metal working first appear, and how did it spread?’ The second one concerns technology: ‘How were the metal objects made?’ This includes all stages of the ‘chaîne opératoire’ and therefore questions on the nature of the ore (or native metal) used, on smelting and alloying techniques, including the type of furnaces or crucible used, and on the shaping of the finished objects whether it is in moulds or by hammering or forging.

To answer the question of the origin, scholars have attempted to discover the oldest pieces of worked copper for example, or the oldest evidence of iron smelting. This approach aimed to set a structure for our perception of early metal technology. It was accompanied by the idea that the succession in which different metals were used could help describe chronological sequences. Indeed in 1836 the Dane C.J. Thomsen established the three-age system, reusing the old idea of a division of prehistory in ages described by the use of different material (Thomsen, 1836). He was able to demonstrate that artefacts could be classified into types, some of which corresponded to the predominance of the use of stone, bronze or iron. The system is still in use nowadays but has regularly been corrected and refined for different regions of the world.

Scholarly interest in the questions of when and where new metals first started to be used was soon followed by studies on how these metal objects were being produced. The first excavation report to present a scientific appendix on ancient metallurgy is Layard’s

“Discovery in the ruins of Nineveh and Babylon” published in 1853. In this volume Dr. John Percy reported on the composition and method of manufacture of four Assyrian bronzes (Layard, 1853, p.670). The study of metallurgical techniques has not always been easy as it requires a thorough understanding of metallurgical remains for which archaeologists are not always fully trained. Since the 1960s however, great advances have been made in the area of ancient metalworking technologies with important contributions by Tylecote with his volume “A History of Metallurgy” for example (Tylecote, 1976). Although now slightly outdated it has been a longstanding reference in the field of ancient metal-working technology. However, Tylecote’s “History of Metallurgy” is an example of the limitations of an exclusively technology oriented approach. Indeed, more recent studies have attempted to deal with a third aspect of metal-working: its social and economic settings.

As Killick reminds us:

*“Extractive metallurgy is a human behaviour: metals do not mine or smelt by themselves, though it would appear so in much of the recent European literature on the subject”* (Killick, 2001, p.490).

Therefore it is not only a question of when, where and how, but also who. Examining the human aspect of metallurgy may be more challenging than the questions mentioned above, especially considering the scarcity of the surviving archaeological evidence for some materials like iron, and has its own flaws, but it is also most rewarding and a way to stimulate the field to move forward. This is why Killick strongly defends speculation on the social setting of early metallurgy (Killick, 2001, p.490).

The human aspect of metal-working can be studied at every single stage of the life of a metal object: how are the metal-working communities organised to find the ore or

obtain it -or metal ingots- through trade, what is the scale of the metal-working industry in these communities, why are certain technological choices made, how are the metal objects used and by whom, how are they traded and finally how are they discarded or recycled.

The question of the provenance of the ores has often been at the forefront of archaeometallurgical projects. Muhly's doctoral dissertation in the 1970s (Muhly, 1973) for example was aimed at reviewing the historical and archaeological evidence available at that date to determine possible sources of copper and tin. The research of ore sources has since been one of the driving forces behind analytical studies of the chemical composition of copper objects and more recently behind the lead isotope analysis (LIA) of copper-based artefacts and ore fragments. Chapter 3 of this dissertation is aimed at presenting in detail the many independent analytical projects that have been carried out on Iranian material and material from neighbouring areas in the past fifty years. While these studies without doubt move the field forward in terms of understanding of the ores used in the production of copper-based metals and the evolution of alloying techniques, it is easy to forget the human aspect of metal-working when faced with tables of numbers. The question of intention and level of control of the metal-workers has nonetheless been a recurring one both in the study of copper-alloys and in that of iron metallurgy. Chapter 2 is a review of our current knowledge on the development of metallurgy in Iran and presents in more detail these debates, but as an example, one of the questions frequently addressed is to what degree the addition of arsenic to copper has been intentional. Concerning iron, scholars have discussed the extent of the ancient blacksmiths' understanding of the properties of the metal, such as its ability to be welded for example.

More recently, discussions have started to emerge not only on the level of skill or control of the metal-worker, but on deliberate technological choices, whether these are driven by the availability of resources, by technological performance or by the community for which the objects are made. This idea of community-driven production has been discussed in particular by Thornton (2009b) who pointed out that the Iranian sites of Tepe Hissar and Tall-i Malyan both show two distinct metal-working traditions within the same site, suggesting a possible separation between “elite-driven production and utilitarian production” (Thornton, 2009b, p.319). He also reported a delay in the adoption of tin-bronze at some sites such as Tepe Yahya for example and suggests it is due to a technological conservatism of the societies in question (Thornton et al., 2005). However, he concludes that the data available so far are not enough to explain this conservatism (Thornton, 2009b, p.320).

As mentioned above, the understanding of the relationship between humans and metal doesn't only concern the production of the metal objects, but its life after the production. The assessment of the way metals were used by ancient societies most often derives from typological considerations. Scholars have carried out quantitative surveys of the relative proportions of ornaments, weapons and tools to get an idea of the status of the metal. In essence, when weapons and tools are predominant, the metal has been considered as “utilitarian”, whereas when ornaments constitute the majority of objects recovered, it has been considered as having an “elite” status. Such quantitative approaches are an inevitable first step in the assessment of metal use, but they are almost always biased by the context of the recoveries. For iron for example, almost all of the objects recovered from Iron Age periods in Iran have been found in graves (see Chapter 2). Part of the problem in the case of this metal is its unstable nature, i.e. its tendency to corrode easily making it more likely to be preserved in tombs with a stable

air environment than in occupational contexts. The choice of the objects that a certain society would chose to bury with their dead gives us only a partial view of all the metal that was in circulation within this society. Quantitative approaches are also biased by the geographical variability and the inconsistency of archaeological activity from one area to another, be it for political or geographical reasons or by chance. This geographical variability can cause us to artificially assume a wider use of metal in some areas than in others (see McConchie's discussion on the difficulties of studying iron (McConchie, 2004, pp.13-14)).

Another aspect of the life of metal objects that has been the subject of scholarly interest is the understanding of the movement of metal between different communities. To date, our understanding of metal trade has mostly derived from typological considerations and from the study of ancient texts. For the area of the Near East, the written sources come mostly from Mesopotamia in the form of ancient cuneiform texts. As mentioned above, Muhly, amongst others, has reviewed the texts dealing with the exchange of copper and tin in the Mediterranean area and the Near East (Muhly, 1973), while Pleiner and Bjorkman have reviewed those mentioning iron (Pleiner & Bjorkman, 1974). As detailed in Chapter 2, these texts mention established trade systems as well as a fair amount of metal movement in the form of booty taking and war tributes. Potts in particular emphasizes the importance of not underestimating the magnitude of what he calls "non reciprocal exchange" (Potts, 1993). Although the ancient texts are an invaluable source of information on trade, they are, in our case, limited to recording the trade that would have affected a certain elite of the Mesopotamian society. Thus, recent attempts are being made to overcome such limitations: as discussed at length in Chapters 4 to 6 of this thesis, scholars are now trying to understand the movement of

copper-based metals (whether by trade or by other means) through the analysis of big datasets of compositional analyses (Bray, 2009; Bray & Pollard, 2012).

The last aspect of the study of ancient metals and the societies making and using them is the end of the life of a metal object. Our understanding of this aspect can tell us more about the status of the metal in the society. However, as we have already discussed above, our evaluation of how objects were discarded or deposited can also be biased by the fortuitous nature of the archaeological excavations. But metal objects that can no longer be used don't always end up in the archaeological record. Indeed, in recent years, attention has been given to the recycling of copper-based items. The nature of copper lends itself to the re-melting of old objects to create some new ones. However, it remains very difficult to assess the proportion of the production made from newly smelted ores as opposed to that resulting of the recycling of old objects. That's why, even if the importance of this practice has been acknowledged in recent works, for example in Hoffman and Miller's work on Indus Valley metalwork (Hoffman & Miller, 2009), it has rarely been studied in detail. But, as discussed in Chapter 6, recent studies have shown that the examination of some key elements in the composition of copper-based artefacts can give us some indication on the level of recycling in use in ancient societies (Bray, 2009; Bray & Pollard, 2012).

To conclude these introductory remarks, let us to go back to the initial question asked at the beginning of this chapter: "Why study metals?" As argued above, discussions on the intention of the metal-workers, the organisation and scale of the metal-working community, as well as the influence of the society on metal-working, are not only intellectually fulfilling and essential to make the field of archaeometallurgy (and

archaeology in general) move forward, but they also take part in building a global awareness of how technologies evolve and impact societies. However, while emphasizing the social aspect of metallurgy, it must not be forgotten that the choices of the metal-worker are not only a result of social pressure: they are highly dependent on the properties of the ores and the metals themselves. The variations of chemical and physical properties from one metal to another, in impacting the actions of the metal-worker, will undoubtedly have affected not only the organisation of the society but also its perception of the metal (Bray, 2009, Ch.1).

It is our belief that, when put together, all of these considerations will allow us to better understand the relationship between human behaviour and technological evolution - an understanding that can be extremely valuable in our modern society, where technology is always in movement.

As we have seen, the sheer range of questions relating to ancient metallurgy is extraordinary. Many would doubtless be of great interest in the study of Iranian archaeology; however it is not possible to address them all in this thesis. The choice has been made here to focus on the metal itself and try to better understand how it evolved between the end of the Bronze Age and the Iron Age. The key problems addressed in this work are presented in the next paragraph (I.2). The following section (I.3) develops on its geographical setting, while section (I.4) is a short outline of its historical setting.

## 2. General concept of the DPhil

As mentioned above, this thesis aims to reassess the analytical data published over the years on Iranian objects. The intention behind this project is to rethink the changes that

have occurred in metallurgy between the Bronze Age and the Iron Age. Indeed, little attention has been given to how bronze-working evolved during the Iron Age. Recent reviews of Iranian metallurgy have either been limited to the Chalcolithic and the Bronze Age - this is the case of Thornton's excellent review of bronze metallurgy in Iran that finishes with the beginning of the 2nd millennium BC (Thornton, 2009a) or, when they have dealt with the Iron Age such as Pigott's 1999 review, they have been limited to mentioning bronze production centres and the impressive Luristan tradition (Pigott, 1999b, pp.90-92). Perhaps the most complete attempt at describing bronze metallurgy in Iron Age Iran goes back to 1982 with Moorey's review of pre-Achaemenid metalworking (Moorey, 1982). In this article he describes the typology of Iron Age objects from north-western Iran and Luristan, discusses the parallels with neighbouring metal-working traditions and examines the tin-contents of different types of bronze objects of this period (Moorey, 1982, p.94). In this thesis, we argue that understanding the nature of bronze metallurgy during the early centuries of the Iron Age is essential to understanding the development of iron metallurgy and ultimately answer questions such as 'why was a new metal needed or desired?'.

As shown in Chapter 2, our current understanding of the development of iron metallurgy is fairly limited. This is in good part due to the fact that no production sites have so far been recovered in Iran. Metallographic analyses have been carried out on some artefacts (see Chapter 7) but these are still far from giving us a complete understanding of the work of the ancient blacksmiths. So far our knowledge of the development of iron-working is therefore mostly based on a description and quantitative analysis of the iron objects recovered at Iron Age sites. Pigott in his PhD (Pigott, 1999b) has reviewed these artefacts for western Iran and proposed a chronology for the development of iron metallurgy that has not evolved much since then (see Chapter 2). In

broad terms, it portrays the beginning of the Iron Age as almost iron-less. It is not until the end of the 2nd millennium that iron starts to appear regularly, and it is not until the 7th century BC that it replaces bronze for most uses. It is clear that the transition from the Bronze Age to the Iron Age in Iran has little to do with a drastic change in metal use. For that reason, the other changes in material culture occurring in this period are discussed in section 4 of this chapter. But if copper and bronze were not immediately and radically replaced by iron, did their metallurgy continue to evolve, and if so how? What this thesis seeks to achieve is a more detailed description of the changes in metallurgy between the Bronze Age and the Iron Age, where bronze still has a role to play.

To achieve this goal, all the compositional data published to date on bronzes from Iran and a significant amount of data from neighbouring regions have been reviewed and assembled in a single database. The idea is to let the metal speak for itself with a regional approach, rather than focusing on site by site differences and isolate patterns in production and inter-regional or international trade.

It is clear that this approach alone is not enough for a complete understanding of bronze metallurgy during the Iron Age. The models that emerge from this analysis have to be tested against the historical and archaeological data available. Finding and publishing some data on mining sites and production sites, would also be crucial to the development of this field. However, we suggest that the metal in itself had much more to offer than is currently believed, especially if all of the available data is put together.

The secondary aim of this work is to test the strength and limitations of this type of analytical work. The difficulties of integrating a great number of projects using a range of different methods into one big dataset, and to do so in an area where the chronology

is not very detailed are discussed in Chapter 3. But this thesis aims to provide a better idea of the extent to which it can be used in archaeometallurgy.

### 3. Geographical scope

#### a. Why Iran?

Initially, this project was meant to focus mainly on the land that lies within the borders of what is now modern Iran. A quick glance at a map of the Near East will be enough to notice Iran's strategic position within the area. Located between the Caspian Sea to the north and the Persian Gulf to the south, it is an obligatory passage for all overland routes linking eastern and central Asia to the Mediterranean, such as for example the Silk Road. Its landscape of high mountain ranges and deserts might appear less inviting than the fertile plains of its Mesopotamian neighbours, but its vantage situation and its geology consisting of rich mineral resources have meant that it has been inhabited by many great civilisations throughout history, including the Achaemenid and Persian empires. Of particular interest for this project is the fact that Iran is very rich in copper resources (see Chapter 2). The idea behind this project was therefore to study the role played by this rich point of contact between east and west in the history of metallurgical development.

This thesis focuses mainly on the changes that occurred in the production and use of metals at the end of the Bronze Age. It was believed that Iran could be a key in understanding the mechanism for these changes in the greater Near East. The first reason for this is the role it might have played in the trade of tin and the possible modifications in the access to tin that could have brought on the advent of iron. Indeed,

as most archaeologists today agree to say that Afghanistan is likely to have been the source of tin for the production of bronze in the ancient Near East at least from the 2nd millennium onwards, Iran located between the mines and the consumers of tin can be expected to have played an important part in its trade.

To the north and the east, Iran is also in close contact with the people of the “Central Eurasian Culture Complex”. These people are thought to have played key role in the economy of the Silk Road (and its ancestries) and to have been constantly interacting with the “peripheral empires” of the easternmost and westernmost parts of Eurasia through commerce, migrations and military actions (Beckwith, 2009). It is remarkable to note that two very different iron-working traditions emerged at either end of Central Eurasia: in the east, in China, bloomery iron was virtually absent in the antiquity and cast iron prevailed from very early in the Iron Age (Bronson, 1999). In the west, Europe and the Mediterranean area in particular, bloomery iron was the only type of iron produced for a very long time. Cast iron only appeared in mass production in the 1860s AD. Although these two regions are separated by a huge distance, they were both in contact throughout the Bronze Age and the Iron Age with people of the Central Eurasian Culture Complex amongst which Iranian people. From this perspective, it is interesting to study whether Iranian metallurgy bears any traces of being in a region where western and eastern influences came together, ideas were shared and goods were traded.

#### b. Broadening of the geographical scope

Although initially our attention was restricted to Iran, it quickly became obvious that in order to understand the movement of metals within the country it was necessary to

understand where the metal came from and therefore to include in our study the countries bearing the potential sources of tin: mostly Afghanistan and ancient Bactria. Our initial results showed that eastern Iran used tin-less copper throughout most of the Bronze Age, which pointed towards an important Persian Gulf tin trade also described in literary sources (Muhly, 1973). That is why the potential actors of this trade: the Harappan civilisation, the Oman peninsula and Bahrain were also added to the area of study. Finally, as ancient texts indicate that Mesopotamia was at the receiving end of the Persian Gulf trade and was probably an important consumer of metals, it was decided that Mesopotamia would also be considered in this project.

Our centre of interest remained Iran and an effort was made to assemble an exhaustive database of metallurgical analyses for this country. However we also collected as much data from the neighbouring countries as was possible within the time frame of this study. In the end, the total geographical extent of this project encompassed a large portion of the ancient Near East defined by the modern political borders of Iran, Iraq, eastern Turkey, Afghanistan, Pakistan, southern Turkmenistan and Uzbekistan, north-western India, Oman, the UAE and Bahrain.

### c. Landscape and resources

During the Bronze and Iron Ages this area was the scene of peaceful commercial contacts as well as hostile military interactions. Some battles were most likely fought for the access to natural resources and the control of their trade routes (Muhly, 1973, pp.324-25). Ancient cuneiform texts inform us that goods travelled by the means of organised trade agreements, sometimes on a wide international scale through the Persian Gulf in particular (Muhly, 1973), but also by the means of booty taking in defeated

cities (Potts, 1993). It is clear that the natural resources offered by such a wide and varied area have modelled the cultures that have developed there, and influenced their interactions.

The Tigris and Euphrates rivers flowing through Mesopotamia turn the land in their vicinity from a desert to highly fertile plains (Van De Mieroop, 2007, p.7). Although ideal for the development of agriculture this area has little else to offer in terms of resources. Mesopotamia could offer food, mud for mud-brick constructions and ceramics and textiles, but had to rely on import for much of the other resources. Timber, stones and metals in particular, as discussed at length in this thesis, were amongst the sought-after goods that Mesopotamia needed to acquire from elsewhere (Potts, 1993, p.383). Overland caravan transport is attested in ancient texts and would have been necessary to cross the high mountainous range of the Zagros running diagonally from the north-west to the south-east of Iran (Muhly, 1973, p.292). Fluvial and maritime trade, however are likely to have been preferred whenever possible as they would have allowed for a greater bulk of goods to be transported at once, and been a faster and cheaper than overland trade (Potts, 1993, p.396). The Euphrates and the Tigris would have provided routes to the west and the north towards Syria and Anatolia (see a map of Iran and the neighbouring areas in Figure I-1). The Greater Zab and the Lesser Zab, tributaries of the Tigris in the north, were probably used to join Assyria from the regions surrounding Lake Urmia in north-western Iran after crossing the Zagros. Further to the south, the Diyala River would also have aided transport between the highlands of Iran and Mesopotamia whilst the Kerkheh and the Karun rivers could have been used to reach the Susana plain in the Khuzistan region. Today, this lowland area lies within the political borders of Iran, but it can geographically be considered as an extension of the Mesopotamian flood plains. The inhabitants of Mesopotamia, as discussed below, also

engaged in maritime trade through the Persian Gulf. From as early as the 5th millennium BC, they would have sailed along the coasts of the Gulf in primitive crafts (Van De Mieroop, 2007, p.9). As sailing developed, the Gulf enabled communication between far spread regions, putting in contact inhabitants of the Arabian Peninsula, the Indus Valley, Iran, Mesopotamia, etc.

The Iranian landscape is dominated by its two main mountain ranges. As mentioned above, the Zagros range, born from the collision of the Arabian and Eurasian tectonic plates and extending through most of the western and southern part of the country would have constituted a challenging natural obstacle to the passage of people and resources. Yet the range comprises of numerous mountain valleys, adapted to herding and small scale agriculture, that were inhabited throughout the Bronze Age and the Iron Age. The region of Luristan for example, as discussed below, was home to a people with a remarkable metallurgical tradition. At the southern end of the Zagros, the highland region of Fars, where the site of Anshan (modern Tall-i Malyan) can be found, together with the lowland region of Khuzistan, were home to the ancient Elamite civilisation.

The other Iranian mountain range is the very high Elburz range stretching along the southern edge of the Caspian Sea. The Elburz creates a barrier between the coastal plains of the Caspian Sea, in particular the Amlash region in the west and the Gorgan plains in the east, and the Central Plateau.

The Central Plateau, with the Qazvin plain in north-central Iran, the Tehran area and the central regions have also yielded many archaeological sites that are of interest in the study of Iranian metallurgy, such as Tepe Ghabristan, Tepe Sialk or Arisman. The Iranian plateau is rich in resources: coal, salt and metals, such as zinc in particular, but

most importantly for the subject at hand, copper and to a lesser degree iron. As shown in the following chapters, it provided the necessary metallic resources for an early development of metallurgy.

To the east lie two high deserts: the Dasht-i Lut and the Dasht-i Kavir. At the border between the Dast-i Kavir and the south-eastern edge of the Zagros, in the provinces of Kerman and Sistan, a number of sites have been recovered, like for example Tepe Yahya and Shahdad (in Kerman) or Shahr-i Sokhta (in Sistan) which have all yielded metallurgical artefacts of interest for this dissertation (see Chapter 3). Eastern Iran shares borders with Afghanistan and Pakistan which also present a very varied landscape combining inhospitable areas, such as the extremely dry Sistan Basin and the high mountain ranges of the Hindu Kush and Himalayas, with more hospitable areas like the Indus Plain in Pakistan and northern India where the Harappan civilisation flourished and the plains in north-eastern Afghanistan that extend into Turkmenistan, Uzbekistan and Tajikistan: the area known as ancient Bactria.

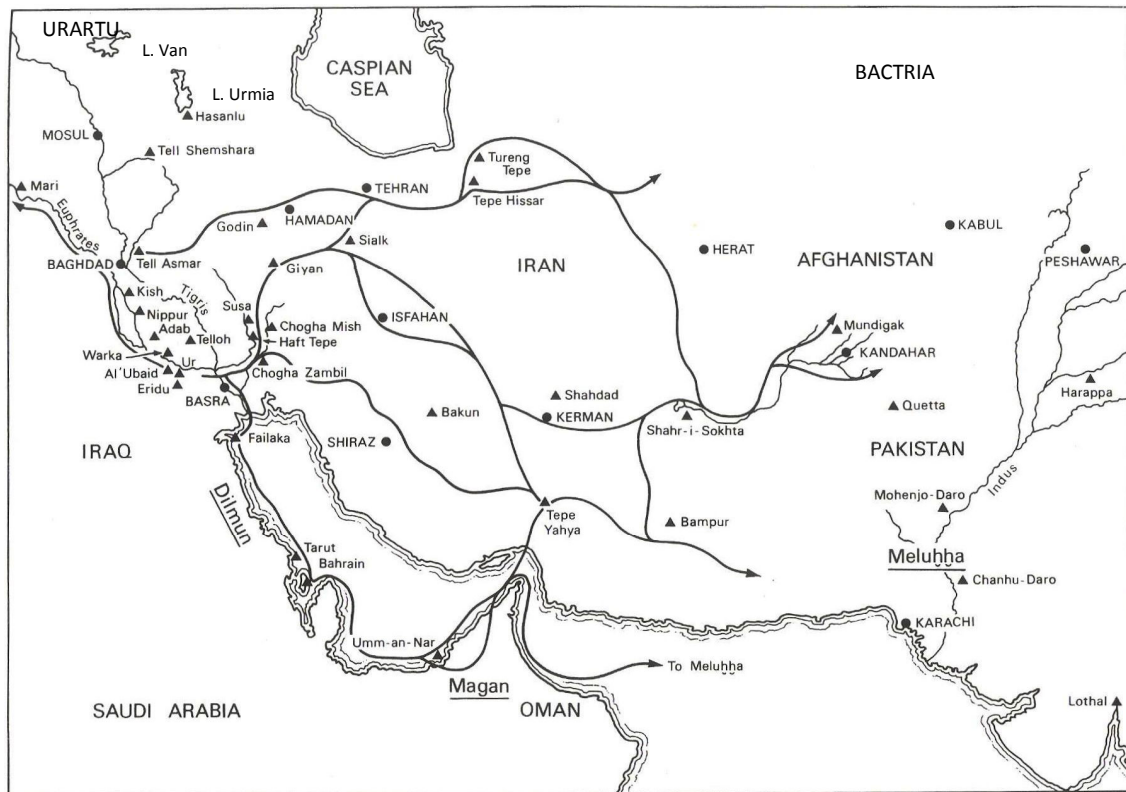


Figure I-1. Lines of contact between Iran and the neighbouring regions. After Moorey (1994, p.xxi Fig.2 )

The southern border of Iran is delimited by the Persian Gulf, a maritime corridor allowing a connection between Mesopotamia, the Indus Valley, Iran, but also the Arabian Peninsula and in particular the Oman Peninsula which occupies a strategic place in the Strait of Hormuz: the narrowest area of the Persian Gulf. The Oman Peninsula comprises of the modern countries of Oman in the east and the United Arab Emirates in the west. Its topography is varied (coastal plains along the Gulf of Oman, Al Hajar mountain range and high desert plateau) and as shown in the following chapters, it possesses copper resources that have been exploited since the Bronze Age.

This complex and diverse landscape, constituted of mountains, deserts, river valleys, fertile plains, marshes and the drainage basins of eastern Iran has created what has been described by Thornton as numerous highland-lowland relationships (Thornton, 2009b).

The dynamics of these interactions are mostly governed by the distribution of the resources: the lowland areas are fertile but lack in mineral resources while the highlands are rich in minerals and can be suited to pastoral or small-scale agricultural societies. In the west for example, the many highland valleys of the Zagros have seen the development of various local communities that interacted with the lowland plains of Mesopotamia and Khuzistan (Thornton, 2009b, p.306). On the eastern part of the Iranian Plateau this distribution of resources has generated the need for localized exchange systems despite the natural barriers that sometimes separated the highland and lowland regions. As an example Thornton describes how the site of Tepe Hissar was, during certain time-periods, more closely connected to the sites of the Gorgan Plain on the other side of the Elburz than to other sites of the plateau such as Tepe Sialk (Thornton, 2009b, p.307).

The next section is an attempt at summarizing the main cultures that flourished in this area in the Bronze and Iron Ages and how they interacted.

#### 4. Historical scope

In order to understand the changes occurring between the Bronze Age and the Iron Age, it was deemed necessary to collect data not only from the late stages of the former and the early stages of the latter but of the entirety of these two periods. In fact, for Iran, all of the data available from the appearance of copper metallurgy up until the late 1st millennium BC was considered. Although there is some analytical data available for as early as the 5th millennium BC, the bulk of it is for objects dating from the early 3rd millennium BC to the mid 1st millennium BC. Of course, a detailed review of the

history and archaeology of such a vast region over three millennia would be far beyond the scope of this thesis. However, an overview of a few key elements will help understand the dynamics of the regions involved in metal-working and metal trade. The main cultural periods and archaeological sites have been summarized in a timeline given in Appendix A based, for the earlier periods, on the timeline provided by Thornton in his PhD amongst others (Thornton, 2009a, pp.xv-xix), but also including Iron Age periods.

As mentioned previously, this thesis focuses on the changes that occur in metallurgy between the Bronze Age and the Iron Age. In this section we will therefore attempt to give a context to these changes by summarizing the history and the archaeology of the Late Bronze Age and Early Iron Age in Iran and the surrounding regions.

In eastern Iran, Pakistan and south-eastern Arabia, the second half of the 2nd millennium BC is marked by a decreased density of settlements. The reasons for this phenomenon are uncertain: some theories suggest a collapse of the “global” economic system occurring some time in the middle of the 2nd millennium. With reference to metals in particular, this economic system was notably described by Muhly in “Copper and Tin”, where he speaks of an “organised foreign trade” and an “international age of metallurgy” that developed in the second half of the 3rd millennium (Muhly, 1973, p.315). His work, based on documentary and archaeological evidence, suggests a period of close contact between Mesopotamia and the Indus Valley beginning around 2500 BC and ending with the close of the Isin Larsa period in Mesopotamia and the collapse of the Harappan civilisation around 1900 BC. During this period, trade through the Persian Gulf flourished. Mesopotamia was importing goods from Meluhha, a land thought to be located somewhere in the Indus Valley, and from Magan, that scholars consider as being the Oman peninsula. The island of Dilmun (modern Bahrain) also played an

important role in the Persian Gulf trade as an entrepôt for the traded goods. Following the fall of Harappa in the beginning of the 2nd millennium, trade between Mesopotamia and Meluhha is no longer supported by textual and archaeological evidence. Trade with Magan, via Dilmun appears to continue briefly but the Persian Gulf trade gradually lost its importance in the 2nd millennium BC and was probably replaced by overland trade.

While the collapse of this economic system is likely to have had an influence on south-eastern Iran, the Indo-Iranian borderlands and eastern Arabia, another potential explanation for the decrease in settlement density in the middle of the 2nd millennium BC is a change in climate (Lamberg-Karlovsky & Magee, 2004, pp.77-81). Lückge et al. (2001) have observed an increase in rainfall between 1900 and 1100 BC and Magee has argued that this might have led to a more dispersed settlement system, less archaeologically visible than a settlement system based on irrigation. The beginning of the 1st millennium BC is marked by an increase in settlement density in the area and a reoccupation of some sites, notably Tepe Yahya in south-eastern Iran. This could be linked to an increased aridification and a reliance on qanat irrigation leading to bigger settlements and the emergence of elites (Magee, 2005). However, until more Iron Age sites are excavated, south-eastern Iran will remain practically silent for the Iron Age. At present evidence is too scarce to grasp the organisation of the region. The only available information is the lack of mention of this area in the Neo-Assyrian and Neo-Elamite texts, suggesting a potential “polycentric settlement system” as in south-eastern Arabia at the time (Lamberg-Karlovsky & Magee, 2004).

A similar phenomenon is observed in southern Turkmenistan where, following a peak in urbanization during the Namazga V period (2100-1850 BC), there was a significant change in settlement pattern. In the Namazga VI period (1600-1000 BC) Antyn Tepe, one of the two main cities of the previous period, was abandoned and the other one,

Namazga Tepe, declined. Biscione described the phenomenon as a collapse of the socio-economic organization due to a high concentration of population in the cities, resulting in a segmentation of the settlement pattern: a shift from a city to an oasis system accompanied by an eastward migration into Bactria. However, this change did not correspond to a drastic modification in the material culture (Biscione, 1977).

Much more is known from the Iron Age in north-western Iran. In the 1960s Young and Dyson proposed a chronology in three phases based on ceramics: the Early Western Grey Ware (EWGW), the Late Western Grey Ware (LWGW) and the Western Buff Ware (WBW). These were initially equated to Dyson's Iron Age I, II and III (Dyson, 1965, p.211) but with the revision of the chronology of Tepe Hasanlu, the absolute dates of the ceramic phases had to be redefined (Young, 1985, pp.362, note 1). It remained nonetheless clear that at the end of the 2nd millennium BC, the Late Bronze Age culture of north-western Iran was replaced by a new culture characterised notably by EWGW type ceramic, replacing the painted wares of the Late Bronze Age, and the introduction of cemeteries outside the main settlement sites. The exact circumstances for this change remain unknown. In 1965, Dyson suggested the arrival in the region of a completely new people:

“The culture of Period V plainly represents an intrusion into the area flanking Lake Urmia on the west, south and east of a new group (or groups) of people who used burnished monochrome pottery, who buried their dead in the cemeteries, who employed stemmed goblets for drinking purposes, who had virtually no iron and who settled on scattered mounds throughout the area” (Dyson, 1965, p.197).

But he recognised that “the question of the origin of these settlers remains to be solved [...]” (Dyson, 1965, p.197). Young proposed a migration from the east to west along the Elburz range (Young, 1985, p.373) possibly following the decline of north-eastern settlements. This theory is supported by the presence of a grey ceramic possibly related to EWGW at north-eastern sites from the period of the Late Bronze Age (Tepe Hissar, Tureng Tepe) and the appearance of EWGW first at sites in north-central Iran (Gheytaryeh, Khorvin, Tepe Sialk) (Young, 1967, p.24; Young, 1985, p.373). Characteristic EWGW shapes then occur simultaneously at several sites of north-western Iran such as Hasanlu, Dinkha Tepe, Sialk, Geoy Tepe and Marlik, but remain confined to that region (Young, 1985, p.367). It has been suggested that these migrations corresponded to the arrival of Indo-Europeans or Indo-Iranian people on the Iranian plateau (Young, 1967; Deshayes, 1969), but there is at present too little evidence to prove such a theory (for a discussion on the subject see Young 1985).

Moving west of Iran, the end of the 2nd millennium BC corresponds to a resurgence of Assyria after a period of Mitanni domination. In the 13th century BC in particular, the Assyrian kings carried out military campaigns in the Zagros. The presence of ceramics with very close Assyrian parallels at north-western Iranian sites such as Hasanlu and Tepe Giyan is likely to be the result of these intrusions (Dyson, 1965, p.195).

Later in the Iron Age, the early to mid 1st millennium BC was marked by the intrusion in north-western Iran of another kingdom: Urartu. This kingdom located around Lake Van in eastern Anatolia first appeared in the Assyrian records in the 13th century BC (Rigg, 1937). At the peak of its expansion, in the early 8th century BC, the borders of Urartu reached Lake Urmia in north-western Iran, illustrated notably by the establishment of an Urartian fortress at the site of Hasanlu IIIB. It is uncertain whether the Urartians were responsible for the sack of Hasanlu IVB in the preceding period

(Thornton & Pigott, 2011, pp.174-76) but what is certain is that the total destruction of the citadel by fire around 800 BC provides a rare insight for the archaeologists into an occupation site of the Iron Age. Two groups of objects have been found in the ruins of Hasanlu IVB: some of local manufacture and some of Assyrian types, either copies or imports attesting of strong links between north-western Iran and their neighbours to the west: the Assyrian Empire (Dyson, 1965, p.199). However north-western Iran seemed to have had only limited contacts with their neighbours to the south, as there is almost no evidence in Hasanlu IVB of material that could be related to Luristan (Dyson, 1965, p.200). Surprisingly, a recent study of the Hasanlu IVB corpus of weaponry by Thornton and Pigott (2011) has showed that the metallic weapons, unlike some other types of objects, showed not only limited parallels with Luristan but also limited parallels with Assyria: the objects seemed more akin to weaponry of Transcaucasia and of Gilan on the Caspian littoral. As discussed in the following chapter, the burnt buildings of Hasanlu yielded an incredible collection of metal objects, both bronze and iron, providing important data for the study of north-western Iranian archaeometallurgy.

The Grey Wares and the pottery shapes characteristic of north-western Iran were not found in the early stages of the Iron Age in Luristan or at least in the Pusht-i Kuh region, excavated in the 1960s and 1970s by Vanden Berghe (Overlaet, 2005, p.7). Vanden Berghe therefore proposed an independent chronology for this region based on the result of his excavations: Iron I 1300/1250 – 1000/900 BC, Iron II 1000/900 – 800/750, Iron III 800/750-600 BC. Little is known from the Late Bronze Age in the Pusht-i Kuh where only one LBA tomb was excavated. However, this tomb shows similarities with the LBA culture of another region of Luristan: the Pish-i Kuh (Gilan III-II culture). At the end of the Bronze Age, the Pish-i Kuh and the Kangavar valley (Young, 2002, pp.424-26) experienced changes that can be compared to the ones that

occurred in eastern Iran: settlements became smaller (Tepe Guran) or were completely deserted (Tepe Baba Jan) without signs of military destruction. Again, the increased rainfall in the Near East towards the end of the Bronze Age has been proposed as a potential explanation for the changes (Overlaet, 2005, p.9). The Early Iron Age material culture of Luristan however exhibits some continuity with the Late Bronze Age culture: the toggle pins and pitchers found in burial goods are continuation of Bronze Age shapes. This has lead Overlaet to suggest an adaptation to new “living circumstances” (environmental/climate changes) rather than the arrival of a completely new population. He also suggested that the changes visible in the archaeological record reflect the expansion of minority groups that were less archaeologically visible in the Bronze Age, perhaps due to a nomadic life style, but were better adapted to the new conditions of the beginning to the Iron Age (Overlaet, 2005, p.10).

In 2005, Overlaet reviewed Vanden Berghe’s data and published a refined study of the different cultural phases of the Iron Age in the Pusht-i Kuh which we attempt to summarize here (Overlaet, 2005). In the Pusht-i Kuh, the Iron Age IA period is characterised by strong links with Kassite Mesopotamia as attested by the presence of “carinated beakers”, faience vessels and shell finger rings of Mesopotamian import amongst others. However, these imports stopped at end of Iron Age IA phase, probably with the defeat of the Kassites by Elam. In terms of metallic objects, bronze anklets, incised arrowheads and flange-hilted daggers were common in the beginning of the Iron Age and phases IA and IB probably saw the beginning of the production of the famous Luristan bronzes (highly decorated axe-heads, pins and idols, often with animal designs such as lions, swimming ducks or ibexes). The Iron Age IB/IIA was marked mostly by a gradual increase in the occurrence of iron and the continuation of the production of the bronzes. However, it would appear that the production of these bronzes ended or at least

significantly decreased in the Iron Age IIB (900-800/750 BC). This period was also marked by a replacement of the communal tombs that were characteristic of the beginning of the Iron Age by individual tombs. The larger settlements of the Pish-i Kuh were in addition reoccupied, possibly as a result of a new change in the environmental conditions. An increase in the number of tombs from the Iron Age III period suggests increase in population at that time. Other characteristics of this period are a renewed contact with Mesopotamia and Elam and, as seen in more detail in the next chapter, the use of iron for all types of weapons and tools.

Throughout the Bronze Age and the Iron Age the plains of Khuzistan and the highlands of Fars in south-western Iran were the home of the Elamite civilisation. This civilisation has alternated between periods of close relationship with its Mesopotamian neighbours and periods of relative isolation. Its influence stretched quite far into eastern Iran to sites such as Shahdad, Tepe Yayha or Shahr-i Sokhta. In the second half of the 3rd millennium BC in particular, all were linked and part of a civilisation that Amiet described as Trans-Elamite (Amiet, 1993, p.27) and that was in contact with the Indus Valley and Bactria. The period of the transition between the Bronze Age and the Iron Age, i.e. the second half of the 2nd millennium BC corresponds in Elam to the Middle Elamite Period. In the 13th-12th centuries BC, it reached the height of its power with rulers capable of regularly challenging the Kassites and who were undertaking architectural projects on a considerable scale (Curtis, 1995, pp.16-17). Little is known from the period of decline that followed the Middle Elamite period at the beginning of the Iron Age other than probable conflicts with the Assyrians and the Medes, an Iranian tribe based in west-central Iran (Godin Tepe, Nush-i Jan, Baba Jan) that first appeared in the historical record in Assyrian texts in the 9th century BC (Roaf, 1995, p.58). The Medes became an important power in Near East in the 7th and 6th centuries BC before

they got overthrown by Cyrus the great in 550 BC who established the Achaemenid Empire. Elam underwent a last period of resurgence in the 8th and 7th cent BC with the Neo-Elamite Period before the sack of Susa by Assyria in 646 BC that marked the end of Elam as a “major political power” (Curtis, 1995, p.21).

In summary, it would appear that the period of roughly 2500 to 1600 BC was marked throughout the whole geographical extent considered in this thesis, by a period of high urbanisation and of increased international relations. This period was followed by an episode of decreased density of population, especially in the east, where a number of Bronze Age settlements were abandoned. The causes for this are yet to be fully understood, but could be the result of climatic changes (increased rain-fall) or the collapse of an over-urbanised system. The changes observed in the archaeological record may also merely reflect the return to a more nomadic lifestyle less visible than the sedentary settlements of the previous period. The turn of the 1st millennium BC was marked by a renewed increase in population density or visibility. In north-western Iran, no real hiatus was observed around the transition into the Iron Age; however a change in material culture is evident from the excavated remains. This change may correspond to the arrival in the region of a new people. All of these changes have to be considered to understand the changes in metallurgy between the Bronze Age and the Iron Age.

## 5. Summary of objectives and methods

To conclude this introductory chapter, we summarize the objectives of this thesis and the steps taken to reach these objectives.

### Objectives:

- Improve the current state of the knowledge on the movement of copper and tin in and around Iran between the 3rd and the 1st millennium BC.
- Arrive to a better description of bronze metallurgy in Iran in the Iron Age to improve our understanding of the setting within which iron was adopted and perhaps the reasons for its adoption.
- Test the strengths and limitations of the use of a synthesis of old datasets in the study of Iranian metallurgy.

### Method:

- Review the current state of the knowledge on copper and iron metallurgy in Iran (Chapter 2).
- Assemble a database of compositional and lead isotope data for copper-based metal from Iran and neighbouring regions (Chapter 3).
- Identify and interpret structures within the data that can indicate technological changes and patterns of trade or recycling (Chapters 4, 5 and 6).
- Identify changes and continuities in bronze metallurgy between the Bronze Age and the Iron Age (Chapters 4, 5 and 6).
- Discuss the adoption of iron in the light of the bronze analysis (Chapter 7).



## Chapter II. Review of the development of metallurgy in Iran

Iran has been the subject of considerable archaeological interest from as early as the 17th century, but most of our modern understanding of the history of Iran derives from the considerable archaeological work undertaken in the country from the end of the 19th century onwards. Throughout the 20th century a number sites have been excavated first by French and American expeditions and then with a greater Iranian and international involvement.

Iran, with its abundant mineral resources and a long history of metal working (copper objects being known from the 7th millennium BC) is an important area for the study of the development of metallurgy and has therefore been dubbed “the heartland of metallurgy” by Pigott (1999a). Scientific research in this field started with the analysis of a few metal objects in the late 1930s (Halm, 1935; Riesch & Horton, 1937; Desch, 1938; Halm, 1939), but it is only in the 1960s that scholarly interest in metalworking in Iran really became significant with the contributions in particular of C.C. Lamberg-Karlovsky, R. Moorey, R. Pleiner, C. S. Smith and T. Wertime. In the following three decades, political unrest and war meant that the involvement of western scholars in Iran had to be greatly limited. In that period however, catalogues of museum and personal collections of Iranian metalwork were published and included a great number of elemental analysis of the objects (Ashmolean collection (Moorey, 1971), Louvre collection (Tallon, 1987), Metropolitan Museum of Art collection (Muscarella, 1988) and H. Mahboubian personal collection (Mahboubian, 1997)). These catalogues along with a couple of Ph.D. dissertations specifically focused on the subject of Iranian

metallurgy (Vatandoost-Haghighi, 1977; Heskell, 1981) attested of a growing interest in understanding copper metallurgy. However, this work was paralleled only by the Ph.D. dissertation of V. Pigott (1981) and a few studies of isolated objects when it came to ferrous metallurgy. Just before the turn of the 21st century, Pigott synthesized the work undertaken until then on both copper and iron in an excellent overview of metalworking techniques and sites bearing archaeometallurgical remains (Pigott, 1999b).

The beginning of the 21st century saw the analysis or re-analysis of archaeological sites presenting an interest for the understanding of metallurgy, notably Tepe Yahya (Thornton et al., 2002; Thornton, 2010), Tepe Hissar (Thornton, 2009), Tall-i Malyan (Pigott et al., 2003a; Pigott et al., 2003b) and the recently published project on Arisman (Vatandoust et al., 2011). Between 1996 and 2010, E. Haerinck and B. Overlaet also published in eight volumes entitled “Luristan Excavation Documents” the results of the work done by the Belgian Archaeological Mission in Iran directed by Louis Vanden Berghe from 1956 to 1979 (Haerinck & Overlaet, 1996; 1998; 1999; 2004; 2005; 2008; 2010; Overlaet 2003). These volumes are invaluable in understanding the metalwork of a region that had for a long time only been known by its looted artwork. These publications gave way to a renewed interest in the metallurgy of Luristan, resulting in the publication of several articles presenting archaeometallurgical analyses of Luristani objects (Fleming et al., 2005; Fleming et al., 2006; Begemann et al., 2008; Frame, 2010) and in research on the mine of Deh Hosein, thought to have been a possible metal source for the metalworkers of Luristan (Nezafati, 2006).

After a little less than a century of research on metalworking in Iran, we have a better understanding of the major steps of its development. However many questions remain unanswered, in particular concerning the locations of the ore sources for copper and tin,

the alloying techniques used for the production of bronze, the forging techniques used for that of iron and the way all of these metals were traded within south-western Asia.

This chapter seeks to offer an overview of the development of metallurgy in Iran. A synthesis of the development of copper-based metallurgy (II-1) is followed by an up-to-date review of our knowledge of ferrous metallurgy (II-2). These overviews present in a chronological format the current accepted paradigm of how metallurgy developed in Iran. It has already been demonstrated that such a global chronological approach has limitations as it appears that different regions had very different approaches to metallurgy (Thornton, 2009). That's why the following chapters attempt to challenge or refine the view proposed here.

1. The development of copper metallurgy
  - a. Sources of copper and mines in Iran

Copper deposits are abundant in Iran: they extend diagonally from the province of Azerbaijan in the north-west to Kerman in the south-east (see Figure II-1).

The mines of Vesnave are located in the north-central area, near the modern cities of Qom and Kashan (Holzer & Momenzadeh, 1971) (for a location of the mines and sites mentioned in the text see Figure II-3). These mines, characterised by oxide (malachite) and sulphide (chalcocite) deposits, bear traces of exploitation during the early to mid 2nd millennium as indicated by radiocarbon dates (Stöllner, 2004). An earlier exploitation of surface ores in the beginning of the 3rd millennium is also possible as indicated notably by pottery sherds (Stöllner & Weisgerber, 2000). Excavations have

proved that activities such as the sorting and concentrating of the ore took place in front of the mine.

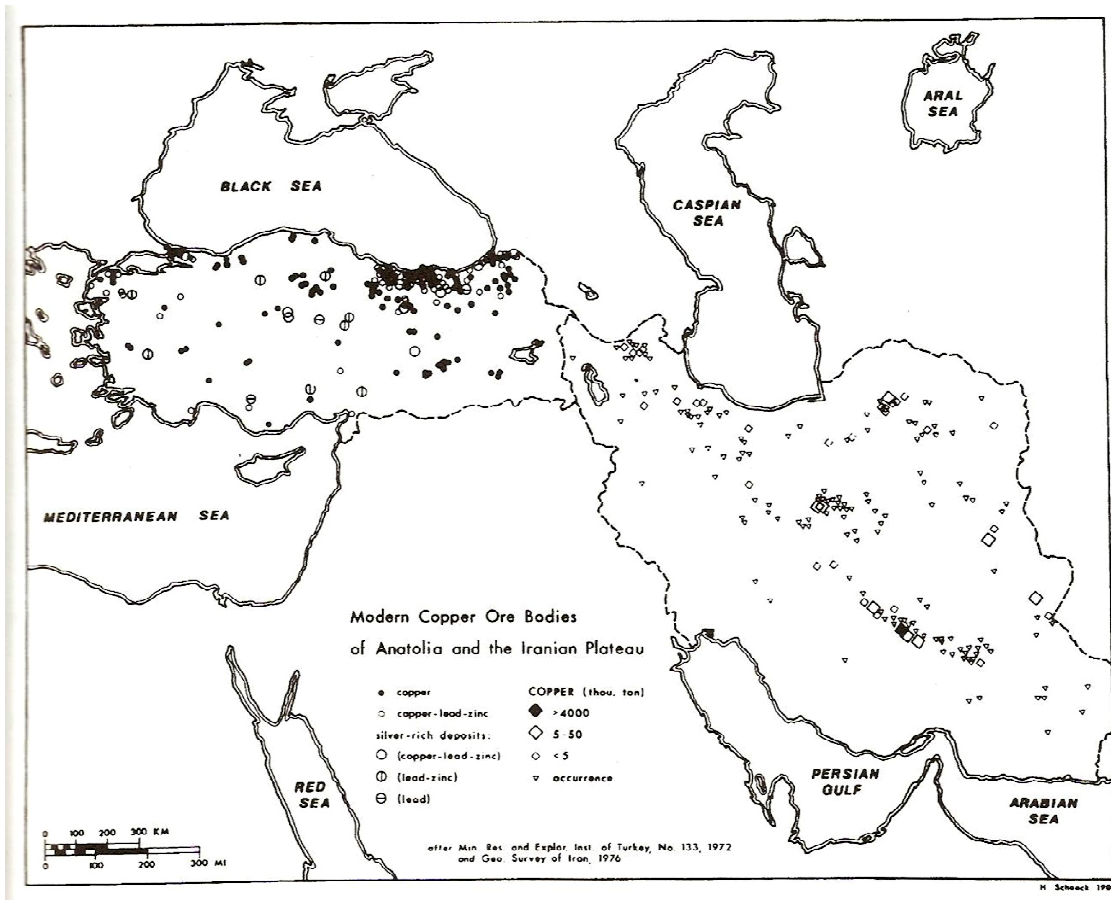


Figure II-1: Modern copper ore bodies in Anatolia and on the Iranian Plateau. From Pigott 1999, Figure 4.6, p.83

Recently Stöllner et al. (2011, pp.607-08) have attempted to determine the volume of copper extracted at Vesnave. Their calculations have resulted in an estimate of 500 to 1000 t of copper exploited in the mines, which is strikingly low when compared to some European mines (the Bronze Age mine of Mitterberg, Austria for example with its estimated 7000 to 10000 t of copper extracted). Although Stöllner reminds us that more research is needed to understand precisely the scale of exploitation of Vesnave, he concludes that it was probably an important regional centre rather than being used for a

long distance trade of copper. However, one has to wonder whether or not the volume extracted at a particular mine can necessarily be directly linked to the distance across which the metal was being traded. Pigott noted that the combination of copper oxides and sulphides at Vesnave would have been well suited for the production of copper by co-smelting in a crucible (Pigott, 1999b, p.78). He also noted the absence there of any major arsenic mineralizations, meaning that it is unlikely that this source would have been used for the production of arsenical copper during the Bronze Age.

Another possible source for copper ores is the Anarak-Talmessi complex. There, traces of ancient mining activities, if there were any, have been destroyed by the modern exploitation of the mines. However it is often considered to have played a major role in the supply of copper to different areas of Iran. Indeed, the two adjacent deposits of Talmessi and Meskani are particularly rich in arsenical mineralizations, in particular copper nickel cobalt arsenides and copper sulphides (Pernicka et al., 2011, p.635). The relatively high content of cobalt and nickel in the ores has lead scholars to suggest that it could have been a likely source for the production of the copper objects found in Susa that have particularly high amounts of nickel (Berthoud et al., 1982). The co-occurrence of these copper arsenides with native copper in the Anarak area also makes it a likely candidate for the early production of arsenical copper in the 4th and 3rd millennium BC (see below) (Pigott et al., 2003a, p.95).

The recent work of Nezafati has revealed a third area of importance in the debate for the source of early copper: on the eastern border of Luristan, the mine of Deh Hosein presents occurrences of both tin and copper minerals and shows traces of an early exploitation (Nezafati, 2006). There, polymetallic mineralizations including native copper, copper sulphides and cassiterite occur in veins and veinlets. Ancient workings were found in an area of 4.5x6 km<sup>2</sup> in the form of more than 75 ellipsoidal depressions

up to 70x50x15 meters in size. Radiocarbon dating has shown that the mine was exploited from the mid 2nd millennium to the early 1st, if not earlier. An analysis of the ore has also shown elevated concentrations of arsenic (Nezafati et al., 2006; Nezafati et al., 2009). Having conducted compositional and lead isotope analysis of Deh Hosein ore samples and Luristan artefacts, Nezafati suggested that it could well have been a major supplier of tin for Iran and even perhaps Mesopotamia and further west, possibly alongside other mines, so far undiscovered, in the same area (Nezafati et al., 2009). Begemann studied the arsenic and tin levels in Luristan objects that had lead isotope ratios corresponding closely to those of the Deh Hosein minerals. The bi-modal distribution of the tin content of these objects has led him to argue against an indiscriminate smelting of the polymetallic ores of Deh Hosein (Begemann et al., 2008, pp.29-30). If, as Begemann believes, the tin ores were recognized and separated from the copper ores, then we have to consider these two metals separately in the discussion of provenance. Begemann suggests that copper “ores from these occurrences in the eastern part of the Central Zagros Mountains were quite important providers for Luristan but did not play any visible role in the Mesopotamia” (Begemann et al., 2008, p.38). The copper from Deh Hosein and possibly other such sources in the area of the modern city of Arak and in the Sanandaj-Sirjan zone might have not been of much more than local importance, but this is not to say that the tin was not traded further or that western Iran did not also mix imported copper with local tin.

The ancient land of Magan is another potential source of copper. This area was referred to in ancient cuneiform texts as having supplied Mesopotamia with considerable amounts of copper directly during the 3rd millennium BC and probably through the emporium of Dilmun in the early 2nd millennium BC (Muhly, 1973, pp.221-25; Potts, 1993, p.391). The Oman Peninsula, identified as being the location of ancient Magan,

and in particular the Al Hajar Mountains hold an estimated 50 major copper deposits and 100 minor ones (Weisgerber, 1983, pp.270-74). Although most of these were exploited in the Islamic period, some prehistoric exploitation is also attested. The German excavations in the Oman Peninsula have indeed provided some evidence of copper production on a large scale, starting in the 3rd millennium BC. The amounts extracted there have been evaluated to about 2000-4000 t for the Bronze Age, enough to be traded via the Persian Gulf with nearby countries (Hauptmann, 1985, pp.113-15). Based on the amount of extraction waste and of smelting slag recovered for the Iron Age, it was estimated that the production of copper increased during this period. Weeks suggested a minimum production in the Iron Age of 7000 to 10000 tonnes (Weeks, 2003, p.26). The largest copper deposits in Oman are sulphide deposits with low concentrations of arsenic, nickel and antimony. Much smaller deposits of oxidized copper presenting higher levels arsenic, nickel and cobalt are also present and could have been used to produce some of the high arsenic and high nickel copper analysed from Oman (Weeks, 2003, p.110).

The copper from Oman was probably at least partly traded in the form of ingots as attested by the presence of bun-shape ingots at 3rd and 2nd millennium Omani sites such as Maysar (Hauptmann, 1985, p.80) or Al-Aqir (Weisgerber & Yule, 2003). However, this form of ingots (also called plano-convex ingots) isn't necessarily indicative of an Omani production as it is common across most of the ancient Near East (as well as Europe) as attested by examples found in 3rd millennium Susa (Moorey, 1994, p.244), at Qala'at al-Bahrain and Al-Nasaria in Bahrain from the late 3rd to early 2nd millennium BC (Højlund & Andersen, 1994, pp.377-80), at Mohenjo-Daho and Lothal in the Indus Valley (Rao, 1965, p.36) and from the 13th-12th century BC

shipwrecks of Cape Gelindonya and Uluburun in the eastern Mediterranean (Bass et al., 1967, p.78; Hauptmann et al., 2002).

The mountain range of Oman might have provided copper not only for Oman and Mesopotamia, but also for the metal-workers of the Indus Valley (Kenoyer & Miller, 1999). Links between the two regions are notably attested by the presence of Harappan artefacts in Oman (Kenoyer & Miller, 1999, p.116). However Kenoyer and Miller have suggested that the Indus Valley drew on three potential other copper sources: the regions of Baluchistan and Afghan Seistan, the region of Rajasthan, and perhaps eastern Iran (see Figure II-2). In the Afghan Seistan, where there are deposits of copper rich in arsenic, copper slag has been found in association with Helmand tradition pottery dating from the mid 3rd millennium BC. In Rajasthan and northern Gujarat, copper deposits rich in arsenic and nickel are found in the Aravalli Mountains. There, evidence of old workings has also been recorded in numerous places (Law, 2008, p.690). Copper smelting slag dating probably from the early 2nd millennium BC has for example been found at the site of Ahar (Kenoyer & Miller, 1999, p.116) and numerous slag heaps have been noted at Singhana, near Khetri (Law, 2008, p.692). Law, in a dissertation detailing potential sources for different rock and minerals found at Indus valley sites, adds another possible source of copper for Harappa: the Himalayas (Law, 2008, p.692). This region had previously been mostly overlooked by scholars, but the many copper deposits and the traces of ancient mining and smelting reported by geologists call for more work on characterising the potential sources in that area (Law, 2008, p.693). The lead isotope analysis carried out by Law does not at present support this possibility, but his study included only two of the many deposits of the Himalayas, and many others would need to be characterised to come to conclusion concerning this region.

Written records indicate an interruption of the direct trade through the Gulf between Mesopotamia and Meluhha identified as being somewhere in the Indus Valley, in the early 2nd millennium BC (Muhly, 1973). Moorey noted that this corresponds very closely with the earliest mentions of copper coming to Mesopotamia from Cyprus via Mari (Moorey, 1994, p.246).



**Figure II-2.** Sources of copper, tin and gold likely to have supplied the Indus Valley civilisation (Figure 5.1 in Kenoyer & Miller, 1999, p.108)

Another region of interest for the study of copper resources is Anatolia. There, the abundance of deposits and the existence of early traces of mining and ore processing have been the subject of much scholarly interest. In his review of potential copper sources for the production of Mesopotamian objects Moorey listed the mines of Kozlu

in central northern Turkey and the Ergani mine in south-eastern Turkey. Both are sulphide deposits that are thought to have been exploited from an early date (3rd millennium BC at least for Kozlu) (Moorey, 1994, p.247). Expeditions lead in the late 1980s (Wagner & Öztunali, 2000) revealed many more copper deposits with traces of ancient mining in various areas of Turkey, including the site of Murgul presenting traces of 4th millennium copper smelting. We cannot here give a detailed description of Anatolian metallurgy but it must be noted that there is abundant literature on the subject, notably the “Anatolian Metal” series of seminars and publications, edited by Ünsal Yalçın (2000; 2002; 2005; 2008; 2011).

Ancient texts also discuss the place of Anatolia in the trade of copper: Old Assyrian texts indicate that Anatolian copper was traded by Assyrians within Anatolia, but there is no evidence indicating that this copper would also have been exported to Assyria (Muhly, 1973, pp.206-08). Much later texts from the Neo-Assyrian period in the 1st millennium BC however indicate large quantities of copper reaching Assyrian kings from Urartu in eastern Anatolia (Muhly, 1973, p.290).

Finally, although these regions are further away from our area of interest, it is worth mentioning the Levant and Cyprus as two areas where copper is known to have been mined in antiquity. In the Levant copper deposits are known on either side of the Wadi Arabah that runs between the Dead Sea and the Gulf of Aqabah. The ancient mines of Timna in Israel and Feinan in Jordan have been the subject of much fieldwork and study since the 1960s and 1980s respectively. Both mines were exploited from the mid 4th millennium BC to the Mamluk period with an apparent interruption in the Byzantine period. Evidence for metal production in that region includes mining galleries and shafts of varied size and depth, mining tools, remains of furnaces and slag and moulds, for ingots in particular (Weisgerber, 2006; Hauptmann, 2007). Hauptmann studied the

chemistry of the copper from the sites and found a relatively pure copper. He states that the slag heaps found in Feinan are too big to consider a solely local production, and he suggests that the copper was exported towards the north and north-west as well as towards Egypt. He also states that the export of Feinan copper further to the east towards Mesopotamia is possible, especially given some lead isotopes results by Begemann and Schmitt-Strecker (2009), but difficult to trace (Hauptmann, 2007, pp.202-03).

Cyprus is also very well known for its copper. Classical sources, for example, mention the mining of copper there, and Egyptian and Syro-Babylonian texts from the Middle and Late Bronze Age mention Alashiya (generally accepted as being the ancient name for Cyprus) as a source for copper (Muhly, 1973, p.193). Copper has indeed been mined on the island periodically from the Bronze Age to modern times. Sites across the island have revealed remains of ancient mining in the form of slag heaps and mining shafts many of which have disappeared due to the modern copper exploitation (Muhly, 2005). The copper sources of Cyprus have been studied in particular by Gale and Stos-Gale who have spent many years studying their lead isotopy and comparing it to that of copper ingots and artefacts of the region (Stos-Gale et al., 1997 for example). They have notably argued that the mine of Alpiki, in Cyprus, was responsible for the production of all oxhide ingots of the eastern Mediterranean post 1400 BC (Stos-Gale & Gale, 2009). This statement has been critiqued by several scholars as it seemed difficult to reconcile with the economic and social model of copper production known on the island (see a review of the criticism in Kassianidou, 2009). Although the extent of the ingot production on Cyprus is obviously still a matter of debate, it is nonetheless clear that the island was an important producer of copper from the second half of the 2nd millennium

and it is in particular quite likely that some of the copper produced there would have found its way to Mesopotamia and perhaps further east to Iran from that period onward.

b. Early stages

The inhabitants of Iran were probably familiar with copper ore from as early as the 9th millennium BC, as attested by a copper mineral pendant found in the Zagros Mountains (Solecki, 1969). However, the first known worked artefact dates from the end of the 8th or the beginning of the 7th millennium BC (see Thornton, 2009, p.22, footnote 1 for a discussion on its dating). It is a rolled bead of cold-worked native copper found in Tepe Ali Kosh in south-western Iran (Smith, 1969). Evidence suggests that the cold-working of native copper prevailed throughout the Neolithic across Iran until the 4th millennium when smelting became a common practice: small objects made of native copper such as beads, pins and tacks were found in 6th millennium contexts in the south-west (Chagha Sefid, Tepe Sabz and in the Marv Dasht), the south-east (Tepe Yahya in particular) and the north (Tepe Sialk I) (Thornton, 2009, pp.22-54). While 5th millennium copper objects are rare in the south-west, they are still present in the north at Tepe Sialk II where native copper continues to prevail, and at Tepe Yahya in the south-east where a tack analysed by Thornton shows a very skilful working of the native copper (Thornton, 2001, pp.94-95).

Early evidence for the use of copper is less obvious in the north-west where there are to date no known copper fragments recovered from Neolithic periods. In the north-east evidence is also scarce and the contexts in which the objects of native copper were found at Yarim Tepe and Sang-i Chakhmaq East for example is unclear (Thornton,

2009, pp.47-48). These two regions will have to be the subject of further excavations and research in order to understand how and when copper was adopted there.

c. Chalcolithic developments: the smelting and casting of copper

It is difficult to assess precisely when the use of native copper made way to the smelting of copper ores, as distinguishing the two by analytical techniques is nearly impossible (Maddin et al., 1980; Moorey, 1994, p.250). However, the presence of a great number of crucible fragments containing copper slags in the Period I levels (roughly 5th millennium) of the site of Tal-i Iblis in south-western Iran could indicate an early adoption of smelting in this area (Dougherty & Caldwell, 1967). Although this has been contested due to the unclear chronology of the site and suggestions notably by Moorey (1982, p.83) that the crucibles were used for the melting of native copper and not the smelting of ores, a recent study by Lesley Frame (2004) would suggest that the crucibles were indeed used for the smelting of copper carbonate ores.

Another region where copper was smelted and cast from an early date is the north-east. There, the presence of objects such as tanged daggers, pyramid-headed pins and flat axes suggests that the casting of copper was in use in the late 5th millennium BC (Thornton, 2009, p.48).



Figure II-3: Location of the main sites mentioned in the text.

Everywhere in Iran the appearance of copper smelting is accompanied by a diversification of the repertoire of artefacts. In the necropolis of Susa in particular, mirrors and flat axes most likely cast in open-faced moulds are found in the late 5th and early 4th millennia in addition to small ornaments, pins and needles (Tallon, 1987, p.311). However only about 70 copper objects have been found in approximately 2000 tombs excavated in the necropolis (Tallon, 1987, p.314) suggesting that copper was still a precious commodity reserved to the elite. The end of the Susa I period in the first half of the 4th millennium BC shows a further expansion of the repertoire towards larger objects and in particular shaft hole axes, probably still cast in open-face moulds, similar to those found in Ghabristan (see below) (Tallon, 1987, p.315).

If the recent re-dating of the tumuli of Se Girdan (Azerbaijan) to the early to mid 4th millennium BC (Lyonnet, 2007) is correct, shaft-hole axes were also present in northern Iran around the same time as in Susa. The objects present at the site of Sialk III also attest to the use of smelting and casting in the early 4th millennium BC in northern Iran (Moorey, 1982, p.85). This was confirmed by the discovery at the site of Ghabristan II (early 4th millennium) of furnaces, a typical style of crucibles, copper ores prepared for smelting, and moulds, most of which were for the casting of shaft-hole tools (Madjidzadeh, 1979). Ghabristan-type crucibles were also found at other sites of the region, including Tepe Sialk and Arisman, attesting of an important metallurgical activity in the region (Thornton, 2009, p.43).

#### d. 4th and 3rd millennium BC: arsenical copper

Analyses of Iranian copper based objects carried out by many different scholars since the 1930s to determine their chemical compositions have shown that the prevalent metal produced during most of the 4th and 3rd millennia BC was arsenical copper. The amount of

arsenic is variable from object to object, but a weight percentage between 0.5% and 5% is typical of the period (see Chapter 5). Whether the elevation of arsenic content was intentional or not, it would certainly have modified the properties of the objects, improving the ease of casting and if in a sufficient amount also improving the mechanical properties. Northover notes that:

“Adding arsenic or antimony to copper rapidly lowers the temperature at which the alloy melts, reduces the viscosity of the melt and expands the freezing range, making good casting alloys.” (Northover, 1997, p.327)

And also:

“Small quantities of arsenic, say up to about 2%, offer very little improvement over pure coppers and it is only at about 4% and over that a good balance between strength and toughness, with properties approaching those of medium tin bronzes, can be obtained.” (Northover, 1989, p.113).

Lechtman, in an experimental study of the properties of arsenical copper and tin bronze, argues that an arsenic content of 0.5 to 1% produces a significant increase in hardness and tensile strength when the metal is heavily cold-work. This property, along with the high ductility of copper-arsenic alloys would have been sought after especially in the production of sheet metal objects (Lechtman, 1996).

Pigott suggests that at least three methods could have been used to obtain the arsenical copper found in Iran, deriving his reflection from the work of Budd et al. (1992, p.679) on copper-based metallurgy in the British Isles (Pigott, 1999b, pp.86-88; Pigott et al., 2003a, pp.95-99). The first method consists in the addition of native arsenic or of copper arsenides to molten native copper or the melting of arsenic rich native copper. This process could have been achieved in a crucible buried in a bed of charcoal (to ensure reducing conditions necessary to

prevent the loss of the highly volatile arsenic) and heated at a temperature of about 900°C. While we have almost no information on the location or exploitation of native arsenic in Iran, the presence of native copper in association with copper arsenides in the mining district of Talmessi is well documented. Whether this mine could have been the sole source for the production of arsenical copper by this simple method for sites as far as Malyan to the southwest or Shahr-i Sokhta to the east is unknown but not impossible given the importance of this deposit and the scarcity of other arsenical copper ores in the country.

The second method proposed is the smelting of sulfarsenides (also called grey copper ore or fahlerz). This would have been achieved by the smelting of the ore at a temperature of at least 1300°C. However, two arguments challenge the possibility that this method was frequently used in Iran. Firstly, it relies on the presence of weathered copper sulphide deposits where the fahlerz ore can be found. Such deposits containing substantial amounts of sulfarsenides haven't so far been detected in Iran. The second limitation is the fact that the smelting of sulfarsenides doesn't directly produce copper, but results in "matte" a copper sulphide that must be re-smelted to obtain copper. It would have been possible to roast the ore beforehand to obtain a higher proportion of copper to copper sulphide. However an alternative approach to this second method has been proposed by Rostoker et al. (1989). It consists in the "co-smelting" or "mixed smelting" – a term usually used when the intentionality is uncertain - of oxides and sulphides (such as the arsenic- rich fahlerz for example). This could be achieved in a single step by smelting the ores in a crucible or furnace. In 1999, Lechtman and Klein published the results of modern co-smelting experiments of copper oxides with arsenic bearing sulphides in both crucibles and furnaces. They were successful in obtaining high arsenic (7 to 26% As) alloys in both cases and with both enargite and arsenopyrite, though the crucible smelted ingots were cleaner (lower iron content and less iron-rich phases). The authors argued that this technique might have been wide-spread because of its success with

many different types of ores and operating conditions (Lechtman & Klein, 1999). Evidence for the mixed smelting of oxides and sulphides, rather than the smelting of sulphidic ores, is available at Shahr-i Sokhta where the matte produced from the smelting was discarded with the slag rather than processed to recover the copper (Hauptmann et al., 2003). The analysis of the artefacts from 4th millennium Susa showed important levels of arsenic, silver, antimony and bismuth leading Berthoud and Francaix (1980) to suggest the exploitation of fahlerz, but the sources for such a production remain unclear. The presence of a great quantity of slags at Tepe Hissar and numerous arsenical copper objects could also result from the use of this second method (Pigott et al., 2003a, p.96).

The third method is the smelting of copper arsenates: “arsenic- and antimony- bearing secondary minerals from the oxidized zones of cupriferous base metal orebodies” (Budd et al., 1992, p.681). This method would have been relatively easy to carry out: it simply requires the smelting at relatively low temperatures (700°C) of the ores, which can easily be mistaken for oxidic copper minerals such as malachite. This type of ore can again be found in the Talmessi-Meskani complex. This third method is characterised by the absence of slag production. The resulting arsenical copper is low in iron and has an arsenic content of less than 5% that is independent of the ratio of arsenic to copper in the ores. Although it is not always easy to distinguish which method is most likely to have been used for the production of a metallic assemblage, Pigott suggests that this third method should be considered when the arsenic content is consistently lower than 5% (Pigott, 1999b, p.88).

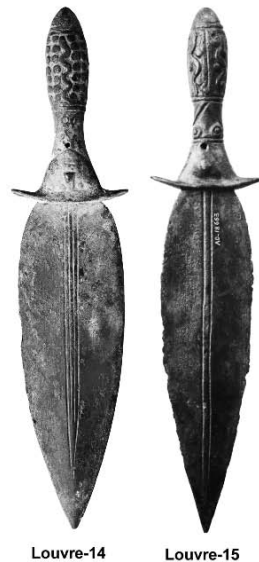
The relatively recent discovery of speiss (an iron-arsenic alloy) at several sites across Iran has started to shed a new light on the production of arsenical copper. Speiss slag was found at Tepe Hissar, in north-eastern Iran (late 4th to early 3rd millennium BC) (Thornton et al., 2009) and at Arisman in central Iran (late 5th/early 4th millennium BC to mid 3rd millennium BC) (Rehren et al., 2012). At Arisman, the excavators found large quantities of copper slag

and speiss slag attesting that both must have been produced on a large scale. Upon examination of the slag, Rehren et al. concluded that the slag, produced by an ore rich in arsenopyrite smelted separately from the copper, was intended for a controlled production of arsenical copper. Thornton et al. similarly suggested that the Tepe Hissar speiss was produced to obtain a reliable source of arsenic to make arsenical copper. Speiss has also been found at the site of Shahr-i Sokhta (first half of 3rd millennium BC) (Hauptmann et al., 2003, p.200). Given the composition of the Shahr-i Sokhta fragment, in which copper was almost completely absent, Hauptmann et al. suggested that it was unlikely to have been the by-product of copper smelting. They believed that it was smelted arsenopyrite and “can only be interpreted as a material in its own right; whether as a raw material for the production of arsenical copper or completely independent of copper metallurgy” (Hauptmann et al., 2003, p.200).

In the 4th and 3rd millennium BC, leaded-copper objects also appear in many Iranian regions: Moorey (1971) and Begemann et al. (2008) analysed several leaded-copper objects from Luristan dating to the 3rd millennium BC. Malfoy and Menu (1987) recorded comparatively high levels of lead in some objects from Susa from the late 4th millennium BC (up to 14.3%). In the early 4th and the 3rd millennium, leaded-copper objects also occur at Susa but the lead percentages rarely exceeded 2%. At Tall-i Malyan five objects from the Banesh period (3400-2800 BC) were found to have more than 1% Pb with a maximum at 5.8% (Pigott et al., 2003a). In the east leaded-copper occurs at Tepe Yahya in the late 4th and 3rd millennium (Heskel, 1981; Thornton et al., 2002; Meier, 2008), at Shahdad in the 3rd millennium (Vatandoost-Haghighi, 1977; Hakemi, 1997), at Shahr-i Sokhta in the early to mid 3rd millennium BC (Hauptmann et al., 2003) and at Tepe Hissar especially in the second half of the 3rd millennium BC (Thornton, 2009). However, as discussed in Chapter 5, leaded-copper objects remain relatively rare compared to arsenical copper objects or later tin bronzes.

In most regions, the period of the 4th and 3rd millennium BC corresponds not only to an increase of the arsenic content of copper objects but also to a general increase in the amount of copper produced, and a change in the metalworking techniques resulting in a diversification of the type of objects that could be produced. Several metalworking sites are known from this period.

In Susa, for example, techniques such as the use of lost-wax casting and bivalve moulds, as well as the use of soldering and the production of sheet-metal objects appear during the Periods II and III (ca. 3500-2800 BC) (Benoit, 2004, pp.186-87). Also in the south-west, in Malyan, Pigott has observed a small-scale production or refinement activity in the Banesh period (ca. 3400-2800 BC) (Pigott et al., 2003a). To the north of Susa, in Luristan, the objects of the late 4th and early to mid 3rd millennium BC that are not alloyed with tin (see following paragraph) present a typical arsenic content of about 1% to 3% (Fleming et al., 2005; Begemann et al., 2008). A series of daggers, that were sadly not recovered in controlled excavations but probably by the looting of graves, with a leaf-shaped blade and a patterned grip (Moorey, 1971, p.69; Begemann et al., 2008) present a higher content of arsenic (between 1.2 and 8.2% with several examples around 4%) possibly attesting of an intentional use of arsenic to improve the ease of casting or maybe change their aesthetical appearance (arsenic would have given a silvery coloration to the copper objects).



**Figure II-4. Two of the arsenical copper daggers from the Louvre collection presenting respectively 4.5 and 4.1% As. (Pl. 14 in Begemann et al., 2008, p. 61)**

Further to the north, the site of Godin Tepe provides the only clear indication of metalworking in the north-west in that period: a small-scale processing and production of copper-based metals. There copper-arsenic alloys are present from period IV (ca. 3000-2500 BC) (Frame, 2010). In north-central Iran, recent work at the site of Arisman has also shown traces of metallurgical production: slagheaps and furnaces for the production of copper and traces of production of silver by cupellation (Vatandoust et al., 2011). In the north-east, the production of this period is characterised by copper-arsenic alloys but also copper-lead alloys. So far, only the sites of Tepe Hissar and Shah Tepe have yielded evidence of metalworking (Thornton, 2009, p.50).

At the eastern site of Shahr-i Sokhta ore fragments, slag, ingots and finished objects attest of some metalworking activity. Most finished objects from the 3rd millennium have been found to be arsenical copper (Heskel, 1981, pp.97-120) and Heskel has suggest a possible production by the co-smelting method described above (see also Pigott (1999b, p.86)).

In the south-east changes in copper production also took place during this period. The repertoire of objects became more varied for example at Tepe Yahya towards the end of the 4th millennium, where heavier tools like a spatula and a “wood chisel” are produced (Thornton et al., 2002, p.1453). The presence at Tepe Yahya of arsenical copper from a very early date (late 5th millennium BC) and the lack of traces of metallurgical activities there has lead Thornton to tentatively suggest that an intra-regional trade was in place in the south-east, with Yahya possibly importing its arsenical copper from Tal-i Iblis (Thornton, 2010). Another site in the region that has yielded clear signs of metallurgical production is Shahdad, where large amounts of slag and metalworking workshops have been recovered. Unfortunately, the chronology of the site is confused and the methods of copper production remain unclear (see Pigott, 1999b, p.89 for an overview of the findings).

e. The appearance of tin bronze

Arguably, the last important step in the development of copper metallurgy is the appearance of tin bronze. While this new alloy started to occur with some regularity from the 3rd millennium BC, arsenical copper remained the dominant metal up to the mid 2nd millennium BC and it was really only towards the beginning of the Iron Age that tin bronze was consistently used for almost all copper based artefacts. The region of Luristan and the site of Kalleh Nissar in particular show the first traces of tin use. There, in the graves of area A-I, elevated amounts of tin have been detected in objects from as early as the late 4th or early 3rd millennium BC (Fleming et al., 2005; Begemann et al., 2008). Interestingly, these objects also contain some arsenic which isn't frequently found in elevated quantities in tin bronzes. Similarly in Susa a few objects found in mid-3rd millennium contexts show elevated levels of both tin and arsenic (Tallon, 1987). Following these first occurrences, a better controlled

bronze making technique was achieved and the quantity of bronze objects produced increased dramatically. The question of how these changes occurred in the different regions of Iran is studied in detail in Chapter 4. From a stylistic point of view however, it is worth mentioning here the rise of several metal working traditions (Moorey, 1982, pp.88-97): perhaps most impressive is the bronze work from Luristan with its highly decorated objects, such as spike-butted axe heads, zoomorphic finials and pin-heads, intricate horse-bits and spouted vessels. Unfortunately, a great majority of these objects have been purchased by museums or individuals on the art market and are most likely to come from the looting of Luristani graves, making their exact dating and the determination of their provenance almost impossible. The work of the Belgian excavations in Luristan has shed some light on this matter, but much is still left unknown. Therefore, it is still mostly on typological grounds that these objects are attributed to the 2nd or early 1st millennium BC (see Moorey 1971 for example).

In the north-west, a well developed tin-bronze tradition was also in place during the Iron Age but is characterized by plainer decorations, a more realistic representation of animals and humans and the use of a high repoussé technique on sheet-metal (Moorey, 1982, p.94). In Azerbaijan in particular, the site of Hasanlu has yielded not only an incredible amount of iron objects (more than 2000) but also a similar number of bronze objects and the indication that bronze-working was practised there, attested notably by crucible fragments and moulds (Pigott, 1999b, p.91).

Also noteworthy is the development in Susa of a type of monumental bronze work in the second half of the 2nd millennium BC, of which the Statue of Queen Napirasu and the monumental bronze gates kept in the Louvre are great examples (Moorey, 1994, p.272). In this region, metal production is attested at the site of Haft Tepe where an unusual kiln dating from the 13th century BC was excavated. It was probably used, amongst other crafts, for the production of bronze objects (Negahban, 1991, p.18).

Tin-bronze remained extremely rare in the north-east and in the south-east until the 2nd millennium BC. From the early 2nd millennium however, the sites of Tureng Tepe, Hissar, Shahdad and Tepe Yahya in particular present objects with elevated levels of tin. They are for the most part comparable to objects from the Bactra-Margiana Archaeological Complex in Central Asia (Thornton, 2009, pp.35, 53). Thornton considers that the late appearance of tin-bronze in these regions might reflect a conscious choice to keep using other alloys but was “overcome by the influx of BMAC peoples at the very end of the 3rd millennium BC” (Thornton, 2009, p.53). He goes further in suggesting that the tin sources of Afghanistan might have remained unexploited until the arrival of BMAC people in this region around 2000 BC, but emphasizes heavily on the purely speculative character of this suggestion until more research is possible in Afghanistan (Thornton & Giardino, 2013).

Iron artefacts started to appear in the middle of the 2nd millennium BC, but it is only toward the turn into the 1st millennium that iron objects were produced to fulfil the same functions bronze implements. As we have seen, bronze remained extensively used for a wide range of functions well into the Iron Age. Although it is difficult to assess exactly when bronze eventually starts to decrease in importance, it is usually considered that by 600 BC iron-working was a well established industry and that iron had replaced bronze for the manufacture of many types of objects and in particular tools and weapons.

f. The problem of the origin of tin

Tin was used extensively in all of the Near East from the 3rd millennium BC to produce bronze, an alloy stronger and harder than unalloyed copper or arsenical copper. But in spite of its presence in bronze objects from this period onwards, its provenance remains somewhat of a mystery. The absence of known tin sources in Iran or Mesopotamia until very recently, and

the evidence provided by ancient texts seemed to indicate that it was traded over long distances.

Although the origin of tin has been the subject of much debate over the past decades, we still rely largely on speculative theories as both textual and archaeological evidence remain sparse. Today though, most scholars seem to agree that Afghanistan is a likely provenance for Near Eastern tin. There, a Soviet geological survey and a French survey by Cleuziou and Berthoud have indicated that tin is present in two main geological formations. One extends from the north-east diagonally to the south of Kandahar. The second is located in western Afghanistan, to the south of Herat and in the Sarkar valley where tin-bearing sands are found in association with copper ores (Cleuziou & Berthoud, 1982). It is worth mentioning here that tin-bronze, lapis lazuli and gold appear simultaneously in Mesopotamia in the Royal Cemetery of Ur and at Ebla notably (Muhly, 1985). These three minerals all occur in Afghanistan, notably in the north-east of the country which is the most likely source region for Near-Eastern lapis (Beale, 1973, p.137). It has therefore been suggested that they might all have been traded from Afghanistan to Mesopotamia along the same routes (Stech & Pigott, 1986). However, although lapis was probably worked in eastern Iranian sites such as Tepe Hissar and Shahr-i Shoktha, very little tin-bronze has been found there (see Chapter 4).

In an expedition of the 1970s, Wertime and colleagues found cassiterite mines in Egypt in close proximity to inscriptions dating to Pepi II (22nd century BC). There is however at present no evidence that this tin would have been exported to other areas of the Near East (Wertime, 1978), and given the general lack of tin bronze in Egypt before the 2nd millennium BC it is doubtful that these sources would have been exploited before that time (Muhly, 1978). Similarly tin is present on the Arabian Peninsula but does not appear to have been exploited before the 1st millennium BC (see summary in Weeks, 2003, p.167).

In the 1970s, scholars became aware of the existence of tin deposits in Central Asia (Crawford, 1974): in the region of the Zeravshan valley (modern Uzbekistan and Tajikistan) the sites of Karnab, Lapas and Mushistan provide evidence of the mining of tin during the Andronovo-Tazabag'jab period in the first half of the 2nd millennium BC (Alimov et al., 1998; Boroffka et al., 2002; Parzinger, 2002; Parzinger & Boroffka, 2003). Although it has been estimated that the production at Karnab exceeded that of local need, it is not known how far and to what extent the tin from the Zeravshan valley could have been exported.

K. A. Yener and G. J. Laughlin have suggested that the mine of Kestel with the processing village of Göltepe in the Taurus Mountains of southern Turkey could also be a viable source of cassiterite tin ore in the 3rd millennium BC (Laughlin & Todd, 2000; Yener & Vandiver, 1993). The true importance of this mine has been the subject of much discussion (Weeks, 2003, p.168): the relatively low concentration of tin in the ore (0.1-1% Sn) and the complex concentrating processes that would have been necessary to produce tin at this site has lead scholars to wonder whether the mine was in fact exploited for gold or lead rather than tin. However, more than 20 years after the discovery of the mine, most objections have been addressed and Kestel/Göltepe still stands as an important clue for the question of the origin of tin especially in the 3rd millennium BC, as the Central Asian mines probably weren't in use until the 2nd millennium (Yener, 2009).

As seen above, a source of copper and tin has recently been found on the eastern edge of Luristan at the mine of Deh Hosein. This source and possibly other deposits in this area that have yet to be discovered, would probably have played a crucial role in the production of the famous Luristan bronzes. In Chapter 4 we attempt to assess the importance of this source in supplying tin for other regions.

Written sources also give us some indication on the provenance of the tin. These sources, compiled by Muhly in the 1970s, would appear to indicate that the tin used in Mesopotamia came from somewhere further east (Muhly, 1973). Most of the documentary evidence dates to the Old Assyrian and Old Babylonian periods in the early 2nd millennium BC. It suggests that the tin came to sites in Mesopotamia either from north-western Iran via passes through the Zagros or up the Euphrates from Susa or the Persian Gulf (see Figure II-5). From Sippar and Eshnunna, it was shipped to Mari that appears to have acted as an entrepôt for tin trade with western sites in Syria, the Levant and maybe Egypt (Muhly, 1973, pp.292-301; Maddin et al., 1977, p.41). An important series of Assyrian texts found in Kanesh documents the trade of large quantities of tin and textiles between Assur and Anatolia (Larsen, 1976, pp.85-92). The original source of tin for this trade however remains unclear. Muhly suggested the possibility of a source in north-western Iran, although at the time he wrote, this wasn't supported by any geological evidence. He listed several trade routes between Iran and Mesopotamia possibly used for tin: the tin from north-western Iran could have reached Assyria via passes in the northern Zagros at the level of Hasanlu and Dinkha Tepe. Passes further south at the level of the Diyala River, might also have been used either to trade the tin from north-western Iran or from further east possibly alongside lapis using the Khorasan Road. Finally it might also have reached Mesopotamia sent from Susa to Larsa along the Karkheh or Karun Rivers (Muhly, 1973, p.328). The movement of tin between Susa and Mesopotamia, although clearly attested, for example in texts from Mari from the 18th century BC reporting that most of Mari's tin was obtained through diplomatic exchanges with Susa (Potts, 1999, p.169), is likely to have been frequently interrupted due to the fluctuating relationship between Elam and its neighbour.

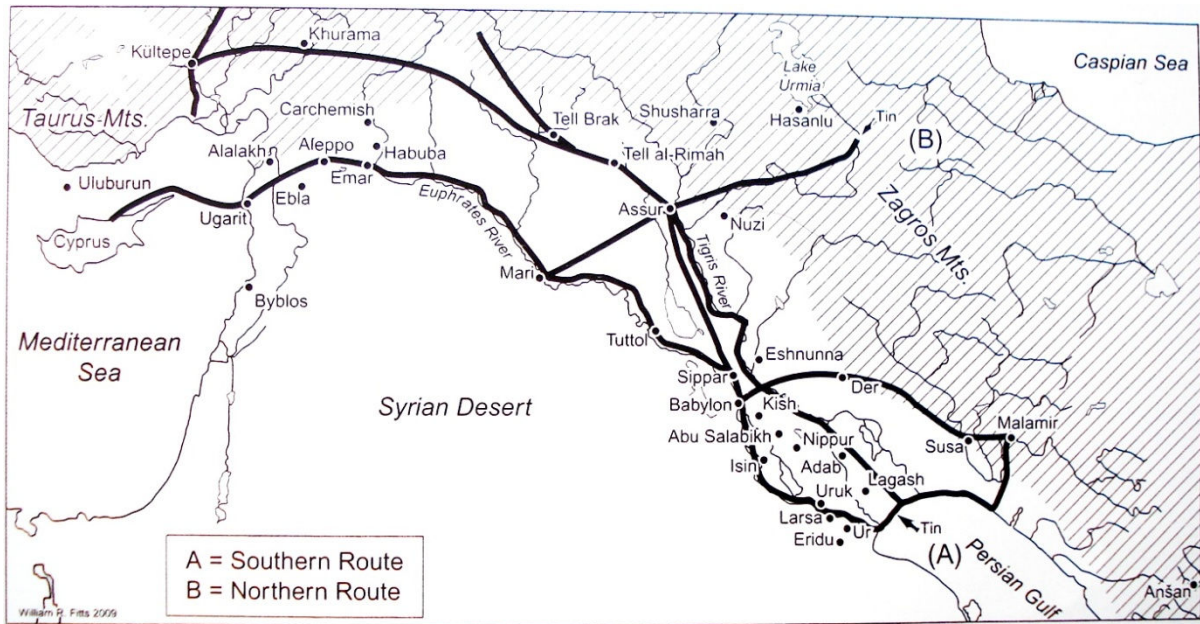


Figure II-5. Map showing the possible trade routes used to bring tin into Mesopotamia and to redistribute it to the west (Fig 27.2 in Pigott, 2011, p.278)

Documentary and archaeological sources also indicate strong contacts between Mesopotamia and Meluhha, between ca. 2500 and the fall of Harappa around 1900 BC. Passing points for this Persian Gulf trade were Magan and Dilmun. This route could have been used for trade of eastern goods such as lapis, carnelian and tin. However there is no indication to what the original source of the tin might have been (Weeks, 2003, pp.179-80).

Another written source comes from the Greek Strabo. He says of the people of Drangiana (modern Sistan in south-eastern Iran) that they have ‘only scanty supplies of wine, but they have tin in their country’ (*Geography* 15.2.10 see Cleuziou & Berthoud, 1982). Modern research has indeed revealed traces of tin in the Dasht-i Lut near Sistan (Stocklin et al., 1972) but these are not very significant compared to the deposits of Afghanistan (Pigott, 1999b, p.81).

Although evidence of tin trade in the late 2nd millennium BC is scarce, it is reasonable to suppose that at least some of the tin either originated in Iran or transited via Iran before it

reached Mesopotamia and was redirected to the west probably as far as the Eastern Mediterranean, making the study of Iranian metallurgy a key to understanding the dynamics of copper, bronze, tin and iron production and trade in the Near East.

The form in which tin was traded is also unclear as very few tin ingots or tin objects have been found in our region of interest (Moorey, 1994, p.301). This had led Cleuziou and Berthoud to suggest that it was traded mostly in the form of ores (Cleuziou & Berthoud, 1982, p.16). One has to wonder however if the high volume of ore compared to metallic tin wouldn't have deterred merchants from the trade of ores. Moreover, although uncommon, metallic tin objects and ingots are not unheard of in the Mediterranean and the Near East. In the late 1970s Wertime organised a seminar to discuss the available evidence concerning source, trade, and use of tin in the Near East and Eastern Mediterranean. Tylecote reviewed the evidence of tin finds in Europe and the Mediterranean (Tylecote, 1978), while Selimkhanov presented the surprisingly relatively high number of analysed tin objects from the Caucasus (Selimkhanov, 1978). Muhly discussed the discovery of two Bronze Age tin ingots in the port of Haifa, Israel (Muhly, 1978), which added to the evidence provided by 8kg of a white material tentatively identified as tin found on the Cape Gelidonya shipwreck from the 12th century BC (Bass, 1967, Maddin et al. 1977, p.44). In the following decades, more discoveries in the Mediterranean provided striking evidence for the trade of tin ingots: approximately a ton of tin ingots alongside about 10 tonnes of copper ingots were recovered on the Uluburun ship-wreck off the coast of Turkey, dated to about 1300 BC (Hauptman et al., 2002). Lead isotope studies have been carried out on these ingots and it has been suggested that the tin came from at least two different sources (Pulak, 2000), but the location of these sources remains elusive (Pigott, 2011). A number of other wrecks off the Carmel coast, Israel, are also reported to have yielded metal ingots. The Hishuley Carmel wreck in

particular, dated to the mid to late 2nd millennium BC, carried 206 kg of tin in ingot shape (Galili et al. 1986, Galili et al. 2013).

A small number of tin objects were also recovered in the Near-East. A tin bracelet was found in Thermi IV and a ring and a flask were recovered in Egypt and dated to the 14th century BC (Maddin et al., 1977). Closer to our area of interest, five bracelets and two rings from the 19th century BC were found in Tell ed-Der, in Mesopotamia (Van Lerberghe & Maes, 1984). In Kültepe, where tin was a common trading item between Assyrians and Anatolians, three flat circular objects that might have been tin ingots were found, but this identification hasn't to our knowledge been verified by chemical analysis (Özgüç, 1986, pp.77-78; Dercksen, 2005, p.21). Finally Weeks reports a tin ring from Tell Abraq in Oman dated to the late 3rd millennium BC. This is to our knowledge the first (and only) known tin artefact from the region (Weeks, 1999, p.59).

Another possibility could be that part of the tin was being traded as pre-alloyed ingots of bronze. However, as shown in Chapter 4, evidence for tin-bronze ingots in our area of interest, doesn't exist from periods earlier than the Iron Age.

#### g. Production processes

Three main processes have been proposed as plausible ways in which ancient metal-smiths could produce tin bronze (Bray, 2009, Ch.5). In 1979 Shell suggested that bronze could be made either by mixed-smelting of copper ores with cassiterite (tin ore) or by separate smelting of copper and tin and then mixing of the molten metals (Shell, 1979, p.252) which would have been a possible way of obtaining high tin bronzes (Northover, 1988, p.52). He also mentioned the possibility of accidentally producing an alloy with a very small tin content (around 2%) if

cassiterite is added to a copper smelt. In 1988 Northover (1988, pp.51-52), building on the experimental work of Charles, added another possible process: the reduction of cassiterite in contact with molten copper. He suggested that this method would result in the production of relatively low tin bronzes (around 6 to 8% tin); however, if this technique was repeated by putting cassiterite in contact with these low tin bronzes, higher tin contents could be obtained.

Bray (2009, Ch.5) proposed a way of distinguishing between these methods by looking at the chemical content of the objects. This work is based on the fact that the addition tin by putting cassiterite in contact with molten copper or bronze is the only one of the three methods for which the amount of tin added depends on the length of time for which the copper was molten. In the case of co-smelting or addition of tin metal to copper metal, time shouldn't affect considerably the eventual amount of tin in the resulting bronze. Bray explained how, when copper is heated or worked, its chemical content changes (see also Bray & Pollard 2012). The levels of arsenic and antimony in particular tend to decrease as these elements are lost in an oxidising atmosphere. Therefore, as cassiterite is reduced in contact with molten copper, the level of As and Sb in the copper decreases. If the addition of tin requires several successive heating cycles, the copper will be gradually depleted in As and Sb at each cycle. Bray was able to show that for a well defined metal group, this depletion can be made evident by plotting the average arsenic or antimony levels against the tin percentage of these objects, thus proving the use of this third alloying process.

The oldest textual reference to the production of bronze in the Near East comes from the Early Dynastic III period (mid 3rd millennium BC) in Mesopotamia. It gives a "standard" recipe for the proportions of tin and copper:  $13^{1/3}$  shekels of tin for 80 shekels of copper (ie. 13.9% Sn. The percentage tin needed to produce bronze, as indicated by recipes, doesn't change much in the following centuries: ratios of copper to tin of six to one, which translates to about 14.3% tin, are common from the pre-Sargonite period to the Neo-Babylonian Period

and a ratio seven to one (12.5% Sn) is also known from the Ur III period (Muhly, 1973; Eaton & McKerrell, 1976, p.179; Moorey, 1994, p.252). Texts from Ebla, in western Syria, on the other hand, indicate a relatively wide range of tin contents, from 0.83 to 21.55 % Sn, but the very low and very high tin values are not the norm and most types of object are produced with between 11 and 17% Sn (Archi, 1993).

As shown in Chapter 3, relatively few metallographic analyses and even fewer hardness measurements have been carried out on Near Eastern metalwork. This means that it is very difficult for us to assess the amount of smithing to which Iranian objects were subjected. Although studying the development of smithing would undoubtedly help understand the development of metallurgy across our region of interest and the transition into the Iron Age, the lack of data has driven this thesis to pay little attention to techniques such as cold-working, annealing or the working of sheet-metal that would probably have been in use in the Near East from an early date (Moorey, 1994, pp.269-76).

## 2. The appearance and development of ferrous metallurgy

### a. Ore sources and mining

Iron is a common element in the Earth's crust (4th most common after oxygen, silicon and aluminium). It is notably present more abundantly than other metallic ores, including those used by humans from an earlier date such as copper, silver, gold, lead and tin (Killick & Fenn, 2012, p.561).

In Iran the major sources of iron ores are situated in central and south-eastern Iran with minor occurrences in the regions of Amlash and Luristan (see Figure II-6). However a great number of iron artefacts have been recovered the north-western part in the country and in particular at

the site of Hasanlu in the province of West Azerbaijan where there are no iron ores. Iron might therefore have been the subject of an organised trade network either within Iran or with nearby countries. In Anatolia for example, iron ores are fairly common and this region could have provided western Iran and Mesopotamia with ore, metal or finished products. On the other side of the Caucasian range, in regions on the coast of the Black Sea, iron rich sands appear to have been smelted since the Iron Age and could also have been a source for Iranian iron. However the use of small low-grade iron deposits that would not be considered economically viable by today's standards is also very likely (Moorey, 1994, p.280) and makes the identification of the sources used for the production of ancient iron difficult.

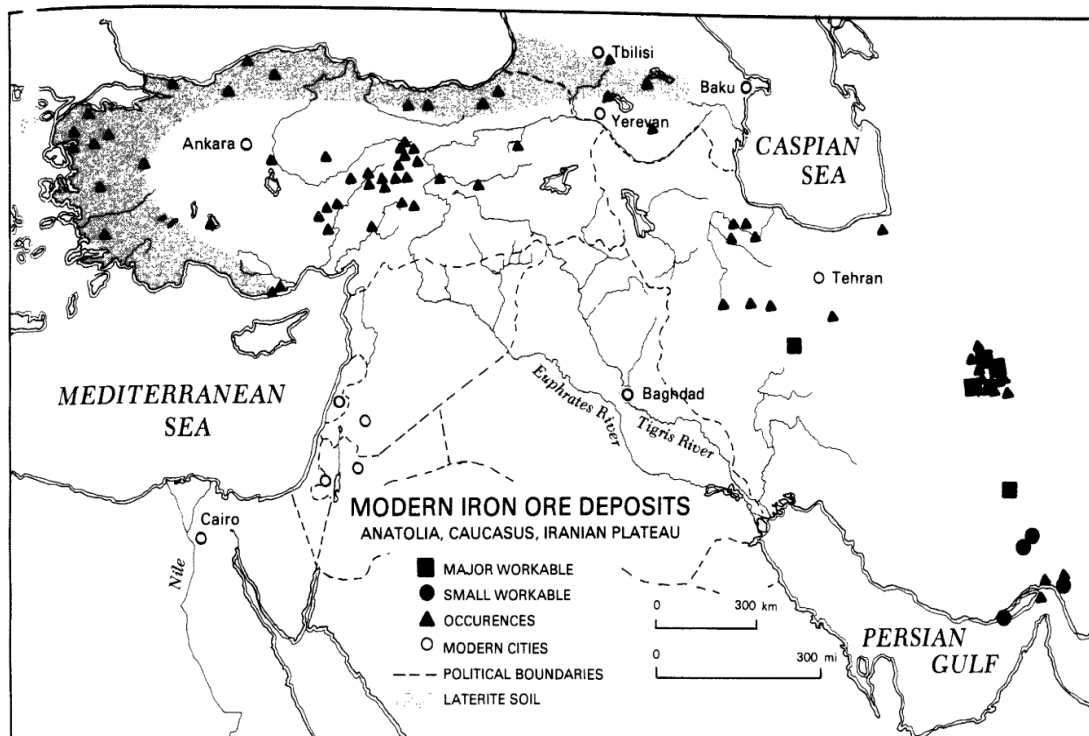


Figure II-6: Location of modern iron ore deposits in Anatolia, Iran and the Caucasus. (Drawing by Jon Snyder after Pigott 1981:Fig.2) (Pigott 1989, p.69)

Assyrian textual sources reviewed by Pleiner and Bjorkman (1974) mention iron received from many different locations to the west and north-west of Mesopotamia and in particular

from Anatolia and Syria. Their review as well as other studies on the subject found that there was no mention of iron coming from Iran (Moorey, 1994, p.281).

b. Chronological outline of the development of ferrous metallurgy

Our understanding of the development of iron metallurgy in Iran is greatly hindered by the fact that, so far, we haven't recovered any sites featuring remains of iron smelting or forging. The instability of iron, i.e. its likeliness to corrode easily, doesn't help the matter, as it means that iron finds are few, fragile and not always well recorded. Metallographic examinations of the artefacts are only rarely undertaken as they are unfortunately destructive and the amount of uncorroded metal left in the objects is often low, making it difficult to interpret the microstructure. However, the excavations in Iran in the past century have yielded enough iron objects to at least get an idea of how common the metal was at different stages of the Iron Age and what it was used for. Moorey and Pigott in particular have participated in establishing an outline of the chronology of iron use in Iran (Pigott, 1981; Moorey, 1982; Pigott, 1999b).

Early occurrences

The first use of iron ores by humans in the Near East is likely to date back as early as the 9th millennium BC when they were used as red ochre pigments, long before the appearance of smelting (Schmandt-Besserat, 1980). This practice, as well as the use of iron as a fluxing agent during the smelting of copper and the possibility of accidentally producing iron as a by-product of copper-smelting (Moorey, 1994, p.279), probably familiarized the early miners with the different iron ores. But it is not until the middle of the 2nd millennium BC that regular smelting of iron started to occur.

The discovery of speiss slag at three Iranian sites (see above) attests to the production as early as the 4th millennium BC of an iron-arsenic alloy. Although in all cases it was concluded that speiss was probably produced to obtain a reliable source of arsenic to make arsenical copper, Thornton et al. pointed out that it was nonetheless the earliest trace of intentional smelting of iron ores. They suggested that, if this technique of producing arsenic for copper metallurgy became widespread, and if attention was given to the unintentional “iron-like” alloy accidentally obtained in these smelts, then Iran could also have been a “heartland” of iron metallurgy (Thornton et al., 2009, p.315). Interestingly, a fragment of löllingite (an iron arsenide) was also found at Harappa amongst a horde of copper-based objects (Hoffman & Miller, 2009, p.241) attesting again of a certain awareness of iron ores.

Other indications of an early familiarity with iron include the presence of iron at the site of Geoy Tepe dating from the late 3rd millennium BC (Burton-Brown, 1950), the mention of an ‘iron dagger from Anshan’ in a Mesopotamian text from the same period (Moorey, 1994, p.287) and the presence of iron rings in the early 2nd millennium levels of Tall-i Malyan (Thornton et al., 2009, p.315). In Mesopotamia, iron similarly occurs very occasionally in 3rd millennium contexts and rare textual references from the same period also exist (Moorey, 1994, p.287).

#### Iron Age I: 1450/1350 – 1100 BC

In the period between 1450/1350 and 1100 BC, defined by Pigott (1981) as Iron Age I, iron occurs very rarely: to our knowledge only four objects have been recovered in Iran that can be dated somewhat securely to that period (see Figure II-7) (Pigott, 1981; Pigott, 1977). A ring has been found at the site of Hasanlu in Azerbaijan. A dagger and a punch have been found at Tepe Sialk in the centre of the country and an iron dagger has been recovered in Giyan in

Luristan. All four objects come from graves, which could suggest that iron was a precious good in that period, but might also be due to better conditions of conservation in graves or simply be coincidental. In addition to these four objects, tombs from the Pusht-i Kuh graveyards excavated by Louis Vanden Berghe in Luristan as part of the Belgian Archaeological Mission in Iran (BAMI) have yielded iron objects that could possibly be as old as Iron Age I. However, the graves from these Iron Age cemeteries have often been reused, making it difficult to date with confidence the burial goods. In the recent publications of the BAMI excavation reports, Overlaet indicates that “Not a single one of these [the iron objects] could be dated with absolute certainty to the Iron Age IA.” (Overlaet, 2003, p.151). The chronology used in this report is a revised chronology for the Pusht-i Kuh area, based mostly on distinctive pottery groups (Overlaet, 2003, pp.8-10). Iron Age IA is defined as the period between 1300-1250 and 1150 BC. In the following period, Iron Age IB - IIA, dated to roughly 1150-900 BC, iron jewellery starts to appear in the tombs. However, as this period corresponds for the most part to what Pigott calls Iron Age II, these objects are treated in the following paragraphs and maps as Iron Age II objects.

Given the dearth of objects dating from the first three centuries of the Iron Age, hardly anything can be said about the nature and magnitude of iron-working in this period. It appears to be merely a prelude to its development on a bigger scale at the very end of the 2nd millennium and mostly at the beginning of the 1st millennium. One is forced to notice that although this period is called “Iron Age”, the transition from Bronze Age to Iron Age has little to do with iron and in fact probably refers mostly to changes in other aspects of the material culture (see Chapter 1).



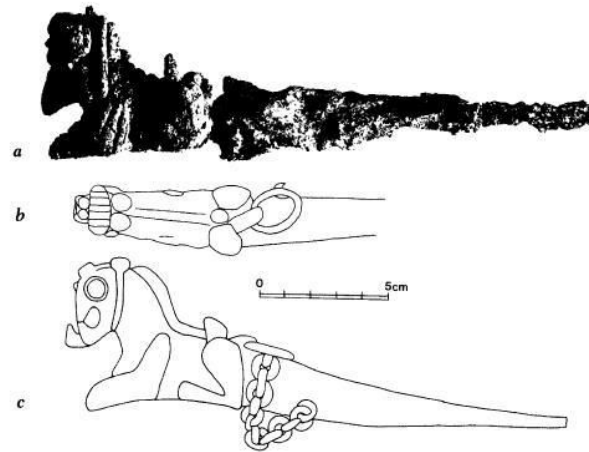
**Figure II-7: Iron finds in Iron Age I contexts (1450/1350 BC - 1100 BC)**

### Iron Age II: 1100 – 800/750 BC

The number of objects deposited between 1100 to 800/750 BC (Iron Age II) is indeed much higher. Iron becomes more widespread and the repertoire of objects is wider. From this period, the iron yielding sites known to us so far are concentrated in three main regions: Azerbaijan in the north-west, Talish and Gilan in the north, near the shores of the Caspian Sea, and Luristan in the West. In Azerbaijan, the site of Hasanlu excavated by Dyson in the 1960s (Dyson, 1965), is without doubt one the richest of the region in iron with over 2000 iron artefacts. Its citadel was destroyed around 800 BC. Most of the artefacts found in Hasanlu come from this destruction level (IVB) and a small number come from the graves outside the citadel and from the Iron Age III levels of the citadel. The number and diversity of artefacts found in Hasanlu, as well as the fact that most of them derive from an occupation context, mean that this assemblage is of particular interest for the study of ancient iron.

Unfortunately, the objects recovered are for the most part in a relatively bad state of conservation making metallographic examinations delicate. Weapons constitute the biggest part of the iron objects recovered there. Arrow-points and spearheads in particular (about 700 and 500 respectively), but swords, daggers, mace-heads and armour plates are also very common (Pigott, 1989; Muscarella, 1989). All of these types were however still made in bronze too and swords and daggers were sometimes produced as bi-metallic objects (in both metals). Although there is no direct evidence of smelting or smithing in the archaeological record at Hasanlu, the sheer number of artefacts recovered there suggests that it could have been an important production centre and potentially a distribution centre on a route linking Assyria to central Iran (Pigott, 1989, p.71). This hypothesis is supported by the presence of magnetite boulders used in the wall foundations and in the floors indicating the presence of iron ore either locally or imported to Hasanlu. Another element supporting a local production is the typology of the objects, a number of which are unknown from Assyrian reliefs. In some cases they appear to be “mass produced and deliberately standardized” (Pigott, 1981, p.71). A typical example of this “standardization” is the series of 86 small tanged iron knives found at Hasanlu but also at the nearby site of Dinkha Tepe.

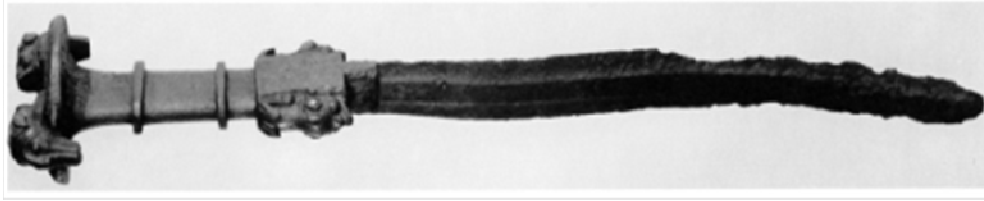
At Dinkha Tepe, about a hundred iron objects were recovered in 10 of the 68 graves excavated. They were mostly ornaments (pin, bracelets and rings) and a few weapons and tools (Muscarella, 1974). Also in Azerbaijan, iron ornaments were recovered in women burials at the site of Haftavan (Burney, 1970; Burney, 1972).



**Figure II-8. Bi-metallic lion pins recovered in Hasanlu (bronze body cast onto and iron shaft). (Fig. 13 in Pigott, 1989, p. 74)**

In the regions of Talish and Gilan, also in northern Iran but closer to the Caspian Sea, a significant number of iron objects have been recovered in the cemeteries explored in 1901 by the expedition directed by H. De Morgan. In the dolmens excavated at Agha Elvar, Chir-Chir, Chagoula Derre and Khalil Dehlil, the excavators found, amongst others, swords, spear-points, and knives made of iron (De Morgan, 1905), now kept in the Musée des Antiquités Nationales in France. Two other sites from the area south-west of the Caspian Sea have also yielded iron objects: at Ghalekuti and Lasulkan, excavated by a Japanese team in 1960, a few iron weapons and tools have been found in burial contexts (Egami et al., 1965).

In Luristan, the most famous group of iron artefacts is without doubt a series of decorated swords. In 1989 Muscarella had listed 88 examples (Muscarella, 1989, p.349), all of remarkably similar typology: a relatively short blade, a circular pommel with zoomorphic and anthropomorphic decorations and, in most cases, crouching lions on the guard (Figure II-9).



**Figure II-9. Luristan short-sword from the Metropolitan Museum of Art. (Pl. 23, Fig. 1 in Leffert (1964, p.63))**

These swords have aroused the interest of many scholars both from a stylistic and a technical point of view. Indeed, multiple studies have shown that they are made of many different pieces assembled together with rivets in a very singular fashion, unexpected for a metal such as iron (see Figure II-10 and Chapter 7). However, none have been found in controlled excavations: discovered by looters, they started to emerge on the antiquity market in the 1920s. Therefore, their provenance and age remain uncertain and have been the subject of much discussion (see for example Moorey 1991). Interestingly, the radiocarbon dating of two examples has placed them at the very beginning of Iron Age II: between 1100 BC and 1000 BC (Rehder, 1991). This early date and their surprising technology, showing an amazingly dexterous forge-work coupled with an apparent ignorance of the properties of iron, have lead scholars to consider them as representative of a transitional period between the use of bronze and the use of iron. This idea is studied in more detail in Chapter 7.

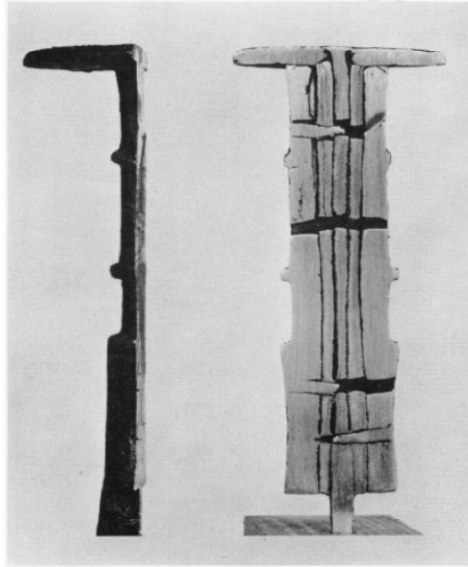


Figure II-10. Hilt of a Luristan sword cut for examination. (Fig. 4 in Bird & Hodges (1968, p.217))

As mentioned above, Vanden Berghe's expedition in the Pusht-i Kuh in the 1950s is another source of information on Luristani iron. Iron started to appear in the tombs in the Pusht-i Kuh Iron Age IB-IIA period (ca. 1150-900 BC) in the form of simple finger-rings, bracelets and bi-metallic pins (with a bronze-head and an iron shaft – see Figure II-11). Bronze was still very common in this period, as attested by the spike-butted axe-heads, bronze idols, flanged daggers, arrowheads and bracelets, pins and rings also found in Iron Age IB-IIA tombs. By period IIB (ca. 900-800/750 BC), iron daggers, knives and swords began to appear but arrowheads were still exclusively made of bronze (Overlaet, 2003).



Figure II-11. Bi-metallic pins and bracelet attributed to Luristan (Ashmolean Museum n° 1931.33, 1951.277 and 1951.329)

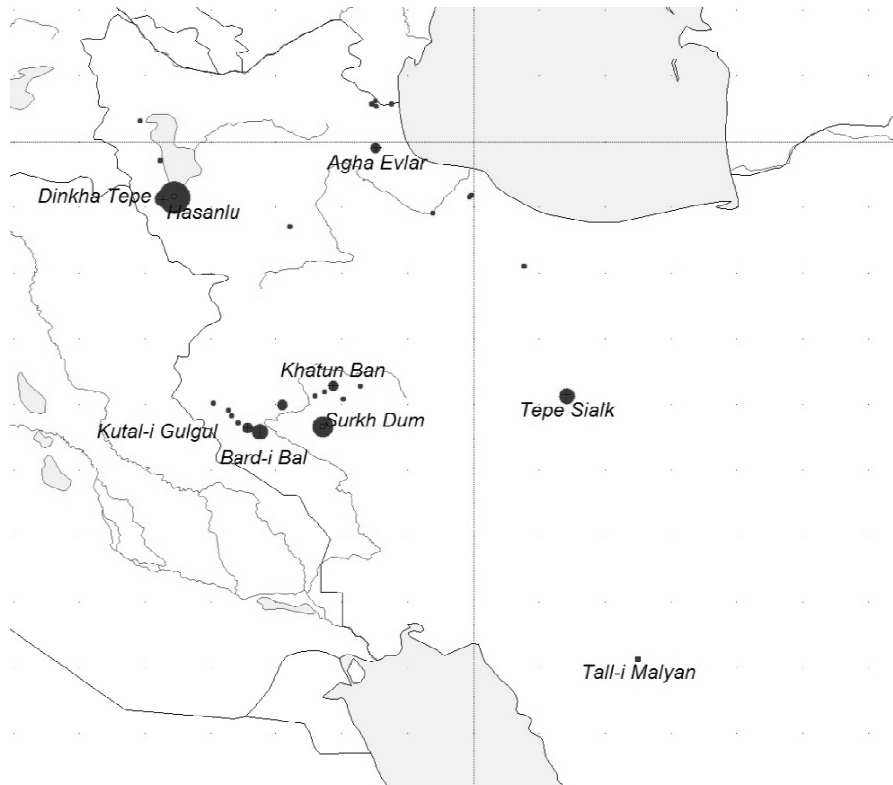
Iron was also found in some cemeteries located in the Pish-i Kuh, another part of Luristan, such as the sites of Kazabad (Stein, 1940) Giyan (Contenau & Ghirshman, 1935), Guran (Thrane, 2001) Khatunban (Schmidt et al., 1989), Cheshmeh Mahi (Maleki, 1964) and in the sanctuary of Surkh Dum (Schmidt et al., 1989). Interpretations are made difficult by the fact that many of the sites had been looted before the arrival of the archaeologists, and that in some cases the excavation reports are incomplete or unclear. However, in most of the tombs attributed to an Iron Age II context, the iron finds seem to be comparable to those found in the Pusht-i Kuh: mostly bracelets and rings and a few blades and daggers. The settlement of Baba-Jan (Goff, 1978), also in the Pish-i Kuh, is so far the only site in Luristan where iron objects have been found in an occupation context. Although most iron objects probably come from an Iron Age III context, a sickle, an arrow-point and a nail are of possible Iron Age II date and would represent the only examples of tools for that period in Luristan. Also remarkable is the Pish-i Kuh site of Kamtarlan, which is possibly the only place in Iran to present evidence of iron smelting in a level of Iron Age II or III context (ca. 1000/750-550 BC):

“Area 3 had a pavement of iron slag and may have been an iron smelter. Room 1 was also floored with iron slag. [...] There was a bin in the floor and a cubicle (room 2) adjacent to room 1. Room 1 contained a red-burned-clay oven.” (Schmidt et al., 1989, p.16)

In addition to the slag, three iron knives were found at Kamtarlan. These finds do not permit to assess the scale of the iron-making activity carried out in Kamtarlan, if any; however they are truly unique in Iran so far and could be an interesting starting point for further excavations in the area.

Finally, in central Iran, a total of 80 iron artefacts were found in the Iron II graves of Sialk B (Ghirshman, 1939). Some iron tools and implements (forks, fragments of horse bit) were

present, but they are far outnumbered by weapons (mostly simple tanged daggers or bimetallic daggers) and personal ornaments (bracelets, anklets and even a fragment of torque).



**Figure II-12: Iron finds in Iron Age II contexts (1100-800 BC)**

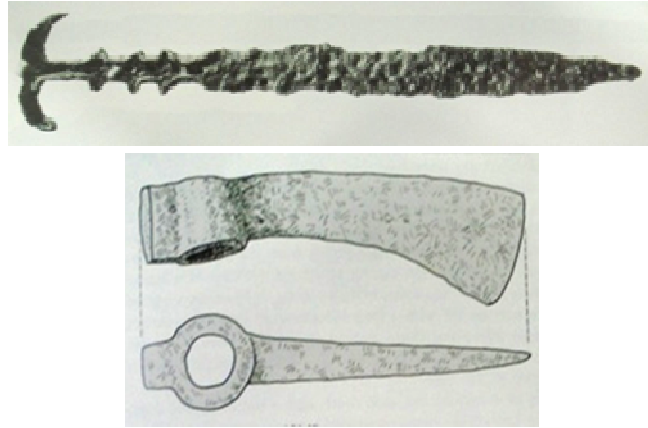
### Iron Age III: 800/750 – 600 BC

While in Iron Age II, iron objects are mostly limited to ornaments and blades, at least in tombs, and still found alongside a great variety of bronze objects, in Iron Age III (800/750 - 600 BC), iron seems to be the metal of choice for certain categories of objects, armament in particular.

While no sites of the magnitude of Hasanlu's Iron Age II destruction levels have been found in Azerbaijan for the Iron III period, it would appear that iron was then regularly used, especially for the production of weapons: in the small Urartian fort of Agrab Tepe, iron was recovered mostly in the form of weaponry artefacts assumed to date from the Iron Age III

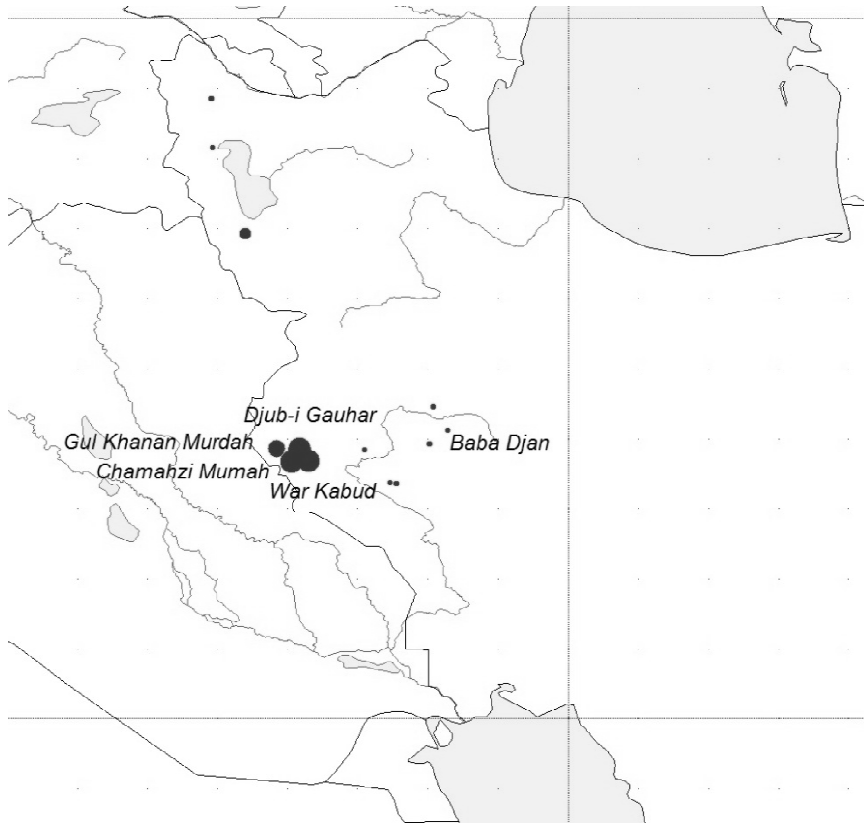
occupation of the fort (Muscarella, 1973). At the site of Haftavan, at the northern end of Lake Urmia, excavated by Burney in the 1970s, the Iron Age III levels are occupation levels of a citadel probably destroyed in 714 BC by Sargon II of Assyria or by the Cimmerians. They have produced only weapons, notably eight arrowheads. Finally small amounts of tools and weapons were recovered at the site of Bastam, another Urartian fortress excavated by Kleiss and probably destroyed around 590 BC.

For Iron Age III Luristan, the most detailed source of information on the use of metals is again the Belgian expedition in the Pusht-i Kuh (Overlaet, 2003; Haerinck & Overlaet, 1998; Haerinck & Overlaet, 1999; Haerinck & Overlaet, 2004). Given that only tombs were excavated in this expedition, we still have very limited information certain types of objects, such as tools. However it appears that at that point, iron had completely replaced bronze for the production of blades. The handles of the Iron Age III daggers are quite typical in that they have three cross ridges close together compared to the two separate ridges of earlier periods (Figure II-13). While in Iron Age II bronze was still the material of choice for arrowheads, in Iron Age III, they are replaced by iron ones. Iron axes, much simpler and heavier than the bronze spike butted axe heads from the beginning of the Iron Age are also commonly found exclusively in Iron Age III graves. On the other hand, iron jewellery becomes rare again: no iron rings and only a few iron anklets or bracelets have been found in the Iron Age III tombs. Instead, bronze is used for the production of ornaments (bracelets, rings, fibulae or rarely pins) and vessels. The fact that iron is no longer chosen to make jewellery indicates that by Iron Age III it was no longer a luxurious metal, but a more practical one.



**Figure II-13. Iron Age III sword (WK 66/467) and axe-head (WK 65/190) from the cemetery of War Kabud, Luristan. (Pl. 121 and Fig. 16 in Haerinck & Overlaet, 2004, p.49)**

As mentioned above, most artefacts from the occupation site of Baba Jan in the Pish-i Kuh are considered to be from an Iron Age III period. A few tools (knives, hoe, spatula, sickle...) have been found alongside a couple of bi-metallic daggers, two iron arrowheads and two iron spearheads (Goff, 1978), indicating that iron was also common for the production of tools in Luristan, even though it was not made obvious by the excavation of the Pusht-i Kuh graves. At Godin Tepe, a major trade centre situated slightly to the north of Luristan on the High Road connecting the Iranian plateau with Mesopotamia, iron was found only in Period II (800-600 BC). Frame states that “Iron completely replaced copper alloys for use in the chisels and blades, but the arrowheads as well as the decorative items continued to be made of copper alloys” (Frame, 2010, p.1709).



**Figure II-14: Iron finds in Iron Age III contexts (800-600 BC)**

Well documented Iron Age sites are extremely rare in southern and eastern Iran. To our knowledge, the only iron object found outside western and northern Iran is an iron nail found in Tall-i Malyan and dating from ca. 1250-1150 to 1100-1000 BC (Carter, 1996, p.34). Only further research in these areas will enable us to tell whether iron was produced or used there in the Iron Age, and if so to what extent.

c. Smelting and forging: technological background

Producing an iron object from the ore is a relatively complex task that involves a great number of independent processes: the mining, the preparation of the ore, the collecting of fuel and fluxes, the smelting, the consolidation of the bloom and finally the forging into a finished

object. Unfortunately, as mentioned above, almost no traces of any of these steps have been recovered in the Iranian archaeological record so far. Two notable exceptions are the site of Geoy Tepe in West Azerbaijan and the site of Kamtarlan in Luristan mentioned above.

At Geoy Tepe, T. Burton Brown's excavations in the 1940s have yielded hematite ores and iron slag (Burton Brown, 1951, pp.198-203). Unfortunately, little significance can be granted to these finds as it is unclear whether the slag really comes from the smelting or smithing of iron or if it is in reality iron rich copper slag. Moreover, the chronology of Geoy is not well established meaning that even if they are indeed iron slags, these remains do not provide much information to refine the chronology of iron-working (Pigott, 1981, p.124).

This incredible scarcity of evidence makes it difficult to know how iron was produced in Iran, or indeed if it was produced there at all or imported from neighbouring regions. Some or all of the steps involved in the production of iron may well have been carried out elsewhere, and iron could have made its way to western Iran in the form of ore, ingots or bars or finished objects. Until convincing remains of furnaces and smelting or forging debris are recovered in Iran, we have to rely on the location of the iron finds, the typology of the objects and the metallurgical analysis of the metal to understand patterns of production and trade for iron.

The first step in the production of iron, after the mining, is the preparation of the ore. It is likely that throughout the Near East, the ore would have been manually sorted in order to be cleaned of all the obvious non metallic content. It would then have been crushed to reduce the size of the fragments, effectively increasing the area exposed to reducing conditions for a better smelting. Finally the ore might have been roasted to eliminate the water and carbon dioxide present in it and increase its porosity and friability. These steps require little organisation and are likely to have taken place near the mining or the smelting sites.

The next step, which is the actual smelting of the ore, not only requires a higher degree of organisation, but also leaves significantly more remains for us to recover. The fuel commonly

used throughout antiquity was charcoal. For the smelting of iron, a large charcoal consumption is likely to have been necessary to attain the required temperatures and reducing conditions in the furnace. Therefore, charcoal production may have developed as a separate activity and become part of the economy of the area where the metal was smelted. In addition to the ore and the charcoal, a flux can be introduced in the furnace to facilitate the separation of the gangue by lowering the temperature at which this separation occurs. This practice was probably far less common for iron smelting than for copper smelting but could be achieved with a mixture of limestone or bone and clay for example.

The evidence for iron smelting installations in the Near East to date is relatively scarce. Several sites have however been uncovered in Anatolia: a furnace has been found in Arsameai and a large amount of iron smelting slag has been found in Sirzi. A workshop thought to have been used for the production of copper and iron artefacts is also currently excavated in Tell Tayinat (Roames, 2009).

In the Levant, recent researches have revealed traces of a certain number of smelting or smithing sites (Tell Hammeh, Tel Beth-Shemesh, Mugharet el-Wardeh, Tell Dor) dating from the beginning of the first millennium BC (Al-Amri & Hauptmann, 2009; Eliyahu-Behar et al., 2009; Veldhuijzen, 2009).

A very important group of about 400 smelting sites, possibly dedicated to iron smelting, has been found in Georgia (ancient Colchis) near the coast of the Black Sea. The results of the excavation of 26 of these sites and the description of 35 furnaces, thought to have been in use between the beginning of the 1st millennium BC and the 6th century BC have been reported in 1987 in Russian by Khakhutaishvili, but remain relatively unknown to western scholars as they have only been translated to English very recently (Khakhutaishvili, 2009). The furnaces found there were of shaft type, of variable sizes and presented a slag pit.

Another smelting complex has been reported at Metsamor in Armenia and has been dated to the Urartian period (8th - 7th centuries BC). At least 24 furnaces were identified at this location. However, the analyses of the slag have failed to reveal if iron was indeed smelted in those furnaces (McConchie, 2004, p.53).

The smelting takes place in the furnace at a temperature of about 1150°C. This is below the temperature of fusion of iron (1536°C), but it is required to allow the gangue to separate and the slag to flow. To reach this temperature, ancient metalworkers would have used tuyeres and bellows or blowpipes to force the draft in the furnace.

During the smelting, the charcoal present in the furnace is partially oxidized and the iron ore is reduced. The particles of iron created by this process agglomerate into a spongy mass called the bloom. The chemical reactions taking place in the furnace can be roughly summarized by these equations:



The smelting of iron requires a constant air input to attain the required temperature; however it also requires a proper ratio of carbon monoxide to carbon dioxide. If too much air is induced in the furnace by the bellows, the carbon monoxide is oxidized into carbon dioxide, modifying the ratio, and therefore the smelting conditions ( $2CO + O_2 = 2CO_2$ ).

After its extraction from the furnace, the bloom is porous and still contains remnant slag. It would have been reheated at a temperature of about 1000°C and hammered to evacuate the trapped slag and to be consolidated. The carbon content of the bloom can be quite uneven depending on its position in the furnace. At this stage of the process, several small blooms

could have been agglomerated to form a bigger one. In certain cases it would have been forged into an ingot or bar to facilitate its transport or trade (Moorey, 1994, p.283).

Objects in the shape of bi-conical bars or “fish-shaped” have been found in Assyria (see Figure II-15). More than a hundred have been found in the ruins of the 8th century BC palace of Sargon at Khorsabad, two have been found in Nimrud and one in Susa in south-western Iran (Pleiner & Bjorkman, 1974; Pleiner, 1979). They provide the only potential the evidence to date of a trade of iron into Iran in the form of ingots.

The blooms or ingots formed by the smelting operations then need to be forged into final products. The forging could usually take place with a simple bellow driven fire at a temperature of 700°C or more. These activities would leave some debris such as hammering scales, and pieces of scrap iron, none of which have been found so far in Iran.

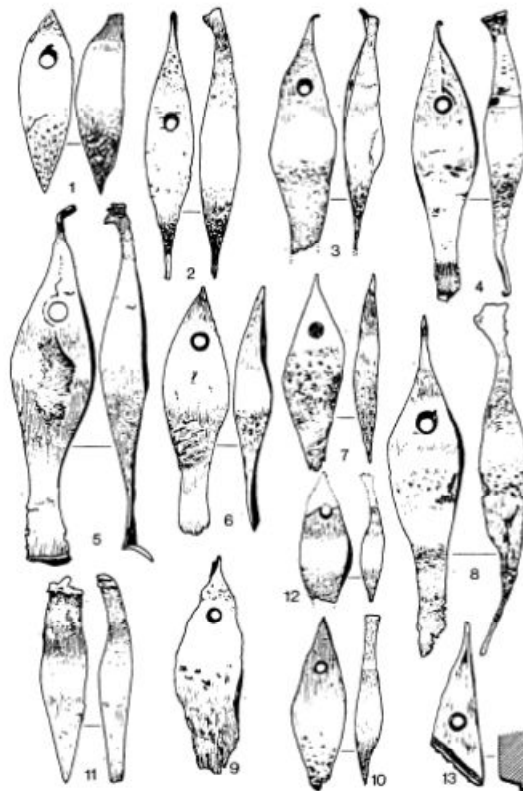


FIG. 8. Iron bars: (1-10) Khorsabad; (11) Susa; (12-13) Nimrud. (1-8) courtesy Louvre: (1) AO 24106, (2) AO 24107, (3) AO 24111, (4) AO 24110, (5) AO 24112, (6) AO 24108, (7), AO 24109, (8) AO 24113; (9-10) courtesy Oriental Institute Museum, Chicago, A 12261 and A 12462; (11) courtesy Louvre S<sup>b</sup> 9159; (12-13) courtesy British Museum, N-1963, N 962 (chopped piece). Scale: 1:10.

Figure II-15: Bi-conical bars possibly used for the trade of Iron. (Fig. 8 in Pleiner & Bjorkman 1974, p.295)

During the forging, the ancient smiths could subject the objects to a certain number of heat-treatments and mechanical treatments that would have modified the microstructure of the steel and ultimately the properties of the finished objects. The most common treatments would probably have been quenching (cooling the metal at a very fast rate by plunging it into water or oil), and work hardening (forging of the object at a relatively low temperature). Both would have resulted in an increased hardness, but also an increased brittleness of the objects which could have been partially relieved by a gentle reheating (Moorey, 1994, p.283).

However both quenching and work hardening can be carried away without any knowledge of their effects, simply to achieve a certain shape or for safety reasons to cool the object rapidly. It is therefore challenging to know to what extent they were used intentionally in the aim of improving the properties of an object (Moorey, 1994, p.284).

#### d. Metallurgical analysis of iron objects

From the late 1950's onwards, archaeologists interested in iron-working carried out metallurgical analysis of ancient iron objects to try and understand the development of blacksmithing technologies. The analyses completed to date on iron artefacts from Iran and the neighbouring regions are reviewed in this section.

##### Iranian tools and weapons

All of the results of metallographic analyses on Iranian iron objects are summarized in Table II-1.

In 1972 Tylecote analysed a punch, a spearhead and a bi-metallic dagger from the site of Marlik and dated to about 1000 BC in north-western Iran (Tylecote, 1972). The dagger and spearhead both presented a low carbon content and slag inclusions while the punch was cleaner and had areas with slightly more carbon.

In 1981 Pigott analysed an array of ten tools and weapons from the citadel of Hasanlu, all presenting a cutting edge: four spearpoints, two swords, an arrowpoint, a celt, a knife and a chisel. Although the preservation of the objects was in general quite poor, relict structures and islands of metal surviving in the oxide matrix have been studied (Pigott, 1981). Pigott concluded that, in Iron Age II, iron in the region of Hasanlu had a relatively low carbon content and was not yet "homogeneously carburized", showing perhaps, as does the presence

of bimetallic artefacts, a transitional phase between the predominance of bronze and the use of both metals. Pigott considered that, although the carbon content of the objects was probably high enough to confer a reasonable degree of resistance for their use as tools or weapons, they might not have had a performance comparable to worked tin bronze. This would justify the presence in abundance of bronze artefacts in the ruins of Hasanlu.

In 1969 France-Lanord published the results of his analyses of fourteen Iranian iron objects as well as a few Luristan swords (France-Lanord 1969). These objects are from the Luristan region, but their exact origin is unknown. The first three objects are interesting examples of the simultaneous use of bronze and iron: they are all bimetallic with an iron blade and a cast-on bronze hilt or handle. The author pointed out that they are a display of a remarkably good forge work as their structure is quite complicated and very-well executed, but there doesn't seem to be any particular attention directed at improving the mechanical properties of the iron. Nine other objects are made of iron of relatively low carbon content. Typological considerations lead the author to estimate their origin to approximately the 8th or the 7th century BC. Compared to the three bimetallic objects presented above, they seem to show a better understanding of the properties of iron: the small decorated knife has a very low carbon content which would have made the decorating easier, while the three single-edged blades have a very simple shape but a higher carbon content which seems to be adapted to their function. The horse bit however shows a lack of knowledge of proper welding techniques which is something that is also striking on Luristan swords (see paragraph below). In general these objects have a remarkably high hardness given their relatively low carbon content, this might be due to a high phosphorous content but the author wasn't able to carry out chemical analyses on these objects to confirm this hypothesis. Finally France-Lanord presented the results of analyses of two Iranian "steel" objects that he distinguished from the others by their exceptionally high carbon content. These objects are relatively corroded fragments of blades.

The author places the second one to around the 7th or 6th century BC and considers that it might come from the south-west of the Caspian Sea or Kurdistan. Its microstructure, with areas of higher carbon content arranged in layers, reminds him of a Wootz steel or a co-fusion steel. Such steels are thought to have originated further east at the end of the 1st millennium BC. Smith, unconvinced by the fact that this object was made by a Wootz or co-fusion process, has commented on this object:

"However, it seems to the writer that the making of "natural" steel by operating the refiner's hearth under highly reducing conditions must at least occasionally involve the transitory production of some liquid cast iron, and structures like that of France- Lanord's No.14 could easily result from the irregular distribution of liquid metal within a poorly consolidated sponge." (Smith, 1971, p.52)

From research, France-Lanord outlined the evolution of iron-working in Iran in the following fashion: the iron used before and around 1000 BC was relatively inhomogeneous and low in carbon. The forging, although very skilful, used techniques adapted to the work of bronze. Later on, the shapes were "freed from their bronze heritage" and the metal used was of better quality. Finally the smiths were able either to recognize and use consistently iron of higher hardness and carbon content or to developed techniques to carburise objects. This led to another modification of the shapes, midribs disappearing and lighter blades being manufactured.

In 1971 Smith analysed three Luristan maces, one dagger suggested to be of Median type, two daggers from the regions of Talish or Gilan and one Luristan short-sword (Smith, 1971). His work was focused on understanding the techniques used by the smiths who forged these objects. They show the use of a relatively pure metal. After a careful observation of etched sections and micrographs, Smith noted that the smiths of the beginning of the 1st millennium (roughly between 1000 and 600 BC) had acquired a high level of skills that enabled them to

forge in iron wonderful decorative shapes that resembled those cast in bronze. Nevertheless, as many others have remarked, he suggests that they seemed to be unaware of the possibility of welding iron. The joints are all ingenious mechanical ones. He also observed little grain deformation near holes or edges that had been cut, pointing towards the used of drilling and abrasives to achieve such results. The carbon content of the objects varied greatly in different parts of the objects (between 0 and 1%). This variation, probably coming from an inhomogeneous iron sponge, was also accompanied by variations of hardness. The hardness had not been improved by any heat-treatment (all the samples appeared to have been air-cooled) or by cold-working. The author remarks that cold-working was however used at the time for bronze objects. He suggests that there might not have been sufficiently powerful hammers or sufficiently resistant anvils to cold-harden iron. Smith's conclusions concerning the lack of welding and the inhomogeneity of the carbon used in the beginning of the 1st millennium concur with the result of the other authors presented above. Interestingly, however, his paper gives new information on the techniques used to shape the objects, such as drilling or grinding with abrasives rather than the use of a cold chisel.

Object	Provenance	Date	Carbon content	Hardness (HV)	Other observations	Reference
Dagger blade	Marlik	Iron Age II	0.1 to 0.2 %	160 to 240	Fine grains, elongated slag	Tylecote 1972
Spearhead	Marlik	Iron Age II	0.1 to 0.2 %	182	Fine grains, spheroidised carbides	Tylecote 1972
Punch	Marlik	Iron Age II		202 / 257	Low slag content, high hardness indicates possible phosphorous	Tylecote 1972
Spearhead	Hasanlu	ca. 800 BC	Wrought iron with some carbide		Elongated slag	Pigott 1981
Spearhead	Hasanlu	ca. 800 BC	Pure with spot concentration of high carbon			Pigott 1981
Spearhead	Hasanlu	ca. 800 BC	Pockets of pearlite. Carburisation at the edges		Slow cooling	Pigott 1981
Spearhead	Hasanlu	ca. 800 BC	Carburisation in centre of mid-rib, unevenly distributed			Pigott 1981
Bi-metallic sword	Hasanlu	ca. 800 BC	High (>0.8%) carbon zones		Many slag particles, slow cooling	Pigott 1981
Sword	Hasanlu	ca. 800 BC	Evenly distributed pearlite in mid-rib and edges		Slow cooling	Pigott 1981
Arrowhead	Hasanlu	ca. 800 BC			No surviving metal found	Pigott 1981
Celt	Hasanlu	ca. 800 BC	0.1 %		Fine grains	Pigott 1981
Knife	Hasanlu	ca. 800 BC	Very low (ferrite)		Elongated slag	Pigott 1981
Chisel	Hasanlu	ca. 800 BC	0.25 to 0.3 %. Evenly distributed pearlite		Fine grains	Pigott 1981
Bi-metallic dagger	Luristan		0.1 % irregular		Many elongated inclusions, cold-work on edges	France Lanord 1969
Bi-metallic dagger			0.1 to 0.2 % homogeneous	172 to 225	Very fine grains	France Lanord 1969
Bi-metallic adze			Very low		Fine grains, very well executed object	France Lanord 1969
Spearhead	Luristan	8th-7th cent. BC?	0.3 % Quite homogeneous	206		France Lanord 1969
Axe	Luristan	8th-7th cent. BC?	0.5 % Quite homogeneous	205 average	Fine grains, no additional carburisation on edges, good metal but mediocre implementation	France Lanord 1969
Horse bit	Luristan	8th-7th cent. BC?	0.1 to 0.2 %	138	Overheated, mediocre metal but skilful forging	France Lanord 1969
Knife	Luristan	8th-7th cent. BC?	<0.1 %	193	Very fine grains deformed on the edge (cold-working)	France Lanord 1969
Short-sword	Luristan	8th-7th cent. BC?	0.5 % Very homogeneous	225	Very fine grains, no defects	France Lanord 1969
Dagger (tang)	Luristan	8th-7th cent. BC?	0.1 to 0.2 %	190	Very fine grains	France Lanord 1969
(point)			0.1 %	181	Cold-working in the centre, overheating on edges	France Lanord 1969

**Table II-1. Summary of metallographic analyses carried out on Iranian iron objects.**

Object	Provenance	Date	Carbon content	Hardness (HV)	Other observations	Reference
Blade	Luristan	8th-7th cent. BC ?	0.4 % Higher on the edges	224	Very fine grains, some martensite	France Lanord 1969
Blade (tang)	Luristan	8th-7th cent. BC ?	0.3 to 0.4 %	203	Very fine grains	France Lanord 1969
(point)				190 to 200	Fine grains, homogeneous, no inclusions	
Blade (tang)	Luristan	8th-7th cent. BC ?	0.3 % Very homogeneous	206	Fine grains	France Lanord 1969
(point)				175 to 206	Bigger grains, overheating and air cooled	
Blade (blade)	Luristan		0.8% Slightly irregular, higher on edges and tip	297 to 309	Fine grains	France Lanord 1969
(point)				397 to 429	Bigger grains, overheating. Pearlite + cementite on grain boundaries	
Dagger or sword	SW. Caspian or Kurdistan?	7th-6th cent. BC ?	1.37 %	339 to 488	Very fine grains. Pearlite and cementite in layers	France Lanord 1969
Dagger blade	Medean / W. Iran	7th cent. BC	0.57 % avg. Irregular			Smith 1971
Dagger blade	Talish / Gilan	9th-8th cent. BC	0.089 % avg. Irregular			Smith 1971
Dagger handle	Talish / Gilan	9th-8th cent. BC	0.23 % avg. Irregular			Smith 1971
Short-sword (blade)	Luristan	8th cent. BC ?	0.63 % avg. Irregular			Smith 1971
(lion on ricasso)			0.39 % avg. Irregular			Smith 1971
Mace	Luristan	8th cent. BC ?	0.063 % avg. Irregular			Smith 1971
Mace	Luristan	8th cent. BC ?	0.23 % avg. Irregular			Smith 1971
Mace	Luristan	8th cent. BC ?	0.10 % avg. Irregular			Smith 1971

**Table II-1 (continued). Summary of metallographic analyses carried out on Iranian iron objects.**

### Luristan short-swords

As mentioned above, a very particular group of decorated short-swords from Luristan has attracted the attention of many scholars. Many of these have been examined since the 1930s. The resulting publications vary in usefulness as some of the studies have understandably been limited by conservatory reasons given such remarkable objects. France-Lanord and Smith however have both been able to sacrifice completely a sword, giving precious information on their manufacture and their metallography.

In 1991 Rehder reviewed the results of the analyses previously undertaken on thirteen of these Luristan swords (Ellis, 1961; Maryon et al., 1961; Damien, 1962; Lefferts, 1964; Ternbach, 1964; Maxwell-Hyslop & Hodges, 1966; France-Lanord, 1969; Smith, 1971). His conclusions are as follows (Rehder, 1991):

The swords were made of many different parts (from eight to fifteen including the rivets) assembled together by mechanical means and never by welding, in a technique that seems more appropriate to the working of wood for example than to that of iron. They were all made of bloomery iron, with a carbon content varying considerably from one sword to another, but also from one part of a sword to the other. The measures however have to be understood with caution because the blooms used to forge the swords appear to have been themselves of inhomogeneous carbon content. This variability would have resulted for the swords in disparities in their mechanical properties such as their hardness and strength, and for the blacksmiths in differences of forgability. The decorated figures, for example, are not always of lower carbon content which would have made them easier to engrave. The slag content is also quite variable from part to part.

Some of the researchers have also been able to conduct chemical analyses of the iron. Their results showed low levels of manganese, sulphur, and phosphorous indicating the use of ores of high purity.

It is interesting to note that all the blades have most likely been treated by a very slow cooling, which is indicated by the presence of spheroidized cementite. As a result the blades are extremely soft and ductile and of considerably lower hardness than worked tin bronze would be. There is no evidence of quench hardening in any of the examined swords.

From a morphological point of view, they seem to be well balanced, however the hilt and the blade are made from separate pieces assembled with rivets, resulting in a weak joint that probably wouldn't resist a blow. As many scholars have already observed, this data seems to indicate that the blacksmiths had incredible forging skills enabling them to achieve intricate mechanical joints while coping with iron of very variable content, but they didn't seem to realize the full potential of the metal and especially its weldability.

Another aspect that Rehder points out and that had not been commented on before is the presence within the metal of traces of internal welds coming from the consolidation of small blooms in a large one after the smelting process. Some of the welds seem to be of good quality (Damien, 1962; Maxwell-Hyslop & Hodges, 1966; France-Lanord, 1969), while others appear badly made (Smith, 1971), and, in one case, a longitudinal flaw, also likely to come from this consolidation stage, has been reported (France-Lanord, 1969). The author suggests the possibility that smelting and forging were not done at the same place (possibility supported by trace elements analysis showing at least two sources for the ore), which could explain why there was an apparent knowledge of

welding during the consolidation of the bloom, but a surprising lack of welds to assemble the different pieces of the swords.

#### Analyses of iron of neighbouring regions

In Assyria, the appearance of iron is evidenced by written records as well as archaeological finds, both of which were reviewed in detail by Pleiner and Bjorkman (1974) (see Chapter 7).

In 1979 Pleiner published the results of the analysis of two Assyrian tools found in Khorsabad dating from the reign of Sargon II (late 8th century BC): a hoe and a sickle. The objects have a low carbon content and are both relatively impure. Pleiner noted a "striking" lack of improvement of the working edges. In his opinion the Mediterranean world was more advanced in terms of iron technology at that time (Pleiner, 1979).

Pleiner also analysed a fish-shaped ingot from the palace of Khorsabad now kept in the Oriental Institute Museum in Chicago. The observation of two samples from opposite tips of the object led him to think that it was probably made of inhomogeneous steel, because the areas of higher carbon content seemed to be rather randomly distributed. There was no evidence of quenching in his samples.

Also in 1979, Curtis et al. published the results of their research on Assyrian iron-working, including the analysis of eight objects from Nimrud. The authors noticed in these objects relatively low carbon content and a lack of evidence of quenching. However, they remarked that since the working edges were not always available for study, it was difficult to conclude on the processes used to increase the quality of the objects (Curtis et al., 1979).

Curtis et al. also studied four fish-shaped ingots (two from Khorsabad and two from Nimrud) and observed regions of relatively high carbon content and regions almost free

of carbon. There appeared to be no consistency between the samples concerning which areas presented more carbon. However, they seemed to present more carbon than the finished objects also studied by Curtis et al. The authors suggested several possible explanations for that. First, the ingots were much less corroded than the other objects they studied so the edges were available for analysis. It is possible that Assyrian ironsmiths could only obtain a carburised iron on the outer layers of their objects leaving the core to be almost pure iron. Secondly it is possible that the ingots, because of their rather complicated shape would have required a longer time for forging and therefore would have had more time to absorb carbon during the heating of the object. The ingots presented some evidence of quite rapid cooling (some dispersed pearlite and bainite) suggesting that they had probably been quenched. However as mentioned above, it is not clear whether this was intentional or not. One of the ingots also presented a porous structure, suggesting that the iron sponge had not been completely forged which is coherent with the hypotheses of a semi-finished product.

The kingdom of Urartu to the north-west of Iran is another example of an area where the appearance of iron is documented by textual as well as archaeological evidence (see Chapter 7). The results of analyses of Urartian iron objects were reviewed in 2004 by McConchie who concluded: "Archaeo-metallurgical evidence does not support the theory that there were attempts to deliberately harden iron during the Urartian period" (McConchie, 2004, p.27).

McConchie also carried out her own analysis of objects from five Urartian sites: Büyüktepe Höyük, Sos Höyük, Van Kale, Karayündüz and Ayanis in an attempt to understand the acceptance of iron by the Urartian society (McConchie, 2004). One of the general characteristics of the objects she analysed was that they were made of

inhomogeneous metal and rarely subjected to heat-treatments, with a few notable exceptions. She however noticed an increase in the range of manufacturing techniques and a slight increase in the consistency of the quality of the metal produced in the late Iron Age / Hellenistic period compared to the Urartian period.

A common theme across all of the analyses presented above is the inhomogeneous carburisation of the objects, especially for the earlier specimens, and the lack of preferential choice of high carbon contents for working edges. Clearly, in the beginning of the 1st millennium BC, the level of control of the smelting conditions was generally poor, which resulted in inhomogeneous blooms. The metal smiths were nevertheless very skilled in their forge work, as attested by the intricate shapes of some of the objects. However, they seem to have had a fairly poor understanding of the properties of iron and its ability to be welded, or for steel to be quench-hardened for example. These aspects of early iron metallurgy are discussed in more detail in Chapter 7.

### 3. Understanding the fundamental differences between bronze and iron metallurgy: a key to understanding the “bronze-iron transition”

This chapter has underlined the discrepancies in our understanding of ferrous metallurgy as opposed to copper based metallurgy. Most striking is the complete lack of information concerning the mining and smelting of iron ores in Iran. For copper, this type of information is not overly abundant, but heaps of slag, furnaces and crucibles attesting of the production of copper objects are not uncommon (Tepe Hissar, Tal-i Iblis, Shahdad, Shahr-i Sokhta) and a few mines have been identified as potential sources for the ore (Talmessi, Vesnave, Deh Hosein). While for copper metallurgy the

chronology of its development is marked by a relatively well understood sequence of technological changes (very schematically: native cold-worked copper followed by smelting and casting, followed by the use of ores rich in arsenic, followed by the development of tin bronze), the chronological outline of iron-work is mostly based on the number, typology and geographical distribution of the recovered iron objects. Of course some iron objects have been analysed (France-Lanord, 1969; Smith, 1971; Tylecote, 1972; Pigott, 1981), giving us some insight into how they were forged, however one can't help but notice that the chronological and technological framework established thus far for iron remains weak. Moreover, it is focused almost exclusively on north-western Iran. The reasons for such a discrepancy are obviously numerous, but understanding them fully is essential to a well-grounded discussion on the transition between the use bronze and that of iron.

As a preliminary remark, it must be noted that the time-frame for which the copper development "sequence" is discussed is much longer than the duration between the first appearance of iron in Iran and its wide-scale distribution (seven millennia compared to just one millennium). With such a difference, one cannot expect to have the same amount of information for both metals.

That said, understanding the very nature of both metals is probably a good starting point for a discussion on the bronze-iron transition. From a chemistry point of view, the greater resistance of copper to corrosion means a greater availability in the archaeological record and a longer history of scholarly interest for the metal (iron having been disregarded for some time due to its degraded state when corroded). This means that simple quantitative surveys of the ratio of copper to iron recovered in the archaeological record must be considered with caution (Moorey, 1994, p.289).

From the point of view of the smelting, iron requires a better control of pyrotechnology. Indeed, the output of an iron smelt would have depended quite heavily on the temperature of the furnace amongst other variables. Producing iron of consistent carbon content and properties would have necessitated a much better control of the smelting conditions than for a successful copper smelt (Moorey, 1994, p.282).

From the point of view of the production of finished objects, the fact that every single iron object would then have had to be individually forged means that bronze would have been better adapted to mass production (Moorey, 1994, p.283).

Also interesting is the discussion on the level of sophistication of the production. While the traditional view is to consider small-scale ‘independent’ production as technologically less advanced than the production of specialized workshops, the bronze data from the site of Tepe Hissar, showing sophisticated production from independent craftspeople has led Thornton to challenge this model (Thornton, 2009). The idea that sophistication isn’t necessarily linked to the scale or locus of production is to be kept in mind when discussing the quality of the early iron. For example, the fact that, in Hasanlu, there is no clear indication of where the iron was produced, while three different bronze workshops have been found (Pigott, 1999b, p.91), doesn’t necessarily mean that the metal objects weren’t produced with a similar level of specialization, possibly by the same craftsmen. Indeed, as we have seen above, bi-metallic objects are a very common occurrence in north-western Iran during the Iron Age attesting of a strong relationship between the two metals.

The following chapters detail the analyses undertaken on copper-based objects and the conclusions that have been derived from these concerning the evolution of copper metallurgy. Unfortunately, the fundamental differences in the nature of both metals mean that the number of analyses done so far on bronze objects greatly exceeds that of

iron objects. The quantity of uncorroded metal in ancient iron objects is usually quite low, and getting to this metal to examine its microstructure often requires cutting into the core of the metal. Conservation considerations have understandably restricted the use of such destructive methods. Another important difference between the bronze and iron is the information held within the metal. In both cases, metallographic observation can give us some information on the way the objects were worked and shaped, and in the case of iron the quantity of carbon can be estimated, giving us an insight into the smelting and possible carburisation techniques. However, while for copper the chemical composition gives us some information on smelting and alloying techniques but also on the possible provenance of the ore, its trade and recycling (see following chapters), such trace element analyses are much less effective for iron, making discussions on provenance in particular much more difficult.

Finally, another aspect that has been at the heart of the discussions on the appearance of iron is its properties compared to the properties of bronze. Iron is generally thought to be a harder and stronger metal than bronze. However, as seen above, analyses of early iron objects have shown that these objects were generally inhomogeneous and weren't subjected to treatments aimed at "improving" the properties of the metal. Therefore, they would have presented little mechanical advantage over a work-hardened bronze. Nevertheless, another property of iron might have been considered an advantage over bronze: its higher ductility meant that if an iron blade was bent or dented, it would have been possible to repair it fairly rapidly. Bronze on the other hand, depending on its alloying and manufacture, will sometimes have had a limited ability to deform and would have broken quite easily. In that case it would have been necessary to entirely recast it making repairs more difficult (Moorey, 1994, p.283). McConchie argued that

this property of iron might have been interesting for military powers such as Assyria or Urartu as it made iron weapons easier to repair in a battle context (McConchie, 2004).

#### 4. Conclusion

The following chapters of this dissertation are focused on presenting the analyses undertaken so far on copper-based objects (Chapter 3) and developing a new methodology to reanalyse the available data (Chapters 4, 5 and 6). The method chosen for the study of copper and bronze attempts to use a systematic methodology, to an extent making the numbers speak for themselves, in order to free ourselves from possibly erroneous preconceptions. However the chronological and technological background and the reflection on the differences between bronze and iron presented in this chapter are of prime importance to discuss the results obtained with this method and to anchor them in a coherent archaeological setting (see Chapter 7).

## Chapter III. The bronze data

As explained in Chapter 1, this thesis aims to reassess the changes that occurred in the production of metals at the end of the Bronze Age and the beginning of the Iron Age by focusing on changes in the composition of the metal itself. To do so, all the previously published chemical compositions of copper alloys have been assembled in one database. Whenever they were available, the results of lead isotope analyses were also collected to aid our interpretation of the provenance of the metal. For Iran alone, this has meant putting together the results of 38 analytical projects carried out in the past century. With such a large and fragmented dataset, it is crucial to assess how compatible these studies are and how to bind them into one coherent chronological and geographical framework. This chapter presents a brief overview of these publications, defines the geographical and chronological scales that are used in the rest of this project and discusses the strength and limitations of assembling such a varied dataset.

### 1. Published data

#### a. A large and fragmented dataset

As mentioned in Chapter 1, the original focus of this project is the land that lies within the borders of what is now modern Iran, but it was considered necessary to add some data from a number of neighbouring countries. For Iran, the collection of old datasets was made as exhaustive as possible, while for the neighbouring countries, the timeframe

of this project did not allow such a thorough investigation of previous publications. As a result, for Iran, the chemical composition of 2133 different artefacts, from 38 different publications, was collected. For the neighbouring regions, another 2897 artefacts from 13 studies were added to the Iranian dataset<sup>1</sup>. As shown in Figure III-1 and Figure III-2 the publications vary a lot in the number of objects they analysed and the analytical methods used: for some, the publication of the chemical composition of a limited number of objects was just a side note to an excavation report or article focused on other things than metallurgy, while for others the composition of metals was the main focus of a systematic metallurgical program. Techniques vary from Optical Emission Spectroscopy (OES) used widely before the 1980s to more modern techniques such as Proton Induced X-ray Emission (PIXE) or Inductively Coupled Plasma Mass Spectrometry (ICP-MS). But the differences don't end there: as shown in the following section, most studies focus on a different region and a different period. As a result, the dataset is very fragmented and there isn't one particular study that provides a good crossover of the whole geographical and chronological range, helping to bind the dataset together. This is why it is particularly crucial in this chapter to define the best possible geographical and chronological divisions to try and overcome this fragmentation and to assess to what extent the results of all of these different studies can be used in one single dataset.

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<sup>1</sup> Since the submission of this thesis, the following publications containing data for Iran and the surrounding regions have been brought to my attention:

- Objects from Oman, Bahrain and Susa (Prange, 2001). Some of these had previously been analysed by Hauptmann et al. (1988) and Tallon (1987)
- Sangtarashan (Luristan) vessels (Oudbashi et al., 2013)
- Nimrud (Assyria) Iron Age metal (Ponting, 2013)
- Masafi (U.A.E) ingots (Goy et al., 2013)

The data from these publications are not included in any of the graphs, tables or calculations in this thesis but a few footnotes indicating some their characteristics have been included where relevant.

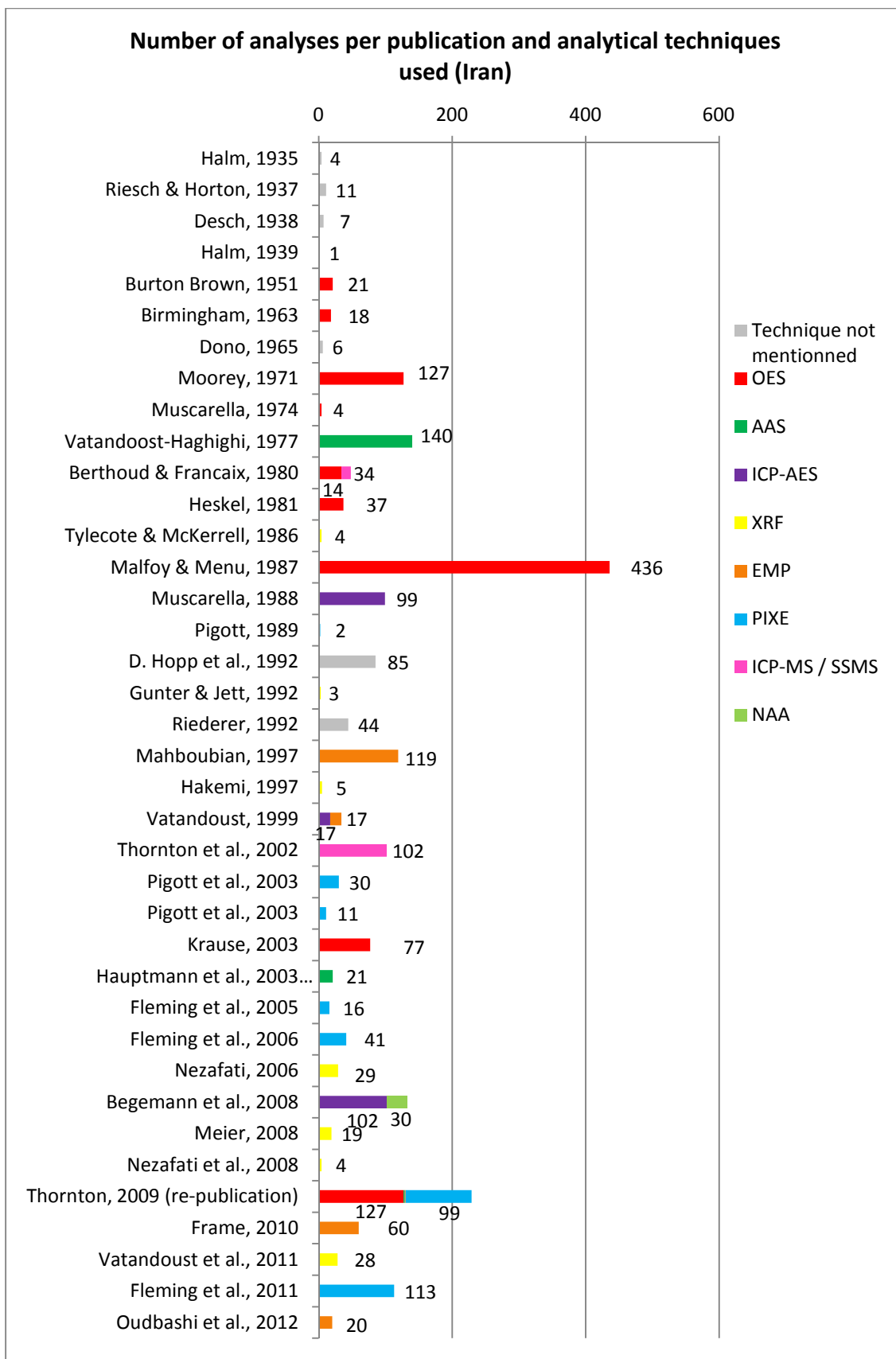


Figure III-1. Number of analyses entered in the database for each publication and analytical techniques used in these publications (Iranian sites)

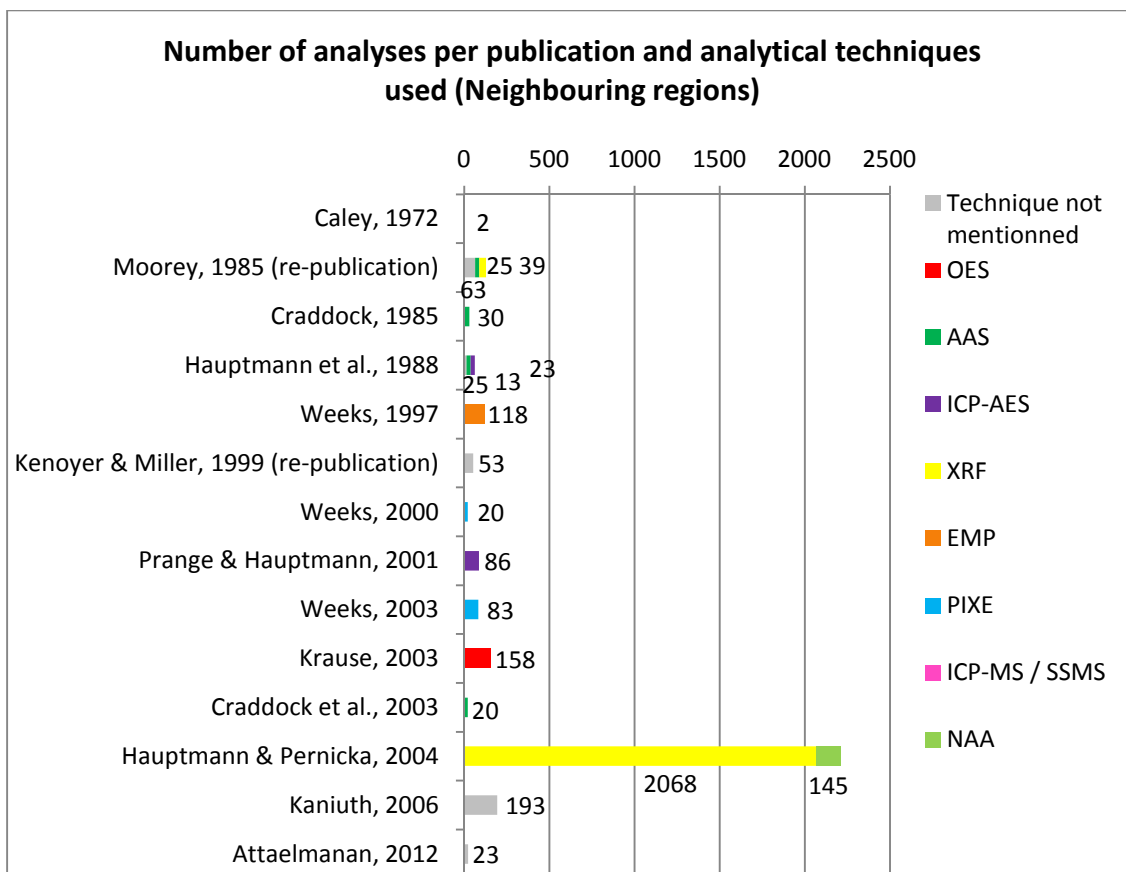


Figure III-2. Number of objects entered in the database for each publication and analytical techniques used in these publications (Oman, Bactria, Indus Valley and Mesopotamia)

#### b. History of the publications

As we have seen in the previous chapter, interest in the metallurgy of Iran has been considerable since the late 1960s, but the analysis of the chemical composition of Iranian copper-based objects had already started three decades earlier: for Iran, the earliest publications used in this thesis are the work of Halm, Riesch and Horton and Desch in the late 1930s. Since then, many more objects have been analysed and this section provides an overview of data provided by each of them, the methods used and the questions they attempted to answer.

### Early reports

The earliest analytical work was often limited to very few objects and published as an appendix to an excavation report. This is the case of two reports written by Halm in 1935 and 1939 in Contenau and Ghirshman's excavation reports of the sites of Tepe Giyan and Tepe Sialk (Halm, 1935; Halm, 1939). In the first one, Halm reports on chemical and metallographic analyses of four copper-based objects, two of which turned out to be tin bronzes (11 and 13% of tin) and two unalloyed copper with elevated amounts of nickel (1.35 and 1%). In the second one, she examined three copper-based objects (two pure copper and one alloyed with around 2% of lead and 2.5% of tin) as well a lead ring and ore fragments. While she did not mention the analytical technique used to obtain these percentages, she explained that a calculation was made on the measurements to account for the corrosion of the samples and give a result closer to what the original composition would have been. In the 1937 excavation report of Tepe Hissar by Schmidt, the appendix, written by Riesch and Horton, presents the analyses of 11 copper-based objects (Riesch & Horton, 1937). The copper, tin, lead and iron contents were "analysed chemically" primarily to assess whether they were copper or bronze. The results showed mostly relatively pure copper, but six objects presented between 1 and 2.5% tin and three objects between 1 and 2% lead which the authors considered as coming from the association of these elements with the copper ore. The next series of analyses was published the following year in the chapter on Luristan Bronzes of Pope's volume on Persian Art (Desch, 1938). The copper, tin, nickel, lead and antimony content were investigated for eight copper-based objects. The high percentages of tin observed in this study (8.8 to 25.2% in the true tin bronzes) contrasts with the percentages observed in later studies for Luristan bronzes, generally centred on

8 to 10% (see below). This study is also the first one to comment on the technology of Iranian bi-metallic artefacts, in this case iron-bronze pins.

In 1951, Burton Brown published the results of the 1949 excavations of Geoy Tepe in the province of Azerbaijan in north-western Iran. This publication includes a report on the spectrographic and metallographic analysis carried out by Dr. Voce on 21 of the objects excavated in Geoy Tepe and dating from the 3rd and 2nd millennium BC (Burton Brown, 1951, pp.179-97). The objects examined were divided into four groups based on tin content: copper objects, low tin bronzes (approximately 0.5% tin), medium tin bronzes (approximately 5% tin) and high tin bronzes (approximately 10% tin). However, it was recognized that the spectrographic method used for the analysis could not give reliable absolute values for tin in true tin bronzes: “It must be remembered that spectrographic analysis gives approximate rather than precise values, especially when the concentration of the element under consideration exceeds a few hundredths of one per cent” (Burton Brown, 1951, p.187). These results were then compared with analysed objects from Greece, Anatolia, Egypt, Mesopotamia, etc. A significant increase in the quantity of arsenic in copper-based objects around the middle of the 3rd millennium, suggesting its use as an alloying element rather than an impurity was noted, as well as the appearance of tin bronze in all regions in the later part of the 3rd millennium BC.

The 1964 excavation report of the chalcolithic mound of Tall-i Nokhodi in Fars included the result of the spectrographic analysis of two copper based objects performed by C.S. Smith. The semi-quantitative results cannot be used in this thesis, but a remark by C.S. Smith is worth noting as it is an early mention of recycling, a practise that is rarely acknowledge even in more recent publications: “Tin is not enough to call it a bronze perhaps, and I suspect that the amount came in accidentally through the use of

scrap at a time when bronze of higher tin content was being intentionally used” (Goff, 1964, p.49).

The 1965 report on the Japanese excavations in the Dailaman (Dono, 1965), the 1986 report on Tepe Yahya (Tylecote & McKerrell, 1986) and the 1997 volume on Shahdad (Hakemi, 1997, pp.109-14) all included the chemical analyses of less than 10 objects. These analyses are of little use on their own, other than to tell the excavators if the metal excavated was bronze or copper, but they can contribute to building a database as exhaustive as possible for Iranian metallurgy.

#### Analyses of museum or private collections

Analyses of bronze objects have also been published as part of the publications of museum collections or private collections. As we have seen, this was already the case of Desch’s note the analyses of Luristan objects in 1938 (Desch, 1938). A couple of decades later, in 1963, Birmingham apologized for publishing “yet more Luristan bronzes of well-known types without known provenance” and hoped that her paper was justified by the presentation of the spectrometric analyses of these 18 objects from the Nicholson Museum in Sidney (Birmingham, 1963). Her analyses revealed a higher tin content for axes and daggers (up to 13.9%) compared to ornamental pieces. It also showed that the axes from the 3rd and early 2nd millennium had less tin than the axes from the late 2nd or early 1st millennium BC. She therefore suggests a comparatively late use of high tin bronzes in Iran. The other elements were characterized by low and constant amounts of arsenic and nickel making unlikely the use of ores from nickel rich deposits in Turkey or northern Iran. The lead content on the other hand was highest for the two “most ornate” objects, a standard top and an ibex mounting (respectively 3%

and 5%) suggesting that, for these two objects, lead was perhaps added intentionally to improve the details of the decorations. Birmingham states that in all of the other objects and possibly even in these two, the occurrence of lead was possibly the accidental result of the use of lead rich copper ores from the 12th century BC onwards.

The first large scale publication of analyses of museum collections was carried out by Moorey of Oxford's Ashmolean Museum in 1971 (Moorey, 1971). About a fourth of the Iranian bronzes of the collection of the Ashmolean Museum at the time (127 objects), most of them of probable Luristan provenance, were analysed by spectroscopy by Mrs. A. Millet of the Research Laboratory for Archaeology and History of Art in Oxford. The interpretations of these analyses however are the work Moorey himself. He distinguished six main groups of chemical composition. These groups and the number of objects in each are summarized in Table III-1 below. Although he considered that the number of analyses was too low to obtain proper statistical results, he noticed the following tendencies: the majority of the bronzes fall in a range of tin content of about 4% to 13% and any percentage higher than that is very rare. However, contrary to Birmingham, he noticed no patterns concerning the type of objects. Indeed in this study, the ornamental objects have comparable or even higher tin percentages than the weapons and tools.

<b>Group</b>	<b>Number of late 3<sup>rd</sup> - early 2<sup>nd</sup> mill. objects</b>	<b>Number of late 2<sup>nd</sup> - early 1<sup>st</sup> mill. objects</b>
Copper		5
Arsenical Bronzes	6	
Tin Bronzes		
With Nickel	12	99
With As, No Ni	1	2
No As, No Ni		3
High Zinc		4

**Table III-1. Moorey's grouping of Ashmolean copper-based objects**

The nickel content on the other hand seems to be consistent with Birmingham's observations. There is a “regular occurrence of small percentages of nickel” (typically less than 1%). Just like Birmingham, Moorey suggested an accidental occurrence of this element. He proposed a mining region in the “west Caspian region”.

In 1974, analyses by semi-quantitative emission spectrography of the composition of four Iranian bronze beakers from the Metropolitan Museum of Art in New York were published as an appendix to a stylistic discussion of such beakers by Muscarella (Muscarella, 1974). The note, written by Mishara and Meyers was primarily aimed at determining whether or not one of these vessels was a modern forgery. It was concluded that the beaker was most likely ancient. More relevant to our discussion is the fact that the four beakers examined had a remarkably consistent composition, with tin ranging between 5.6% and 8.3%, nickel between 0.24% and 0.37%, arsenic between 0.2 and 0.3% and lead no higher than 0.3%.

In 1987, the Louvre museum published a detailed study of the metalwork of Susa from the 5th millennium to the 18th century BC. The typological study of over a thousand objects mostly from the Louvre and the museum of Tehran objects was accompanied by the study of their chemical composition by Malfoy and Menu (Malfoy & Menu, 1987). Some of these analyses had already been published a few years before by Berthoud and Françaix as shown below (Berthoud & Françaix, 1980). Of the 479 objects analysed by what we understand was optical emission spectrometry using UV lines, we were only able to use the 436 objects that were dated in the catalogue. Malfoy and Menu, building on the work of Berthoud, observed the variation of the arsenic, tin, and lead content as well as trends for trace elements allowing them to suggest an outline of the evolution of copper metallurgy of Susa during the 4th and 3rd millennia BC. The first phases of this period are characterised by the predominant use of arsenical copper possibly with

arsenic rich copper minerals from the region of Anarak. Later, the origin of the ore might have shifted towards Oman. They noted that the production of tin bronze became important at the end of the 3rd millennium BC. The authors however made no attempt to solve the issue of the provenance of the tin. Interestingly they noted in period VB a differentiation of the type of metal used in daggers between the blades and the handles: the blades are made of tin bronze, sometimes also with arsenic while the handles are made of copper, attesting of a good understanding of the properties of the metals.

In 1988, Muscarella published a complete survey of the Near Eastern bronze and iron objects held in the Metropolitan Museum of Art (Muscarella, 1988). The entries for these artefacts are accompanied by 99 chemical analyses. These had either been conducted over the years by the laboratory of the Metropolitan Museum or in a number of cases by P. Meyers of the Los Angeles County Museum of Art who analysed many samples in 1986 by induction-coupled plasma emission spectrometry. There is however no discussion or summary of this data attached to the catalogue.

The third large scale publication of museum bronzes compositions is that of the Altenessen Museum in Germany. In 1992, Hopp et al. published Riederer's analyses of 87 bronze artefacts mostly from Luristan and northern Iran (Hopp et al., 1992). Unfortunately the analytical method was not given in the publication. Eight main groups of compositions were isolated (Table III-2). The average percentages of each element across the whole range of objects were also calculated. This data is given in Table III-3. The conclusions drawn by Riederer were not unlike Moorey's remarks. Indeed, Riederer also noticed no obvious correlation between the type of objects and their tin levels. The distribution of the tin is also relatively similar. Most of the objects fall in a range of 4% to 10% of tin with a peak at around 9 to 10%. Objects with a higher percentage of tin occur only rarely. The similarities between these two studies are certainly correlated to

the fact that they cover similar areas and periods. In both cases, the objects are attributed to Luristan and northern Iran and a significant number of them are in fact of uncertain provenance. In terms of chronology, most of the dating in both catalogues is deduced from typological considerations, but both collections probably range from about the late 3rd millennium to the early 1st millennium BC.

Group	Composition	No. of obj
Pure copper	Cu > 98%	7
Arsenical copper	As > 1%	6
Low tin bz	Sn 1-5%	16
Moderate tin bz	Sn 5-10 %	30
High tin bz	Sn 10-20 %	14
Leaded low tin bz	Sn 1-5%	5
Leaded middle tin bz	Sn > 5%	9
Leaded copper	Sn < 1%	2

Table III-2. Riederer's grouping of Altenessen objects

Cu	Sn	Pb	Zn	Fe	Ni	Ag	Sb	As	Co
90.62	6.32	0.50	0.013	0.17	0.06	0.06	0.06	0.44	0.009

Table III-3. Element averages for the Altenessen Museum objects

In 1992 Riederer also published an article with the analyses of a series of 44 daggers and swords from north-western Iran, mostly from the 1st millennium BC, kept in another German museum: the Deutsche Klingmuseum in Solingen (Riederer, 1992). Again, he does not mention the analytical method used for this study. Most of the objects were found to be tin bronzes, with a tin content of ranging for the most part between 4 and 12%. Riederer discussed the variations in tin, lead, iron, nickel, silver, antimony and arsenic contents between the seven different regions of northern and western Iran represented. He also compared the averages of these elements for the Luristan objects to the averages in the Ashmolean and the Altenessen collections. The

three studies were found to be in good agreement which lead him to suggest that a relatively homogeneous material was used in the production of the Luristan bronzes.

In 1992 A.C. Gunter and P. Jett published a catalogue of the Iranian metal objects of the Sackler and Freer Galleries in Washington D.C (Gunter & Jett, 1992). The catalogue includes a large number of analyses of silver objects from the Achaemenid, Seleucid, Parthian and Sasanian periods. Three bronze objects of the Iron Age and two from later periods have also been analysed by Energy Dispersive X-Ray Fluorescence (EDXRF). The averages of two measurements were taken. Only the three earlier objects will be considered for the following chapters of this thesis.

Finally, in 1997, the analyses of 128 objects coming mostly from Elam, Luristan and northern Iran were published in a very nicely illustrated catalogue of the bronze and iron objects of the Mahboubian family collection. Northover examined 142 samples by electron microprobe with Energy Dispersive Spectrometry and presented the mean of three measurements (Mahboubian, 1997). He also carried out some metallographic examinations of the objects. The compositions of these bronzes fall into five main categories, summarised in Table III-4. The results show slightly higher amounts of tin than in the Ashmolean or Altenessen collections but are still remarkably consistent with the previous results: the majority of the bronze objects have a tin content between about 8% and 13% and the peak is once again situated around 9-10%. Northover notes that this amount of tin would give the metal a "great hardenability and a pleasing gold colour". A few objects have a tin content comprised between 2% and 5%. These percentages are not very frequent, but Northover thinks they may reflect the recycling and mixing of other alloys rather than an intentional addition of tin.

<b>Group</b>	<b>Composition</b>	<b>No. of obj</b>
Copper & Arsenical copper	Cu, Cu/As	15
Bronze	Cu/Sn	97
Leaded Bronze	Cu/Sn/Pb (Pb>2%)	9
Gunmetal	Cu/Sn/Zn	3
Brass	Cu/Zn	2
Silver alloy	Ag/Cu	1
Iron	Fe	1

Table III-4. Northover's grouping of the objects from the Mahboubian collection

#### Archaeometallurgical projects

In parallel to the publication of collections catalogues, a number of programs which primary goals were the understanding of Iranian metallurgy were undertaken from the 1970s. Vatandoost-Haghighi's PhD from 1977 is one of those (Vatandoost-Haghighi, 1977). He analysed 131 objects by Atomic Absorption Spectrometry mostly from the north-west, but also from Shahdad and Susa. This work enabled him to propose a chronology of the development of copper metallurgy, in particular for the use of arsenical copper and tin bronze. A few attempts were also made to find trends in the provenance of the objects, but the results are rather unconvincing. These analyses and those of a further 30 objects analysed by SEM-EDS and ICP-AES were then published as part of the proceedings of the 1995 "Beginnings of Metallurgy" conference in Bochum (Vatandoust, 1999).

In 1980 Berthoud and Françaix published the result of a collaborative work between the Laboratoire de Recherche des Musées de France and the Commissariat à l'Energie Atomique (Berthoud & Françaix, 1980). This work was primarily intended as a

methodological study on how to characterise ancient copper based objects by the analysis of trace elements. To this end the authors analysed 179 objects mostly from Susa but also from Tepe Sialk and Tepe Giyan located in the Louvre Museum. Most of the analyses from Susa objects had already been entered in our database as part of Malfoy and Menu's publication (see above). The objects were analysed by emission spectrometry and some were also subjected to spark source mass spectrometry. The methods were compared and the authors concluded that emission spectrometry was a good method to analyse a large number of copper objects but mass spectrometry with its lower detection limits enabled them to observe differences in the composition of groups of objects that were undetectable with emission spectrometry. From an archaeological point of view, the authors observed a change in the composition of the copper objects from Susa in the 3rd millennium BC and suggested a change in the provenance of the copper possibly from a provenance on the Iranian plateau to sources in the Arabian Peninsula.

In 1981 Heskell in his PhD thesis reported on the spectrographic analysis at the MIT of 83 objects from Tepe Hissar, Susa, Tepe Yahya and Shahr-i Sokhta from the 4th and 3rd millennia BC (Heskell, 1981). He also assembled results from other researchers: the SAM project, Dyson from the University of Pennsylvania Museum, Amiet from the Louvre and other published analyses for a total of 303 objects. He then performed a cluster analysis on this data but found the result of this clustering disappointing due to a great similarity in trace element composition for most of the samples. Unfortunately Heskell had great difficulties in detecting arsenic: the detection limit for this element was 1.2% (Heskell, 1981, p.286). Moreover, the labelling of the samples and of the analytical results in his thesis was unclear, making it difficult to extract the data in order

to use it in the present work. As a result, only 37 objects, most of them from Tepe Yahya were inserted into our database.

The SAM (Studien zu den Anfängen der Metallurgie) project, based in Stuttgart is best known for the OES analysis of an extraordinary number of European copper-based objects (Junghans et al., 1960; 1968), but the publication of these analyses by Krause (2003) also included some material from the Near-East, notably 200 artefacts from Tepe Hissar, re-used by Thornton in his PhD as discussed below, 46 objects from Susa, 22 from the Amlash region and 23 from Hasanlu. From countries around Iran, it included 75 analyses from Tepe Gawra in Assyria and 76 objects from Bahrain. The publication also included a few analyses carried out by other teams, in particular a few of the Luristan objects from the Ashmolean Museum published by Moorey, and some analyses of objects of Mesopotamian sites carried out by Berthoud. A total of 235 objects from the 2003 publication were entered in our database.

The beginning of the 21st century was marked by a reanalysis of a certain number of Iranian sites. In 2002, Thornton published an article on the metallic material of Tepe Yahya, a site excavated from 1967 to 1975 (Thornton et al., 2002). As seen above, objects from Tepe Yahya had previously been analysed by Heskell and by Tylecote and McKerrell, who analysed only four objects by arc spectrography and XRF (Heskell, 1981; Tylecote & McKerrell, 1986, p.214). In his 2002 article, Thornton presents the results of the compositional analysis of an additional 105 non-ferrous metal objects from the Peabody Museum. The objects, mostly pins, awls, needles and jewellery were analysed by Inductively-Coupled Plasma Mass Spectrometer (ICP-MS) and Electron Microprobe (EMP). They date from all periods of occupation of the site, i.e. from the 6th millennium BC to the Iron Age and allow a good overview of the development of copper metallurgy as viewed from this site. A remarkable aspect of Thornton's results is

the prevalence of arsenical copper for about 3000 years after their first appearance in the late 5th millennium: even during the Late Bronze Age and the Iron Age, Cu-As alloys are more common than tin bronzes. A second noteworthy result is the presence at Tepe Yahya of some of the oldest brasses. Three jewellery fragments from the 15th century BC have an elevated amount of zinc (17-20%) shedding new light on the origins of this alloy.

For his PhD completed in 2009, Thornton also studied the North-Eastern site of Tepe Hissar (Thornton, 2009). This project did not include any new analyses of copper objects from the site, but reviewed and listed in an annexe 228 analyses carried out by different groups up to that date. It included the OES work of the SAM program, the analyses carried out by Dennis Heskell (1981) by OES and metallography (already mentioned above), the metallographic and compositional analysis by Michael Notis and Heidi Moyer from Lehigh University of three objects by EMPA and two ingots by AAS (Pigott et al., 1982, p.229), and the PIXE analysis of Charles Swann and the MASCA (only part of the data obtained by this program was made available and could be used by Thornton). As shown further in this chapter, 65 artefacts have been analysed by more than one group allowing a comparison of the different analytical programs. Thornton noted in general a good agreement between the different programs, with the main discrepancies occurring when lead or arsenic were present in more than trace amounts, in which cases the Thornton considered the SAM data should be considered as semi-quantitative. Heskell also had difficulties detecting arsenic and Thornton chose to “approach his chemical data with great caution”. The result showed that the main elements present in copper-based objects from Tepe Hissar were lead and arsenic. Only two objects were tin-bronze. One is of no interest for this project as it is dated to the Sassanian period, but the other one, a spear-head with 3.6% tin, was found in the Hissar

IIIC level (late 3rd to early 2nd millennium BC) in association with objects of Bactria-Margiana Archaeological Complex connections proving, for Thornton, that tin bronze came to Iran from Turkmenistan. Thornton found that the levels of lead and arsenic were significant enough to consider that the alloying was intentional. Whereas the amount of lead in the objects fluctuated over time, the amount of arsenic, after the first appearance of arsenical copper, stayed roughly the same, showing that the production of copper-arsenic alloys was a standard practice not affected by changes in social contexts. Thornton's analysis of the slag remains added to the understanding of metal production at Tepe Hissar. It appeared that the main variations in production were not chronological but spatial: a small-scale 'traditional' yet innovative production of arsenical copper on the Main Mound and a large scale 'standardized' production of copper and copper-lead alloys on the South Hill.

In 2003, Pigott and the MASCA (Museum of Applied Science Center for Archaeology at the University of Pennsylvania Museum) published two articles on the copper artefacts of the site of Tall-i Malyan (ancient Anshan) in Elam: one presented the artefacts from the Banesh period (3400-2800 BC) (Pigott et al., 2003a) while the other one consisted of the analysis of the Kaftari period copper-based objects (2400-1600 BC) (Pigott et al., 2003b). In both cases, the objects were analysed by PIXE. Unfortunately only metal processing remains could be sent abroad so the analysed objects are mostly bar and sheet metal stock, casting debris, rods, etc. The analyses are therefore not likely to be representative of the entire Malyan assemblage that also included tools and personal ornaments. Although the composition of these objects can still give some information on the kind of alloys used at Malyan in each period, the predominance of metal processing debris will have to be kept in mind when discussing the region of Fars, as the

objects from Malyan constitute almost the entirety of the objects in our database for this region.

The 30 artefacts from the earlier period turned out to be mostly arsenical copper with an arsenic content ranging typically between 1 and 5% and trace levels of other elements, notably nickel and antimony. The authors confronted these results to the three possible methods of arsenical copper production proposed by Pigott (see Chapter 2) and concluded that the most likely candidate was the low temperature smelting of copper arsenates possibly from the Talmessi complex. Indeed this process is non-slagging which corresponds well to the little quantity of slag recovered at Malyan and produces copper artefacts with typically less than 5% of arsenic, which is consistent with the composition of the Banesh period artefacts.

Of the 11 objects from the Kaftari period, five presented over 2% of tin (up to 16.8%), all of which had less than 1% of arsenic, while two objects had almost no tin and 1% As or more. Again, the authors focussed on the process used to make these artefacts and in particular the tin bronze ones. They suggested that the Kaftari bronzes might have been produced by the co-smelting of copper and tin ores in one crucible or furnace as this method would explain the presence of sizable bronze slags on site and the high variability in the amount of tin found in the objects. Interestingly, they pointed out that the small quantity and size of bronze objects recovered at Malyan do not argue in favour of the remelting of scrap bronze to produce the Malyan bronzes as the quantity of metal available would not have been sufficient to support recycling.

For both periods, the objects were also subjected to metallography, but as the artefacts were for the most part unfinished objects, the metallography did not give much

information on the metalworking operations used at Malyan other than the impression that it was carried out on a relatively small scale.

Also in 2003, Hauptmann et al. published an article aimed at reassessing many aspects of the copper production of the eastern site of Shahr-i Sokhta (Hauptmann et al., 2003). It re-examined the results of analyses carried out up to that date on ores, crucible fragments, slag, metal objects, etc. In particular, it presented the results obtained in 1986 by Helmig on 21 copper objects (prills and lumps as well as ingots) analysed by AAS and wet chemical analysis (Helmig, 1986). Their chemical composition is quite variable. The concentration in arsenic in particular varies from only trace amounts to almost 6%. It is interesting to note the presence of a few samples of speiss that could potentially have been used for the production of this arsenical copper and can explain why arsenical copper objects were produced even though almost all of the ore and slag samples had low concentrations of arsenic. None of the copper-based objects however presented tin in concentrations higher than a few ppm, indicating that tin bronze was not in use at that time (approximately 2700-2500 BC) in Shahr-i Sokhta. The analysis of other aspects of the copper production (ore, slag, matte, crucibles and lead isotope analysis) have lead the authors to postulate a co-smelting in crucibles of copper oxides and copper sulphides possibly coming from the Malik-i Siah mountains west of the site.

The publication by Haerinck and Overlaet between 1996 and 2010 of the work of the Belgian Archaeological Mission in Iran (BAMI) was accompanied by the analysis of some of the material excavated in the Luristan tombs. In 2005, Fleming et al. published a summary of the analysis by Proton-Induced X-Ray Emission (PIXE) spectrometry of 169 objects from the Bronze Age cemetery of Kalleh Nissar and the Iron Age cemeteries of Bard-i Bal, Kotal-i Gulgul and War Kabud in an attempt to create a benchmark against which all the looted items that are now in museum collections

around the world can be compared (Fleming et al., 2005). Although that article contained only a very small part of the raw data, another article published in 2006 presented the raw data for the analysis of the 48 War Kabud objects (Fleming et al., 2006). The analysis of the Early Bronze Age copper-base material from Kalleh Nissar has shown a co-occurrence of tin and arsenic, which was also common during this period in Mesopotamia. This fact, and the absence of known copper deposits in Luristan at the time, have led the authors to suggest Elam or Mesopotamia as the source of the metal used in Luristan. The Iron Age material was characterised by a prevalence of tin bronze as the arsenic and the iron contents were diminished. Many of the typical Luristan bronzes were made in this period, probably representative of a localized production. For all periods, the absence of correlation between the type of object and the tin content was again noted.

An important new contribution to the understanding of Iranian metallurgy was made in the beginning of the 21st century when a tin-copper mine was discovered at Deh Hossein in west central Iran (Momenzadeh et al. 2002). In 2006, in his doctoral dissertation, Nezafati described the mine and provided compositional and lead isotope analysis of its ore. He also presented the result of the EDXRF analysis of 29 Luristan bronzes from the Iranian National Museum, the Tehran Money Museum and a private collection coming from both secure (BAMI expeditions) and insecure contexts and dated from the early 2nd to the 1st millennium BC. It was suggested that the absence of correlation observed between the nature of the objects and the tin content could be the result of the use of a “naturally mixed source of copper and tin” such as the Deh Hosein mine (Nezafati, 2006, p.89). The percentages of other elements in the Luristan bronzes were also deemed compatible with the composition of the Deh Hosein ore.

Research on the Pusht-i Kuh bronzes excavated by the BAMI continued as Françoise Tallon needed a control group to which she could compare the unprovenanced objects from the Louvre that she was analysing as part of her research on Iranian bronzes. This resulted in the publication by Begemann et al. in 2008 of the compositional analyses of 58 Pusht-i Kuh objects and 48 Louvre objects (Begemann et al. 2008). The analyses were performed by what is described in the publication as “Optical Atomic Emission Spectroscopy” (which we understand must be ICP-OES) and Neutron Activation Analysis (NAA). The results from both methods were found to be in good agreement (Begemann et al. 2008, 11). The main difference observed between the excavated and the Louvre objects was the wider spread of the tin content in the second group. Moreover, for the BAMI objects, the peak in tin content was around 3% whereas for the Louvre objects it was around 10% which to the authors suggested a possible local production in the Pusht-i Kuh where the addition of tin was well controlled and used in smaller quantities. 29 BAMI objects and 40 Louvre objects were also subjected to Lead Isotope Analysis. About a third of these objects were grouped in what the authors called the “Main Isotope Cluster” where the isotopic abundance ratios are compatible with the data from ores in the region of Arak and of the Deh Hosein. Surprisingly this “Luristan signature” is absent from contemporaneous Mesopotamian objects (Begemann & Schmitt-Strecker 2008), suggesting that the eastern central Zagros copper and mixed copper and tin ores wasn’t used for the production of Mesopotamian bronze. This led the authors to conclude that the role of Luristan in metal trade with Mesopotamia and Elam in particular might have been less important than Fleming et al. had suggested based solely on the chemical composition of the objects (Fleming et al. 2005).

More data was collected in two MSc. dissertations: one by Lesley Frame completed in 2007 and the other by David Meier completed in 2008. Frame worked on the metal

objects from the site of Godin Tepe in western Iran, kept at the Royal Ontario Museum in Toronto (Frame, 2007; Frame, 2010). She carried out the electron microprobe analysis of more than 60 objects from different periods of occupation of the site by quantitative wavelength dispersive spectrometry (SEM-WDS) as well as a metallographic study of a few samples. These objects presented a wide range of compositions, with tin bronze appearing in the mid 3rd millennium BC but restricted to burials until the 2nd millennium BC. As the evidence for primary copper production at Godin exists but is limited, Frame suggested that most of the metal found there must have been acquired through trade with other sites on the High Road or other trade routes, therefore representing the metallurgy of the entire region. She observed a lack of uniformity in the production techniques of objects otherwise similar in typology which suggests an independent development of the metalworking techniques over the region.

As part of his master's project on pins from the site of Shahdad in south-eastern Iran, Meier performed the EDXRF analysis of 19 objects dating between 3100 and 2000 BC, three from Tepe Yahya, the rest from Shahdad, from the Iranian National Museum (Meier, 2008). He observed that all of the pins, except for one which a high lead content, were made of arsenical copper. He suggested the use of a source of copper close to Shahdad for their production and remarked that a better geological knowledge of the region would be needed to support this hypothesis.

In recent years, attention was given to central Iran with two large scale projects, the Arisman Project and the Sialk Reconsideration Project, both of which had a great archaeometallurgical component. The re-excavation of Sialk revealed large amounts of metallurgical remains and finished objects, demonstrating the importance of Sialk as a metal production locality during the late 5th and 4th millennia BC. Four objects from different periods were analysed by EDXRF for their composition and also subjected to

lead isotope analysis alongside some ore samples (Nezafati et al., 2008). These results were compared to analytical results published by Schreiner (Schreiner, 2002; Schreiner et al., 2003) on ore samples of the region. This did not allow the authors to postulate a provenance for the copper ore used for the metallurgical activities at Sialk, except for a possible link with the Anarak mining area for the later periods.

The results of the first five years of work on the Arisman project were published in 2011 in a substantial volume presenting new information on the production of silver and copper at Arisman as well as mining on the central Iranian plateau. The copper production at Arisman reached an industrial scale in the Proto-Elamite period (late 4th, early 3rd millennium BC) as attested by the slag heaps, smelting furnaces, moulds, etc. recovered at the site. The metalworkers of Arisman are likely to have engaged in a trade of the finished objects, possibly directed towards Susa and Khuzistan in the mid 4th millennium and shifting to the north at the turn of the 3rd millennium (Vatandoust et al., 2011, pp.523-31). Of particular interest for this chapter are the EDXRF analyses of 28 copper objects and copper prills found in Arisman as well as the lead isotope analyses of copper slags and prills from Arisman and other Iranian mines (Vatandoust et al., 2011, pp.675-78). None of the objects from Arisman were alloyed with tin. They presented a relatively high arsenic content and low nickel content. While the lead isotope analysis of the litharge from the sites of Sialk and Arisman both point towards Nakhlak as a source for the argentiferous lead ores used in the production of silver, the situation seems to be more complicated for the provenance of the copper ore. The high arsenic content and the results of the LIA analysis suggest that the ore did not come from the mines of Vesnave. The low nickel content in turn seems to exclude the possibility of the ore coming from the Talmessi-Meskani mines. The lead isotope data brings little more light to the question as the copper slags and prills plot in different

areas of the graphs and while it would appear that local ores and ores from the Anarak area are more likely than ores from the Kasan region (Vesnave), a mixing of ores from different sources is possible, as is a chronological variation for their provenance.

Also the subject of ongoing research is the site of Hasanlu in the north-west. The most recent publication in the series of Hasanlu Special Studies focuses on the crafts (furniture, textiles, glass and metals) and the people of Hasanlu IVB (de Schauensee, 2011). Of interest here is the contribution by Fleming et al. on the archaeometallurgy of some of the 2000 or more copper-based objects recovered at Hasanlu (Fleming et al., 2011). 106 artefacts and six ingots were analysed using PIXE and 11 objects and one ingot were subjected to a metallographic analysis. Before this project, our knowledge of the composition of Hasanlu copper-based metals was limited to the PIXE analysis two objects: a bimetallic spear-point and a bimetallic lion pin published by Pigott in an article focused mainly on the iron artefacts of Hasanlu (Pigott, 1989). The results of the compositional analyses showed that a vast majority (all but seven) the objects were tin bronzes, with a tin content mostly comprised between 6 and 12% but reaching up to 21.3 and 18.3% in two lion pins. As most of the bronzes analysed were decorative, tin must have been added to give the metal a certain colour, rather than for its mechanical properties. The six ingots were all tin bronzes with tin contents between 8.5 and 14.3%. The levels of arsenic, lead, iron, silver and nickel in the objects were all relatively low. The antimony however was surprisingly high, reaching up to 27.9% and being noticeably elevated in the lion pins in particular. One can only speculate as to whether this addition was meant to change the colour of the pins or to increase the ease of casting crisp details.

The last publication used to build this database is an article by Oudbashi et al. reviewing the archeometallurgy of Iran and including the SEM-EDS analysis metallography of 10

objects from the Middle Elamite site of Haft Tepe in Khuzestan and 10 vessels from Sangtarashan in the Pish-i Kuh region of Luristan (Oudbashi et al., 2012). The composition of the Haft Tepe objects turned out to be quite varied. Three of the objects were tin bronzes (3.2 to 17.9% Sn), and all but two of the objects had more than 1% of lead and one reached 18.2%. The nickel content was also relatively high (all but two over 0.5% and reaching 4.7% in one object). It would appear however that the arsenic content was not measured, making this data difficult to use in our work. The composition of the Sangtarashan vessels was much more homogeneous: all were tin bronzes (7.7 to 13.5% Sn) with relatively low levels of other elements. Arsenic was this time measured, making the Sangtarashan data a valuable addition to our database.

### Neighbouring regions

As mentioned earlier, although the primary focus of this research is Iran, it was considered necessary to collect some data from the neighbouring regions in a hope to better understand the exchange of material through the Persian Gulf and via overland trade. The following paragraph will briefly review the publications that were used for that purpose.

In 1972 Caley examined some artefacts from north-eastern Afghanistan (Caley, 1972). Unfortunately, only two of these objects came from time periods relevant for our project. Caley did not mention which analytical technique was used for his study, but he was able to give weight percentages for copper, tin, lead, iron, nickel and zinc. However, he deplored the fact that most of the objects he was able to analyse were very corroded and although he attempted to recalculate the original compositions from the corroded compositions, some uncertainty remained as to the original nature of the metal.

The next region for which we gathered analytical data was the Oman Peninsula. The first source we used was the work of Craddock published in 1985 on the metal objects from the tombs of Ras al-Khaimah (Craddock, 1985). Craddock performed an AAS analysis of 29 objects and noted high levels of nickel (up to 4.65%) that he attributed to the smelting of nickel rich copper ores. He suggested the nearby copper deposits of Jabal al-Ma'adan as a possible source for the ore used at Ras al-Khaimah.

In 1988 Hauptmann et al. published more data for the Oman Peninsula (Hauptmann et al., 1988). They reported on the ICP-OES analysis of copper bun-shaped ingots, fragments and artefacts from the site of Maysar, where a hoard of 22 ingots was recovered, from the site of Soweiq and from the island of Masirah. They also gave the results of the AAS analysis of tin bronze artefacts from the Selme hoard, some of which were later reanalysed by Prange and Hauptmann (2001) (see below). These objects had tin contents generally ranging between 9 and 13% Sn, which is slightly higher than the ranges observed in most Iranian regions as seen above. From these analyses, they were able to suggest the following chronology for the use of different alloys in Oman: the use of copper-arsenic in the Umm an Nar Period, the use of copper-arsenic-nickel in the later Wadi Suq Period simultaneously with the first appearance of tin bronze and the appearance of “true” tin bronzes (defined in that publication as having more than 9% Sn) after 1200 BC.

Work on the Oman Peninsula continued in the following decade and in 1997 Weeks published an article on the metallurgy of the site of Tell Abraq in what is now the United Arab Emirates (Weeks, 1997). Of the more than 550 metal objects excavated there, he analysed 150 objects (finished objects and metallurgical waste) from different periods of occupation using SEM-EDX and metallography. His main aims were to understand the production processes of the objects and how they evolved over time and

varied between the different types of objects. He also wanted to consider the metallurgy of Tell Abraq in a wider geographical context. The composition of the metallurgical waste as well as the presence of a copper ingot and of metallurgical ceramics pointed towards a secondary processing of copper at the site: the refining of a copper rich in iron and sulfur (matte). The compositional analysis of the finished objects revealed the presence of arsenic and nickel, often as minor elements, and sometimes in amounts over 1%. Weeks considered that this presence was due to the presence of these elements already in the raw copper, not to intentional alloying. Weeks also noted significant difference in compositions over time and between different types of objects: tin bronze was being used more often in the Umm an-Nar period (2300-1900 BC) than in the subsequent Wadi Suq and Iron Age. Vessels and rings were also more frequently made of tin bronze than utilitarian objects, suggesting that the tin was added for the colour it gave the objects rather than for potential improvements in mechanical properties. In general, Weeks noticed an unexpected prevalence of tin bronze in Tell Abraq, quite surprising for the Oman Peninsula. He suggested that this might be a result of the location of the site on the coast, positioning it on potential tin trade routes and certainly being able to interact with other tin-using people around the Gulf.

In 2000 Weeks published more data from the United Arab Emirates, this time from the metal assemblage of the Sharm tomb in Fujairah (Weeks, 2000). He analysed 21 objects using the PIXE technique. Half of the analysed objects were tin bronzes, with a relatively high percentage of tin (around 10 to 15% and up to 22.2%). Also notable is the amount of iron in a majority of the objects, perhaps attesting of a lack of refining during their production. Once again Weeks observed that the tin bronzes were used mostly to make vessels perhaps indicating that tin bronze was initially a “prestige item” in south-eastern Arabia.

The following year, Yule and Weisgerber published a volume on a metal hoard found in the plain of Selme, near Ibri, in the Sultanate of Oman. The hoard is composed of over 500 metal objects: weapons, vessels, bangles, etc. from the Early Iron Age. Prange and Hauptmann contributed to this volume with a chapter on the chemical composition of the hoard (Prange & Hauptmann, 2001). 86 of these, mostly bangles, were sampled and analysed by ICP-OES and one of them was subjected to metallographic investigations. 17 of these artefacts had already been analysed by AAS and the results of these analyses were published by Hauptmann et al. (1988, p.47). All but one of the objects are alloyed with tin, and all but three have tin concentrations between 7.3% and 13%. For Prange and Hauptman, such a small range of concentrations reflected the use of a single recipe for the production of the hoard, possibly coming from one workshop over a short period of time. The percentages of arsenic and nickel in the artefacts are mostly in the 0.1 to 1% range which is similar to the range observed in other Omani metals and compatible with the use of a local ore. The metallurgical analysis of the bangle pointed towards the use of a sulphidic ore and showed that the piece was hammered and annealed a few times after casting.

More Omani data was published in 2003 by Craddock et al. on the settlement site of Ra's al Hadd situated on the coast, at the very east of Oman and dating from the 3rd millennium BC (Craddock et al., 2003). This was prompted by the discovery and analysis of a small ingot of copper and matte with a surprisingly low iron content. 19 other objects, mostly small points and hooks, were analysed using the AAS method. All, including the ingot, had elevated amounts of arsenic (0.5 to 2.8%), higher than usual for objects of the Oman Peninsula and in particular of the neighbouring site of Ras al Khaimah mentioned above. However, the rest of the composition and in particular the relatively high nickel (0.28 to 2.13%) appeared to be fairly consistent with Omani

copper. The authors also discussed the differences in iron levels observed at various sites in Oman and how they can be representative of different levels of refinement.

Also in 2003, Weeks published a book dedicated solely to the metallurgy of the Persian Gulf (Weeks, 2003). Doing it justice here would be impossible as it addresses many aspects of metalworking in Oman and the U.A.E. Of interest for the creation of our database is the compositional analysis by PIXE of 83 objects from four sites, Al Sufouh, Unar 1, Unar 2 and Tell Abraç, dating roughly between 2500 and 1900 BC. Over 40 of them were also subjected to lead isotope analysis. The initial aim of this project was to give more context to the earlier work of Weeks on the site of Tell Abraç. Weeks found that the early use of tin bronze observed at Tell Abraç was not exceptional: the other coastal sites of northern U.A.E displayed similar alloying patterns, illustrating a specific role of this region in a wider trade of metals. As had already been observed, three main types of copper were used there: unalloyed copper, arsenic or nickel alloys (As and/or Ni between 1 and 6%) and relatively high tin bronzes (typically >10%). The arsenic-nickel alloys were used mostly for utilitarian objects either reflecting their use for improved mechanical properties, or as a result of their relative low value compared to tin-bronze that was used mostly for decorative objects. The lead isotope analysis showed a wide diversity of isotopic values indicating multiple sources for the metal used in Oman, but definitive conclusions are made impossible due to incomplete data on isotopic signatures of ores of that region.

The last set of data collected for the Oman Peninsula was the result of the analyses of copper ingots found in an Iron Age building at Masafi in the Fujairah Emirate under the direction of Dr. Anne Benoist (Attaelmanan, 2012). Their composition is unsurprising when compared to the other Omani objects: elevated amounts of arsenic and nickel

most likely deriving from the ore used to make these ingots and also relatively high iron concentrations, probably reflecting their low level of refinement.

In their article on metal working in the Indus Valley published in 1999, Kenoyer and Miller included tables presenting the results of 129 previously published chemical analyses of copper-based objects (Kenoyer & Miller, 1999): artefacts from Mohenjo-Daro originally published by SanaUllah (1931, p.484) and Hamid in Mackay (1938, pp.479-80), artefacts from Harappa (SanaUllah, 1940, p.378; Pigott et al., 1989), from Lothal (Lal, 1985) and Rangpur (Rao, 1963, p.153). We only entered in our database the data from Mohenjo-Daro and Harappa as the other two sites were considered too far away from our region of interest. Kenoyer and Miller observed varying contents of tin and arsenic, but no clear pattern dependent on the category of artefact emerged. They clearly stated some of the problems inherent to grouping old datasets: the lack of stratigraphy for some objects and the differences in sampling and analytical methodologies. Their discussion was also made difficult by the fact that not very many analyses are available for the Indus Valley. Finally they noted the added difficulty induced by the recycling of copper-base metals, when commenting on alloy compositions. For the Indus Valley this practice is indicated by the presence of caches of scrap metal and broken objects on major archaeological sites.

The next region for which data was collected was Mesopotamia. There, the analysis of copper-based objects goes back to the 19th century: in the metallurgical appendix to Layard's 1853 "Discovery's in the Ruins of Nineveh and Babylon", Percy gives the percentage of copper and tin of four Neo-Assyrian bronze objects (Layard, 1853, p.670). Interestingly he also commented on the manufacture of the bi-metallic tripod he analysed stating that the bronze was probably cast-on to the iron. Many then followed Percy's lead and in 1985 Moorey summarized the analytical studies, both qualitative

and quantitative, that had been published up to that date (Moorey, 1985, pp.51-68). We were able to enter 127 objects from these publications in our database. Unfortunately, very few of the analytical methods used are mentioned and for many of the objects the range of elements tested was very limited. As an example, tin was sought for in 121 objects while antimony was tested in only 36 cases.

The next source of Mesopotamian data used was the SAM data (Krause, 2003) which included the OES analyses of 75 objects from the site of Tepe Gawra collected by the SAM and the analyses of 20 artefacts from Ur collected by Berthoud (1979).

Finally, data was collected from a catalogue of 2623 objects from Mesopotamia and Syria published in 2004 by Harald Hauptmann and Ernst Pernicka (Hauptmann & Pernicka, 2004). Most of the objects were subjected to XRF analysis and 145 were also subjected to neutron activation analysis (NAA). Unfortunately, to our knowledge, the second part of this study including the discussions of these results has yet to be published. This data included the re-analysis of 14 objects from Ur previously analysed by Berthoud, 22 more objects from Ur previously analysed by AAS by Craddock (1984) and re-published by Moorey (Moorey, 1985, pp.66-68) and 31 of the Kish and Ur objects previously analysed by XRF by Moorey and Schweizer (Moorey & Schweizer, 1972) and also re-published by Moorey.

These publications provide valuable information on Mesopotamian copper metallurgy as they include a wide range of types of objects across a large number of sites. Unfortunately however most of the objects date from the 3rd or early 2nd millennia BC with only 101 objects from Iron Age periods. The Mesopotamian Metals Projects undertaken from 1980 at the University of Pennsylvania by Fleming, Muhly, Pigott and Stech also analysed about 350 Mesopotamian copper-base objects using the PIXE

method (Stech, 1999), however the raw data for this study remains to our knowledge unpublished.

The last region for which metal objects were entered in our database was Northern Bactria: in 2006, Kaniuth published a book detailing the typology and chronology of metal objects of the Sapalli Culture of southern Uzbekistan (Kaniuth, 2006). 194 objects from the sites of Dzarkutan and Sapalli Tepe, dating from between the 20th and the 15th millennium BC, were analysed. A wide range of alloys were used (tin bronzes, leaded copper and arsenical copper) and different alloys were used for different types of objects. The seals and flacons for examples present a high percentage of lead. As has been noticed in many other places, tools were often made of unalloyed copper, once again showing that alloying practices were probably linked to the outside appearance of the objects rather than to their mechanical properties. In the later periods of the Sapalli Culture (17th-15th centuries BC), both the typology and the chemistry of the objects (high tin bronzes) point towards an influence of the Andronovo Culture in the Eurasian Steppe and possibly towards a use of the tin-rich copper deposits of the Upper Zerafshan valley.

## 2. Integrating the data

### a. The geographical and chronological framework

The data collected represents a total 81 sites in Iran and the neighbouring countries. For most of the objects, the site and context of their discovery are known. The chronology of most Iranian sites however does not allow a very fine dating for the objects, and it is not uncommon to see objects attributed, in their original publication, to a time period of

over 300 years (see Figure III-5). The chronologies of the Qazvin Plain and the central Iranian plateau (notably the site of Tepe Sialk) are currently being revisited by the means of radiocarbon dating (Pollard, personal communication), but this project has not yet affected the sites of interest here.

A limiting problem, however, is that of the many objects of uncertain provenance. The most famous difficulty is of course Luristan, where tombs were extensively looted for their metal objects, a great number of which appeared on the art market in the early 20th century. The Luristani objects aren't however the only problematic ones and other objects, mostly from art or museum collections, are attributed to a region and dated based on typological grounds. In all of these cases, we have no other choice but to rely on the expertise of the person who has studied the objects and proposed a tentative provenance and dating, as a reassessment of all of these unprovenanced objects would be beyond the scope of the present project.

To determine the framework of this research we took into consideration these uncertainties, but also the number of objects necessary for a valid statistical approach to the metal composition: in order to keep enough data per period and region to draw meaningful conclusions it was necessary to define relatively large geographical regions and relatively long time-periods.

### Geographical framework

The sites for which compositional analyses were collected are shown on the map of Figure III-3. They were grouped in 14 regions as shown on the map of Figure III-4. The aim was for each region not only to represent a geographical area, but also a cultural entity.

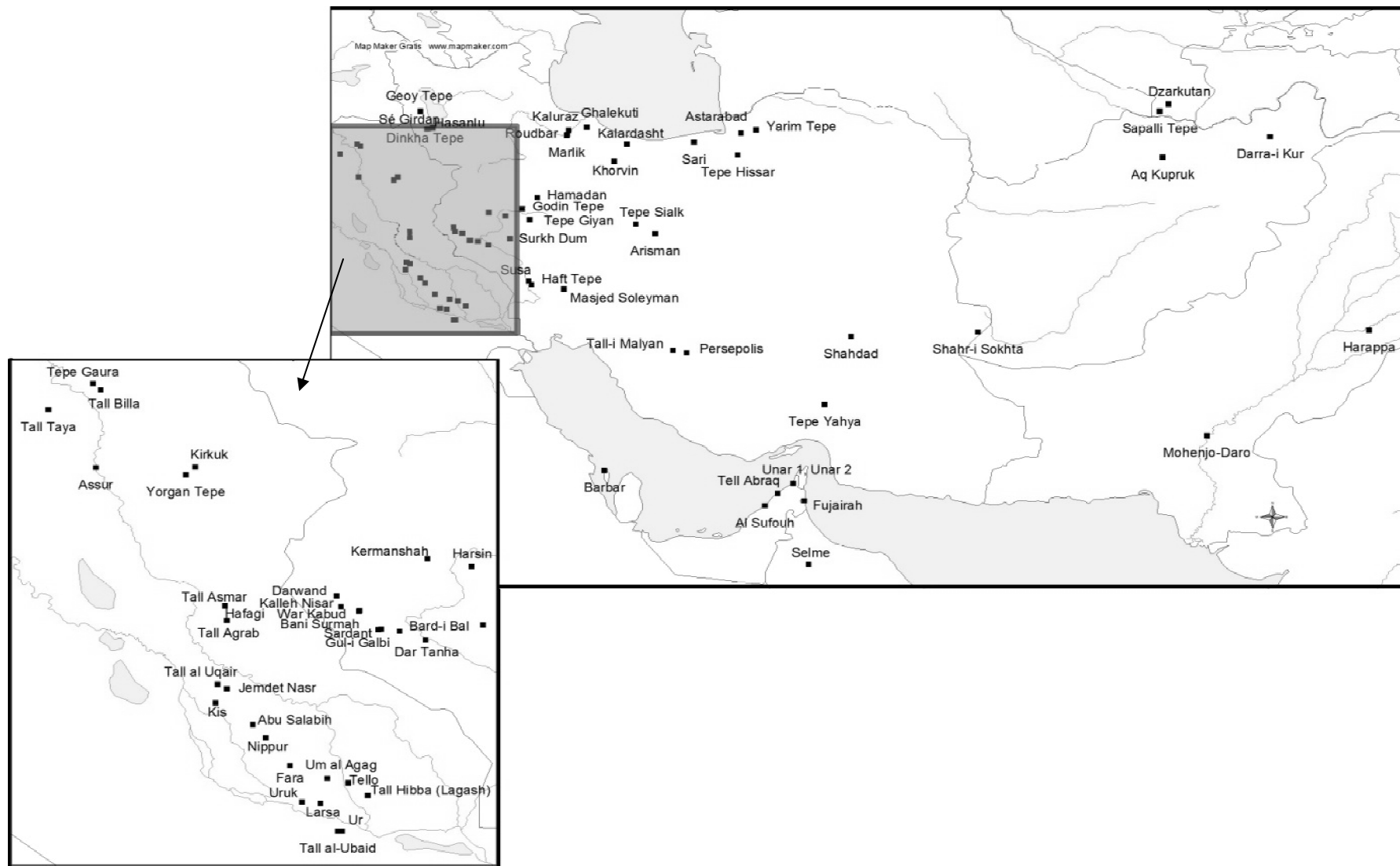
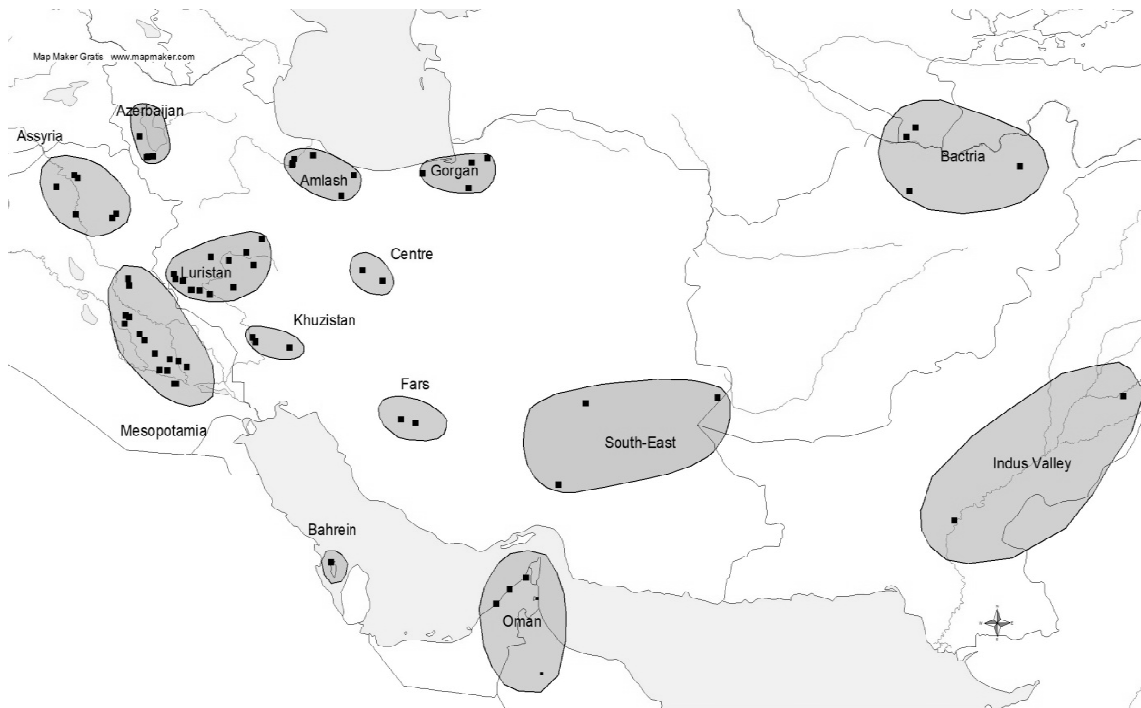


Figure III-3. Map of the sites for which compositional data was collected



**Figure III-4. Map showing the regional divide used in this thesis**

Although the lowland plain of Khuzistan with the site of Susa notably and the highland region of Fars with Tall-i Malyan were both part of the Elamite kingdom, we chose to treat them separately as they span over a considerable area and they are likely to show different influences, Khuzistan being in closer contact with Mesopotamia for a substantial part of its history.

Similarly, it was thought that the highland settlement of Tepe Hissar and the sites located in the Gorgan plain near the Caspian Sea should preferably be treated separately, but the dearth of data for the Gorgan sites meant that both had to be grouped under one region that we called Gorgan. This is not completely absurd, as highland and lowland settlements are thought to have been in contact on a local scale. Thornton has noted that Tepe Hissar was closely connected to the sites of the Gorgan plain despite the Elburz Mountain range between them (Thornton, 2009b, p.307).

It should be noted that the region labelled Oman in this thesis represents the Oman Peninsula regrouping both modern Oman and the United Arab Emirates.

### Chronological framework

We have defined two chronological scales for the analysis of the copper-based data. The first one is a simple division into thirds of a millennium (early, mid and late 2nd millennium for example). Each object was attributed to one of these time periods to the best of our ability based on the dating provided in the publication they came from, even when that dating was only tentative. When objects fell at the interface between two time periods, the earlier one was chosen. For example an object dated “1700-1000 BC” was attributed to the mid 2nd millennium BC. This decision was arbitrary and made for the sake of consistency, but it must be remembered that the interpretations, if anything, might be slightly shifted towards early dates.

Figure III-5 shows the time ranges given in the original publications for the dating of objects, and the time period to which they were attributed in this thesis. We can see that the time ranges given only rarely span less than 300 years, meaning that defining a tighter chronology would have been unwise. A division in thirds of a millennium seems to give a coherent chronology, but it is unsurprisingly far from perfect. Indeed, there are a number of objects that have been attributed to a time period but 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 for the analysis, a second, less detailed, scale was found necessary. We have therefore defined two main periods of analysis based on three criteria, the historical

context presented in Chapter 1, the availability of the data and the problem addressed in this thesis, which is the transition between the Bronze Age and the Iron Age.

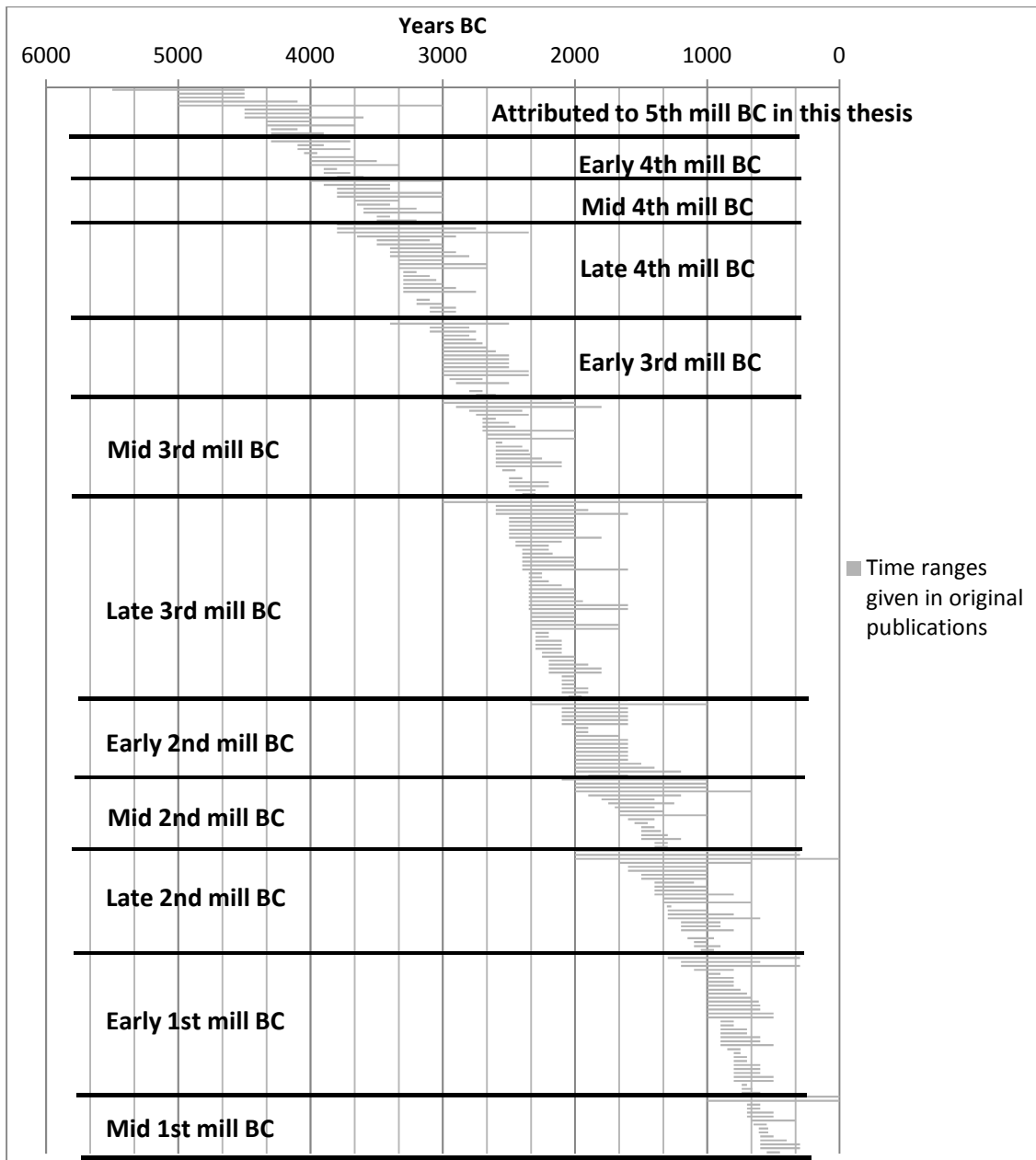
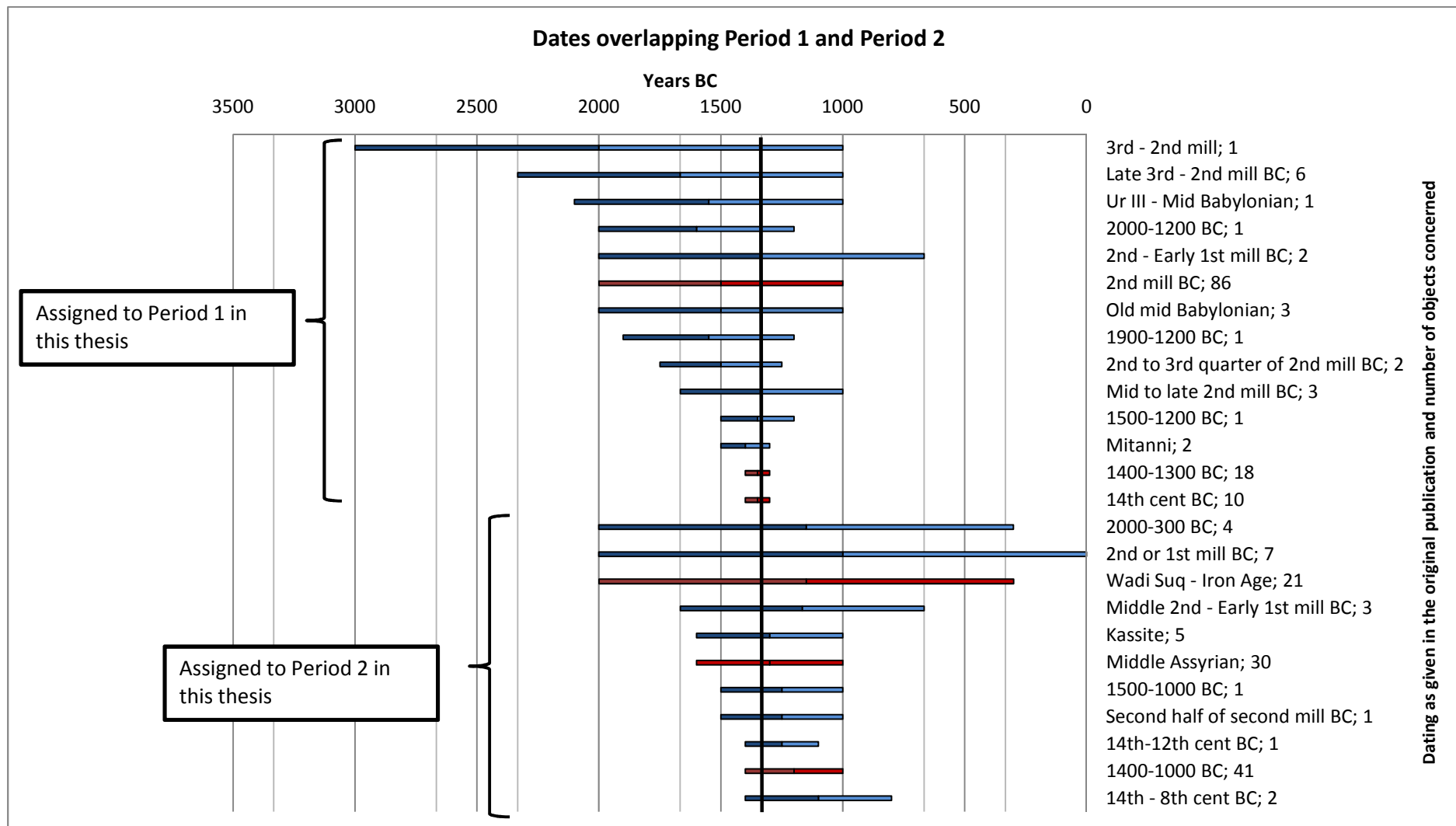


Figure III-5. Time ranges given in the original publications to date the copper objects and time ranges to which they were attributed in the framework of this thesis

Figure III-7 and Figure III-8 show the number of objects analysed for each region in each time period in Iran and in the neighbouring regions. As we can see, only a small amount of data is available for the mid 2nd millennium BC. It is remarkable to note that

this is the case across all of the regions considered in this thesis. However, this is not overly surprising as this period was marked, in general (but most obviously in eastern Iran and Luristan in particular), by a decrease in settlement density throughout the Near-East (see Chapter 1). The mid 2nd millennium also corresponds to the end of a period of international trade notably through the Persian Gulf and to the appearance of iron in Iran. We have therefore decided to define our first time period (Period 1) as spanning between the early 3rd millennium and the mid 2nd millennium inclusive and the second period (Period 2) from the late 2nd to the mid 1st, in other words representing the Iron Age.

Figure III-6 shows the time periods given in the original publications that overlap both Period 1 and Period 2. As we can see, the latest objects of Period 1 could actually date as late as the end of the 1st millennium BC and the earliest objects of Period 2 could date as early as the very beginning of the 2nd millennium BC. Although this is a significant overlap, the number of objects dated to each one of the overlapping periods is on the whole relatively small. The most worrying cases are the 86 objects dated to the “2nd millennium BC”, the 21 objects dating to the “Wadi Suq or Iron Age” period and the 30 “Middle Assyrian” objects. In the first case, the objects come from different regions of western and northern Iran and Mesopotamia. Khuzistan is the region with the most objects dated to the “2nd millennium BC”. There, a maximum of 28 objects may have been misattributed to Period 1. In Oman, 21 of the 213 objects of Period 2 are dated to the “Wadi Suq or Iron Age” and could in fact belong to Period 1. Similarly, the 30 Assyrian objects dated to the “Middle Assyrian Period” and attributed to Period 2 could shift into Period 1, but of these objects only four are given with a measurement of the antimony and nickel content, meaning that it should not have much impact on our assessment of the changes in Assyrian metallurgy (see Chapter 6).



**Figure III-6. Time ranges given in the original publications that overlap Period 1 (early 3rd to mid 2nd millennium BC) and Period 2 (late 2nd to mid 1st millennium BC). The periods in red are those to which 10 or more objects are associated.**

Overall, given the number of objects analysed for each region (see following section), the possibility that some might have been misattributed weakens slightly our analysis but is not an extremely problematic limitation to this study.

Unfortunately, not all regions are well represented over the entire time frame of this project (Figure III-7 and Figure III-8). Very few objects have been analysed altogether for Fars, central Iran, the Indus Valley and Bahrain. Azerbaijan and Amlash, two regions of north-western Iran, are poorly represented for the first time period. In the second period on the other hand, western Iran is well represented with many analyses from Azerbaijan and Amlash, but also from the bronzes of Luristan that have been the subject of much interest over the past century. Very few Iron Age analyses are available however for eastern, central and southern Iran. This can be partially attributed to the abandonment of many settlements from these regions before the 1st millennium BC. Regrettably, Khuzistan is also poorly represented in this period: not enough objects from the end of the Middle Elamite period or the Neo-Elamite period have been sampled for us to be able to comment on the metallurgy of these periods in the following chapters.

As we can see on Figure III-8, the number of analyses from Mesopotamia in the 3rd millennium clearly outweighs any other regions meaning that we will be able to make observations with a good degree of confidence for this region in our first time period. However we only have a small number of Mesopotamian analyses for the Iron Age, therefore any comparison of the two time periods for this region will be difficult. A similar problem occurs with Bactria where most of the data comes from the early 2nd millennium BC. Oman is the only one of the neighbouring regions to be fairly evenly represented throughout the entire time-frame.

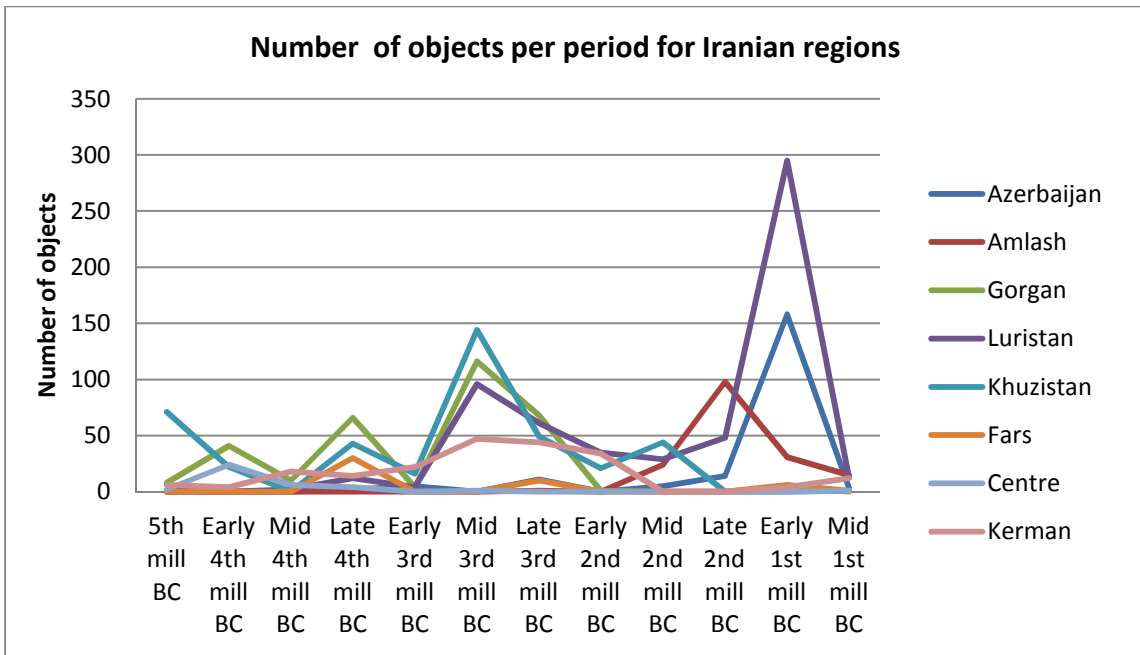


Figure III-7. Number of objects entered in the database for each period (Iranian regions)

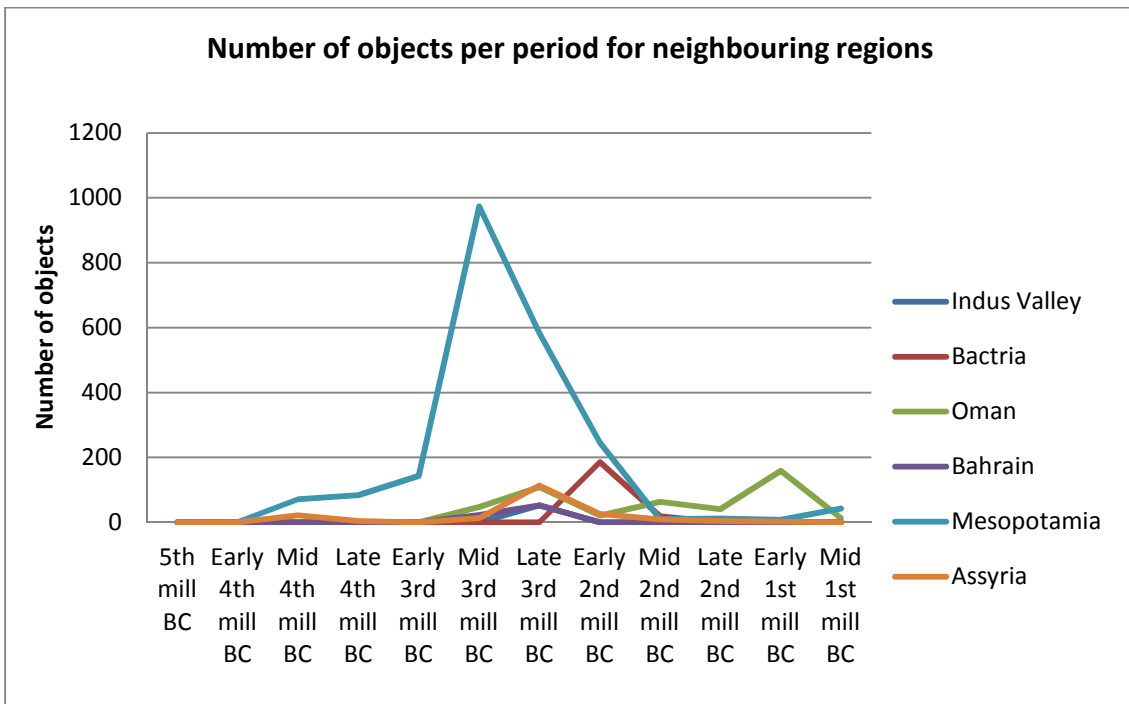


Figure III-8. Number of objects entered in the database for each period (neighbouring regions)

b. Integration of a large and varied dataset

Inevitably, the 51 studies used for this project and conducted over 80 years used a wide range of experimental methods, reflective of the evolution of the techniques used in archaeometry and as we have seen in the previous section, some studies did not even mention the analytical technique used to determine the composition of the objects. In addition they did not all focus on the same set of elements. The precision, accuracy and detection limit of the measurements of course varies from one technique to another. But measurements obtained by a single technique can also be affected by differences in the calibration of the instruments by different groups and by the lack of homogeneity within one object. How then is it possible to directly compare the data of all of these studies?

Element set

Figure III-9 shows for each element the number of measurements entered in the database<sup>2</sup>. As previously mentioned, not all studies have analysed the same suite of elements but 10 elements stand out as being most frequently analysed: tin, lead, arsenic, antimony, nickel, iron, zinc, silver, gold and cobalt. Of these, tin was looked for most often which isn't surprising as the aim of many studies was to differentiate copper from bronze. Gold and cobalt on the other hand were less commonly sought for. In this thesis, we primarily look at the tin content in Chapter 4, the lead and zinc contents are briefly examined in Chapter 5 and the arsenic, nickel, antimony and silver contents are studied in detail in Chapters 5 and 6.

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<sup>2</sup> For some elements the total number of analyses exceeds the total number of objects inserted in the database presented above. This is due to the fact that some objects have been either analysed in multiple spots or by multiple techniques.

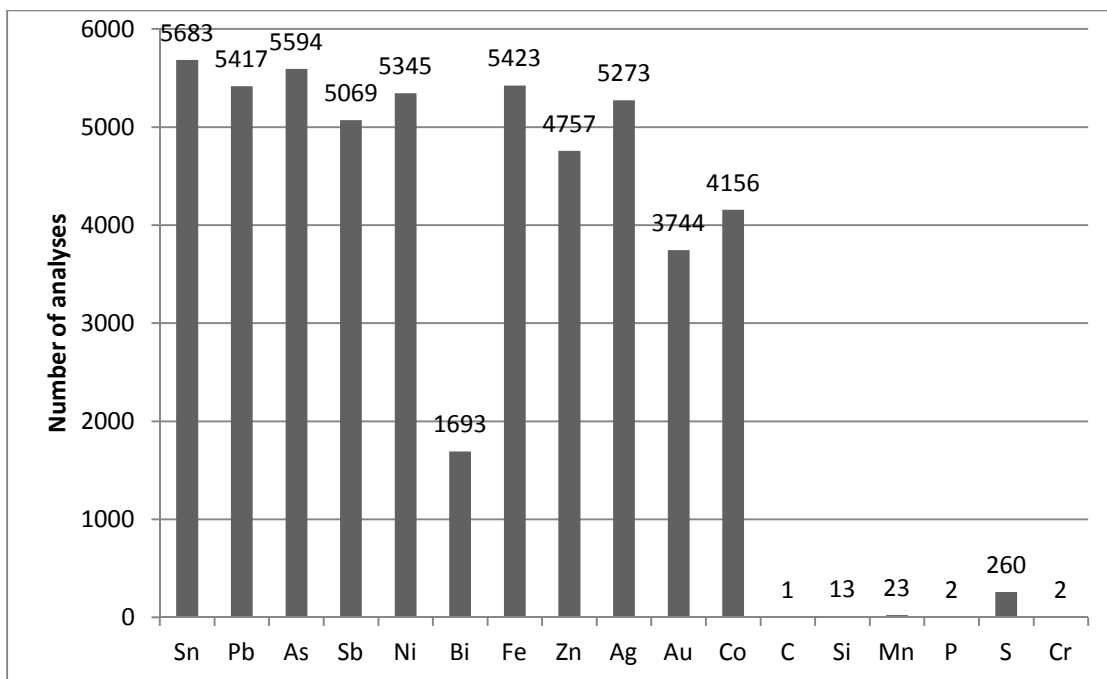


Figure III-9. Number of measurements entered in the database for each element

### Inhomogeneity of the objects

Regardless of the technique employed to analyse an object, a lack of homogeneity due to the segregation of elements within the objects can induce some variability in the measurements. The impact of this lack of homogeneity on the assessment of the composition of an artefact has been discussed notably by Bray (2009). He concluded that segregation was not a critical problem as the absolute variations in element levels are actually very small and several teams had accounted for it by analysing multiple spots for each artefact. In our case, this practise was used for example by Northover in his work on the Mahboubian collection, where he gave for each object the mean of three measurements (Mahboubian, 1997) and by Gunter and Jett who gave for each object the average of two EDXRF measurements (Gunter & Jett, 1992). More studies have possibly used a similar approach but the exact experimental procedure is very rarely published, making the impact of segregation on our work very difficult to assess.

Moreover, many objects of our database are constituted of multiple parts either cast on to each other or assembled by riveting (for example a dagger with a cast-on grip, a vessel with a handle, etc). A look at the database reveals that in several cases these parts are made of metal of significantly different composition. Table III-5 shows the example of a Luristan dagger analysed by OES by the SAM project (Krause, 2003) where the hilt is alloyed with tin, while the blade is made of copper with an elevated amount of arsenic and a percentage of antimony also higher than in the grip, clearly indicating that several different alloys could be available simultaneously to a metal-smith and were indeed used simultaneously.

					Sn	Pb	As	Sb	Ni	Fe	Zn	Ag	Au	Co
<b>Dagger</b>	2nd BC	mill	Luristan	Grip	4.7	0.03	0.48	0.02	0.35	0	0	0.02	0	0
				Blade	0.09	0.03	1.45	0.85	0.22	0	0	0.07	0	0

**Table III-5. Chemical analysis of the grip and blade of a Luristan dagger**

It therefore became obvious that the analysis of one part of an object is not necessarily representative of the entire object. For this reason, this thesis will never focus on commenting on individual objects but on a large sample size as it allows for the inhomogeneities and the discrepancies in metal choices to average out.

Another factor that can not only cause the metal to become inhomogeneous, but also significantly skew the measurements, is corrosion. An example of this phenomenon comes from the site of Tell Abraq where very high tin contents in the archaeological artefacts were interpreted as a result of a preferential corrosion of the copper. Indeed, in the acidic soil of Tell Abraq, the tin remained while the copper corroded and the corrosion products leached to the surface (Weeks, 1997, p.76). In such cases, the data can only be considered qualitatively and rather than looking at the actual percentages of

certain elements in the objects, we look at whether or not an element is can be considered as being “present” in the object (see below).

### Technique-based variability

As we have seen above, many different techniques have been used to collect the data used in this thesis: OES, AAS, ICP- OES, XRF, EMP, PIXE, ICP-MS, etc. We will not embark here on a detailed review and comparison of these techniques as it has been done elsewhere (Pollard & Heron, 2008). This would be of little use in our case, as for some of the analyses in our database, the techniques aren’t even mentioned. We however attempt to assess how much variability is typically linked to the technique in order not to jump to erroneous conclusions later in this thesis.

Three aspects should be considered here: the accuracy of the methods, their precision and their detection limits. The accuracy represents how close a measurement is from the true value, in our case the true weight percentage of an element. The precision represents the reproducibility of a method under fixed conditions. As we have already mentioned, we will not attempt to comment on the composition of isolated objects, therefore the precision of the measurements is slightly less critical for us: the errors will tend to average out over a large sample size. The accuracy on the other hand is more problematic. Assessing the accuracy of each publication is rendered impossible for us due to the fact that most publications do not mention if their instruments were calibrated with standards. Differences in accuracy also induce a technique related bias between the different studies. This wouldn’t be a problem if each study had analysed objects from all the different regions and time periods, but in our case some regions are represented almost exclusively by one publication, e.g. Mesopotamia where almost all of the data

comes from Hauptman and Pernicka's study (Hauptmann & Pernicka, 2004). Therefore, when comparing the metal assemblage of different regions or time periods we are at risk of picking up variations related to offsets between techniques. In the following paragraph, we attempt to assess the order of magnitude of these variations and the impact they have on our discussion.

In his work on the bronze material from Tepe Yahya, Thornton noticed that Heskell's OES analysis gave results for arsenic consistently 1 to 2% lower than his ICP-MS analyses (Thornton et al., 2002, p.1457). We examine here the data of four other sites, where a significant number of objects have been analysed by more than one method to see if they present a similar method-related bias. 56 Tepe Hissar objects were analysed both by OES (SAM project and Heskell) and PIXE spectrometry (MASCA) (Thornton, 2009), 151 objects from Mesopotamia were analysed by XRF and AAS (Hauptmann & Pernicka, 2004), 17 objects from Selme in Oman were analysed by AAS (Hauptmann et al., 1988) and ICP-OES (Prange & Hauptmann, 2001) and for Susa 70 objects were analysed by Berthoud with both OES and spark source and mass spectrometry (Berthoud & Francaix, 1980).

### Alloying elements

*Tin:* Most of the Susa and Hissar objects were not alloyed with tin. For the Mesopotamian sites and Selme the values of both sets of measurements showed a general agreement (Figure III-10 and Figure III-11) with a small bias in the case of Mesopotamia for which the AAS measurements were slightly lower (Figure III-10).

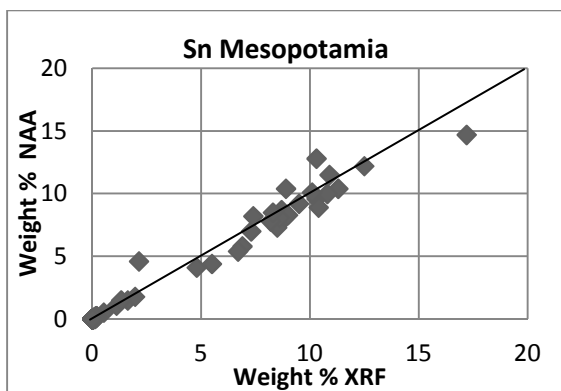


Figure III-10. Comparison of XRF and NAA tin measurements for the Mesopotamian sites

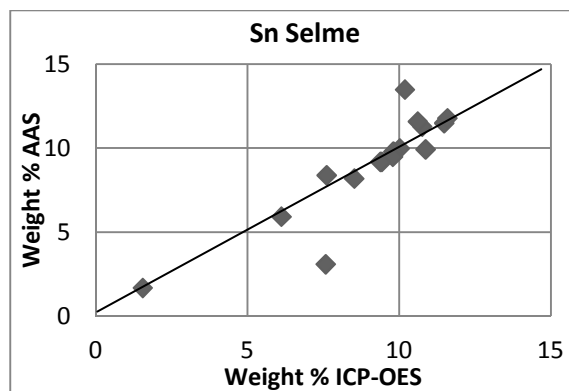


Figure III-11. Comparison of ICP-OES and AAS tin measurements for Selme

Figure III-12 shows for each of our four sites how much difference in weight percentage is typically observed between two measurements of the same object (when tin was detected by both methods). As we can see there are some instances where the measurements vary quite drastically (up to 4.5 wt % difference for Selme), but in most cases the variation remains under 1 wt % and in more than half of the cases it is under 0.25 wt %, which is on the whole relatively insignificant.

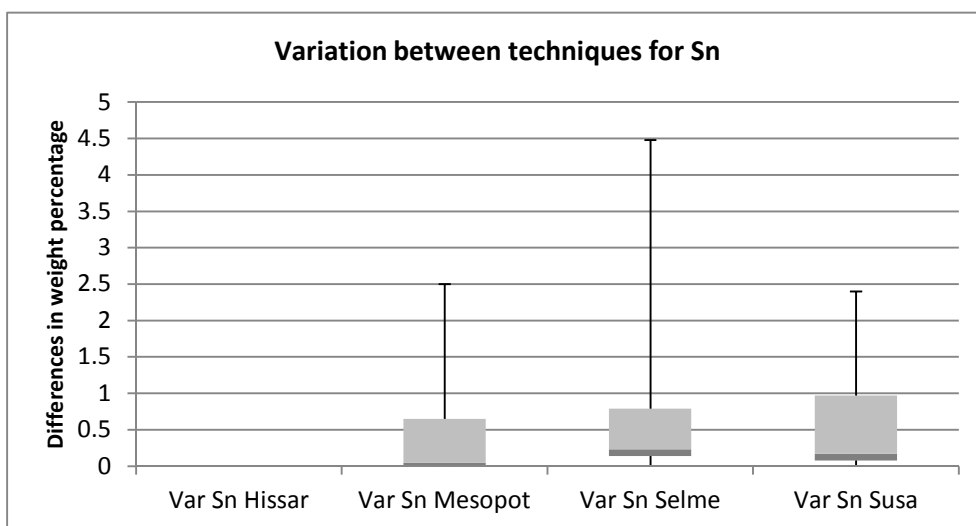


Figure III-12. Spread of the absolute difference in weight percentage of tin between two measures of one object using different techniques

Lead: The agreement isn't as convincing for lead. In the case of Hissar, all of but one of the PIXE values are higher than the OES values. In the case of Mesopotamia, the XRF values are typically only two thirds of the NAA values. Selme has very few objects where lead is present in more than trace quantities. Finally in the case of Susa, most of the values obtained by mass spectrometry are higher than the ones obtained by OES and mass spectrometry detected lead in some objects where OES didn't (Figure III-13).

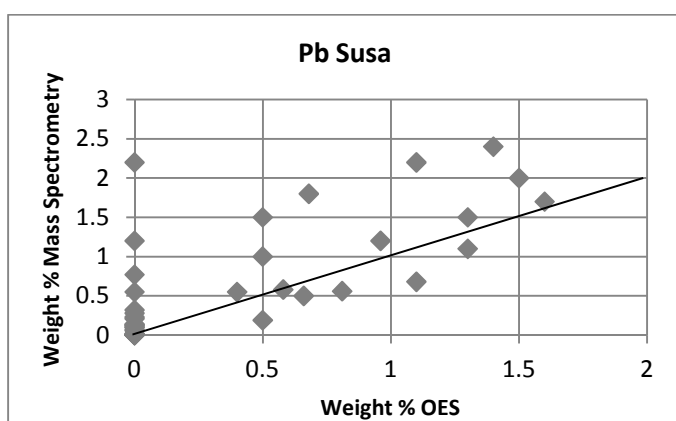


Figure III-13. Comparison of Optical Emission Spectrometry and Mass Spectrometry lead measurements for Susa

Arsenic: At Selme, arsenic was only found in small quantities (<0.5%) and although there is a slight scatter, the AAS and OES values are in general quite close with an average difference of 0.07 wt % between the two measurements. For the three other sites a wide range of arsenic content was observed (Figure III-14, Figure III-15 and Figure III-16). In the case of Hissar, the OES measurements picked up, in some cases, relatively high arsenic contents where the PIXE method only detected traces. The NAA and XRF measurements in Mesopotamia were in good agreement with a slight offset: the NAA measurements are slightly higher. Berthoud's values for Susa are relatively scattered with differences of up to 4.1 wt % between measurements.

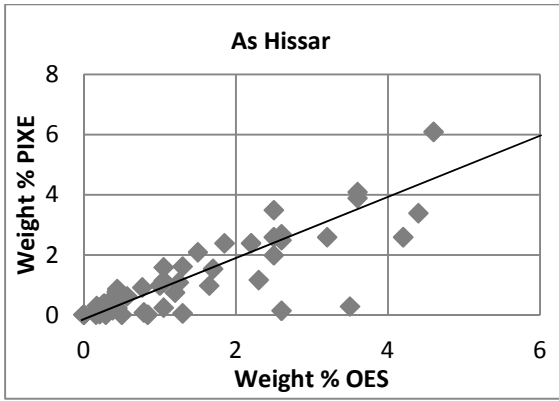


Figure III-14. Comparison of OES and PIXE arsenic measurements for Hissar

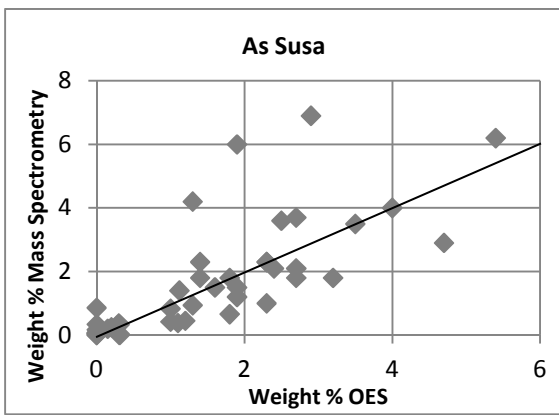


Figure III-15. Comparison of OES and Mass Spectrometry arsenic measurements for Susa

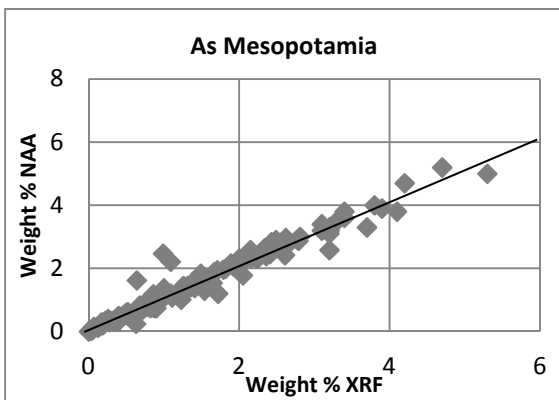


Figure III-16. Comparison of XRF and NAA arsenic measurements for the Mesopotamian sites

## Trace elements

The other elements used in Chapters 5 and 6 of this thesis are nickel, antimony and silver.

Nickel: For nickel, the correlation between the two sets of measurements is in general good (see Figure III-17). The comparison of values for Susa is not as good as for the other sites but for more than 75% of the objects, the difference between the measurements obtained by the two techniques is under 0.2 wt %. This suggests that nickel is fairly easily detected by most of the methods presented here and a certain degree of confidence can be assumed when discussing nickel contents.

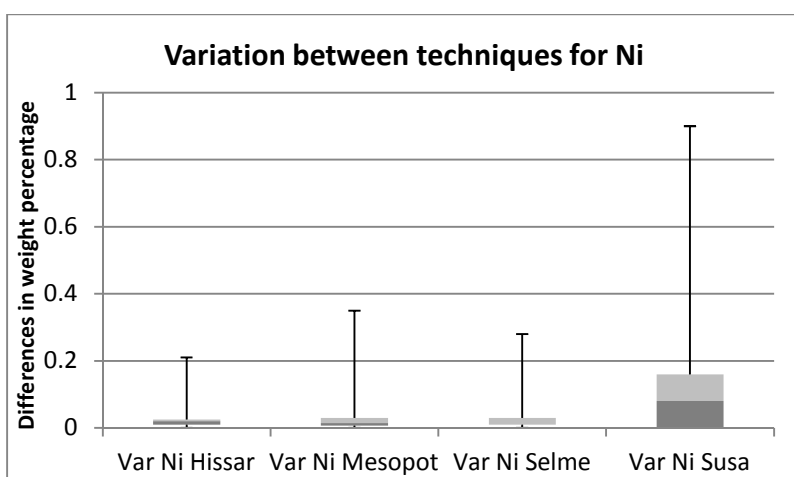


Figure III-17. Spread of the absolute difference in weight percentage of nickel between two measurements of one object using different techniques

Antimony and silver: For Hissar and Selme comparisons of antimony and silver values are difficult as there are less than 10 values available in one case, and in the other case almost all of the values are very small (<0.015%). For Mesopotamia, the correlation between XRF and NAA measurements of both antimony and silver is good (Figure

III-18 and Figure III-19) with a slight bias in the case of silver for which the NAA measurements are slightly lower. The correlation is much worse for Susa and the OES had trouble detecting antimony under 0.1 wt %. The mass spectrometry values for silver are also consistently higher than the OES values. Although this does not allow us to extrapolate on the precision or accuracy of any of the other set of measurements of this database, it is clear that we will need to exercise some caution when dealing with these two elements and especially with the values provided for Susa by Berthoud and Francaix (1980) and Malfoy and Menu (1987).

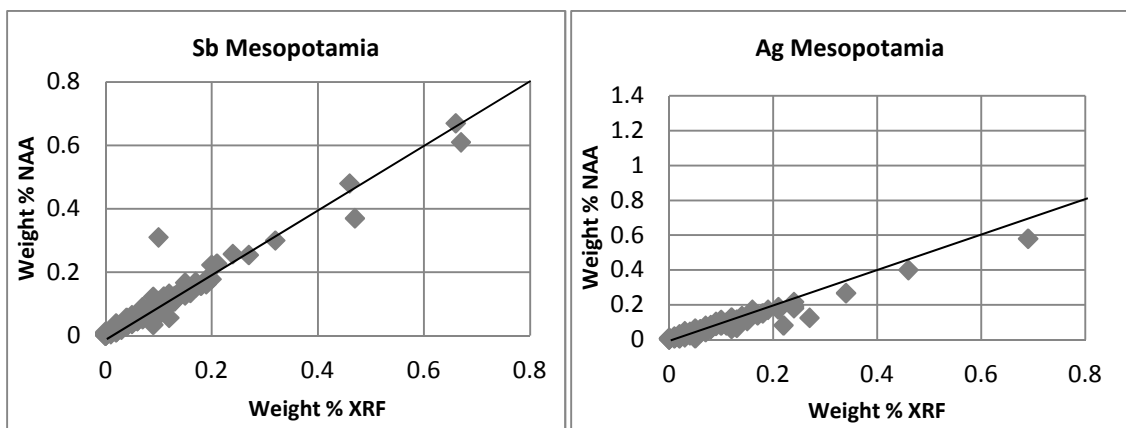


Figure III-18. Comparison of XRF and NAA antimony and silver measurements for the Mesopotamian sites

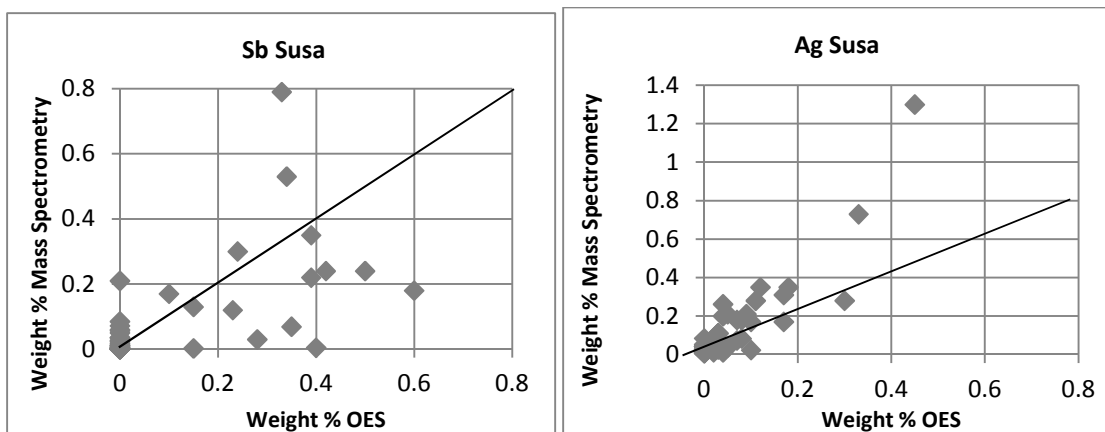


Figure III-19. Comparison of OES and Mass Spectrometry antimony and silver measurements for Susa

This section has demonstrated that even if the data were obtained with different analytical techniques they thankfully show a general agreement, but there are undeniably significant differences in precision, accuracy and detection limits that can not be overlooked. The data from the site of Susa in particular has to be treated carefully as for most elements the values obtained by the two different techniques only show a vague agreement. As already mentioned, we do not focus on the precise chemical compositions of isolated objects as the values are too dependant on the technique used. This section has also highlighted the importance of being very cautious when commenting on periods or regions where not many objects were analysed. What's more, the existence of offsets between two set of measurements of the same group of objects means that is a danger when dealing with averages or medians. For example the arsenic content of the Mesopotamian objects analysed by Hauptmann and Pernicka (2004) averages at 1.49% for the XRF measurements and 1.61% for the NAA measurements: a fairly significant difference of 0.12%. For this reason, we need to be careful when directly comparing averages for different periods or regions and we have to avoid giving too much importance to small variations. It has also dissuaded us from the use of automated methods of analysis of the data, such as clustering or Principal Component Analysis as they are likely to pick up technique related trends rather than archaeological ones. Whenever possible, we prefer the use of graphs showing the distribution of the content of an element, as this is not only less misleading than a single average, but also it tells us more about the metallurgy of the area in question. Because of these technique-related limitations, we often use the simple question: was this element present or absent in these objects? Although this approach is not perfect either it is less affected by slight discrepancies between analytical techniques. For example the tin content of Mesopotamian objects showed a slight bias between XRF and NAA with the NAA

measurements being slightly lower. However, if we define presence of tin as being contents of over 2% tin, then for all of the objects the methods agree on the presence or absence of tin (see Figure III-20).

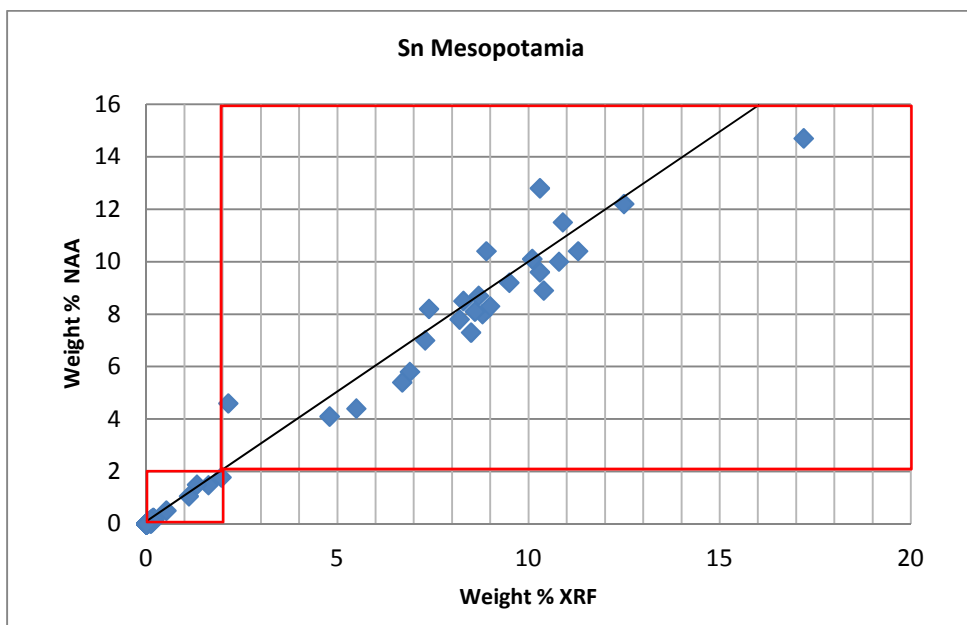


Figure III-20. Comparison of XRF and NAA tin measurements for the Mesopotamian sites. The red boxes represent the zones of the graph for which both techniques agree on the absence or presence of tin (with presence defined here as >0.2% tin)

### 3. Comparison with Chernykh's study of Eurasian metallurgy

Although none of his data have been used in this thesis, it is perhaps useful to mention in this chapter the work of Professor Chernykh, as it is an example of another study attempting to make sense of a large amount of data to understand the history of the development of metal. Moreover, Chernykh's work focused mostly on what was then the territory of the USSR, and as such essentially borders the area of interest of this thesis to the north and east. His work has been presented in many journal publications, most of which are written in Russian, but also in the important volume Ancient Metallurgy in the USSR (Chernykh, 1992), written in English and therefore more accessible to

western scholars. Years of research on the metallic assemblages of this region have enabled him to develop a more global approach than the one used in this thesis, heavily based on the typology of the metal artefacts, but also using the tens of thousands of chemical analyses produced by the Spectrographic Analysis Laboratory of Moscow, of which he is currently director, as well as a number of published analyses from other laboratories, and also looking at the metallography of some objects. From this data he defined a certain number of ‘metallurgical focuses’, often represented by one or more archaeological cultures, and characterised by a similarity in the typology and chemistry of the artefacts and in the methods of production and its organisation. He also defined bigger systems that he called ‘metallurgical provinces’, grouping together related metallurgical focuses.

One of Chernykh colleague in Moscow, L. Avilova essentially applied these concepts to the metal assemblages of the Near-East (Avilova, 2008; 2009). Her area of interest is located slightly to the west of the one described in this thesis: it includes Anatolia and the Levant but leaves out areas surrounding the Persian Gulf, the Indus Valley and Bactria, which we considered of crucial importance for the understanding of trade patterns for both tin and copper. She assembled information on an impressive 60.7 thousands metal objects from 147 sites, but acquired chemical data for only 1672 of these. Her work is also limited to the time period between the late 5th and early 2nd millennium BC and as such doesn’t broach the question of copper metallurgy into the Iron Age.

In many ways Chernyck and Avilova’s approach is similar to the one presented in this thesis, albeit much larger in scale: they defined a certain number of regions and broad time periods (Chalcolithic, Early, Middle and Late Bronze Age), looked at the type and proportion of alloys and metal types present there and revealed pattern of metallurgical

practices and trade, that had previously gone unnoticed or had been poorly described. The scale of their work and their attention to the typology enabled Chernykh in particular to define the different regions based on their metal assemblage. As mentioned earlier, the definition of the regions used in this thesis was driven mostly by the geographical proximity of sites for which data was available, but attention also given to cultural horizons and we believe that in most cases, the metal assemblage of a region is quite homogeneous.

Beyond the geographical framework, there are two fundamental points in which the approach of this thesis differs from Chernykh and Avilova's methodology. First, they rely quite heavily on quantitative analysis, often comparing the absolute number of copper-based finds, or the number of finds per 10000 km (which they call the "saturation indicator") recovered in different regions. Given their thorough investigation of Russian and foreign collection, this approach might well be a good indicator of metal consumption, but, it appears very dangerous to us given the possibility of biases due to differences in the way different regions are excavated, especially in the climate of political unrest that currently exists in the Middle East. For this reason, we prefer to limit ourselves to looking at the representation of specific metal groups or alloys as percentages of the copper-base assemblage, which they also do. Arguably, this approach is still subject to a possible bias due to the type of site excavated in each regions and the subset of objects that have been chosen for chemical analysis, but at least not the number of object recovered.

The second, and perhaps more critical way in which the present work is different from Chernykh's approach, is the way in which the metal itself is considered. Reading Chernykh or Avilova's work, it seems that once it has been smelted and at times alloyed, the metal in circulations remains the same. In particular, Chernykh's discussion

on the difficulties of determining the provenance of copper (Chernykh, 1992, pp.18-23), does not mention the added layer of complexity introduced by changes in chemical composition during the life-time of the metal if it is re-melted, and mixed with different metal. Their more “static” approach is an important first step in the analysis of large datasets and is used at points in the following chapters but it must be recognised that the metal can change and switch from one group to another upon re-melting, as presented notably in Chapter 6 of this thesis.

Nevertheless, there is no doubt that Chernykh’s work remains extremely important in the understanding of Eurasian metallurgy. His Circumpontic Metal Province (CMP) of the Early and Middle Bronze Ages borders our region of interest in the north-west, and of particular interest there is the role of Anatolia and the Trans-Caucasus in supplying metal to the ore poor regions on the other side of the Caucasus that appear to have been great consumers of copper. This is not dissimilar to the role of Iran in supplying metal to their Mesopotamia neighbours on the other side of the Zagros, as described in the following chapters of this thesis. In this sense the Iranian plateau could be described in Chernykh’s terms as a “mining and metallurgical region”, while the Mesopotamian plains are a “metal-working zone”: casting metal in particular types of objects, but without conducting the mining and smelting in the region.

Chernykh’s description of the CMP in 1992 left its southern borders undefined (Chernykh, 1992, p.140). The southern territories that could potentially be included in the province (Anatolia, Mesopotamia, the Levant and Western-Iran) were subsequently studied by Avilova. Her analysis of the composition of copper-based metals remains quite superficial and brings little to the results presented in the following chapters, but her study of other metals and of the typology of the objects complements the present research. A few interesting points are for example the slightly earlier production of

metals in Iran than in the other three regions, the presence of many gold beads in the Early Bronze Age cemeteries of Se Girdan in Iranian Azerbaijan and Tepe Gaura in Assyria, perhaps indicating strong links between the two regions as is also visible in the copper-based data (see following chapters), and the importance of precious metals (gold and silver) in Mesopotamia. Her research also led her to define a culture zone with a single tradition of metal production in the Early Bronze Age (3700-2700 BC) in Northern Mesopotamia, eastern Anatolia, West and Central Iran, the Northern Caucasus and to some extent the Levant, which then no longer existed in the Middle Bronze Age. While there were still contacts between the regions, each displayed more local features, with the Caucasus in particular being more isolated (Avilova, 2008, pp.88-89).

Going back to Chernykh's analysis, he explains that, when the CMP fell apart, it was replaced in the Late Bronze Age by the smaller Caucasian Province bordered in the north by the steppe foothills of the Caucasus and encompassing parts of Anatolia and north-western Iran, its southern borders having also been left undefined (Chernykh, 1992, p.275). This region is remarkably isolated from the steppes to the north when compared to the previous time period. The objects produced there are highly decorated and very varied, especially in the east. Of interest to us is the variety of metal types present there and in particular the presence of antimony alloys. Chernykh also mentions the mining of antimony in the Caucasus, in particular at Zopkhito (Chernykh, 1992, p.276). This is particularly interesting when compared to the high antimony component of alloys in the north-west of our area of interest (see Chapter 6).

Finally, amongst other provinces of the Late Bronze Age, Chernykh defined the Irano-Afghan Metallurgical Province, but admits to only know it from its northern periphery: what has been defined as Bactria for this thesis. Of interest here is the "major fluctuation" in alloying elements and the relatively poor quality of the tin bronzes

(Chernykh, 1992, p.275) and from a typological point of view the presence of many objects of types similar to those of the Eurasian Province which according to Chernykh was of major importance in the Late Bronze Age.

#### 4. Conclusion

This chapter has shown the difficulties associated with binding together such a varied set of data, and especially the integration of data obtained with different techniques. However we have shown that the absolute differences between measurements are in many cases quite small. It seems that the technique-related problems can then be overcome by questioning the database with a cautious approach, giving less importance to the exact composition of individual objects and focusing on comparing patterns observed for a large group of objects. Trends obtained from the database, and in particular averages, clearly have to be analysed sensibly as small variations are likely to come from the technique related offsets rather than actual differences in the metal.

More difficult to overcome on the other hand is fact that the data is very fragmented. Very few regions present a reasonable amount of data for both of the time periods defined in this project, making diachronic considerations difficult. Only Luristan and Oman in fact provide a fairly good amount of analyses across the entire chronological timeframe considered here. For our first time-period however synchronic considerations across nearly the entire geographical span are possible, as most regions (with the notable exception of Amlash and Azerbaijan) present a good number of data. For our second time-period, unfortunately, many regions in the east are poorly represented and we have to limit our reflection to western Iran and the Persian Gulf.

This “patchiness” of the database is undoubtedly a limitation for our study. Yet, grouping all of the published data together enables us to pinpoint exactly the regions and time periods that would benefit from an analytical program. In the meantime, we believe that the data collected so far has more to offer than what has already been proposed by all of the studies summarised at the beginning of this chapter. By grouping the data in one binding chronological and geographical framework, we hope to get a transversal view of the development of metallurgy in this area of the Near-East. And by analysing it with no a priori assumptions on the way metal-working evolved, we hope to offer a critical vision on the current hypotheses for metal sources, metal use and trade.

The following chapter focuses on the practise of alloying copper with tin to produce bronze and attempts to provide a new take on the much debated question of the provenance of this tin. Following this, Chapters 5 and 6 look in more details at the other elements, in particular arsenic, nickel, antimony and silver and what they can tell us about metal movement.



## Chapter IV. The tin story

### 1. Introduction

Tin is of course of great importance in the history of metallurgy as it was one of the first metals to be used extensively as a deliberate alloying element. Added to copper it allows the production of bronze, an alloy stronger and harder than both of the unalloyed metals. But tin doesn't only modify the mechanical properties of copper; it also changes its characteristic reddish orange colour to give it a golden aspect.

As mentioned in the introductory chapters of this thesis, tin has been the subject of much discussion, with questions touching the entire spectrum of archaeometallurgical considerations. In Chapter 2, we mentioned the problem of the provenance and trade of tin, a question that has been thoroughly discussed but still mostly eludes us (Franklin et al., 1978). We have also mentioned research dedicated to the production process of bronze and recent approaches in their identification. In Chapter 3 we have seen that tin has almost systematically been sought for in chemical analyses from the very first investigations in the 1850s (Layard, 1853, p.870). At that time the goal was often to answer the simple question: "What is this object made of? Is it copper or is it bronze?" But behind this straightforward question hides a whole range of more complex deliberations involving the adoption of this alloying technique: when and where did this alloy first appear, who was it subsequently used by and for what purpose. Also fascinating are the questions linked to changes in technological practices. Why did people start to use tin bronze over unalloyed copper or arsenical copper? Was it for its

mechanical properties or perhaps its colour or something else that we still do not fully comprehend? We also wonder what the status of this alloy was and whether it was used at all levels of the society. And finally, why was it replaced by iron about 2000 years after its advent?

a. Tin in the framework of this thesis

Tin was sought for in 5015 of the objects entered in our database, 2330 of which came from Iran and the rest coming from neighbouring regions. In this chapter we attempt to provide a new perspective on these data. As we have seen in the previous chapter, almost all of these analyses have been looked at and commented on before, but only separately. Looking at them together, in the framework defined in Chapter 3, allows us to bring to light differences and patterns that would otherwise have been lost, fragmented across over 50 publications.

We look at the data from the perspective of the Bronze Age / Iron Age transition to try and understand what (if anything) changed with or because of the advent of iron, or perhaps even caused it. This touches many of the questions mentioned above, one of them being the provenance of tin. Did the tin supply change in the middle to late 2nd millennium BC? We also investigate questions such as the practice of recycling and mixing and the status of tin bronze and how it potentially changed in the Iron Age.

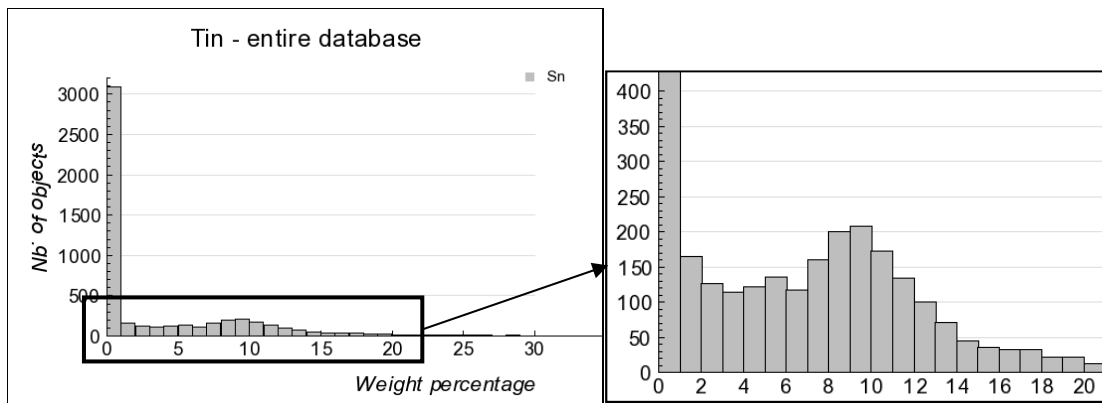
To this end we first look at the chronological evolution of general trends such as the ubiquity of tin bronze, or the quantity of tin in the bronzes. This is followed by a more detailed analysis of the two time periods defined in Chapter 3, independently of chronology this time, as it allows us to work with more data for each region.

## 2. Diachronic analysis

### a. Evolution of the ubiquity of tin bronzes

As can be seen on Figure IV-1 the data contained in our database shows a wide variety of tin contents, with the majority of the objects containing between 0 and 20% of tin. Over 3000 objects however have less than 1% Sn. It should be noted that objects for which the tin content appeared as “trace”, “below detection limit” or “not detected” in the original publications are also assigned to this 0-1% range. For numerical calculations (averages and medians for example), they are considered as having 0% tin.

What distinguishes a bronze with a deliberate addition of tin from a copper object where tin, if present, appears only in trace amounts that were present in the copper ore, is slightly arbitrary (see discussion in Cleuziou & Berthoud (1982, p.15)). Traditionally scholars have used 1 or 2% as a cut-off mark between copper and bronze. To choose a limit, we can consider how much tin is likely to appear as a trace given the composition of copper ores, or the amount of tin necessary to see a significant modification of the hardness or colour of the objects, but this picture is blurred by re-melting and recycling. For example, the mixing of bits of scrap bronze and scrap copper would result in a copper with a lower tin content. Do these objects then qualify as “deliberate” tin bronzes? This question could be discussed at length but for now we simply choose the arbitrary limit of 2% to distinguish what we call tin-less copper from tin bronzes. Tin-less copper is not necessarily pure copper as it can contain elevated amounts of other alloying elements, arsenic for example, as well as trace elements.



**Figure IV-1. Distribution of the tin content for all of the objects of the database**

Figure IV-2 provides an overview of the evolution of alloying with tin for the whole of Iran. It clearly shows that although this practise occurred very occasionally from the 5th to the early 3rd millennium BC, it only really started gaining in importance from the mid 3rd millennium onwards. In the following millennia we can see a very gradual increase of the proportion of tin bronze until the Iron Age when it stabilised with around 90% of the assemblage being tin bronze.

This graph can of course be slightly misleading as it groups under the modern appellation of ‘Iran’ regions where copper metallurgy is likely to have evolved in different ways. In particular, what could be interpreted as a decrease in the use of tin bronze in the mid 1st millennium BC is in fact a reflection of the low amount of tin bronze in the South-East, a region that always had less than 30% of tin bronzes and the small number of artefacts for all of the other regions that still have 100% tin bronzes in their assemblage. If we omit this mid 1st millennium dip, the curve shows an s-shape characteristic of the diffusion of innovations where successive “consumer groups” adopt the use of a new technology, here tin bronze (Rogers, 2003). This curve tells a very believable story for the adoption of tin bronze at the scale of Iran, with an experimental stage from the 5th to the early 3rd millennium BC, then its introduction in the beginning of the 3rd millennium, its growth throughout the end of the 3rd millennium and most of

the 2nd millennium, and finally, its maturity from the late 2nd to the mid 1st millennium BC. Undoubtedly, if this graph showed the proportion of tin bronze in the entire metal assemblage (including ferrous metals) rather than copper-based metals only, it would show a decline from the early 1st millennium BC when iron began to replace bronze.

From this very general plot, it certainly seems that the early appearance of iron in the middle to late 2nd millennium BC in Iran did not deter the development of tin bronze. On the contrary it would appear that the period conventionally called Iron Age in fact corresponds to what we could call a “Mature Bronze Age”. However, as we have already mentioned, the database does not give the same weight to every region in every time period meaning that this curve presents a slightly distorted image of the development of tin bronze. That is why we now take a closer look at regional developments.



Figure IV-2. Chronological evolution of the ubiquity of tin bronze in the copper-based metal assemblage

Figure IV-3 shows the break-down of the evolution of the ubiquity of tin bronze for each one of the Iranian regions. To interpret this plot correctly we need to look at it in parallel to the graph showing the number of objects available in the database for each period and each region (Figure IV-4). Indeed, if the peaks at 83% of tin bronzes for Luristan, and 25% for Gorgan in the 4th and 3rd millennia BC do indicate that tin was present in concentrations higher than 2% in this period, the percentages should not be considered representative of the production of tin bronze since the number of objects sampled in these cases is very low (respectively 12 and 4). Moreover, the amount of tin in some of these objects is just barely above the arbitrary 2% limit (2.06% of tin for example in the Gorgan object).

Nevertheless what we can see from these ubiquity graphs is that the frequency of tin bronzes started to increase for most regions in the beginning of the 3rd millennium BC. A few regions however seem to have had a delayed adoption of the alloy. This is the case for example of eastern Iran: in Gorgan tin bronze is not attested at all, while in the south-east there was an increase in the quantity of tin bronze from 0% to 20% between the late 3rd and the early 2nd millennium but since no data is available for the following periods it is difficult to state whether this increase was in fact truly representative of an adoption of tin bronze.

In Luristan, a region for which we have data across the entire time-frame, the increase in the proportion of tin bronzes in the assemblage is relatively smooth, going from 0% in the early 3rd millennium up to 100% over two millennia with a slower growth and even a slight recess in the first half of the 2nd millennium BC. A high percentage is then maintained in the beginning of the 1st millennium. Although evidence is patchier elsewhere, the data we do have points towards a steady increase in the amount of tin bronze from the early 3rd to the mid 1st millennium BC in north-western Iran and Fars.

Khuzistan could also have had a similar uptake of tin bronze albeit with a slightly slower start, but our data stops in the mid 2nd millennium BC, so, from this graph, it is impossible to determine whether or not the increase in the proportion of tin bronze continued in the Iron Age. However, the presence at Susa of monumental bronze work dating from the second half of the 2nd millennium BC would suggest that tin-bronze was fairly common in Khuzistan at that time.

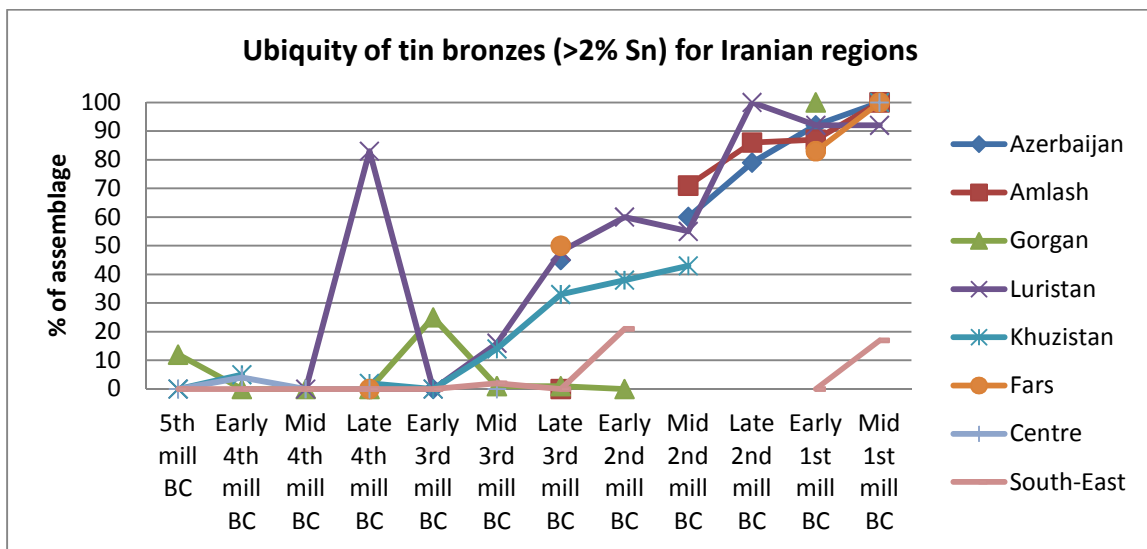


Figure IV-3. Chronological evolution of the ubiquity of tin bronze in the copper-based metal assemblage for Iranian regions

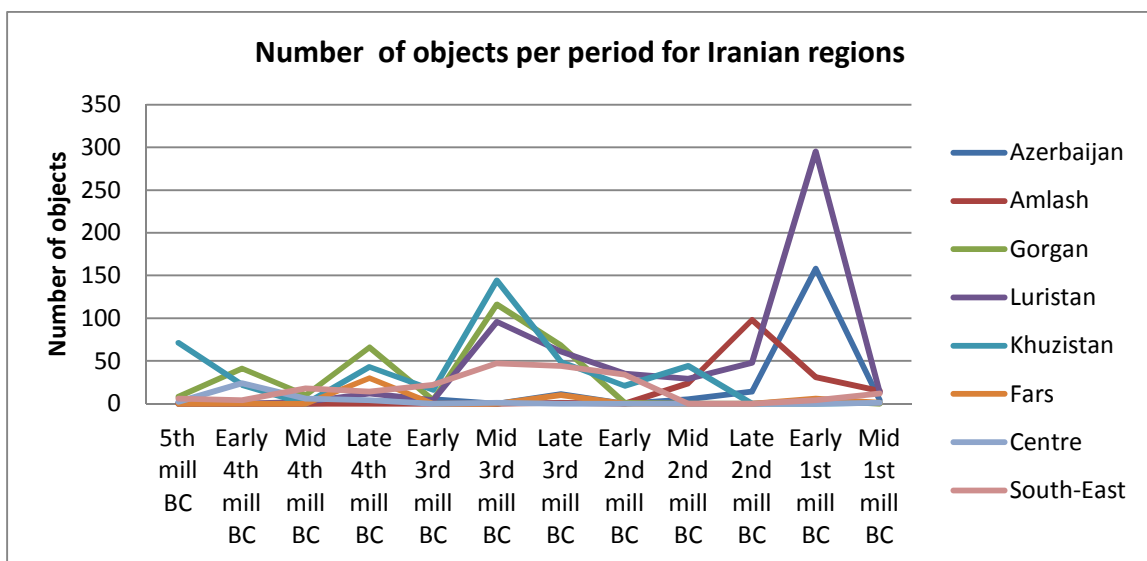


Figure IV-4. Number of objects in the database for each period for Iranian regions

The situation appears to be different in the countries surrounding Iran. As we can see in Figure IV-5 most regions show an increase from around 0% in the early 3rd millennium BC to around 50% quite rapidly but the ubiquity of bronze then remains between 30 and 60% throughout the rest of the 3rd and the 2nd millennium, with an increase again in the early 1st millennium. It should be noted that the 50% ubiquity and 100% ubiquity observed for Assyria in the late 4th and early 1st millennia are derived from only four and two measures respectively and should therefore not be considered of much significance<sup>3</sup>.

This graph shows a decrease in the percentage of tin bronzes first in Oman, from the late 3rd to the mid 2nd millennium BC and about 300 years later in Mesopotamia and Assyria. This decrease is potentially related to the slower growth observed in Luristan in the first half of the 2nd millennium BC, and could be explained by a difficulty in accessing tin but this hypothesis cannot be verified at this stage.

For none of these neighbouring regions aside from Assyria do we see percentages as high as for some of the Iranian regions where over 80% of the assemblage is made of tin bronze in the Iron Age. Potential explanations for this major difference are discussed later.

The uptake of alloying with tin for the Indus Valley, Bactria or Bahrain can unfortunately not be chronologically described as we only have information for very restricted periods of time.

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<sup>3</sup> Since the submission of this thesis, a volume containing chemical data for about 120 objects from Nimrud, Assyria and dated to the Iron Age has been published (Ponting, 2013). 93% of these are tin-bronzes. This confirms the high ubiquity of tin-bronze in Assyria in the beginning of the 1st millennium BC.

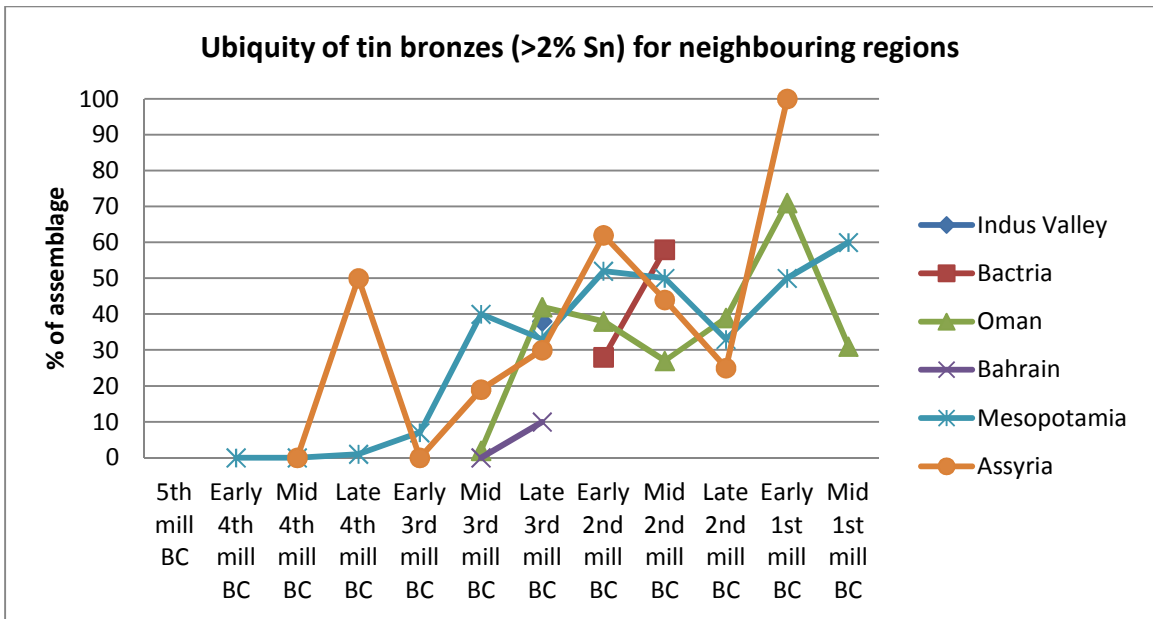


Figure IV-5. Chronological evolution of the ubiquity of tin bronze in the copper-based metal assemblage for neighbouring regions

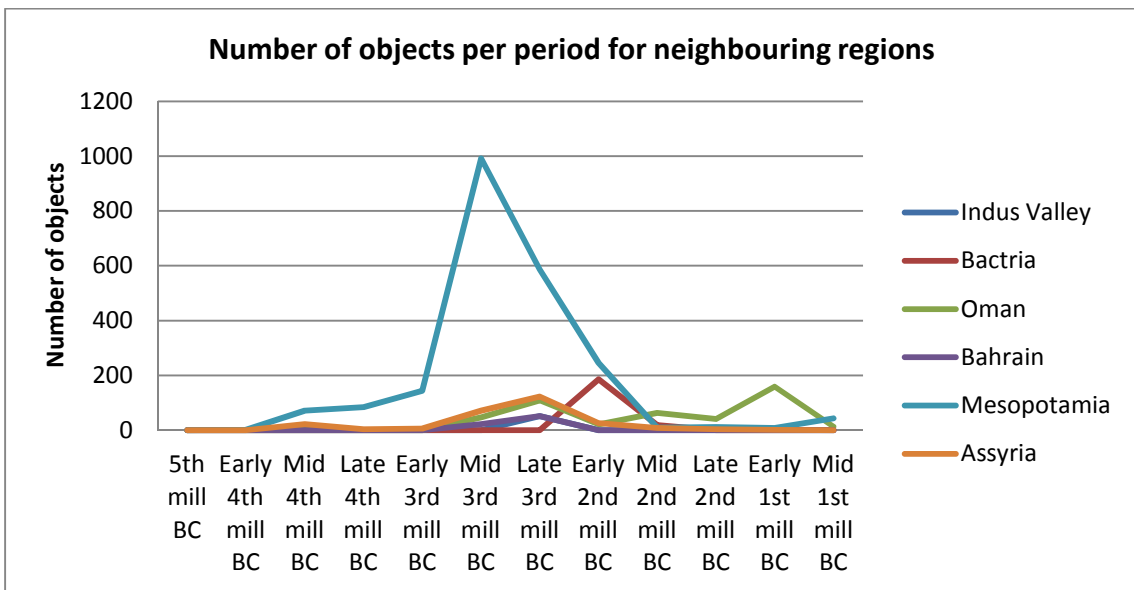


Figure IV-6. Number of objects in the database for each period for neighbouring regions

These graphs have highlighted a certain number of aspects of the adoption of tin bronze in and around Iran, some of which have already been observed before while some other are to my knowledge new observations made possible by the direct comparison of data

from many different studies. The first observation, certainly not new but made clear on these graphs, is that the true development of the technique of alloying with tin occurred from the early 3rd millennium in most regions, potentially a few centuries earlier in Mesopotamia and a few centuries later in Oman. Before then, a few objects with more than 2% tin occur occasionally, often still with a relatively low percentage of tin, attesting of early experimentations with this technique. The regions of Gorgan and the South-East are notable exceptions to this pattern. There, until at least the end of the 3rd millennium, tin-less copper is still almost always preferred to tin bronze.

Secondly, these graphs have showed a respite in the adoption of alloying with tin from the late 3rd millennium BC to the mid to late 2nd millennium BC. In the following sections we attempt to define the metallurgy in this period more precisely to explain this recession and its potential impact on the arrival of iron. Unfortunately this task is made more difficult by the fact that, for most regions, this period also corresponds to the time when the least data is available. This might not be a coincidence: as we have already seen in Chapters 1 and 3, the mid 2nd millennium BC was marked in many regions of the Near East by a decrease in settlement densities and a shift to a less sedentary lifestyle. This means that fewer sites from this period have been excavated and therefore less copper-based objects have been recovered and made available for analysis. But this change in the organisation of society is also likely to have destabilised the progress of metallurgy, possibly changing the main actors in its development.

Finally, we have observed a difference between the proportion of tin bronze reached in north-western and western Iran (over 80% in the Iron Age) compared to the maximum reached outside Iran (70% if we omit the value of 100% obtained for Assyria with only two data points). This discrepancy could be explained by the presence of one or multiple tin sources easily accessible to western Iran but not as easily reached by its

neighbours in the Iron Age. The mine of Deh Hosein at the eastern border of Luristan (Nezafati, 2006) or possibly other similar mines in that area (Nezafati et al., 2009, p.228) would be likely candidates for the supply of tin to north-western Iran. If this is the case we can suppose that the tin was exported to the west in small quantities but that the supply wasn't sufficient for tin bronze to reach the same predominance in Mesopotamia as it did in Iran. Until more data is gathered for the Iron Age in Elam, it will remain difficult to assess if the tin that Nezafati and colleagues believe was mined in western Iran was also exported to the south.

#### b. Evolution of the tin content

The previous section has enabled us to make observations on the development of metallurgy from the very simple distinction between presence and absence of tin. However we have not commented on the actual amount of tin in tin bronzes and whether that amount changed significantly through time. This section explores the chronological evolution of the tin content for the three regions where enough data is available across most of the time-frame considered here: Luristan, Oman and Mesopotamia.

Figure IV-9, Figure IV-10 and Figure IV-11 highlight a certain number of differences between these three regions: first the range of tin content differs greatly. In Luristan none of the values (except for a few outliers not represented on this graph) exceed 17% Sn, while in Mesopotamia the range reaches up to 30% Sn in the mid 3rd millennium BC. The situation in Oman is quite surprising: many objects have very high tin contents. Values range across a very wide spectrum, from 2% up to a tin ring with over 90% of

tin. The range is particularly wide from the late 3rd millennium to the mid 2nd (see also Figure IV-7).

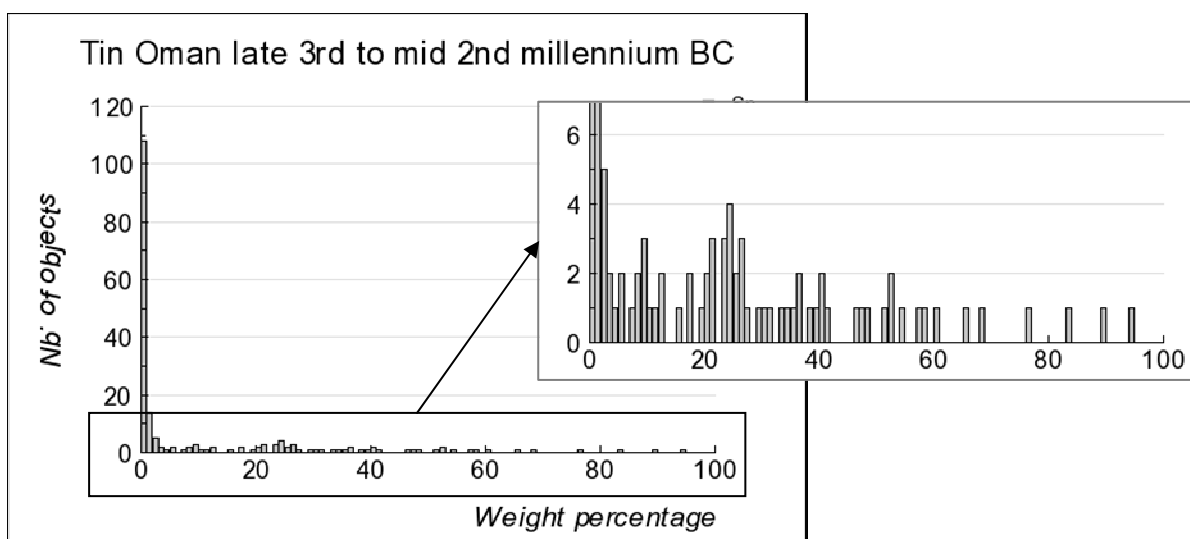


Figure IV-7. Distribution of the tin content for objects from Oman from the late 3rd to mid 2nd millennium BC

The objects with over 20% tin are rings, bracelets, pins, daggers and vessel fragments coming from occupational contexts and tombs at the sites of Tell Abraq and Unar (Weeks, 1997; 2003). Weeks attributed the very high levels measured in Tell Abraq objects to the preferential corrosion of copper in the samples due to the acidic soil conditions of this site (Weeks, 1997, p.76). However, two uncorroded objects showed tin contents of 12% and 20.8%, higher than most of the Luristani objects and at the high end of levels observed in Mesopotamia. Unfortunately, the results obtained for the corroded items have to be treated qualitatively, simply indicating that tin was used as an alloying element in Bronze Age Oman, perhaps, but not certainly, in relatively high quantities. Figure IV-11 shows a much narrower range in the Iron Age. This is confirmed in Figure IV-8 where we can see that in late 2nd to early 1st millennium BC even if there are still a few very highly alloyed objects, most of the objects fall into a

range of 4 to 15% tin with an extremely well defined peak between 9 and 12% tin. Most of the objects from this time period come from the hoard of Selme (Prange & Hauptmann, 2001). This suggests that the metalworkers who made this hoard were able to produce an alloy with a well defined composition. There appears to have been very little mixing of copper and bronze, and probably not much reuse of scrap metal. Indeed, if this had been the case, we would expect to see a wider range of compositions reflecting the use of different means to obtain bronze. Instead, it looks like the metal workers there used a single “recipe” that would almost always result in the production of a bronze with between 9 and 12% tin. It should be noted that this percentage, which is almost certainly indicative of a primary production of tin-bronze, is lower than most of the percentages mentioned in recipes recovered in Mesopotamia or Ebla mentioned in Chapter 2 (Muhly, 1973; Eaton & McKerrell, 1976, p.179; Archi, 1993). It is unclear at this stage whether this represents a difference in the preferences of the Omani metalworkers responsible for this hoard and the Sumerian metal-workers or if it represents a loss of tin during the alloying process resulting in percentages in the finished object differing from the initial proportions of metal added to the mix. In any case, this area, at the specific time of the production of the hoard, must have had a good access to both tin and copper to be able produce bronze from pure raw materials. For the reasons discussed above, this data can unfortunately not be directly compared to the data of the earlier periods in Oman. However, further in this chapter, we endeavour to compare it to the profile of other regions in the same period and to discuss a possible provenance for that tin. For now, all that can be said is that Oman started to use a significant amount of tin bronze objects in the late 3rd millennium BC, a few hundred years later than in Mesopotamia and Iran, but, from that period onwards, it seems to have had a good access to tin.

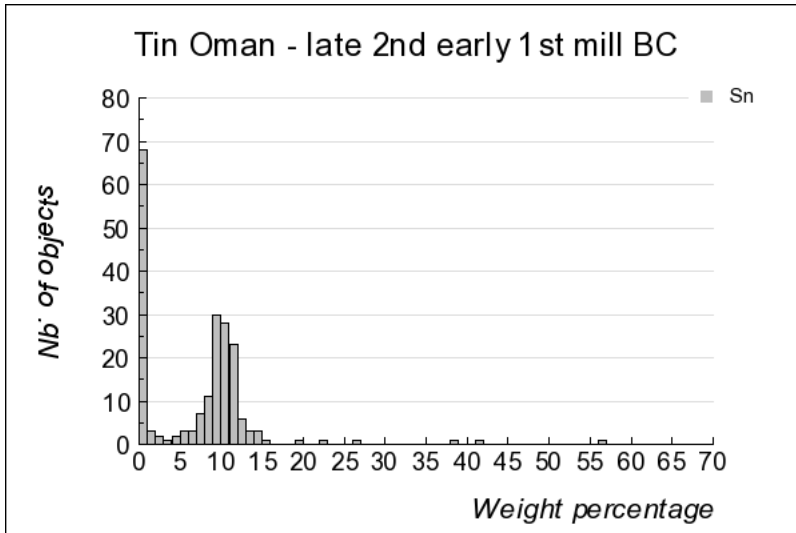


Figure IV-8. Distribution of the tin content for objects from Oman from the late 2nd to the early 1st millennium BC

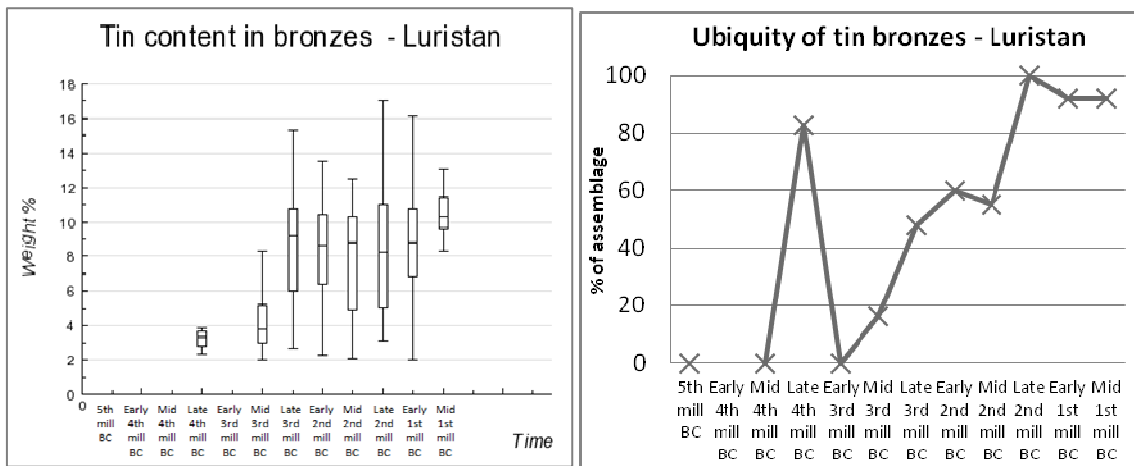


Figure IV-9. Evolution of the tin content in tin bronzes (>2% Sn) and of the ubiquity of tin bronzes in Luristan

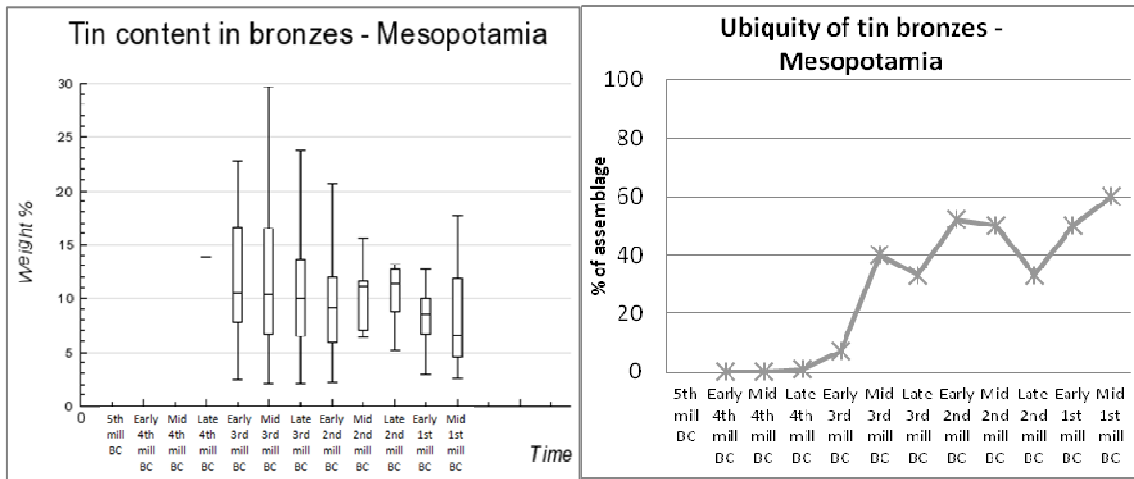


Figure IV-10. Evolution of the tin content in tin bronzes (>2% Sn) and of the ubiquity of tin bronzes in Mesopotamia

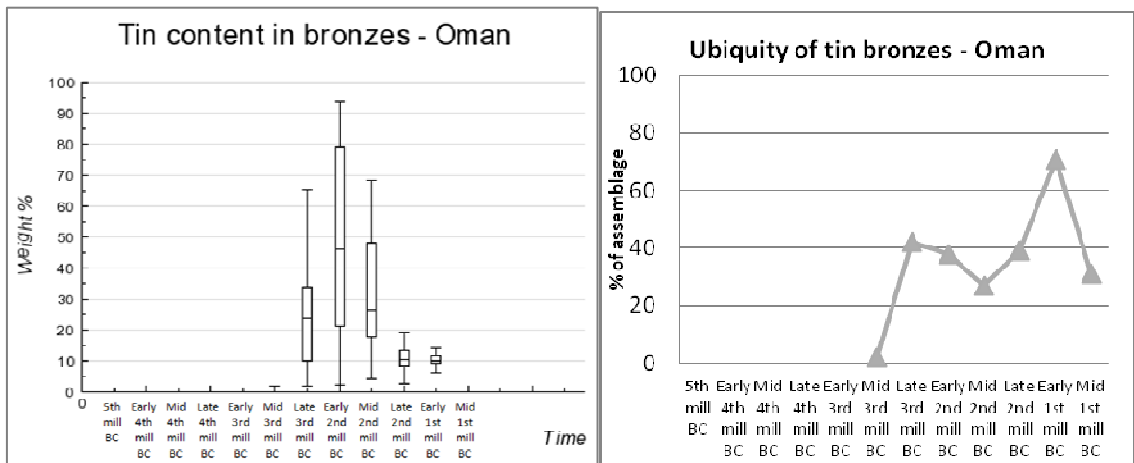


Figure IV-11. Evolution of the tin content in tin bronzes (>2% Sn) and of the ubiquity of tin bronzes in Oman

Figure IV-9 shows that in Luristan, as we have already mentioned, the peak in the ubiquity of tin bronzes in the late 4th millennium BC corresponds to objects that still have quite a low tin content (between 2 and 4%). In the mid 3rd millennium BC, at the beginning of the true increase in the proportion of tin bronze, the tin contents are also relatively low (between 2 and 8% with a median at 4%), suggesting that the technique of alloying was not yet entirely settled or that the early bronze objects are the result of the recycling or mixing of tin bronze perhaps coming from elsewhere. However, by the late 3rd millennium the median level of tin reached 9% with 50% of the objects

containing between 6 and 11%. While the proportion of tin bronzes kept increasing in the following millennia, this level of tin remained relatively unchanged up to the beginning of the 1st millennium BC. In other words, while more and more objects were made with this alloy, the technique of alloying in itself, or at least the proportions of copper to tin used in these objects became standard from very early into the advent of tin bronze.

This trend is even more striking in Mesopotamia where a “standard” level of tin is reached as early as the early 3rd millennium BC, when only 7% of the objects were alloyed. The fact that a standard level of tin is maintained through time is perhaps the result of a control on the proportion of copper and tin by the central authorities in Mesopotamia. Recipes indicating a proportion of tin to copper for the production of tin-bronze are indeed known from the mid 3rd millennium BC and change very little thereafter (see Chapter 2). Again, the median levels seen here are lower than the percentages mentioned in the recipes, indicating losses of tin between the alloying event and the deposition of the objects. The level of tin then slightly decreases throughout the years going from a median of just above 10% in the beginning of the 3rd millennium to about 7% in the mid 1st. The observation of the tin levels in the early 3rd millennium rests only on 10 alloyed objects (see Figure IV-12), but if it is true that as soon as Mesopotamia had bronze, this bronze was “true tin bronze”, alloyed to a level that would then remain unchanged, with no sign of experimentation on the amount of tin necessary to achieve the desired result, then one has to wonder if Mesopotamia imported this bronze from a place where the industry had already matured. Where this place would be however remains problematic as of all the regions considered in this thesis Mesopotamia is the one that presents the earliest adoption of tin bronze. The answer might be further to the north or west but this is beyond the scope of this thesis.

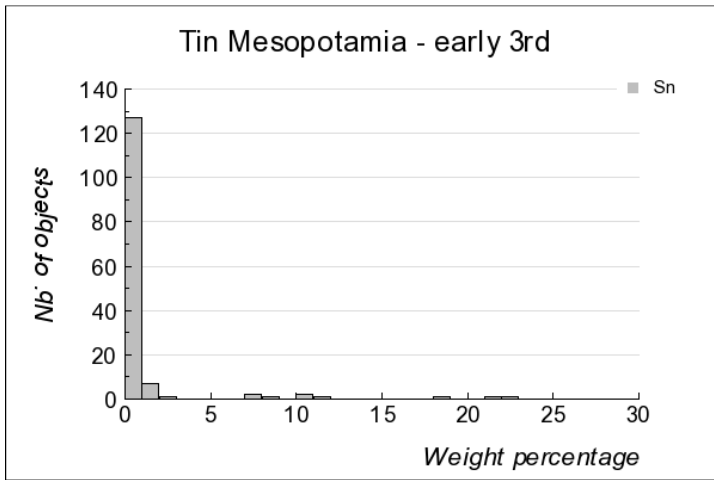


Figure IV-12. Distribution of the tin content for Mesopotamian objects of the early 3rd millennium BC

On the contrary, one could argue that the low tin bronzes present in Luristan in the mid 3rd millennium (see Figure IV-13) were obtained by the recycling or mixing of high tin bronzes obtained from elsewhere, perhaps Mesopotamia. However, as the number of alloyed objects is extremely small in both cases, the question of the level of tin in early tin bronzes and the meaning of such levels will have to remain open until more bronzes from these periods are analysed.

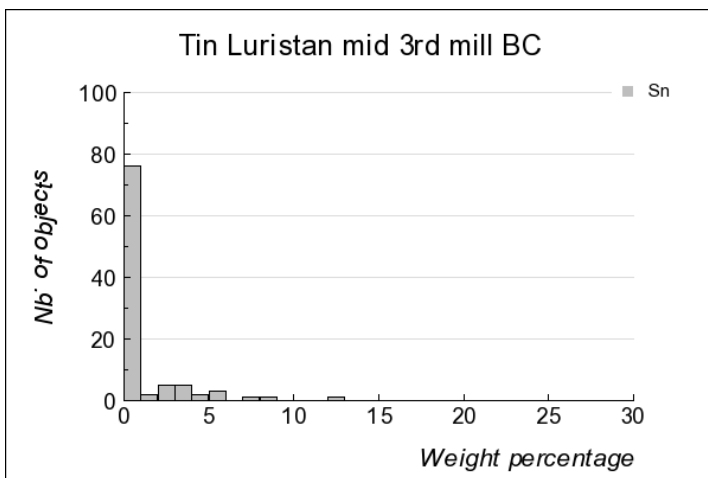
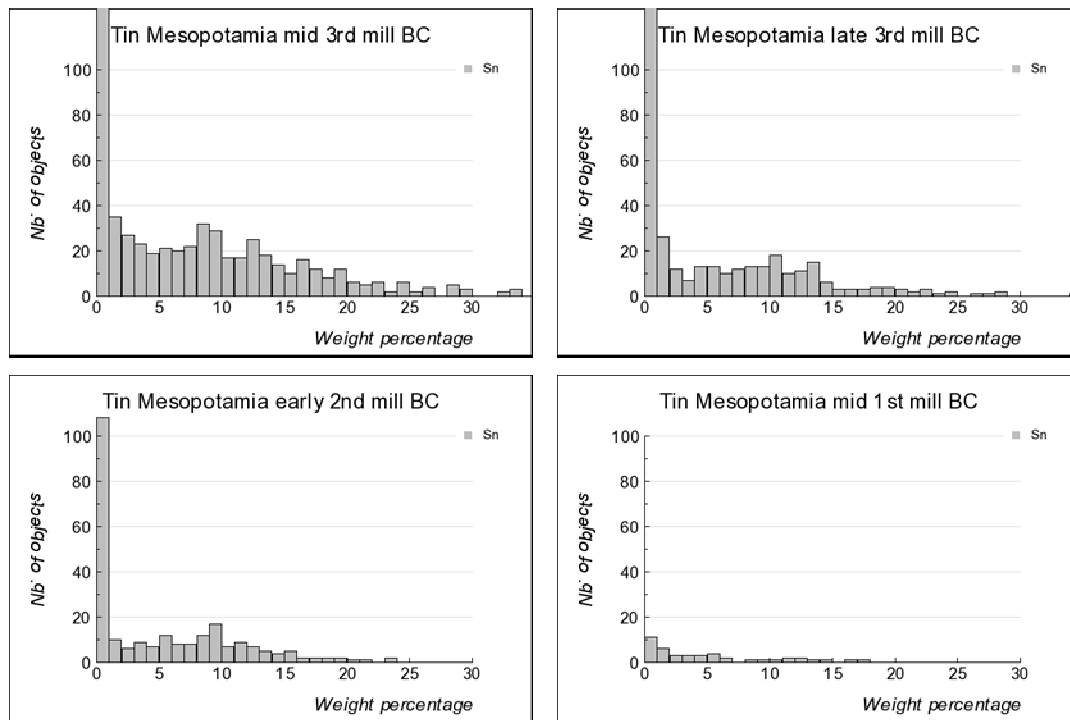


Figure IV-13. Distribution of the tin content for Luristan objects of the mid 3rd millennium BC

In an attempt to interpret the slight gradual decrease in the amount of tin in Mesopotamia observed on Figure IV-10, we have plotted the distribution of the tin content in this region from the mid 3rd millennium BC to the mid 1st, omitting the mid 2nd, late 2nd and early 1st for which respectively only 10, 12 and 8 objects are available in our database (Figure IV-14). On these graphs we can see as expected the preponderance of tin-free objects for all but the mid 1st millennium BC. More striking however is the wide range of tin contents for alloyed objects. The apparent narrowing of this range over time is, in our opinion, mostly due to the diminution of the number of objects analysed across these four periods (respectively 992, 586, 246 and 43 objects), making it less likely in the later cases that “rare” objects with very high tin contents would have been analysed. The distribution plot in all four cases is remarkably flat: the majority of the alloyed objects contain between around 5 and 15% of tin but many objects fall outside of this range, and only a faint peak can be observed around 10% of tin. As we will see, this distribution contrasts greatly with the tin “profiles” observed in Iran or Oman where very clear peaks can be observed. Here we can only say that the slight decrease in the amount of tin suggested by Figure IV-10 can primarily be explained by the shape of the distribution (highly alloyed objects being less common than objects with less tin) added to the fact that the number of objects analysed in the later periods is much smaller than in the earlier periods causing both the apparent range and the apparent median content to artificially decrease over time. The distribution graphs however show very few differences certainly between the mid 3rd millennium and the early 2nd, suggesting that the metalworking didn’t change much during that period in Mesopotamia. Unfortunately the data for the Iron Age is too limited for us to look at potential changes that could have occurred in the late 2nd millennium BC.



**Figure IV-14. Distribution of the tin content for Mesopotamian objects from the mid 3rd millennium BC to the mid 1st millennium BC**

Figure IV-15 shows the same graphs this time for Luristan between the mid 3rd millennium BC and the early 1st. They confirm the gradual shift in the proportion of tin-free copper to bronze that we have already observed in the ubiquity plots. Unlike in Mesopotamia, the profile of the distribution of the alloyed objects changes through time. As we have already mentioned, at the very beginning of the adoption of tin bronze in Luristan, the majority of the objects have low tin levels (2 to 6%). From the late 3rd millennium to the late second the number of alloyed objects analysed is fairly low, but we can just about distinguish two “bumps” corresponding to two groups of tin bronzes with a different tin content. This double peak is mostly visible in the early 1st millennium for which many more objects were analysed (295 in total). The first group is represented by objects containing between about 2 and 6% tin, while the second group is constituted of objects with over 6% tin but mostly between 6 and 15%.

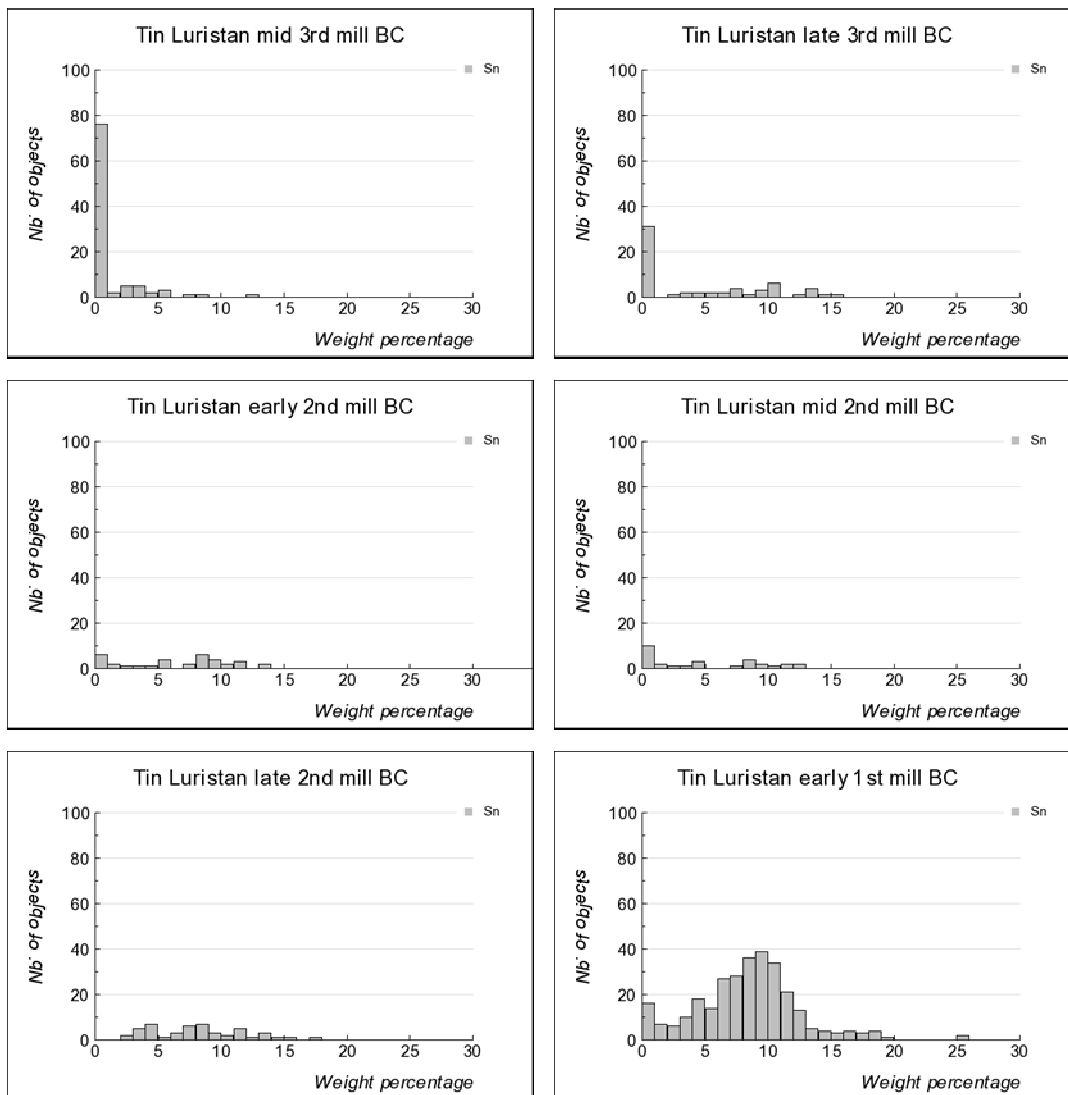


Figure IV-15. Distribution of the tin content for Luristan objects from the mid 3rd millennium BC to the early 1st millennium BC

A similar bi-modal pattern can be observed in other regions, for example it is quite striking in Iranian Azerbaijan in the early 1st millennium BC and it is also visible, even if less data is available, in Amlash in the late 2nd millennium BC (Figure IV-16). From looking at Figure IV-15 it would appear that, in Luristan, in parallel to the change in the tin-less copper to tin bronze ratio, there is a shift in the preponderance of the mid-range tin group and the high tin group, the latter becoming more prevalent in the later periods. We believe that understanding the nature of these groups and the shifts in prevalence of one to the other is a key to our understanding of the metallurgy of the Near-East. That is

why section 4.3 is dedicated to their definition and a chronological study of their preponderance.

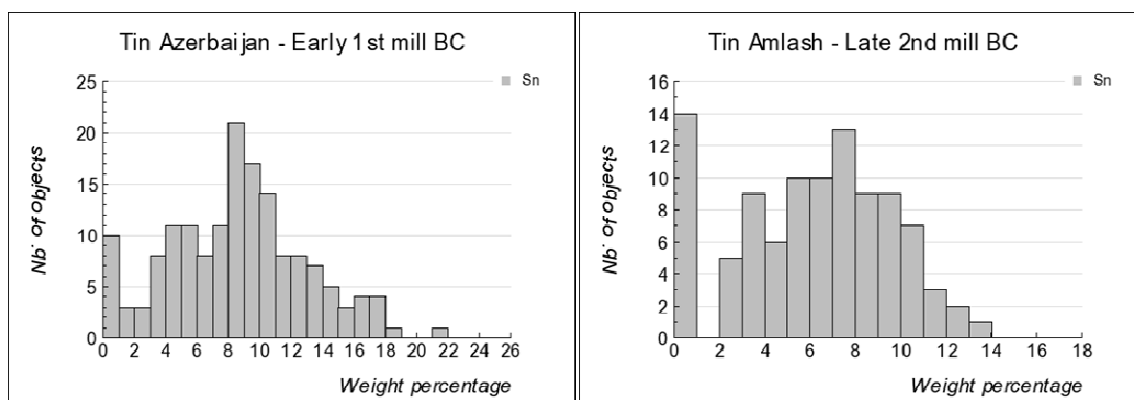


Figure IV-16. Distribution of the tin content for objects from Azerbaijan from the early 1st millennium BC.

### c. Artefact types

In the introduction of his recent review of the understanding of tin sources and trade Pigott stated:

*“This alloy of copper and tin was used initially mainly for decorative items. Over the next two millennia, tin-bronze became the alloy of choice for ornaments, vessels, tools and weapons across the Asian continent –from Beirut to Beijing to Bangkok. This was not to change until iron began to become widely available ca. 1000 BC and in the centuries immediately following, at which point tin-bronze reverted back to being a mainly decorative metal.”* (Pigott, 2011, p.273)

In this paragraph we endeavour to verify how much evidence is actually available for us to compare the tin content of different object types and how these contents changed over time. Figure IV-17 shows for Iran the evolution of the ratio of tin-bronze to tin-less copper in four different types of objects: weapons, tools, ornaments and vessels. We can see that aside from a few tools in the early 4th millennium and a few ornaments in the

5th and late 4th millennium that have low amounts of tin, the ornaments and tools truly start to be made of tin bronze at the same time: in the beginning of the 3rd millennium BC. Tin bronze is then adopted at a very similar rate for tools, weapons and ornaments throughout the 3rd and 2nd millennium BC. Unfortunately not enough Iranian vessels or weapons have been analysed from periods before the mid 3rd millennium to confirm whether or not metal-workers also started producing them in tin bronze at the same time, but from the available data it certainly seems to be a possibility for weapons at least. Figure IV-18 shows that in Mesopotamia on the other hand, vessels were made of tin bronze first, followed by ornaments and a few hundred years later tools and weapons. For other regions, the amount of data collected for this thesis does not allow for a satisfactory chronological comparison of different object types.

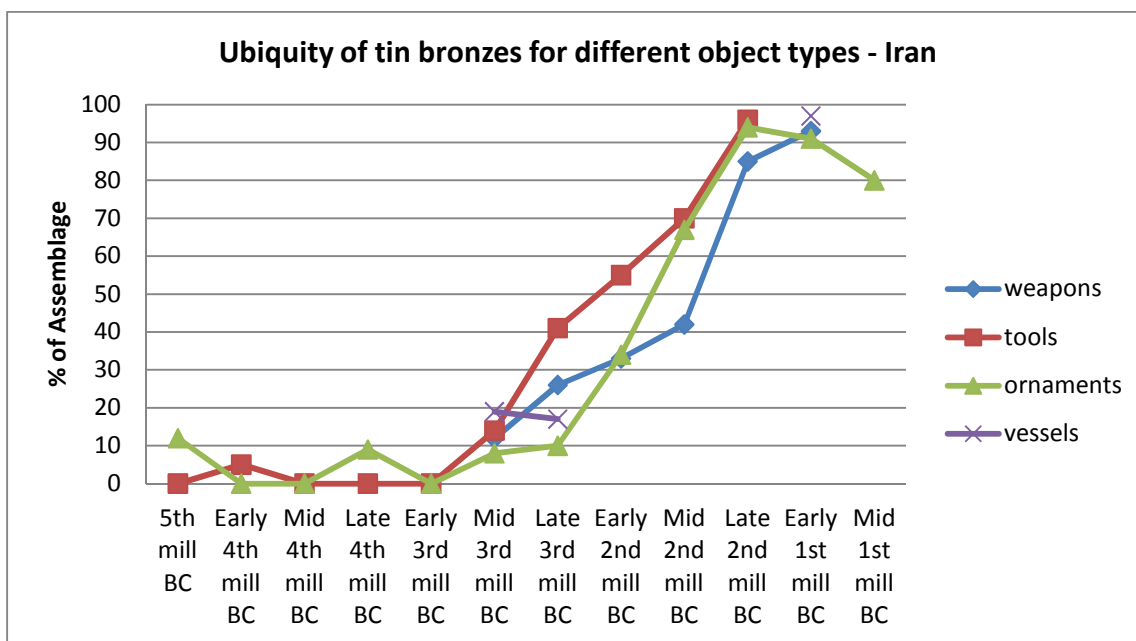
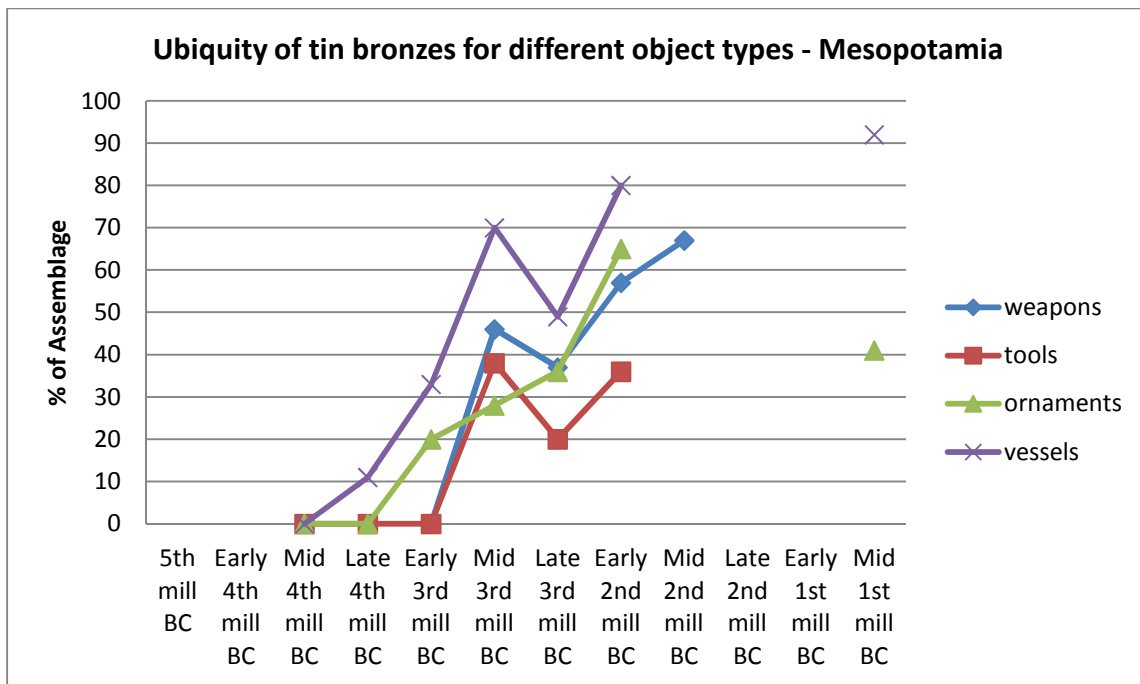


Figure IV-17. Chronological evolution of the ubiquity of tin bronze in the copper-based metal assemblage for different object types in Iran (only periods for which more than 5 objects were analysed have been plotted on this graph)



**Figure IV-18. Chronological evolution of the ubiquity of tin bronze in the copper-based metal assemblage for different object types in Mesopotamia (only periods for which more than 5 objects were analysed have been plotted on this graph)**

While these observations do not fundamentally contradict the accepted understanding of the adoption of tin bronze as summarized by Pigott, they do add detail to it and challenge the notion of a similar adoption pattern in the entire Near East. In particular, we can no longer accept the general paradigm of the adoption of tin bronze first for “decorative” items and then for weapons and tools in Iran, as the data available to date points towards a simultaneous adoption of bronze for ornaments and tools.

Turning back to the Bronze-Age / Iron-Age transition, Figure IV-17 shows that by the late 2nd millennium BC in Iran, all types of copper-based objects were made almost exclusively of tin bronze rather than unalloyed copper. It should be remembered that these graphs represent the ubiquity of tin bronze in the copper-based metal assemblage only, rather than in the entire assemblage of metal objects and therefore do not show how copper or bronze were replaced by iron. It seems however that the appearance of

iron was accompanied by a slight resurgence in the use of tin-less copper for ornaments, but more data would be needed to confirm this trend as there are only 20 Iranian ornaments from the mid 1st millennium BC in our database. If this trend were to be verified, it would be interesting to discuss whether this came from a conscious choice to use copper rather than bronze for some of the ornaments or whether it had to do with the access to tin or with a change in the status of tin-bronze with the appearance of iron.

Changes between the Bronze Age and the Iron Age in Mesopotamia are difficult to assess as we do not have enough data from this transition period to compare different object types. All that can be said so far is that, for ornaments, the ratio of tin bronze to tin-less copper is lower in the mid 1st millennium than in the early 2nd millennium BC (the last period prior to the mid 1st for which more than five objects were analysed). This observation would obviously need to be verified with more analyses, but seems to follow the same trend as in Iran: a slight resurgence of tin-less copper in the Iron Age. For vessels on the other hand, tin bronze was adopted earlier than for the other object types and was still used preferentially in the mid 1st millennium BC when 92% of the analysed copper-based vessels were made of tin-bronze. Tin bronze must have had properties that made it desirable for the production of vessels in the eyes of the Mesopotamians. Since most of the Mesopotamian vessels are made of sheet metal, the ease of casting and the hardness that tin confers to bronze would not have been too important, so it is possible that the Sumerians were trying to emulate the colour of gold for their vessels. Another potential explanation for the apparent earlier adoption of bronze for vessels could be a lag in the deposition of other kinds of objects. This possibility would require further investigation, notably with a study of the context in which the vessels were recovered, compared to weapons and tools in particular, but this is beyond the scope of this thesis.

Figure IV-19 shows the evolution of the percentage of tin in the bronzes for these four categories of objects in Iran. Median levels are for the most part fairly constant and situated between 7 and 9% tin with no significant differences between object types. In the 3rd millennium the levels are slightly more variable for weapons in the mid 3rd for example having high tin levels and ornaments lower tin levels, before it stabilises in the 2nd millennium. In Mesopotamia, the levels are on average higher than in Iran and also more variable (Figure IV-20). These graphs do not show any striking differences in the amount of tin added in different types of objects, but we will look into this question in more details in the following section by studying histograms of tin contents. There is also little significant chronological variation of the tin content confirming what we have said earlier about a “standard recipe” being reached early on and then carried through into the Iron Age.

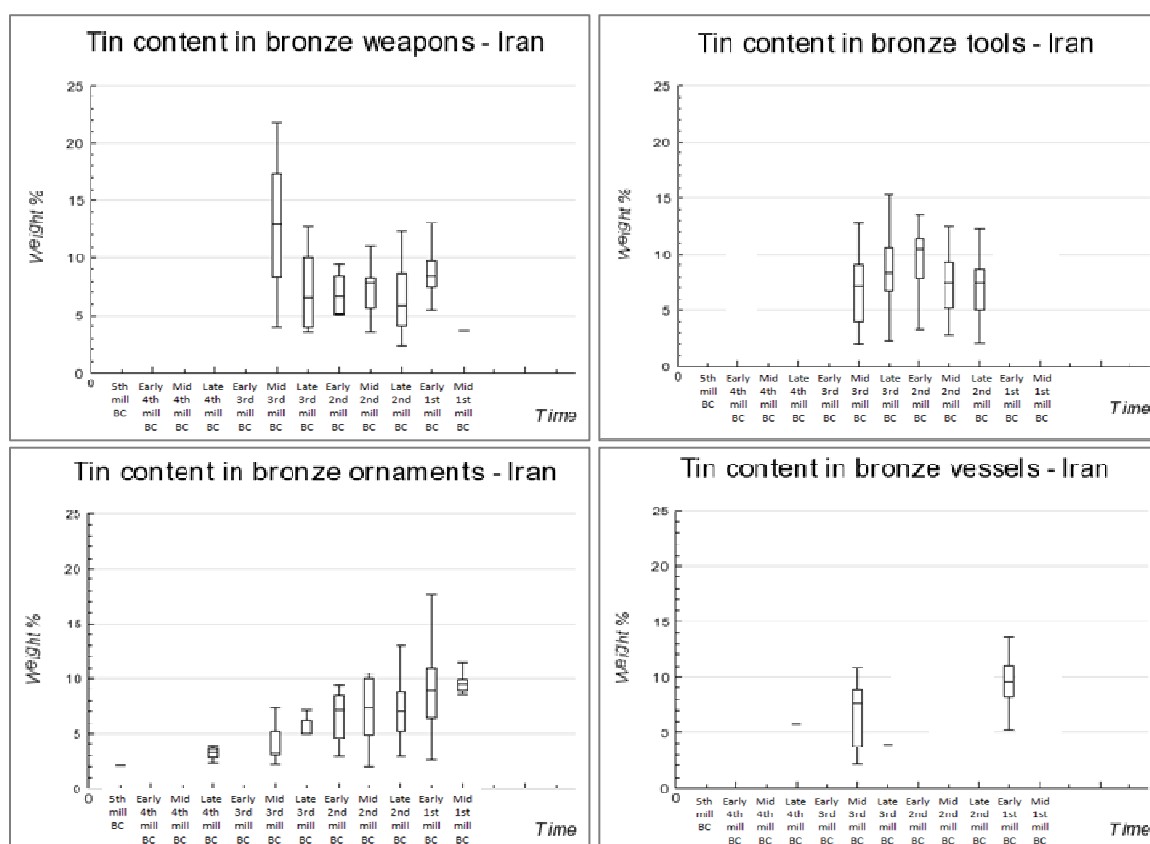
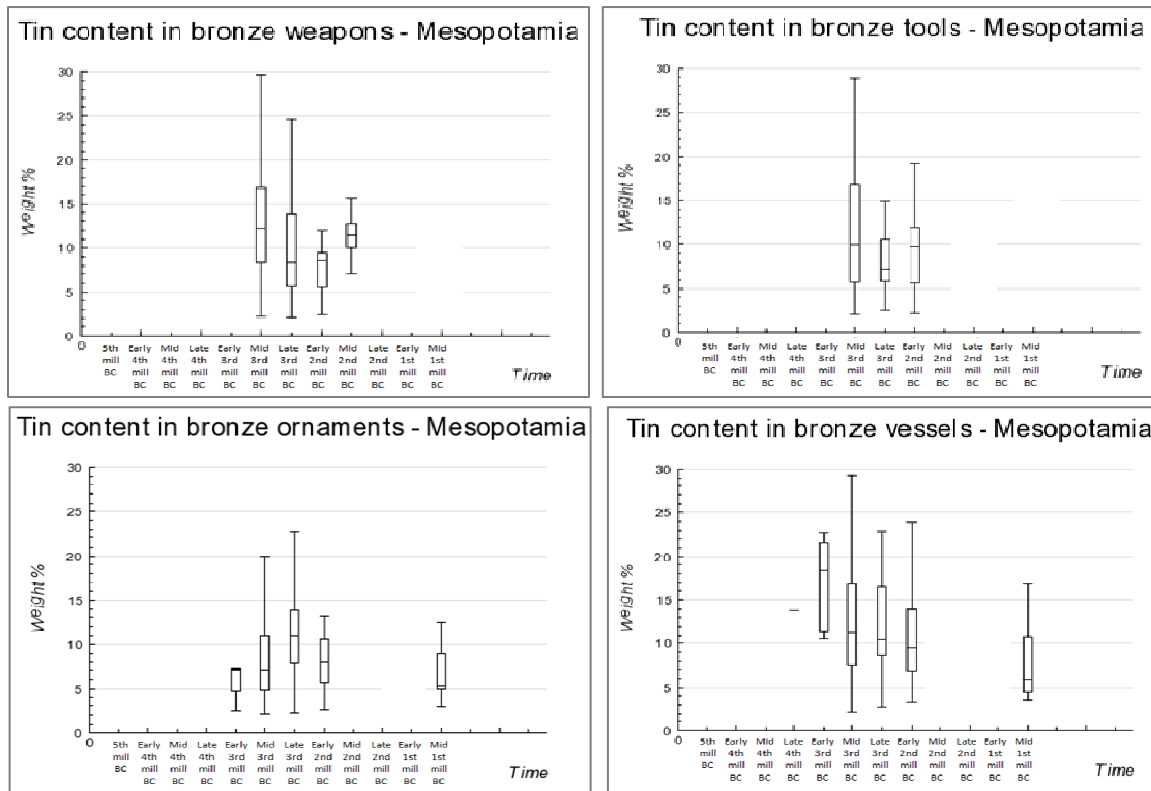


Figure IV-19. Evolution of the tin content in tin bronzes (>2% Sn) for weapons, tools, ornaments and vessels in Iran



**Figure IV-20. Evolution of the tin content in tin bronzes (>2% Sn) for weapons, tools, ornaments and vessels in Mesopotamia**

### 3. Definition of a “mid range” tin content.

We have so far studied the use of tin using a very simple dichotomy, the absence or presence of tin using 2% as a cut-off value between the two categories. However the distribution plots representing the tin content of the objects of this database have showed that for several Iranian regions this dichotomy is probably too simplistic as there appears to be two peaks in these diagrams: a primary peak around 8-9% tin and a secondary peak around 4-5% tin. What do these two ranges of tin content represent? Theoretically, they could result from the use of different alloying processes or indicate that the copper used has been through a different history involving some or all of the processes of smelting, casting, working, re-melting, mixing, etc. But were these different pathways, leading to different tin-contents the result of conscious choices, driven by the properties of the end result (whether these are colour, strength or

something else) or were they driven by practicality, availability of resources or tradition?

First we have examined the distribution histograms in Iran for different types of objects for the two time periods defined in Chapter 3 (Figure IV-22 and Figure IV-23). If the choice of the tin content was driven by the end result, we would expect to see certain type of objects using preferentially one range of tin over the other. The graphs show that all of the different object types have objects falling in both ranges. In most of the cases where enough objects were analysed, the second peak (>6% tin) is predominant. There are however a few exceptions: in the first time period pins and daggers and swords are more often in the 2-6% tin range. In the second time period this is only the case for arrowheads and spearheads and to some extent for pins. Interestingly these two categories are also the only two for which tin-less copper is still also used.

Despite these differences, there doesn't appear to be a straightforward pattern, for example utilitarian objects falling into one tin range and decorative objects into another. This can mean that our categorizing of objects is flawed and we need to reassess the distinction between decorative and utilitarian or that we need to think in terms of different criteria. A closer look at the axes from the second time period for instance makes it obvious that the distinction between ornaments and weapons or tools is not as straightforward as it appears. Many of the axes from Luristan in particular are highly decorated spike butted shaft-hole axe-heads such as the ones represented in Figure IV-21. For some of these, what should have been cutting edges were in fact never ground down after casting making these axes impossible to use as weapons or tools (Moorey, 1971). This means that this type of object could well have been fashioned for ceremonial purposes only and therefore our "axes and adzes" category includes both real tools and weapons and ceremonial one for which different properties were probably

required. This ambiguity is most likely also true for the “daggers and swords” category. This only leaves the “arrowheads and spearheads” category as the only one that we can somewhat securely consider as representative of utilitarian objects. However these have a slightly different status to other weapons: as projectiles, they would have been easily lost and therefore their durability was less important. They probably needed to be produced in greater quantities than swords or daggers for example to renew the lost stock. It is therefore possible that the ancient metal smiths would have chosen quantity over quality for their production. Interestingly, Overlaet and Frame have noted that at Pisht-i Kuh sites in Luristan and at Tepe Godin, in the late stages of the Iron Age when almost all weapons were made of iron, arrowheads were still often made of copper alloys or even of silex or bone (Overlaet, 2003, pp.151, 174; Frame, 2010, p.1709). This indicates that arrowheads were rarely produced in materials that were considered valuable by the community that made and used them. Since for arrowheads and spearheads the 2-6 % tin peak is more important than for other object types it is possible that this range of tin contents was available in higher quantities and cheaper than bronze with more tin.



Figure IV-21. Luristan spike butted axe-heads in the Ashmolean Museum. Top two: personal photography - accession n° 1951.158, 1951.160. Bottom two: the cutting edge was never ground down (Moorey, 1971) - accession n° 1965.188 and 1965.761

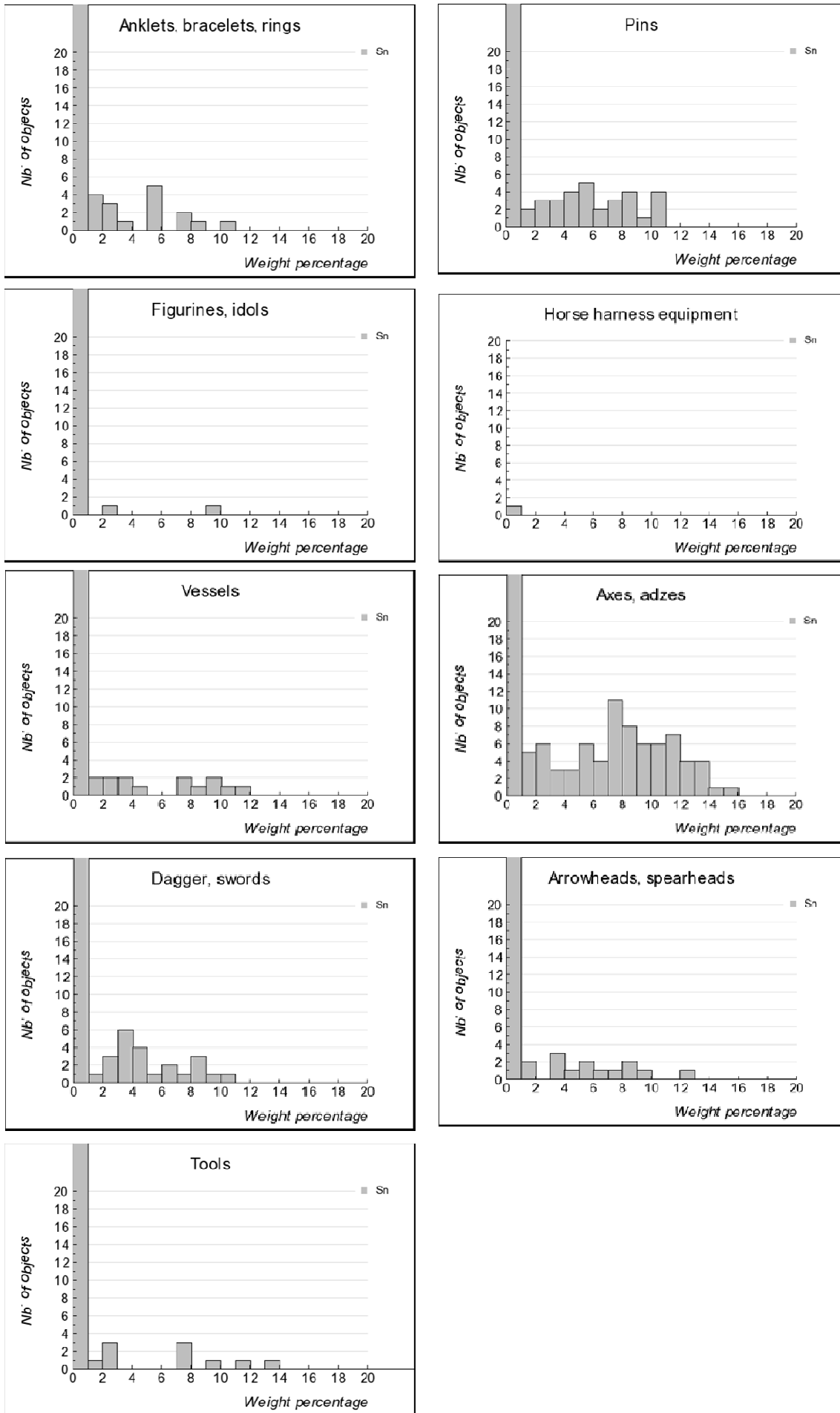


Figure IV-22. Tin content in different objects types in Iran between the early 3rd and the mid 2nd millennium BC

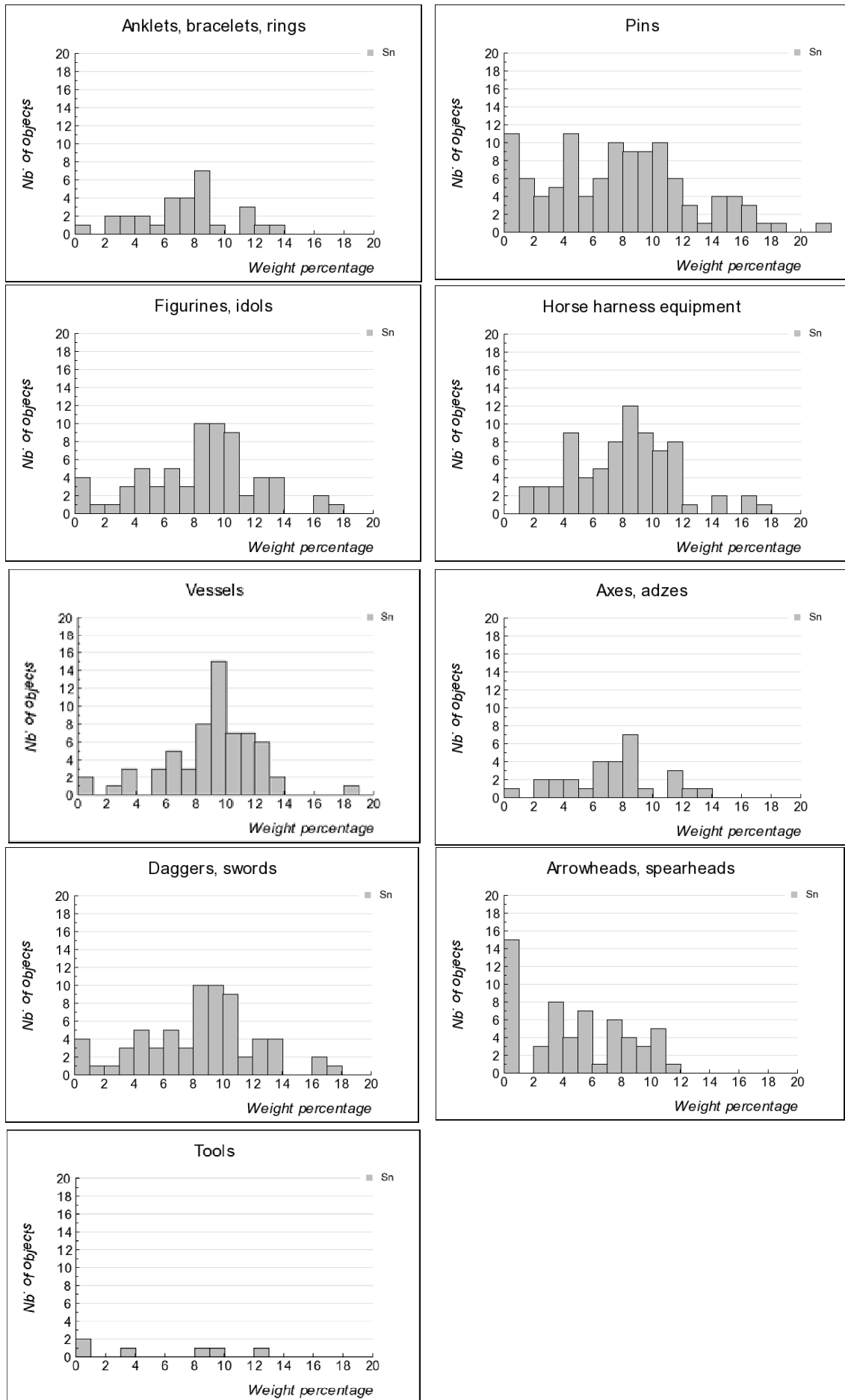


Figure IV-23. Tin content in different objects types in Iran between the late 2nd and mid 1st millennium BC

Some of these objects with lower amounts of tin were probably simply made from a recipe involving less tin. Indeed recipes from Ebla indicate quite a wide variation in recipes, with in particular 200 objects tentatively translated as scissors said to have been made with 3.45% Sn (Archi, 1993). However, the vast majority of the output of the Ebla workshops is represented by objects with around 13% Sn (see Figure IV-24. Percentage of tin in objects produced in Ebla as described in the tablets recovered there (after Archi, 1993)), and these low tin content recipes are probably not enough to explain the graphs described above.

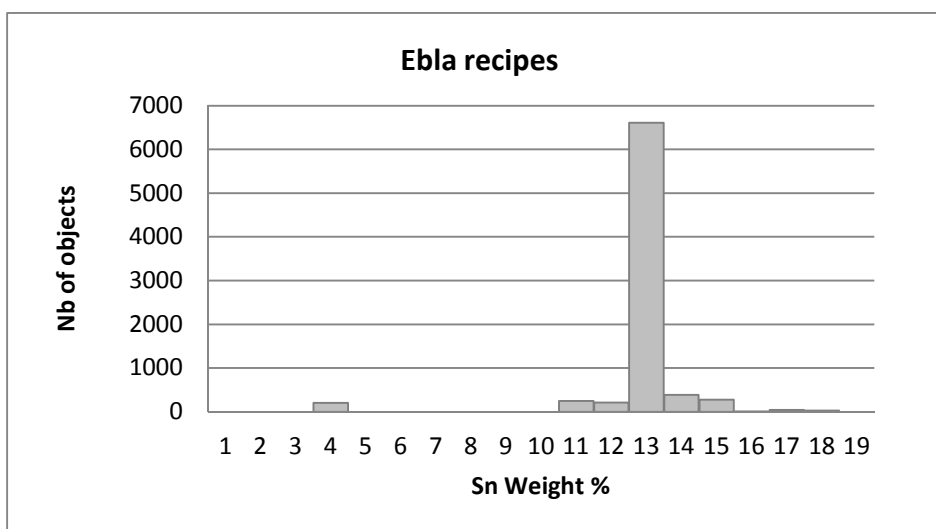


Figure IV-24. Percentage of tin in objects produced in Ebla as described in the tablets recovered there (after Archi, 1993)

Bronze with relatively low amounts of tin can then also have been the result of the recycling or mixing of bronze with higher tin contents. Indeed, we would expect the primary production of bronze from the mixing of copper and tin or cassiterite by one standardised process to produce a tin bronze profile more or less in the shape of a Gaussian distribution with the width of the curve depending on how reproducible and standard the production technique was (and also depending on the precision of the modern measurement techniques). However, the re-melting of copper at a further stage

in a recycling process would have resulted in a loss of a portion of the tin by oxidation (Bray, 2009). A mixing of tin bronze with about 8-10% tin with tin-less copper would also have resulted in bronze of lower tin content. Overall, the bi-modal distribution observed here is probably the combination of the Gaussian type curve representing a primary production of bronze and a second distribution representing recycled and mixed objects.

Figure IV-25 shows for Iran the evolution of the ubiquity of these two tin ranges. As we can see, the bronze with the lower tin percentage appears first, in the late 4th millennium BC. We have considered earlier that this corresponds to an experimentation stage where only a small amount of tin would have been added to copper. It could also be the result of recycling or mixing of imported tin-bronze. However, where this bronze would come from at this very early stage is not obvious. More interesting for our study is the decrease in the proportion of mid-range bronze from the late 2nd millennium BC. If this mid-range does indeed represent the result of bronze being recycled, then we could expect this class of object to increase in importance as time goes. Instead this might represent a good access to tin in Iran in the Iron Age meaning that recycling was not necessary and the primary production of bronze by direct addition of tin by one of the processes described in Chapter 2 was preferred. Figure IV-26 shows that this pattern is observed in all of the north-western regions of Iran and possibly Khuzistan. However, this is not observed in Oman, where the proportion of mid-range objects is always very low, suggesting perhaps that the mixing and recycling of bronze was not a widespread practise (although we have already mentioned that the values for Oman are artificially high due to the corrosion of the samples) (Figure IV-27). It is not observed in Mesopotamia either, where this proportion stays very low until the mid 2nd millennium BC and if we trust the result shown by the small number of analysed objects for the

following time period, then rises up to about 30% during the Iron Age. This could be an indicator of a change in tin supply around the mid to late 2nd millennium. It seems that Mesopotamia might have lost the ease of access to tin they had in the Bronze Age, while Iran and Oman either found a new access to tin or developed the access they already had in the Bronze Age. Thinking back to the potential sources of tin known to us by geological and archaeological research and described by ancient texts (see Chapter 2), we can envisage this change being linked to an interruption in the Persian Gulf trade that was likely to have been the route that got tin into Mesopotamia. Simultaneously, the inhabitants of Iran seem to have increased the importance of local sources such as the mine of Deh Hosein in eastern Luristan. The next sections attempt to describe more precisely the actors of these different potential pathways for the trade of tin.

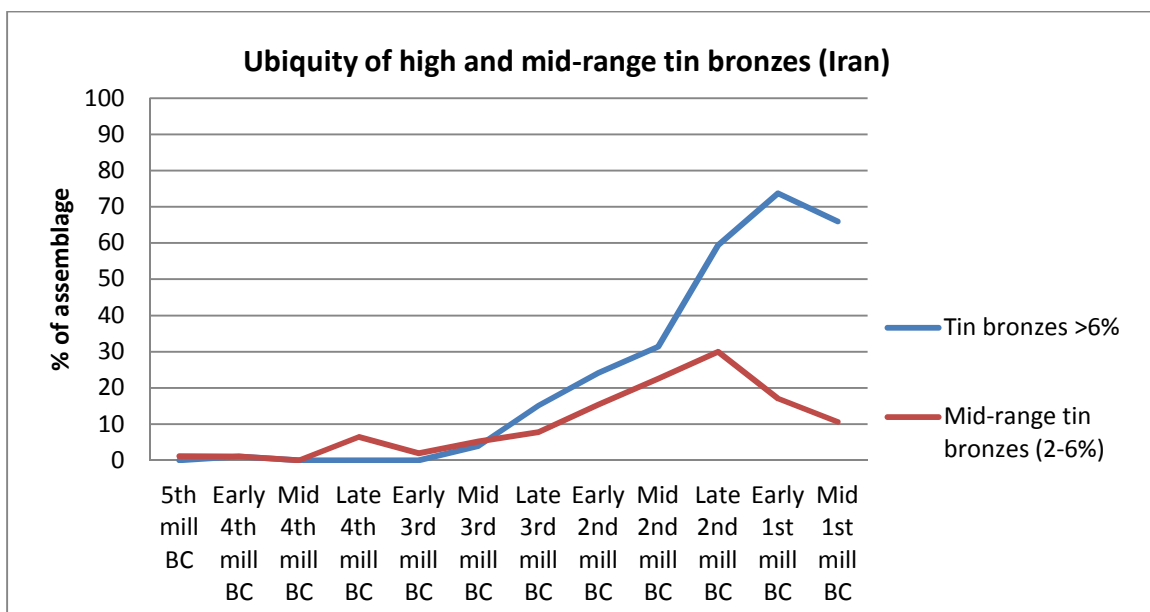


Figure IV-25. Chronological evolution of the ubiquity of tin bronze with over 6% tin and tin bronze with between 2 and 6% tin in the copper-based metal assemblage

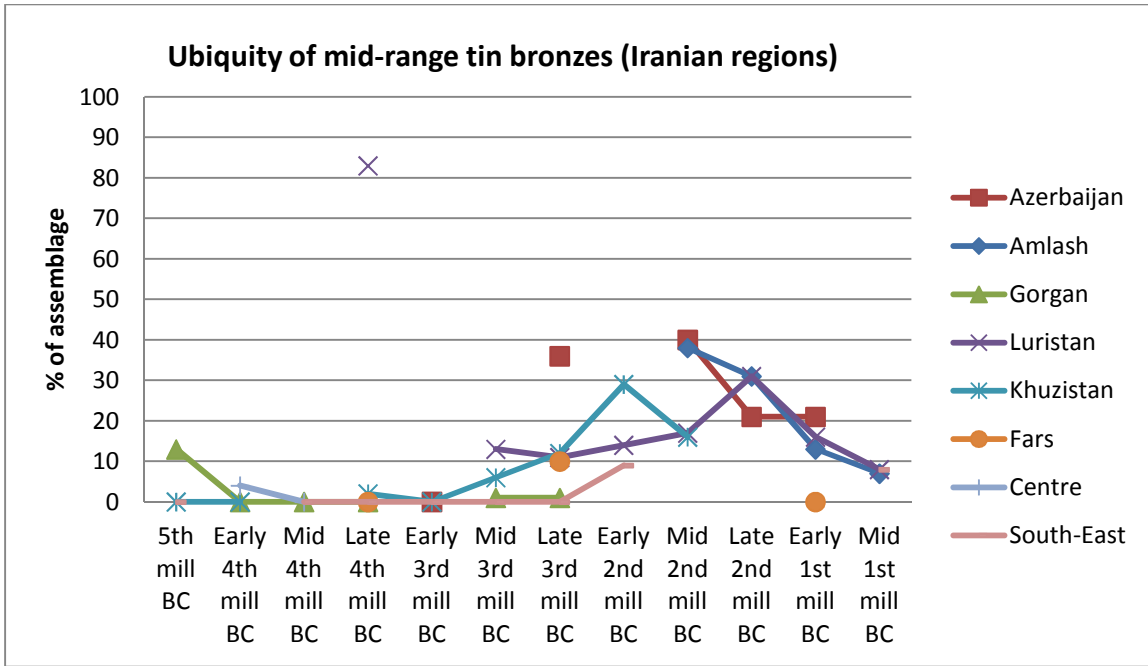


Figure IV-26. Chronological evolution of the ubiquity of tin bronze containing between 2 and 6% tin in the copper-based metal assemblage for Iranian regions. Only data points for regions where more than 5 objects were analysed are included in this graph

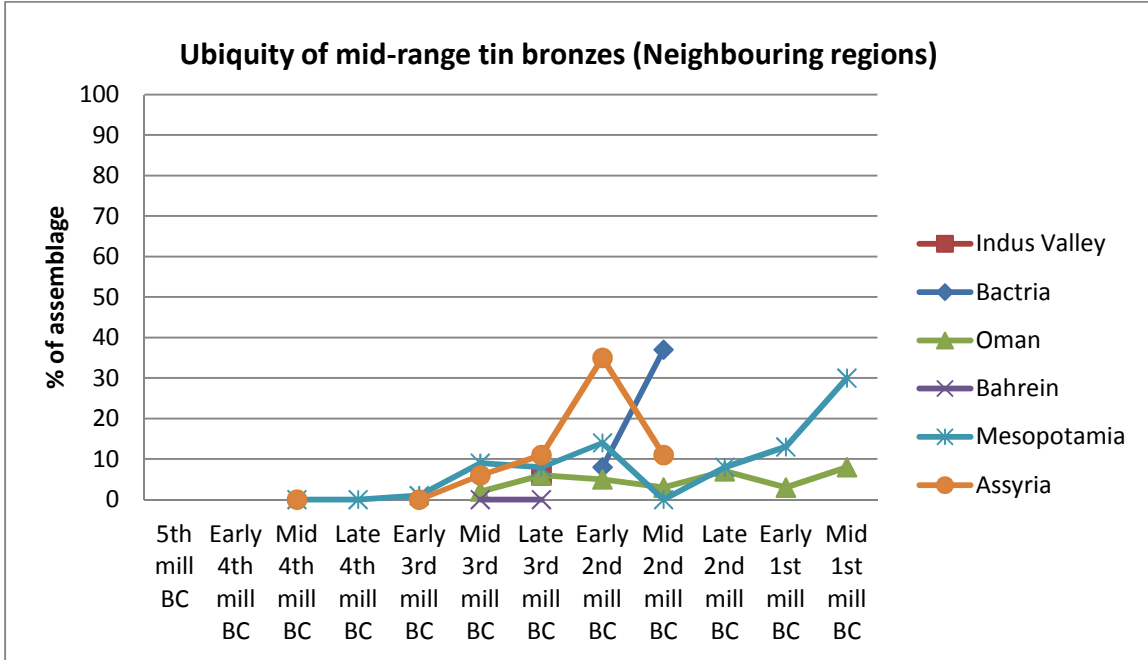


Figure IV-27. Chronological evolution of the ubiquity of tin bronze containing between 2 and 6% tin in the copper-based metal assemblage for neighbouring regions. Only data points for regions where more than 5 objects were analysed are included in this graph

#### 4. Synchronic analysis

The diachronic analysis has showed significant differences in the “bronze stories” of the different regions in and surrounding Iran. In particular we have observed a potential change in the supply of tin occurring around the period of the turn from the Bronze Age to the Iron Age. To try and understand this change, we need to know more about the different actors of the trade in the periods preceding and following it. The approach used in this section is to use the larger time periods defined in Chapter 3. Although we have seen in the previous paragraphs that metallurgy wasn't static during these long periods of time, we haven't observed within each of these periods any real rupture in the development of tin bronze. What we will call Period 1 here (from the early 3rd millennium BC to the mid 2nd inclusive) appears to very much be a time of development for tin bronze. The proportion of bronze in the assemblage steadily rises and aside from the mid 3rd millennium in Iran, the average amount of tin added to copper is very constant. Period 2 (late 2nd millennium BC to mid 1st) corresponds in Iran at least to a period where the production of tin bronze had reached maturity: in most Iranian regions it represented 80 to 100% of the bronze assemblage and the levels of tin added to the objects were still relatively constant. This approach will enable us to deal with a larger amount of data for each region which will be particularly important for the eastern regions of Bactria and the Indus Valley which are possibly key actors in the trade of tin but for which we haven't been able to say much so far.

##### a. Period 1 – Early 3rd to mid 2nd millennium BC

The map represented in Figure IV-28 shows for Period 1 the number of objects analysed for each region, the ubiquity of tin bronzes (defined again as objects having over 2%

tin) in the copper-base metal assemblage and the average amount of tin in these bronzes. A clear geographic divide immediately stands out between eastern and western Iran: both north-eastern Iran and south-eastern Iran have very few bronze objects (respectively 2% and 5% of the assemblage) while for this time period the western regions all have over 20% of bronze objects. Unfortunately not enough objects were analysed from the central regions to assess whether or not tin bronze was present there in significant quantities. Further east however, in Bactria and the Indus Valley, tin bronze is present in proportions comparable to western Iran and Mesopotamia: respectively 31% and 38% of the copper-based metal assemblage. This is also true of the Oman peninsula where tin bronze represents 30% of the assemblage.

The effective absence of tin bronze in eastern Iran has been the subject of many discussions about tin trade routes and technological conservatism. Indeed, if the major sources of tin used for bronze production in the Near East in this period are located in Afghanistan, as seems to be the consensus today (see Chapter 2), then why doesn't eastern Iran, located closest to the sources, use this tin?

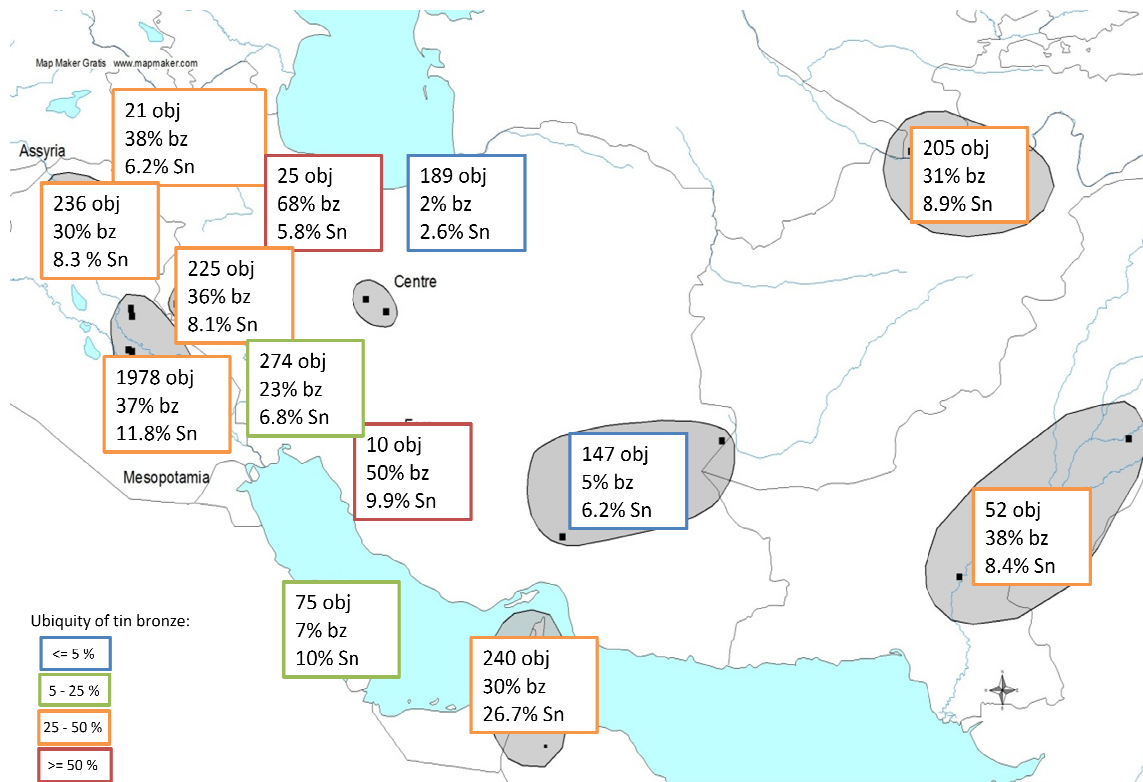


Figure IV-28. Map representing the number of copper-based objects analysed for each region between the early 3rd and the mid 2nd millennium BC, the ubiquity of objects with more than 2% tin and the average tin concentration in the bronzes of these assemblages

Beale studied the trade between Iran and Mesopotamia as seen from south-eastern Iran. Based on evidence from the trade of minerals, in particular chlorite and turquoise to and from Tepe Yahya, he defined different trade mechanisms and attempted to understand the reasons behind changes between these mechanisms. He introduced the idea of a by-pass phenomenon “whereby consumers or individuals who might profit from trade sought consistently to reduce the number of intermediaries along the trade chain and deal more directly with the source area so as to reduce the cost of desired goods” (Beale, 1973, p.144). His work focused mostly on the period between 4500 and 3400 BC, earlier than the period we are interested in here, but this by-pass concept can be applied to later periods, which is what Moorey and Pigott did regarding the trade of tin in Iran in the 3rd millennium (Moorey, 1982, p.88; Pigott, 1999b, p.84). Moorey

suggested that the by-pass of eastern Iran might have to do with the level of complexity of its economy compared to those of Mesopotamia and Elam. He wrote:

*“Kohl has argued, by implication rather than by demonstration, that by the middle of the third millennium B.C. tin had assumed greater significance in the lowland economies of Elam and Sumer than in the less complex economies nearer to the metal's supposed sources; if so, on Beale's "by-pass phenomenon", this may be a case of a trade conducted directly between Mesopotamia and Elamite city-states and those settlements, or rulers, controlling the resource area, by-passing many Iranian regional centre.”*  
(Moorey, 1982, p.88)

For Pigott the by-pass of eastern Iran might be related to differences in the status of tin and in the demand for tin between these two areas:

*“Sumerians were intent on trade and the acquisition of exotic, luxury materials. The rarity of tin may have promoted its status among Sumerians while the peoples of the Iranian Plateau may have remained uninfluenced by such pressures (Stech & Pigott, 1986, p.48). As such, the tin “by-passed” the plateau on its way to Mesopotamia.”*  
(Pigott, 1999b, pp.83-84)

In this article from 1999, Pigott also presented the Iranian plateau as “technologically conservative” since it kept using arsenical copper while tin-bronze developed in surrounding regions (Pigott, 1999b, p.84). As we have seen in Chapter 1, this idea was further discussed by Thornton (Thornton et al., 2005; Thornton, 2009b, p.320) with regard of the copper-base metal assemblage of Tepe Yahya. The absence of tin bronze in eastern Iran may well be a question of choice rather than accessibility. Could it be that tin trade still transited via the Iranian plateau but was simply redirected further west, while the metalworkers of the Plateau preferred the use of arsenical copper?

Alternatively in a recent paper Thornton and Giardino suggest that the tin sources of Afghanistan might have not actually been exploited until the arrival of BMAC people in this region around 2000 BC (Thornton & Giardino, 2013).

Rather than trying to understand the exact cause for the lack of tin-bronze in eastern Iran, others have focussed on defining the trade routes used. As we have seen in Chapter 2, the textual evidence reviewed by Muhly (1973) pointed towards two potential routes: a coastal route passing through the Gulf and an overland route supplying Mesopotamia through passes in the Zagros. Kenoyer and Miller have studied the metal assemblage of the Indus Valley which would have played an important role in the Persian Gulf trade. The land of Meluhha, from which eastern goods possibly including tin were shipped to Mesopotamia, is usually considered to be located in the Indus Valley. From 2600 BC to 1900 BC, during the Harappan Phase, the Indus Valley civilization covered a vast region with several major urban centres. From 1900 BC onwards, the civilization started to decline. In their overview of Harappan metal-working, Kenoyer and Miller suggest that while some sites of the Helmand tradition (Mundigak and Shahr-i Sokhta) were located at strategic points on routes linking Afghan tin resources to the Indus Valley and Iran, the Indus people might have chosen not to rely on these and developed their own system of distribution in which the Harappan outpost of Shortugai in northern Afghanistan could have played an important role (Kenoyer & Miller, 1999, p.118). Unfortunately much more than the 52 analyses of objects from only two sites (Mohenjo-Daro and Harrappa) available to date would be necessary to confirm this hypothesis and understand trade and alloying patterns in that region (Hoffman & Miller, 2009). Data from Afghan sites notably would be particularly precious. Analyses of objects from Mundigak for example would shed some light on the role of the Helmand tradition, while objects from Shortugai would tell us more about the Harappan role in tin mining

and trade. As we have seen in Chapter 3 however we were only able to find the analyses of two Afghan objects from relevant time-periods to enter in our database (Caley, 1972). Yet, given the data shown in Figure IV-28 and Figure IV-29, it is at this stage possible to envisage the tin trade being controlled by the Harappans and shipped by them through the Gulf towards Mesopotamia and Elam, via Oman and by-passing the Helmand tradition and eastern Iran.

Weeks also looked into the nature of a Persian Gulf trade route, this time from the perspective of the Oman Peninsula. Based on his observations on trade of different kinds of goods as well as on his compositional and lead isotope analysis of Omani metals (Weeks, 1997; 1999), it would appear that the Persian Gulf route transporting supplies from the Indus Valley into Elam via Oman and Bahrain was active during the 3rd millennium but collapsed when the Harappan civilization declined around 1900 BC (Weeks, 2003, pp.181-87). Weeks suggests that, at this point, tin could have been imported into the regions of western Iran via a northern overland route and that Susa might have played an important role in redistributing the tin from this northern route to Bahrain and to the west.

The analysis of the data present in our database does not allow us to make any ground-breaking conclusions concerning the trade routes for tin used in Period 1. However a closer look at the histogram of tin distribution in copper-based objects (Figure IV-29) prompts a few remarks:

- The data available so far from Bactria and the Indus Valley is meagre, but we can see that they both have a similar percentages of tin bronze in their assemblages, similar average percentages of tin in the bronzes and a similar histogram profiles: bronzes with compositions anywhere from 2 to 18% Sn were

produced, with no real peak of concentration anywhere. This might suggest that they had a similar access to tin. While this is not enough to prove the importance of the Indus Valley in the trade of northern Afghan tin or possibly even in the sites of the Andronovo culture, it certainly gives credence to this possibility, that will have to keep being explored as new data comes to light. Similarly, although the data from Oman is difficult to interpret due to corrosion problems, it is still clear that Oman had access to tin and produced tin bronze in percentages again quite similar to the Indus Valley and Bactria, giving weight to the possibility that northern Afghan tin then continued from the Indus Valley through the Gulf and was used there with similar alloying practices.

- The histogram of tin concentrations in Mesopotamian bronzes presents a peak around 8-10% of tin, but also tin bronzes of many different compositions. This shows that Mesopotamia was able to procure significant amounts of tin during Period 1, in the form of bronze, cassiterite, or tin ingots, and would then manufacture objects with compositions they deemed suitable for the local demand. It is clear from the texts and from evidence reviewed above that some of this tin would have come to Mesopotamia through the Gulf. Some of the objects with very high percentages of tin might have come pre-alloyed from Oman which also shows very high percentages of tin (although we have seen that these values are biased by corrosion). We argue here that, to satisfy this demand, Mesopotamia had to draw from other sources, one of which might have been located in Luristan. Indeed the histogram of tin concentrations for Luristan indicates that this region too was producing tin bronze in significant quantities. Of course there is a possibility that we are reading the data backwards and that

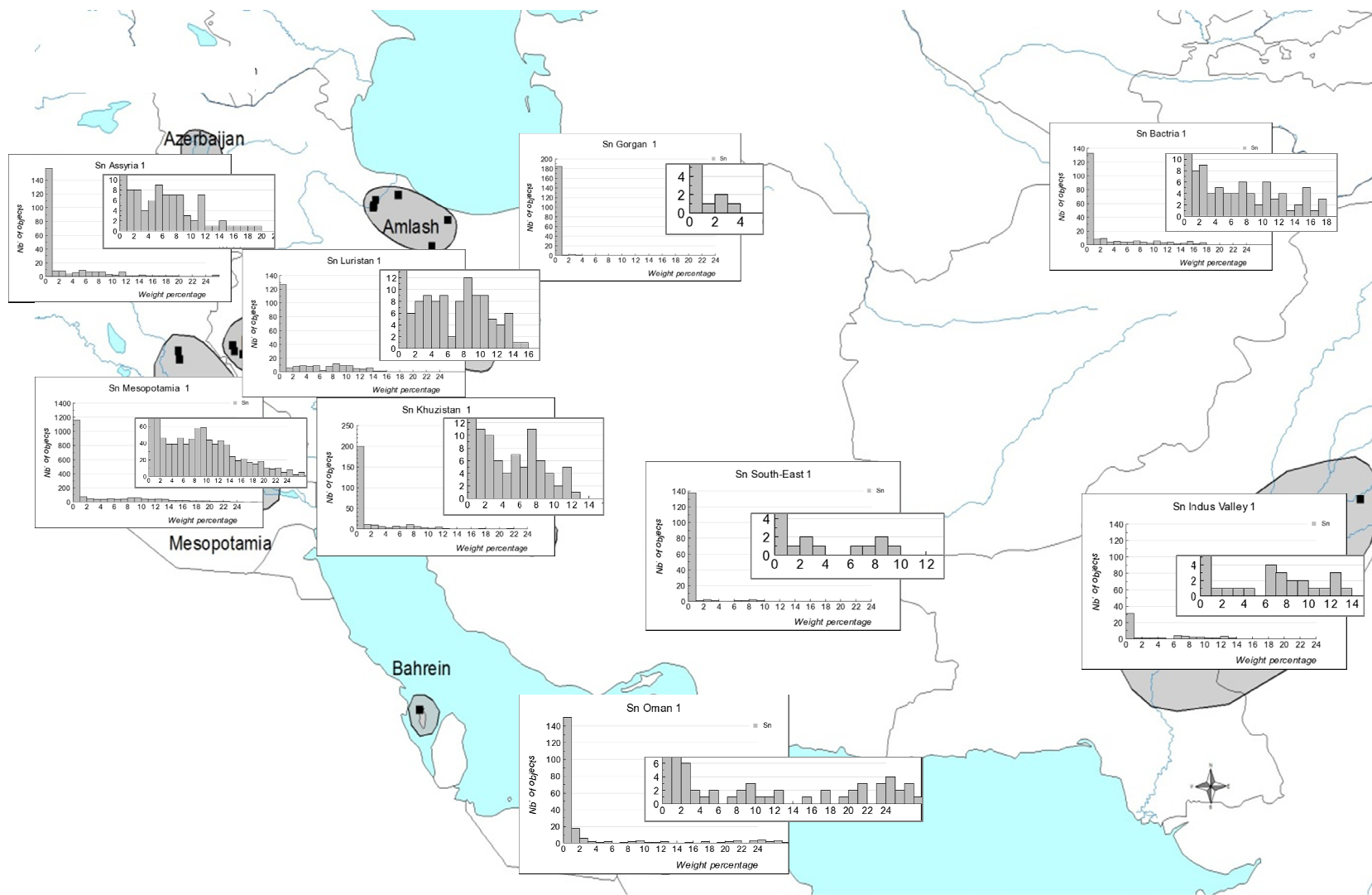
Luristan was at the receiving end of the Persian Gulf trade. However the profile presented in Figure IV-29 with a distinct peak at 8-9% tin suggesting a well defined “recipe” for tin bronze and the regularity of development curve of Figure IV-3 tend to indicate that Luristan had a steady, possibly uninterrupted access to tin. If it came to Luristan from Mesopotamia through passes in the Zagros, we could envisage difficulties to procure tin due to fluctuations in the control of these passes. Added to the discovery of the Deh Hosein mine, and the texts indicating tin arriving to Mesopotamia through the Zagros it appears much more likely that Mesopotamia acted as a “tin magnet”, being the final destination of the Persian Gulf trade as well as attracting tin from western Iran either in the form of tin, or perhaps already alloyed as bronze. We saw above that the tin levels in Mesopotamia reach a “standard level” before they do in Luristan, indicating, in accordance with the texts, that the Persian Gulf trade might have been used first (from the early 3rd millennium BC) while the Luristan source gained more importance towards the end of the 3rd millennium, when the coastal trade lost of its importance. However small amounts of tin (or bronze) coming from Luristan throughout the 3rd millennium are also conceivable.

- Finally concerning the role of Elam and Susa in this trade, Pigott asks:

*“Did the Elamites have a hand in controlling access to nearby tin deposits, as it is hard to imagine a more sought after and strategic material.”* (Pigott, 2011, p.287)

It is clear from Figure IV-29 that Khuzistan did have access to tin, but it doesn’t appear to have a production industry quite as developed as in Luristan or

Mesopotamia: the percentage of tin bronze in the assemblage there is lower than in other regions of that area and Figure IV-29 shows that many objects fall in the 1-4% Sn range and are possibly recycled. Moreover, although Pigott emphasizes the early adoption of tin bronze in Elam, the data we have collected indicates that the real adoption of tin bronze in Elam happens at the same time as in Luristan and its development there is slower than in other Iranian regions (Figure IV-3). Therefore we suggest that in Period 1, Elam might have mostly been a middleman in the trade of tin from Luristan to Mesopotamia, possibly when the access through the Zagros was difficult for political reasons. It is not impossible that it also played a role in the Persian Gulf trade and we can envisage contact between Oman and Elam, in which case it would be interesting to have access to more data from Fars. For Elam, it seems that tin might have been of strategic importance rather than truly sought after to produce bronze objects. To examine this possibility, we have looked at the types of objects made of tin bronze from Elam. Of the 25 objects with over 6% tin from Elam for which we have pictures or drawings, at least half are typologically identical to objects from Luristan or Mesopotamia. Although it is possible that Elam manufactured objects in a very similar style to its two neighbours, it is also conceivable that these objects were traded from Luristan and Mesopotamia into Elam and that Elam produced very little bronze of its own. In Chapter 6, we attempt to verify this hypothesis with a study of the other elements present in the metal.



**Figure IV-29.** Distribution of the tin concentration for each region between the early 3rd and the mid 2nd millennium BC. For each region a full histogram and a zoomed-in version are given. The first one allows to visualize the proportion of tin-less copper while the second one gives a better idea of the profile of the tin-content distribution.

b. Period 2 – Late 2nd to mid 1st millennium BC

Figure IV-30 shows for our second time period the number of objects analysed in each region for which more than ten Period 2 objects were analysed, the ubiquity of bronzes in the copper-based metal assemblage and the average percentage of tin in these bronzes. For reasons we have already discussed at the beginning of this chapter, only very few objects have been analysed in eastern and southern Iran and we have no data at all for the Indus Valley and Bactria.

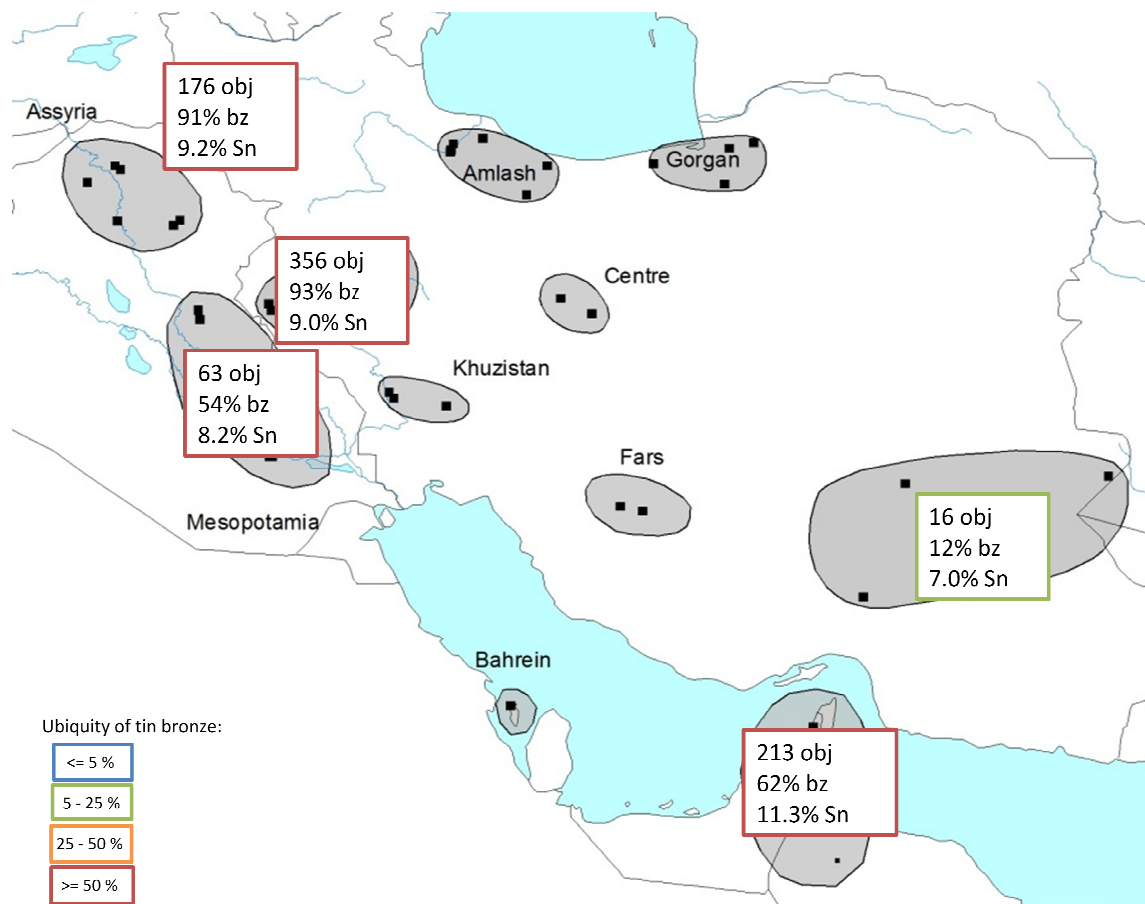


Figure IV-30. Map representing the number of copper-based objects analysed for each region between the late 2nd and mid 1st millennium BC, the ubiquity of objects with more than 2% tin and the average tin concentration in the bronzes of these assemblages

In the regions for which a significant number of objects were analysed: north-western Iran, Oman and Mesopotamia, we can observe a phenomenon already described in our

diachronic analysis. While tin-less copper is almost entirely replaced by bronze in north-western Iran (copper representing only about 10% of the assemblage)<sup>4</sup>, in the other two regions it is still very much in use representing just under half of the assemblage. As we have already suggested, reasons for this discrepancy could be the interruption of the Persian Gulf trade, making it difficult for Mesopotamia and Oman to procure a constant supply of tin and therefore continue to develop the tin-bronze industry, while north-western Iran started or continued to make use of local sources such as the mine of Deh Hosein.

A look at the distribution histograms for tin in these regions in Figure IV-31, gives us new information on the nature of the bronze present in Mesopotamia in this period. The concentration profile for Mesopotamia contrasts greatly with the ones in north-western Iran in that more objects fall in the low-tin bronze range (2-6% tin). If this profile is confirmed when more objects from this period are analysed and if it is true that this range represents a mixing of tin bronze with tin-less copper in recycling operations, then, we can conclude that most of the Mesopotamian bronze was recycled. Three possible sources for the bronze used for this mixing and/or recycling are conceivable. Most of it must have been either local bronze produced in previous periods, or must have come from north-western Iran given the ubiquity of tin bronze in that area and the clear peak around the 8-10% tin concentration, indicating that they were regions of primary production of bronze. However some of the bronze (or tin) might still have come from Oman. We must remember that the tin bronze from Oman present in our database for this period comes almost exclusively from one hoard. Nevertheless it indicates that at least from time to time, Omani metalworkers were able to lay their hands on substantial amounts of tin. Where this tin would have come from is an open

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<sup>4</sup> And Assyria, as attested by the recently published volume on Assyrian Iron Age metalwork where tin-less copper represents only 7% of the analysed material (Ponting, 2013).

question and will probably remain so until more analyses of Iron Age objects from eastern regions are available. It is possible that the coastal trade of tin from Afghanistan continued, perhaps more sporadically than in the previous period, and if so might have delivered small amounts of tin or bronze to Mesopotamia.

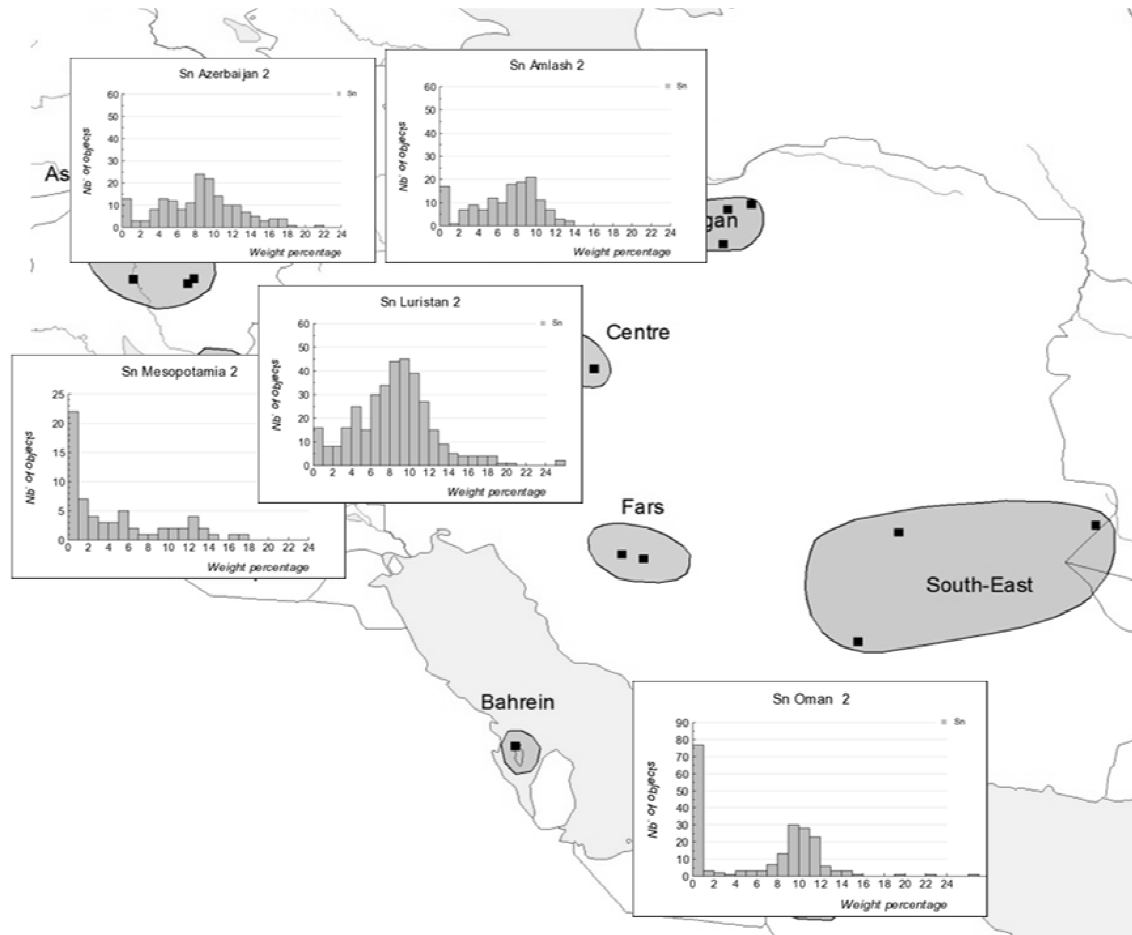


Figure IV-31. Distribution of the tin concentration for each region between the late 2nd and mid 1st millennium BC

## 5. Discussion

### a. Benefits of using a large dataset

In this chapter we have set out to examine the problematic “tin question” from a new perspective, looking simultaneously at the evidence from 5015 Near Eastern objects.

This has enabled us at first to let the numbers speak for themselves. Only subsequently have we looked at how that data compared with ancient texts and our knowledge of geological deposits. We have therefore been able to verify, from all the literature available on the subjects, how much of it can be confirmed, how much needs revision or is founded on too little evidence and how much new information we can bring to the table.

Although it is not the main focus of this thesis, we have been able to clarify some aspects of the early adoption and development of tin bronze. We have seen in particular that despite the traditional view of bronze being adopted first for ornaments and then for utilitarian objects, bronze weapons, tools and ornaments appeared simultaneously in Iran. In Mesopotamia bronze vessels appeared first, followed about a third of a millennium later by ornaments and another three centuries or so later by tools and weapons. This gives us some information on the different status of bronze in these two countries and the different reasons for adopting it over copper. The story we would like to propose here is that Mesopotamia started using this metal because of the rarity of tin and the valuable status that it conferred to bronze, as Pigott wrote, because they were “intent on trade and the acquisition of exotic luxury materials” (Pigott, 1999b, p.83). Only later did they realize the other properties of tin bronze notably its greater mechanical strength. Iran however possibly only followed the trend set by its powerful neighbours and started using tin bronze 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 intermittent Mesopotamian presence in Khuzistan must notably have played a role in the adoption of tin-bronze there. But the adoption of tin bronze in Iran and in western Iran in particular might also be due to the role it played in supplying tin for Mesopotamia whether it was as a source region (Luristan) or as a middleman in the

trade (Elam). If the impetus for the adoption of bronze came from following the example of Mesopotamia, Iran however subsequently developed a bronze industry of its own, eventually surpassing that of Mesopotamia.

The second point we would like to make and that has not been abundantly commented on in the literature is the level of tin in the bronzes of the Near-East from the 3rd to the 1st millennia BC. We have observed that the median percentages of tin in bronzes stayed, as far as we can tell, fairly constant over this entire time period, perhaps decreasing slightly for Mesopotamia and increasing ever so slightly for Iran. On the percentages of tin themselves we have not been able to comment on the production processes used by the ancient metal-smiths. This however tends to prove that a “standard recipe” for the production of tin was reached early on in the development of this alloy which is compatible with the existence of written recipes varying only slightly over time (see Chapter 2). But things are not so simple. As we have seen the proportion of objects falling in the 2-6% tin range to those falling with over 6% tin changes over time. We have supposed that the middle tin range corresponds to a mixing or recycling of bronze objects. If we consider, as explained in more detail in Chapter 6, that the further away we get from a source of tin, the more likely it is that the copper will have been recycled after alloying, then looking at the histograms of tin concentration can potentially be a key tool in understanding patterns of alloying and trade. As an example, we have calculated the ratios of objects with over 6% tin to objects with 2 to 6% tin for objects from each region in our Periods 1 and 2 where more than 50 objects were available (Table IV-1). Interestingly, the regions with the higher ratios are the regions located closest to the potential tin sources: the IndusValley and Oman in Period 1 and Luristan in Period 2 (Oman in Period 2 has a very high ratio, but as we have seen this is due to the high number of objects analysed from a single hoard for this period). This

ratio can potentially be used as a crude indicator of the ease of access to tin and in turn to trace the movement of tin, from regions with higher access to tin to region with more difficult access.

Ratio highly alloyed/mid-range	Period 1	Period 2
Azerbaijan		3.4
Amlash		2.6
Gorgan	0.0	
Luristan	1.8	4.2
Khuzistan	1.3	
Fars		
Centre		
South-East	1.7	
Indus Valley	5.7	
Bactria	1.9	
Oman	5.5	13.8
Bahrein		
Mesopotamia	3.3	1.3
Assyria	1.6	

Table IV-1. Ratio of the number of objects with over 6% tin to the number of objects with 2 to 6% tin for regions where more than 50 objects were analysed for Period 1 and Period 2 respectively

#### b. Bronze Age/Iron Age transition

Going back to the Bronze Age / Iron Age transition, it must first be noted that in Iran, the Iron Age really corresponds to the time when the tin-bronze industry reaches its maturity: for all of the western regions and for all of the object types, the percentage of tin bronze in the copper-based assemblage reaches 80% or more in the late 2nd millennium BC and stays around that level until the mid 1st millennium BC. If in terms of the number of bronze artefacts produced the Iron Age is the continuation of the Bronze Age, what then, if anything, changed with the arrival of iron? To answer this question we have had to look outside Iran. Unfortunately, we are severely lacking in data for Afghanistan, Bactria and the Indus Valley for the Iron Age periods. The data

we do have for Mesopotamia and Oman shows a picture very different from what was happening in Iran. There, the proportion of tin bronze never reaches levels quite that high and doesn't change very significantly from about the late 3rd millennium onwards.

This data allows us to propose a new picture of the Near Eastern tin trade. Going back to Beale's typology of trade (Beale, 1973), we would like to suggest the following patterns:

#### Period 1: Early 3rd – mid 2nd millennium BC (Figure IV-32)

This period, and especially the 3rd millennium BC, is as Muhly suggested in the 1970s an "international trading era" (Muhly, 1973). The main way in which tin appears to be traded is through what Beale would call a "Long-distance Organized Trade" passing through the Persian Gulf. The exact origin of the tin for that trade remains obscure and will probably remain so until Afghanistan become more easily accessible to archaeologists and more is learnt about the metallurgy of northern Afghanistan and Bactria but also of sites of the Helmand tradition. Mesopotamia was the ending point of this long-distance route, but once it had reached southern Mesopotamia, the tin was redistributed, as texts from the early 2nd millennium BC suggest, to sites of northern Mesopotamia and Syria via "Regional Organized Trade". We would like to add that it was also potentially redistributed to the east towards Elam and Luristan via a "Local Redistributive Trade" following Beale's typology, perhaps already in an alloyed form, where it kick-started the Iranian bronze industry in the first half of the 3rd millennium having started somewhat earlier for some types of objects (notably vessels) in Mesopotamia. The presence of a very small number of bronze objects in eastern Iran might be the result of a "Trickle Trade" of bronze objects ("a large number of small transactions") from Bactria, Helmand and Indus Valley sites or from western Iran.

Towards the end of this period the Luristan bronze industry had gained in importance and it is possible that Luristan started trading tin or perhaps bronze to Mesopotamia while the long-distance trade was still active.

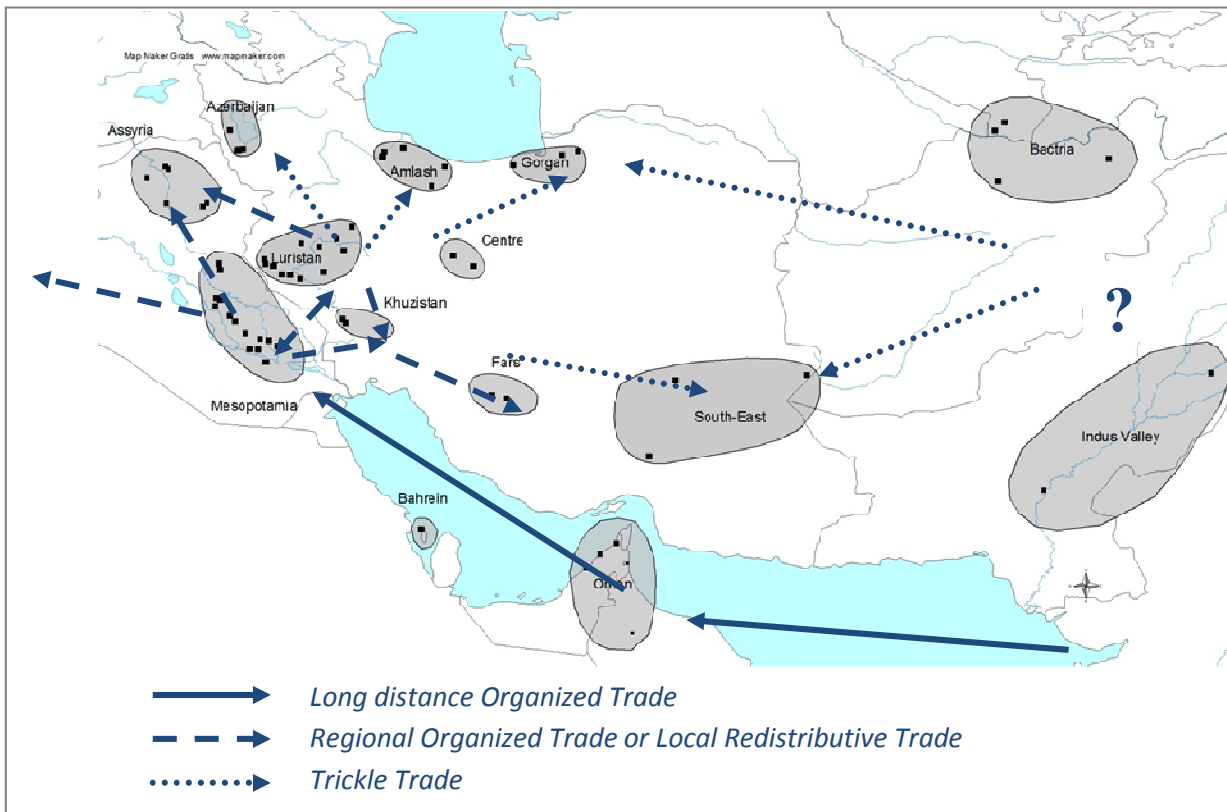


Figure IV-32. Possible tin trade routes in Period 1 (early 3rd to mid 2nd millennium BC)

### Period 2: Late 2nd – mid 1st millennium BC (Figure IV-33)

Towards the mid 2nd millennium the pattern described above started to change: the long-distance trade faded and Mesopotamia was no longer the final destination of tin trading routes. Oman seemed to still have access to tin however, at least from time to time; it is therefore possible that some form of regional trade continued to a smaller scale between Oman and the Indus Valley. We can envisage that the Indus Valley traded tin to Oman in exchange for Omani copper.

In this second period, western Iran gained in importance and Luristan, Azerbaijan and Amlash appear to be regions of bronze production. We know of the mine of Deh Hosein in eastern Luristan, but it is possible that other such mines were active in north-western Iran in that period. We suggest that tin was traded through the Zagros into Assyria, and that tin and probably bronze were traded from north-western Iran to Mesopotamia either directly or via Elam when the trade through the Zagros became difficult for political reasons. We have seen that it is also likely that Mesopotamia would have practiced recycling and mixed bronze with unalloyed copper to produce low tin-bronze objects.

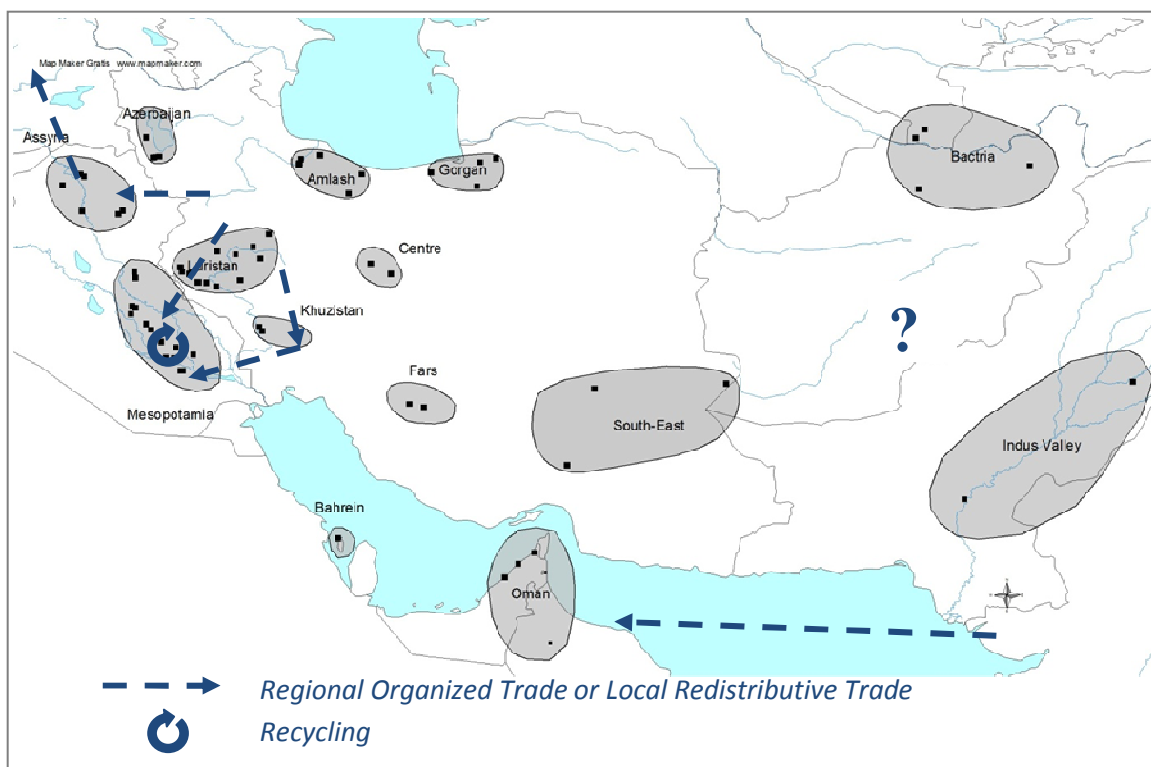


Figure IV-33. Possible tin trade routes in Period 2 (late 2nd to mid 1st millennium BC)

This shift in emphasis of the tin trade is likely to be linked to the changes that occurred during mid 2nd millennium BC and that are so far not well explained as we have seen in Chapter 1. In eastern Iran, Bactria, the Indus-Valley and Luristan, the density of settlements (or at least their archaeological visibility) decreased due to a return to a

more nomadic life style. While tin is still mined in great quantities and indeed available in Iran in great quantities, the dissolution of supra regional structures made it less accessible to Mesopotamia.

Of course, the picture presented above remains very basic and will no doubt have to be corrected when more data becomes available. Our idea of the provenance of tin has indeed changed significantly since Cleuziou and Berthoud's publication of tin deposits in Afghanistan in 1982 (Cleuziou & Berthoud, 1982). Back then, the scarcity of known deposits elsewhere in the Near-East and the correspondence of this source with texts mentioning tin coming from the east, led to a general acceptance that Afghanistan supplied tin for most of the ancient Near-East. In the following 30 years however significant contributions have been made to the subject with the discoveries of the Kestel, Karnab-Mushiston and Deh Hosein mines, and the model of a unique source can thus no longer be accepted. There is no doubt that more discoveries will follow providing evidence for yet more possible tin sources. Indeed, Nezafati et al. reported on a prospect 12km south of Deh Hosein presenting tin and copper as well as traces of ancient mining (Nezafati et al., 2009). In another paper they mention Chah Palang: a Cu-W-Au-Sn mineralisation in central Iran also with traces of ancient mining (Nezafati et al., 2008). To our knowledge these two mineralisations have yet to be fully investigated and the dates at which they operated are still unknown, but their existence goes to show that tin mineralisations are more widespread than was believed three decades ago.

The multiplicity of tin occurrences comes with a whole series of new questions and in particular the scale of the production at these occurrences and the distances onto which their tin was traded. While we do not deny that long distance organised trade was happening in the Near-East as attested by the Mesopotamian texts and mid to late 2nd

millennium ingots found in the Mediterranean, we now believe that, superimposed to that, was a complex network of tin mining and trade originating from many occurrences in Iran, Turkey, possibly India (Kochhar et al., 1999) and, although no deposits have been confirmed to date, we could conceivably suggest the Caucasus, given the high number of tin objects from the 2nd and 1st millennium BC recovered there (Selimkhanov, 1978). How this network operated will remain unknown until more fieldwork is carried out, precise dating is established and more analytical data is obtained. Yener for example has responded to doubts about the production of tin at Kestel for more than two decades, which might be a testament that tin was indeed produced there (Weeks, 2003), but was it really an important source of tin for the whole of Anatolia? The mine probably stopped operating around the turn of the 3rd to 2nd millennium BC and in the beginning of the 2nd millennium Kanesh was getting its tin from Assyria rather than more locally from Kestel, but given the high level of complexity of the extraction of tin there, it is unsure whether Kestel was ever competitive on more than a regional scale. The yield of the mine, estimated to 115 tonnes of tin (Yener, 2009, p.146) does sound impressive but would have to be compared to the yield of other mines of the region to get a better idea of compared output. Similarly, it is undeniable that mining occurred at Deh Hosein but when exactly the mine started operating, how much tin was extracted there etc. is still unknown. Finally, the central Asian mines in Karnab and Mushiston are also said to have exceeded local needs, but was it their tin that was traded all the way to Mesopotamia, tin from yet unknown Afghan mines or Iranian tin? Although the current dating could suggest that these mines operated in chronological succession (Kestel in the 3rd millennium, the Central Asian mines from early to mid 2nd, followed by Deh Hosein in the mid 2nd to 1st millennium), it seems unlikely that this will still stand when/if new

mines are discovered and it seems more believable that tin could be obtained from several locations at one time. In a way, the questions have now shifted from “where is there tin?” to “why was one source preferred to another?” (Yener, 2009, p.144), “how far was the tin from each source used?”, “when exactly did the mines operate?” etc.

All in all, many of these questions are still very much unanswered, but the present chapter has given us a few clues to move in the right direction. In particular it has shown the prevalence of tin-bronze in north-western Iran in the late 2nd and early 1st millennium and has enabled us to suggest that local tin (whether from Deh Hosein, another mine or a combination of multiple mines) must have helped support that industry.

In the following chapters we attempt to define the trade of both copper and bronze by looking at the concentration of other elements in the objects of our database.

## Chapter V. Overview of the copper composition and lead isotope analyses

In the previous chapter we found that changes have occurred in the movement of tin around the time of the transition between the Bronze Age and the Iron Age. We have postulated changes in the scale of tin trade, going from an organized international scale to a more regional trade. In this chapter, we look at changes between these two periods for other elements that are found in the copper. First, we discuss the arsenic levels in the objects, and what they can tell us about the production processes and level of control and intention exercised by ancient metal-worker. Then we present a quick summary of the levels of other elements (lead and zinc, nickel, antimony, silver and cobalt) and how they evolved over time. Finally, we look at general trends in the lead isotope data. This section acts as introduction to Chapter 6 and shows the need for a different approach in the study of our large datasets.

### 1. Correction of the composition to take into account the deliberate addition of tin

In this chapter, we deal with the concentration in elements that are found in the copper before it is alloyed with tin. In order to be able to make observations on these elements independently of the variations in tin concentration, we have corrected the measured values for the addition of tin so that, after correction, the weight percentages of all elements apart from tin add up to 100%. To that end, the following formula has been used:

$$Percentage_{corrected} = 100 \times \frac{Percentage_{measured}}{100 - Percentage_{tin}}$$

What this formula essentially represents is that all the compositions are re-normalised to (100-Sn).

Indeed, if for instance a copper of constant composition was used through time in Iran, the progressive increase in the number of tin bronzes observed in Chapter 4 would mean that we would artificially observe an overall decrease in the average measured weight percentage of all other elements. The use of corrected values means that we do not have to worry about such artificial variations. Although it is not mentioned systematically, it is this corrected value that is used in all the graphs of this chapter and the next and for the assignment of objects to metal groups in the following chapter.

A limitation of this method is that it assumes that only tin is deliberately added as an alloying element and that this tin is “pure”. As explained below, lead and zinc were also sometimes used as alloying elements. However, because the proportion of objects alloyed with tin or zinc remained relatively low and more importantly because it doesn’t show any drastic fluctuations over time, it was decided for the sake of simplicity to only correct values for the addition of tin. The case of the use of arsenic as an alloying element is a more complex problem. As detailed in the following section, it is extremely difficult to distinguish arsenic that came from the copper ore from arsenic that might have been deliberately added at a later stage. For this reason we are unable to correct the composition for the addition of arsenic.

The second point, regarding the purity of the tin, can be addressed by looking at the few analyses of tin ingots known to date. In 1977 Maddin presented the results of the chemical analysis of two of the Haifa tin ingots (Maddin et al., 1977). These were found to be of relatively pure tin, with a high magnesium component probably coming from the sea water. Later, five ingots from the Hishuley Carmel shipwreck were analysed by different

laboratories and also found to be of relatively pure copper. The most common impurity was iron between 62 and 3040 ppm, with the other impurities generally not above 100 ppm. It must be noted that lead was very low and only reached 19 ppm in one sample (Galili et al., 2013). Another 32 ingots, this time from the Uluburun shipwreck, were also analysed for their chemical content. Once again they were found to be of “surprisingly pure tin” (Hauptmann et al., 2002, p.16). Most impurities were indeed present at less than 100 ppm. If we consider a bronze made from the mixing of 90% copper with 10% of this tin, these impurities would only contribute to less than 0.001% in the copper. Given the disparities in precision and accuracy of the different methods used to analyse the copper objects studied in this thesis (see Chapter 3), such low values can be overlooked. The lead content in the Uluburun ingots was however found to be more variable than the other elements and in one case reached almost 1% Pb. In the hypothetical alloy described above this would result in a contribution of 0.1% Pb from the tin. Such a difference could start to have an influence on our interpretations, but this level of lead content seems to only occur rarely.

Very few metallic tin objects have, to our knowledge, been analysed to date. The Thermi bracelet and both of the Egyptian objects (see Chapter 2) have been described as being “pure tin” but their analyses haven’t been published (Maddin et al., 1977). The Caucasian tin objects reviewed by Selimkhanov were also generally quite pure aside from their lead content which was of the order of a few percents in some of the objects. These lead contents might however reflect a deliberate addition of lead as a few “tin-lead pewters” are also known in the region (Selimkhanov, 1978).

All in all, it is obvious that correcting for the addition of tin as described above is not a perfect method and will never reflect exactly the composition of the copper before alloying, both because of the possible adjunction of other elements and because of losses

during the alloying process. However, it is a good way of filtering out the most important fluctuations due to the addition of tin and in particular to prevent us from observing an artificial decrease in the other elements when tin-bronze becomes more commonly used.

## 2. The arsenic content

### a. Properties of arsenical copper

As we have seen in Chapter 2, copper with elevated amounts of arsenic occurs frequently during the Bronze Age in the Near East. This has spurred numerous discussions about the reasons for the occurrence of such a composition, the level of intention going into its production and whether or not we can truly speak of an arsenic-copper “alloy”. In order to start answering some of these questions, Northover (1989) has reviewed the properties of arsenical coppers (see also Moorey 1994, p.250). As with tin, an addition of arsenic into copper will improve its hardness and strength. It will also improve the increase in hardness and strength obtained when an object is cold-worked (Figure V-1). Unfortunately, the limited amount of metallographic investigation carried out on Near Eastern objects means that we can not assess the amount of cold-working to which the objects have been subjected and therefore we are not able to know whether the ancient metal workers would have taken advantage of this property. Under 2% of arsenic, these effects are minimal and it is only with relatively high arsenic contents (4% and over) that Northover noticed true mechanical improvements over pure copper. Such contents can also result in a surface enrichment in arsenic, giving the objects a silver colour that would tarnish into a golden appearance (Northover, 1989, p.115). Yet lower contents of about 2 to 4% As could also have an advantage on pure copper as they improve its castability by reducing the melting point and acting as a deoxidant (Craddock, 1995, p.291), thus giving the objects a better finish (Northover, 1989, p.117).

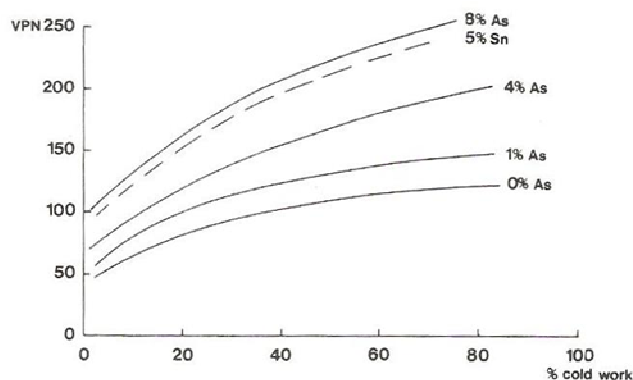


Figure V-1. Variation of hardness with cold work for Cu-As alloys (Northover, 1989, p.113)

Lechtman studied the properties of arsenical copper as compared to tin bronze. She carried out a number of mechanical tests on both types of alloys and found that while the tin bronze samples could be work-hardened more easily than the arsenical copper ones, arsenical copper was more ductile, a property which is interesting for the production of thin sheets of metals as in vessels for example (Lechtman, 1996).

Despite the relatively well understood properties of arsenical copper, it is difficult to assess the actual properties of the Near Eastern objects considered in this thesis. Indeed arsenic is not present homogeneously in the objects. Northover estimated that up to 25% of the arsenic in an object can be isolated in cuprous and arsenious oxides ( $\text{Cu}_2\text{O}$  and  $\text{As}_2\text{O}_3$ ) rather than being present in solid solution. Another cause of inhomogeneity is the phenomenon of inverse segregation that gives AsCu objects their characteristic silvery colour but also means that surface readings of the As content will give much higher results than an analysis of the core of the objects.

Given these limitations, it is difficult to determine which properties, if any, were actively sought after by the ancient metalworkers, leaving the question of whether or not the production of such alloys were intentional or accidental mostly open. We have already discussed in Chapter 2 the different processes by which arsenical copper can be produced.

Weeks suggested that the processes which involve the addition of arsenic metal or arsenic-bearing minerals to molten copper at a relatively late stage of the production suggest an intentional addition of arsenic, whereas the smelting of a charge containing both copper ores and arsenic bearing ores could result of both intentional or accidental production of arsenical coppers (Weeks, 2003, p.113). The production of arsenical copper via the addition of speiss to copper ore or copper metal as described by Rehren et al. for the site of Arisman (Rehren et al., 2012) fits into the first type of processes. It points towards an intentional production with a certain level of control of the process at this site, and most likely at others, including the sites of Shahr-i Sokhta and Tepe Hissar where speiss was also recovered (see Chapter 2). However, since many different types of copper and arsenic ores would have been available to the ancient metal-workers of Iran and the neighbouring areas, many different processes might have been used simultaneously throughout the Bronze Age, and it is impossible to generalise this type of intentional production to the whole of our area of interest.

With those considerations and limitations in mind, the following paragraph presents an overview of the arsenic content of the objects present in our database and its evolution over time, especially around the turn from the Bronze Age to the Iron Age.

#### b. Chronological analysis

Figure V-2 presents the chronological evolution of the arsenic content in objects from Iran, Mesopotamia and Oman. The graphs obtained for Iran and Mesopotamia are strikingly similar. In these two areas, the maximum arsenic content was reached in the early 3rd millennium BC with median contents of respectively approximately 1.8% and 1.4% As. Throughout the 3rd millennium and the first half of the 2nd millennium, the range of

arsenic contents became smaller and the median percentage gradually decreased. By the late 2nd millennium, the arsenic content reached a minimum that would stay more or less constant during the beginning of the 1st millennium BC. By then, almost all objects presented less than 1% As and median levels were under 0.5% As.

In Oman, for the periods for which we do have some information, the pattern is not dissimilar: from the mid 3rd millennium onward, the range of arsenic contents becomes smaller and the median levels of arsenic decrease. However it seems that in Oman, this decrease happens slightly quicker than in the countries discussed above with median levels of under 0.5% as soon as the late 3rd millennium BC.

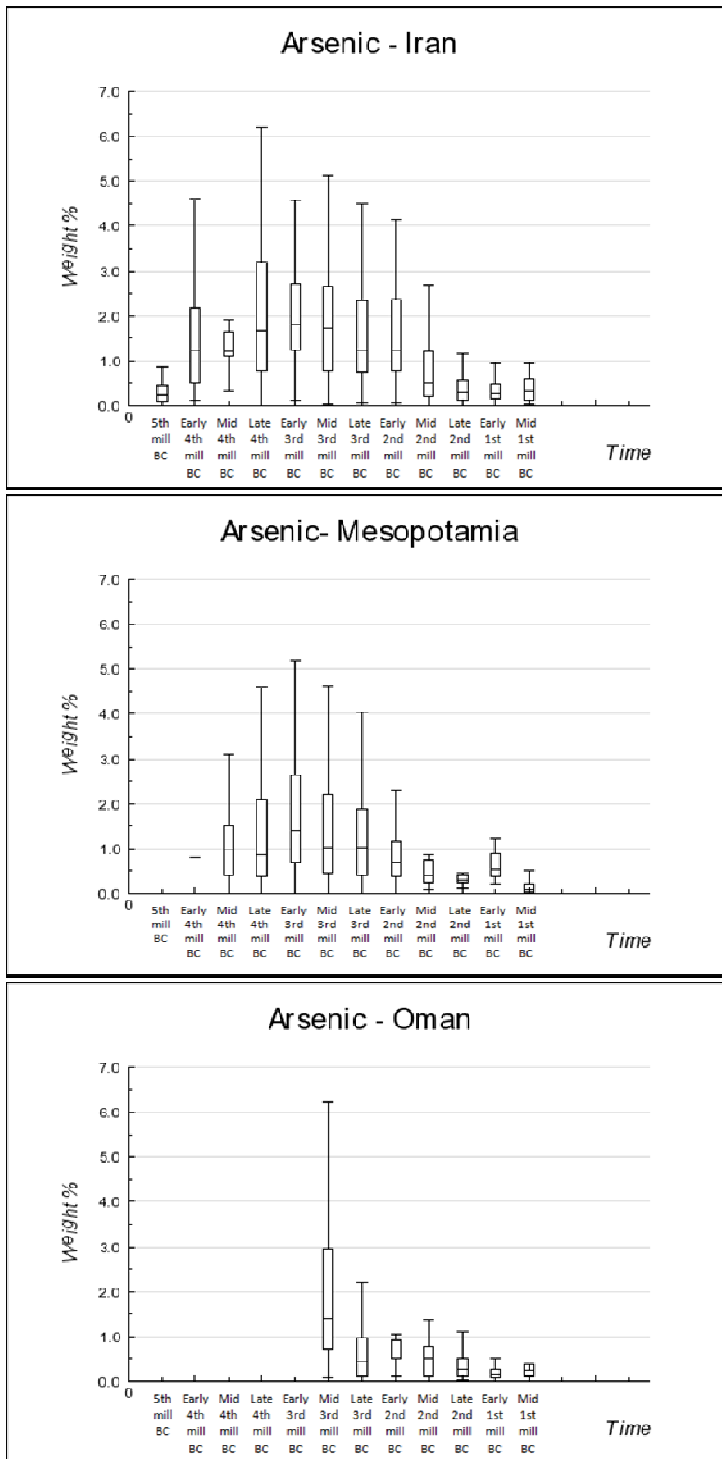


Figure V-2. Evolution of the arsenic content in copper-based objects in Iran, Mesopotamia and Oman

Regardless of the process of production of arsenical copper, the volatility of arsenic meant that it would have been very difficult for the ancient metallurgist to control the exact quantity of arsenic present in an object. It is therefore clear that the decrease in arsenic

content observed here does not reflect a will of the metalworkers to produce alloys with on average say 0.2% less arsenic than in the previous period.

In Iran, the decrease in arsenic content from the 3rd millennium onwards and its stabilisation during the Iron Age (Figure V-3) mirrors closely the increase in the ubiquity of tin bronzes present in the assemblage (Figure IV-2). The decrease in arsenic could then reflect the increase in the number of tin-bronzes if these have a lower arsenic content.

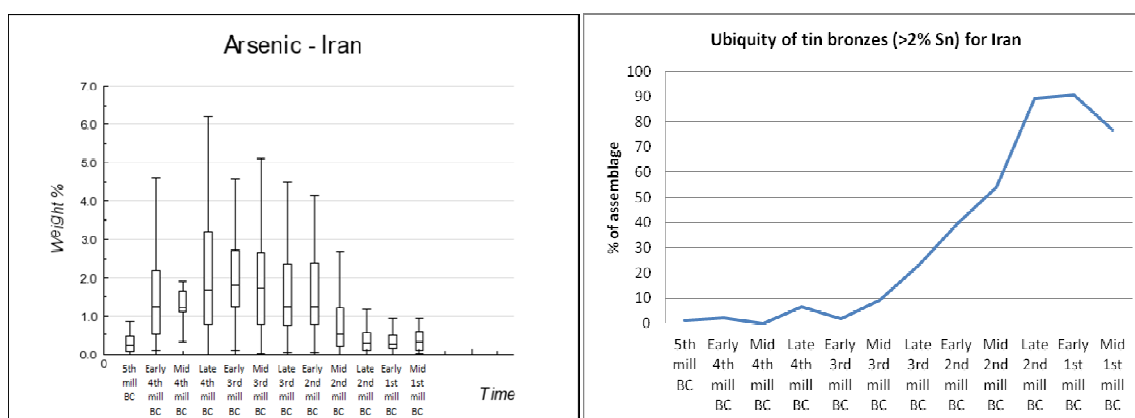


Figure V-3. Evolution of the arsenic content and of the ubiquity of tin bronzes in the copper-based metal assemblage in Iran

- c. Inverse correlation between tin and arsenic and its contribution to the decrease in arsenic levels

We have mentioned in previous chapters that some of the early tin bronzes, in Luristan and Elam in particular, present simultaneously over 2% Sn and over 1% As. Figure V-4 shows the distribution of the arsenic content in tin-less copper (<2% Sn), mid-range bronzes (2-6% Sn) and high tin bronzes (>6% Sn) and confirms the presence of objects with both As and Sn. However, we can see that in general tin bronzes contained less arsenic than tin-less copper objects. This trend is observed not only for high percentages of tin, but also for the mid-range tin bronzes. As we have mentioned above, the values used in these graphs for the arsenic content have been corrected to take into account the addition of tin, meaning

that such a pattern is not just a “mathematical” artefact resulting from a lower overall content in all elements other than tin.

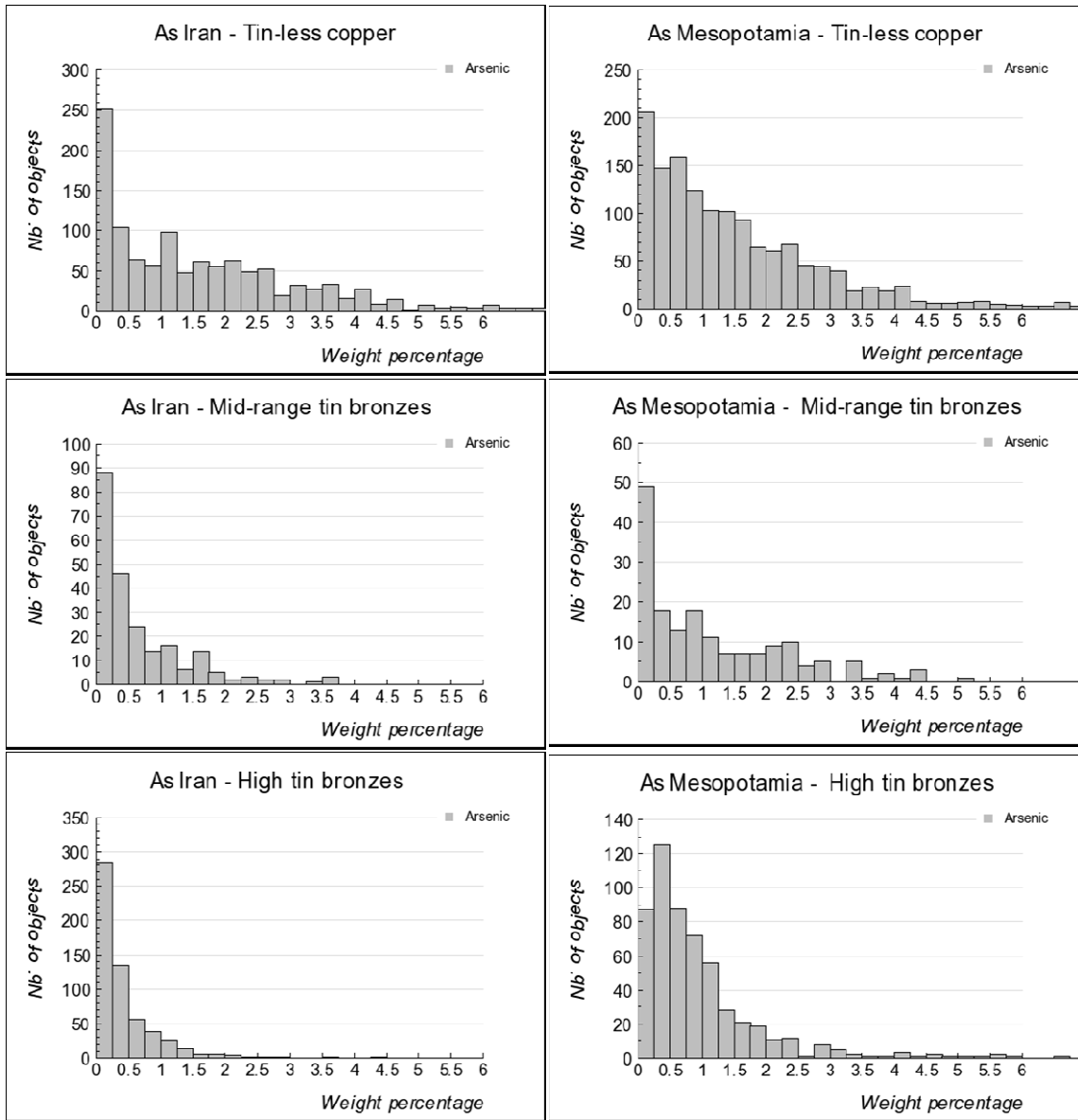


Figure V-4. Distribution of the arsenic content in tin-less copper (<2% Sn), mid-range bronze (2-6% Sn) and high tin bronze (>6% Sn) in Iran (left) and Mesopotamia (right)

The inverse correlation between the presence of tin and the presence of arsenic observed here follows a pattern that has been noticed elsewhere and abundantly commented on. In 1967, Charles concluded that such a separation indicates an intentional production of arsenical copper: the ancient metal smith would have either been able to differentiate

arsenical copper from pure copper and selected only pure copper for the addition of tin, or would have created either arsenical copper or tin bronzes by the addition of one element or the other to pure copper (Charles, 1967). This indicates a certain level of understanding of the properties of these two alloys. Indeed since both elements have similar types of effects on copper (increased hardness, increased response to cold-working and better castability), a metal-worker who understood these properties would have understood that there was little advantage to produce an alloy with elevated levels of both.

Another potential explanation for the inverse correlation between these two elements would be that the tin bronze used in Iran and Mesopotamia was directly imported from a region where arsenical copper was uncommon. This possibility is investigated in Chapter 6.

A third point that probably contributed to the lower levels of arsenic present in tin-bronzes comes from the production process of tin-bronze itself. Indeed, unless copper ores and tin ores are co-smelted (see Chapter 2), the addition of tin requires for copper to be melted (either for the addition of tin metal or for the reduction of cassiterite). The melting of the copper would have resulted in an oxidative loss of arsenic. As explained in Chapter 2, for the reduction of cassiterite in contact with molten copper, the final tin content would have been dependant on the amount of time for which the copper was melted. Therefore, in this case, the higher the tin content, the lower the arsenic content.

However even if there is a correlation between high levels of tin and low levels of arsenic, Figure V-4 shows that the number of objects with over 2% of tin and arsenic levels over 0.25% is not insignificant. Moreover Figure V-5 shows that the reduction of the arsenic range and the decrease of the median arsenic level can be observed even when plotting only objects with less than 2% tin, not particularly in Iran, but certainly in Mesopotamia

and Oman. Therefore, the inverse correlation between tin-bronze and arsenical copper is not enough to explain entirely the diminution of arsenic through time.

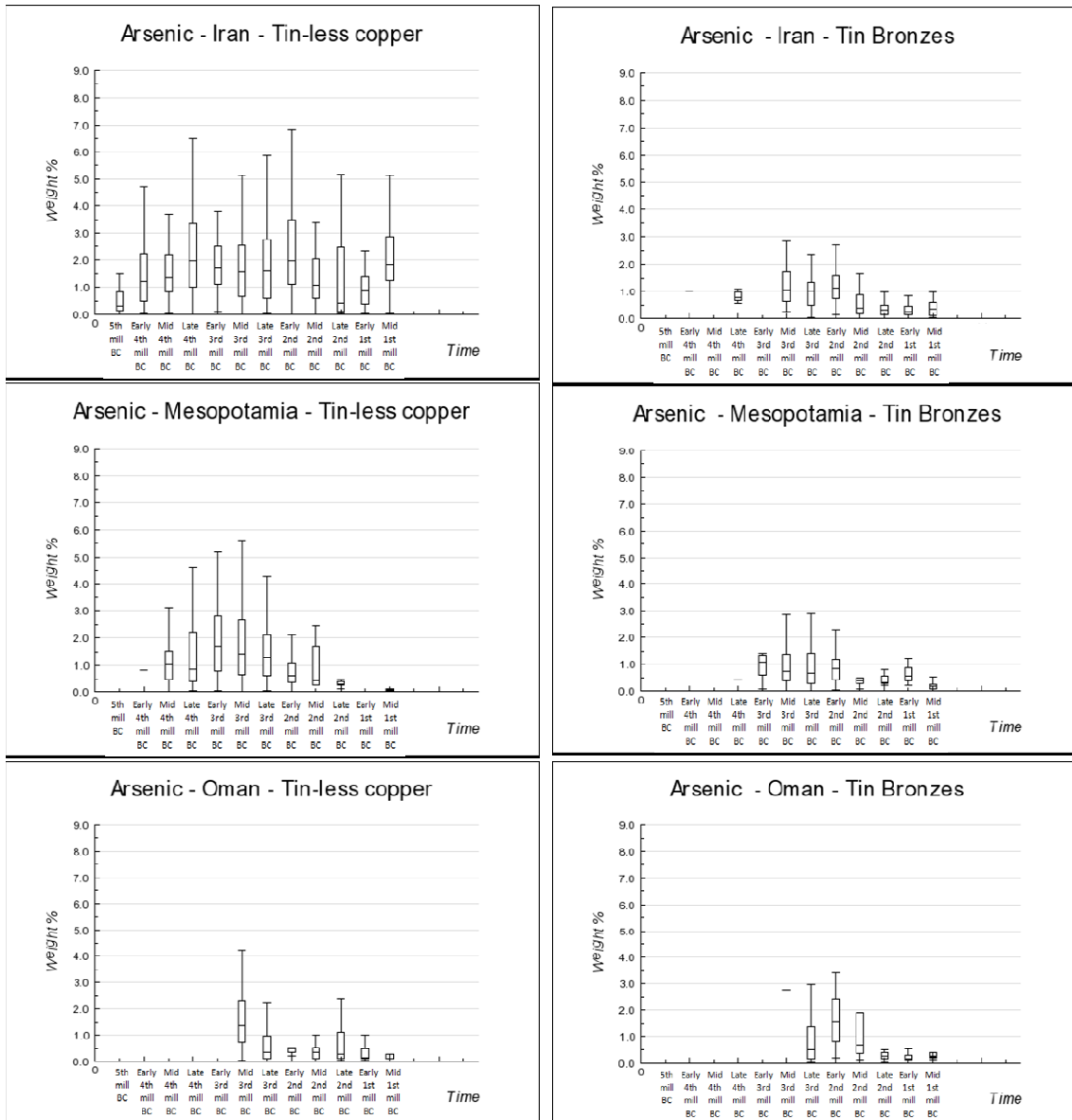


Figure V-5. Evolution of the arsenic content in Iran, Mesopotamia and Oman for tin-less copper objects (<2% Sn, left) and tin bronzes (>2% Sn, right)

It could be argued that this is only due to the fact that the number of tin-less copper objects analysed decreases over time since in Iran, the majority of objects becomes alloyed with bronze and in Mesopotamia we simply do not have as many analyses for later periods (see Figure V-6). Given the shape of the distribution of arsenic content, a diminution of the sample size would mean that the objects with high arsenic content, as they are less

common, are less likely to have been analysed. This would result not only in a reduction of the range of arsenic contents but also an artificial decrease of the median.

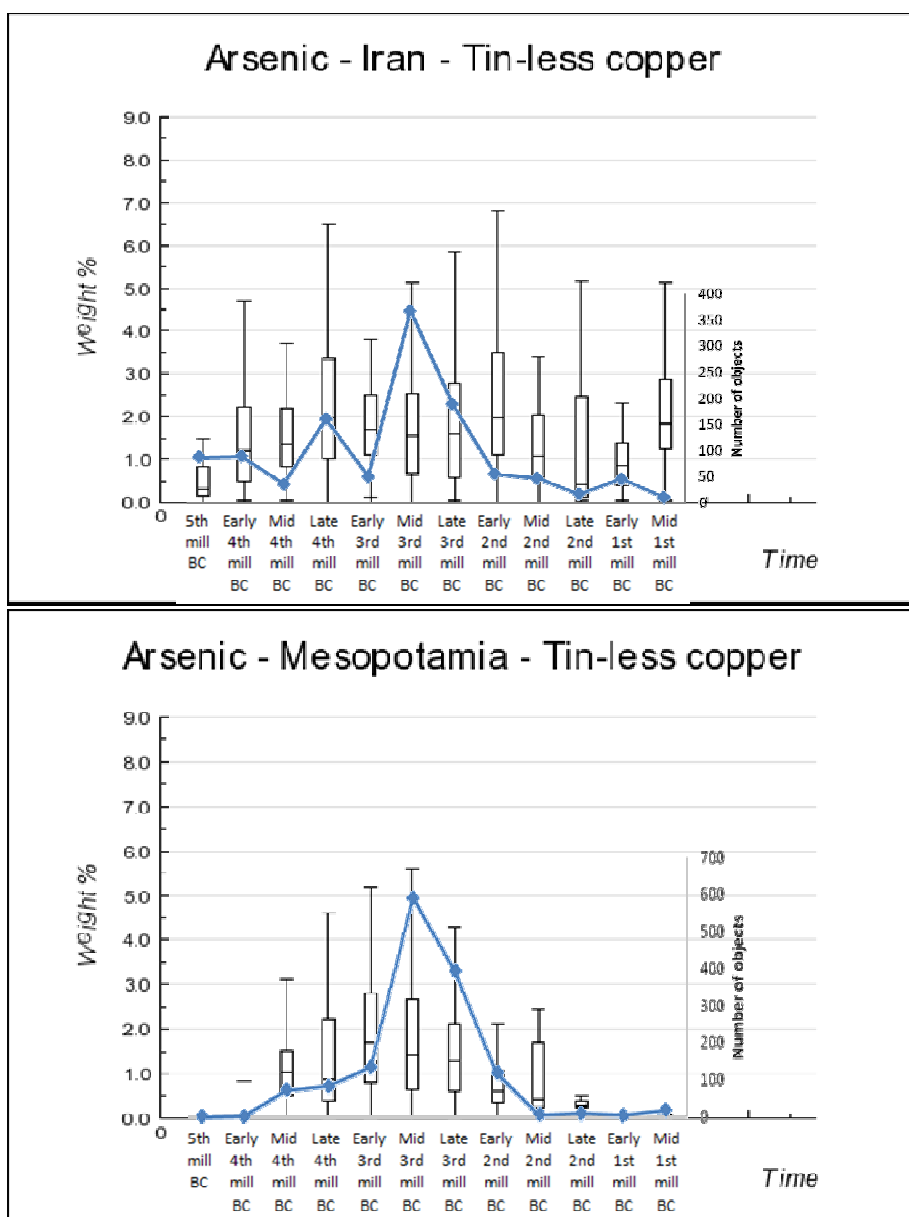
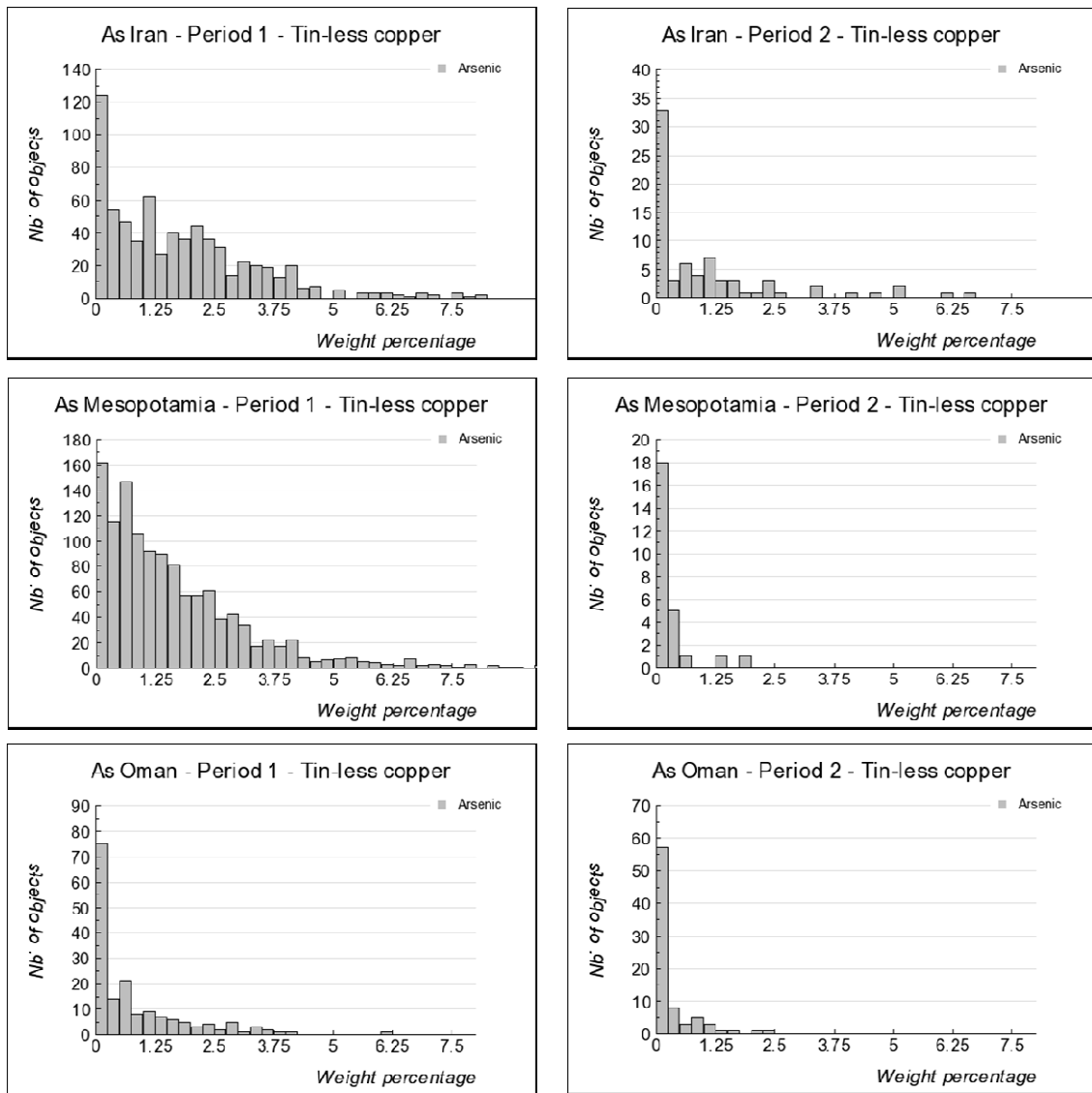


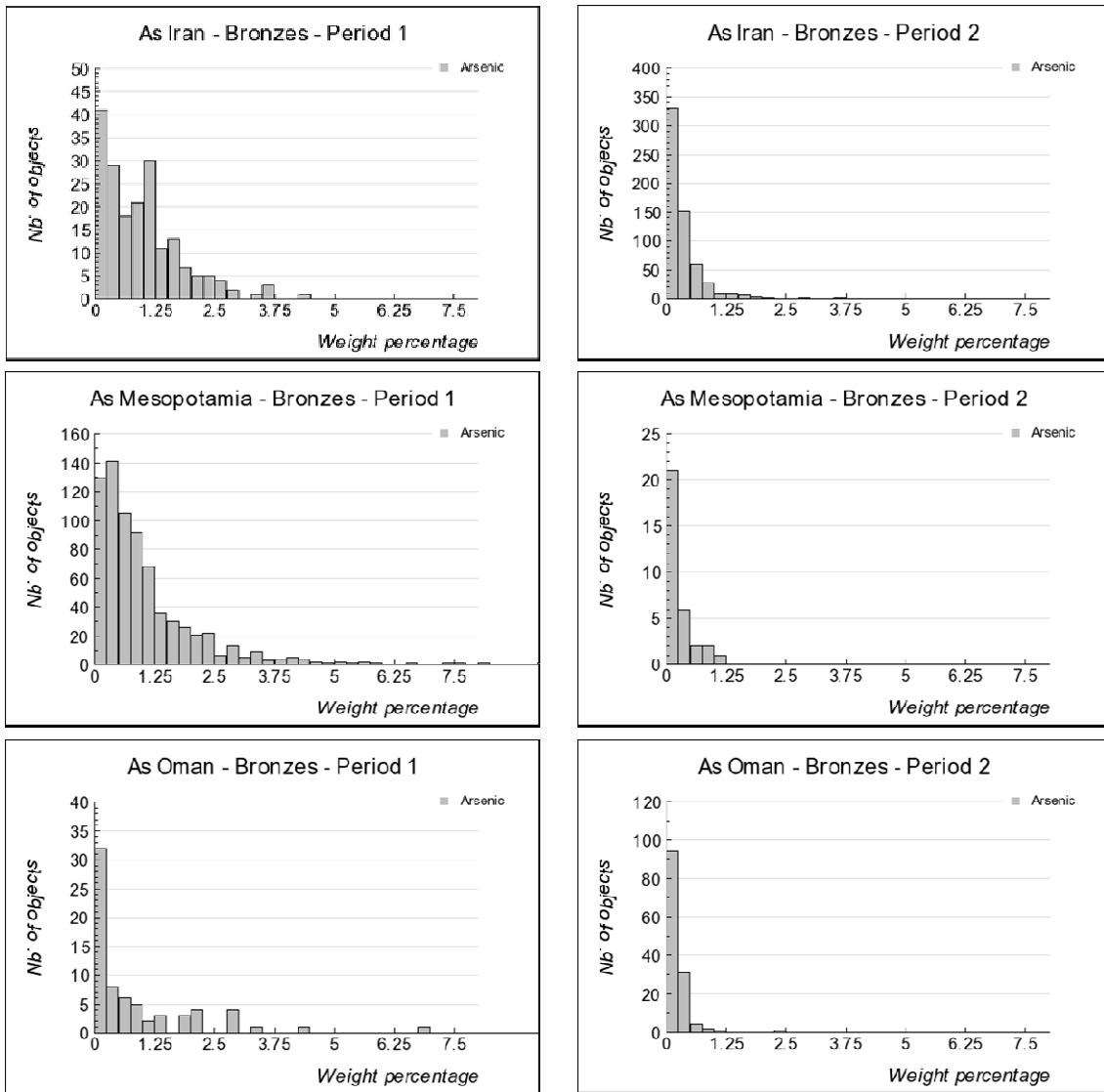
Figure V-6. Evolution of the arsenic content in tin-less copper objects (<2% Sn) and number of tin-less copper objects analysed for each period in Iran (top) and Mesopotamia (bottom)

However Figure V-7 clearly shows that in Iran and Mesopotamia, the diminution of arsenic contents in copper with less than 2% tin is not simply due to a reduction of the sample size. Indeed, the shape of the distribution changes significantly between Period 1 (early 3rd to mid 2nd millennium BC) and Period 2 (late 2nd to mid 1st millennium BC).



**Figure V-7. Distribution of the arsenic content for tin-less copper objects (<2% Sn) in Period 1 (early 3rd to mid 2nd millennium BC, left) and Period 2 (late 2nd to mid 1st millennium BC, right) in Iran (top), Mesopotamia (middle) and Oman (bottom)**

The ratio of objects with under 0.25% As to objects with more that 0.25% As in particular increases significantly in Period 2. Figure V-7 also shows a striking difference between the levels of arsenic in Oman and those of Mesopotamia and Iran. There, levels of arsenic higher that 0.25% are much less common than in the other two regions, even in Period 1.



**Figure V-8.** Distribution of the arsenic content for bronze objects (>2% Sn) in Period 1 (early 3rd to mid 2nd millennium BC, left) and Period 2 (late 2nd to mid 1st millennium BC, right) In Iran (top), Mesopotamia (middle) and Oman (bottom)

Although there is undoubtedly a correlation between high levels of tin and low levels of arsenic, we have seen that there are still a certain number of objects with elevated levels of both. Interestingly, the drop in arsenic levels observed between Period 1 and Period 2 in tin-less copper can also be observed in tin bronzes (Figure V-5). In Iran the median levels drop from around 1% of arsenic in the 3rd and early 2nd millennium to under 0.5% in the Iron Age and similar changes are observed in Mesopotamia and Oman. This pattern is confirmed by Figure V-8. We can see that the arsenic content of bronzes in Period 1 ranged between 0 and 5% approximately, while in the Iron Age there were only very rare

cases in which the arsenic content was greater than 1%. Again, this figure also shows lower levels of arsenic in Oman than in Mesopotamia or Iran in Period 1, with most objects having less than 0.5% As, but the range of arsenic contents is still slightly smaller in Period 2.

#### d. Object types

Not only is this drop observed for tin-bronzes as well as tin-less copper, but it is also observed for all types of objects. Figure V-9 and Figure V-10 show for Period 1 and Period 2, the distribution of arsenic content in different types of objects.

In Period 1, most object types present a similar distribution of arsenic contents: apart from a small number of outliers, the majority of objects fall within the 0 to 5% As range, with more objects towards 0% than towards 5%. Notable exceptions are arrowheads and spearheads, pins and to a lesser degree bracelets and rings. For these three types, objects still mostly fall in the 0-5% As range, but the proportion of objects with under 0.25% arsenic is higher. This distinction, perhaps more than the negative correlation between tin and arsenic levels, may denote some sort of intention behind the production or choice of arsenical copper. The fact that pins and jewellery are less often made of arsenical copper, than weapons (aside from projectiles) and tools might indicate an understanding of the increased hardness and strength that arsenic would have provided.

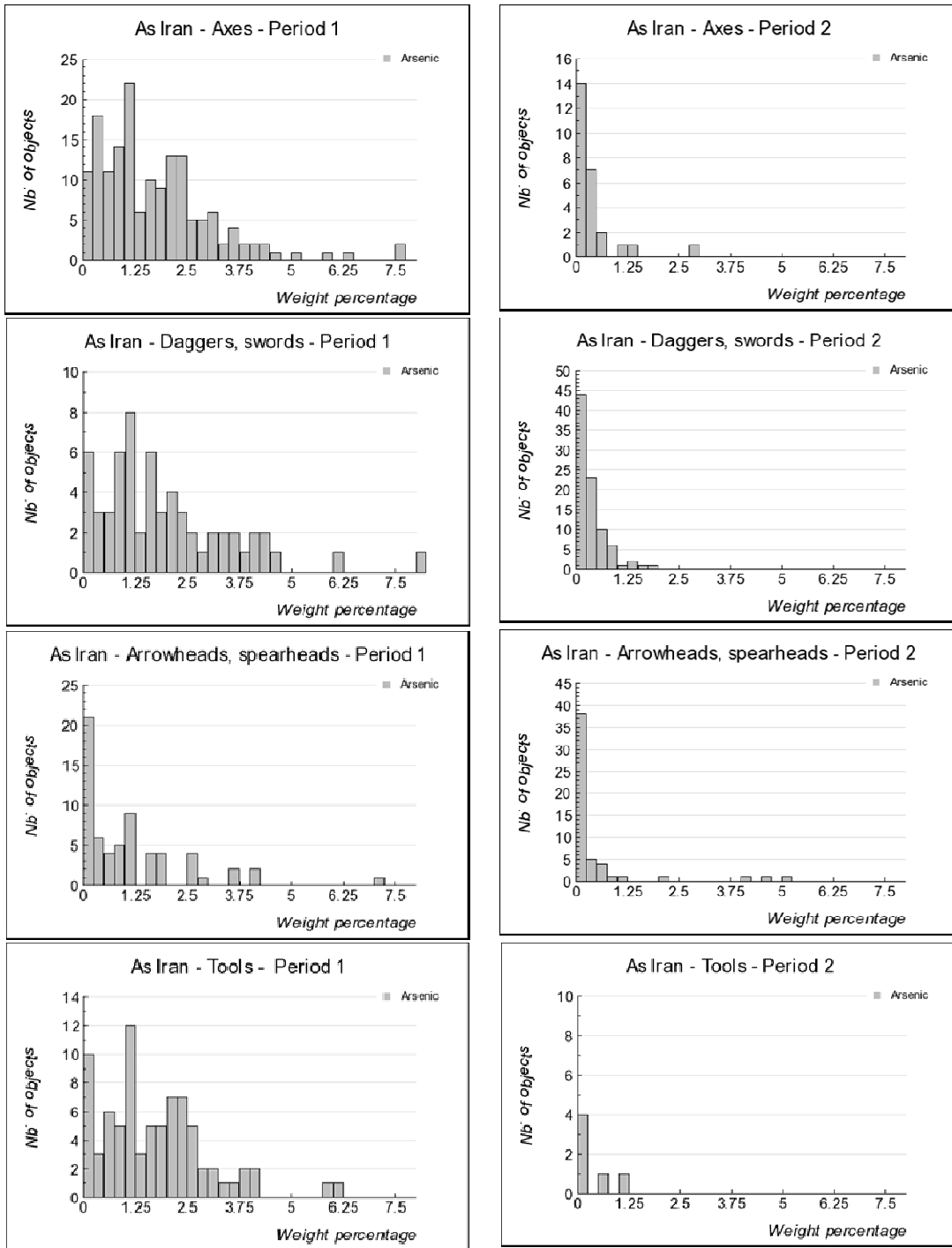
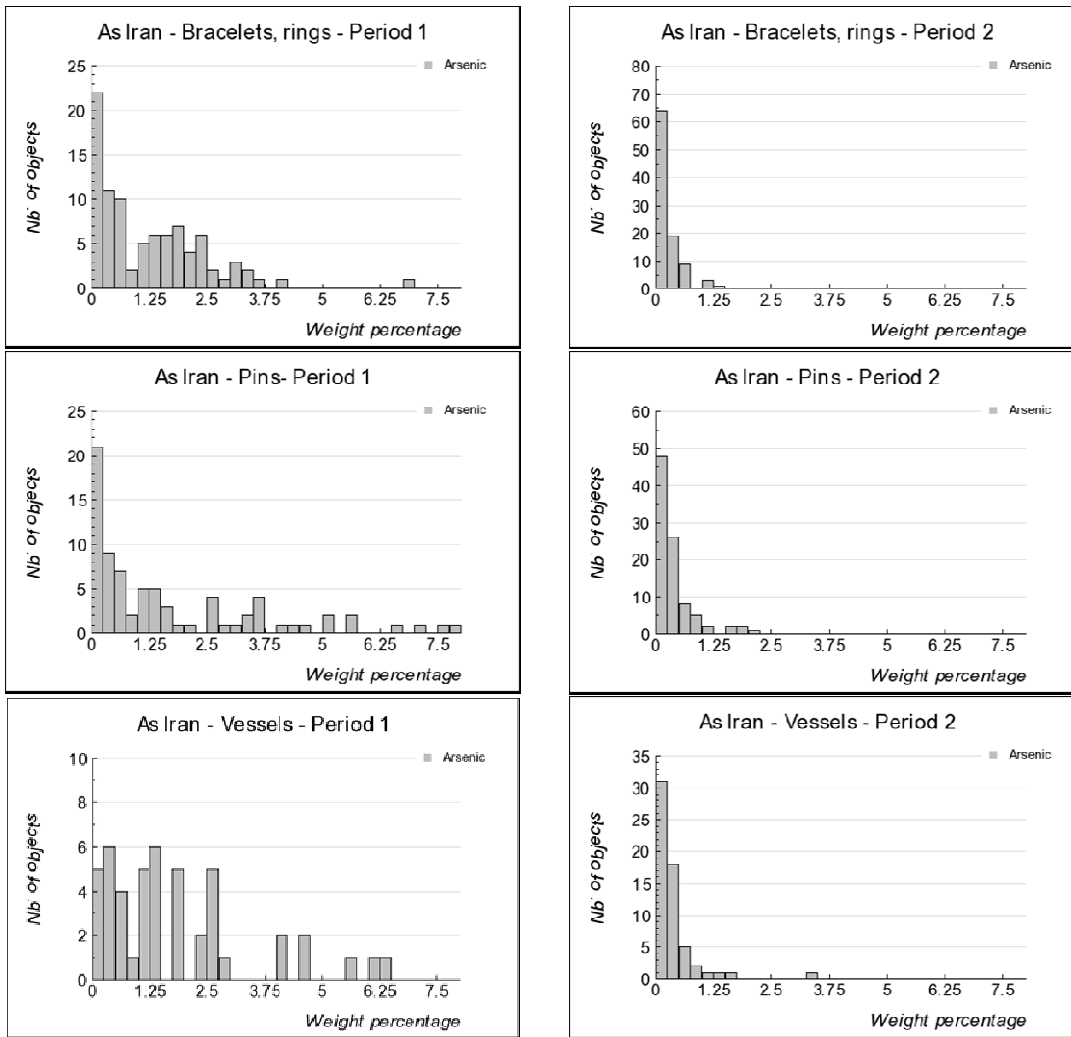


Figure V-9. Distribution of the arsenic content for weapons and tools in Period 1 (early 3rd to mid 2nd millennium BC, left) and Period 2 (late 2nd to mid 1st millennium BC, right)



**Figure V-10. Distribution of the arsenic content for ornaments and vessels in Period 1 (early 3rd to mid 2nd millennium BC, left) and Period 2 (late 2nd to mid 1st millennium BC, right)**

The small number of vessels analysed from Period 1 seems to indicate that arsenical copper was used for their production. Neither hardness and strength, nor increased castability are likely to have been the motivation behind these choices given that most of the vessels were made of sheet metal. It is therefore possible that arsenic was in this case added to give the metal a silvery colour. However the levels of arsenic observed here (under 3% As for all but six of the objects) are probably too low for the surface enrichment in arsenic to make a great difference on the colour of the object (Northover, 1989, p.117).

The particular composition of arrowheads and spearheads, with lower arsenic levels than other weapons can either be explained by their production process (for example casting in an open mould would have resulted in a loss of arsenic by oxidation) or in the same context as the low levels of tin or the late adoption of iron for these projectiles (see Chapter 4). If the latter explanation is valid and they were indeed produced in unalloyed copper because it was better suited to a mass production and a possible loss of many arrows and lances, then this gives us an indication of the status of arsenical copper and the intention behind its production. As summarized by Weeks, the debate on the “intentional” production of arsenical copper is made difficult by the fact that the presence of arsenic in an object can come from choices (deliberate or not) made at many different stages of the production (Weeks, 2003, p.117). It can come from the selective mining of specific ores, the choice of specific smelting and casting conditions, the addition of arsenic or arsenic ores to molten copper after the smelting of copper itself, or even the selection of a certain batch of metal over another after their production. The projectiles in our database could have been made either of arsenical copper that lost its arsenic during the casting, or of freshly smelted unalloyed copper or recycled scrap metal depleted in arsenic. The second and third possibilities indicate that the metal-workers would have been able to produce objects of distinct compositions for distinct purposes or to recognise distinct compositions and use them to different ends even if they didn’t control their production. Unfortunately, we do not have enough information on the typology and production processes of our objects to conduct more thorough assessment of this question.

Moreover, the comparison of different types for all regions taken as a whole, as attempted above, relies on the assumption that, from one type of object to another, the contribution of the different regions (likely to use metal of different compositions) is more or less equal. While this is true for most type of objects here (with the exception of vessels coming only

from Luristan and Khuzistan and perhaps explaining the high proportion of arsenic-rich objects), we can see the limitations of such a generic approach and the need for a more detailed assessment of different metal groups as presented in Chapter 6. What we can say however, going back to Figure V-9 and Figure V-10, is that in Period 2 for all types of objects without exception, the range of arsenic contents is much smaller. Virtually all of the objects fall between 0 and 2% As and a great majority present under 0.5% As. All in all we have observed for both Iran and Mesopotamia a significant reduction of the range of arsenic content and an increase in the proportion of objects with very low arsenic contents between Period 1 and Period 2 for all types of objects and all levels of tin. It might be the case that a similar trend also occurred in Oman, but it is less significant due to the low levels of arsenic observed in Period 1 already. Unfortunately the lack of data for the Iron Age in Bactria and in the Indus Valley makes it impossible for us to determine exactly the extent of this phenomenon.

e. Causes for the decrease in arsenic content

Several possible causes can contribute to such a global pattern. The first one is a progressive abandonment of the production of arsenical copper, whether this was due to a change in the access to arsenic bearing ores, a change in metalworking traditions or in the taste of the people for whom the objects were made, or whether it was due to a conscious choice to use less arsenic perhaps because of its impact on health for example. Indeed we can envisage that since, aside from their colour and ductility, tin bronze and arsenical copper were able to offer a similar type of improvement on pure copper, with tin bronze however improving the hardness to a higher degree and in a more controllable fashion (depending on how tin was added), the need for the production of arsenical copper

diminished as tin bronze became more common. With respect to a potential change in access to ores, we can envisage either a change in the geographical provenance of the ores (say for example a change in the supply reaching Mesopotamia from Omani ores to Iranian ores) that could be due to political reasons like the control of trade routes, or the opening or closure of mines within the same region or even mining area. This second possibility has been suggested notably in Oman. The lower levels of arsenic and nickel observed in the Iron Age compared to the Umm al-Nar period and some archaeological evidence on the location of Iron Age extraction sites have led to the suggestion that there was a shift from the exploitation of the small oxidized copper deposits rich in As, Ni and Co to the exploitation of the massive sulfide deposits poor in As and Ni also found in Oman (Weeks, 2003, pp.118-19). Weeks however advises caution with regard to this conclusion as many other deposits in the Near East and in particular on the Iranian plateau present levels of arsenic and nickel that could also have been used for the production of the Omani Umm al-Nar objects (Weeks, 2003, p.127).

A second cause for the generalized drop in arsenic levels observed in the Iron Age probably has to do with the oxidative loss of arsenic that occurs when the copper is melted. Each successive cycle of re-melting and re-casting of copper-based objects would have resulted in an impoverished arsenic content meaning that the overall arsenic levels of the metal in circulation would have decreased over time (Bray, 2009). This aspect is examined in detail in Chapter 6.

#### f. Regional specificities

Of course, the global picture presented above is blurred by the fact that Iran is presented as a whole even though we have seen in the previous chapters that different regions, and east

and west in particular had very different metal-working traditions. However, this has provided starting point for reflection and emphasized the generalised drop in arsenic levels between Period 1 and Period 2. Regional specificities are studied in more details in Chapter 6, and here we will simply make a few remarks, going back to the question of the intention and level of control of alloying with arsenic. It has been established that noticeable improvements over unalloyed copper occur with content of about 2% As and over and affect mostly the castability of the metal. We would then expect in a region that uses arsenic in a controlled fashion with these improvements as a target to see, on top of a peak of very low arsenic contents representing unalloyed objects, a distribution of arsenic contents approaching a gaussian shape centred on a concentration greater than 2%. This is indeed what we find in Luristan and possibly in Bactria for the tin-less copper objects of Period 1 (see Figure V-11, top).

In Oman, the South-East and Gorgan, objects with significant amounts of arsenic are also common; however the distribution in these regions shows no peak other than the one representing unalloyed objects (Figure V-11). This suggests a lack of control over the arsenic content. Given that, and the great number of arsenic-free objects, it is tempting to interpret these distributions as the result of the use of a mixture of ores, some arsenic free and some containing arsenic. It is worth noting that in these regions, there is evidence of the use of slagging processes. The arsenic-bearing objects may therefore have been produced by the direct smelting of copper sulpharsenides, but this is not very likely given that fahlerz is quite rare in this region and given the example of Shahr-i Sokhta where matte was discarded rather than further processed (Hauptmann et al., 2003). Another more plausible possibility is the mixed-smelting of copper oxides and sulfarsenides, which is consistent with the variable arsenic content observed above. This is not to say that the use of arsenic bearing ores was completely accidental: selective mining may have taken place,

but it would appear that contrary to the situation in Luristan and Bactria the production process wasn't controlled in order to retain a specific amount of arsenic.

A third remarkable type of distribution occurs in Assyria and Khuzistan. In these regions, we can observe a peak of arsenic concentration, but it is centred on a value lower than 2% (Figure V-11). Since there would be very little advantage to controlling the addition of arsenic to such a low concentration, these distributions can be interpreted in two ways: they could be the natural result of the smelting of arsenic bearing copper ores, such as copper arsenates for example, through availability rather than to achieve any particular properties. Alternatively, these distributions could represent an assemblage of objects made from a metal initially presenting a distribution such as the ones observed in Luristan and Bactria, but that have undergone processes that have resulted in a drop in arsenic content such as remelting that would have resulted in an oxidative loss of arsenic. We discuss this second possibility in more details in the section concerning metal flow in Chapter 6.

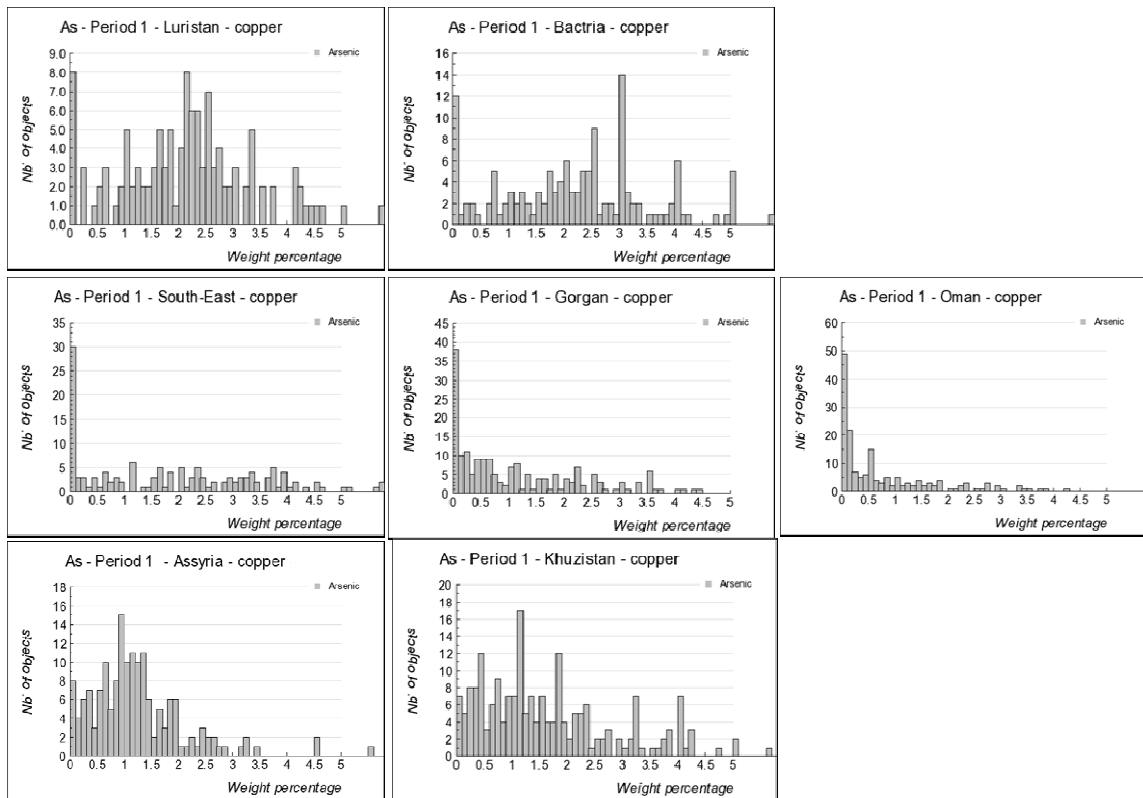


Figure V-11. Examples of different arsenic distribution profiles for tin-less copper objects (<2% Sn) in Period 1

### 3. The lead and zinc contents

Aside from tin and arsenic, lead and zinc are the other two elements that have historically been intentionally added to copper to produce alloys. As we can see on Figure V-12 the practice of alloying copper with significant amounts of either lead or zinc was on the whole relatively uncommon in our region and period of interest. Objects with significant amounts of these elements do occur but are greatly outnumbered by objects with less than 1%.

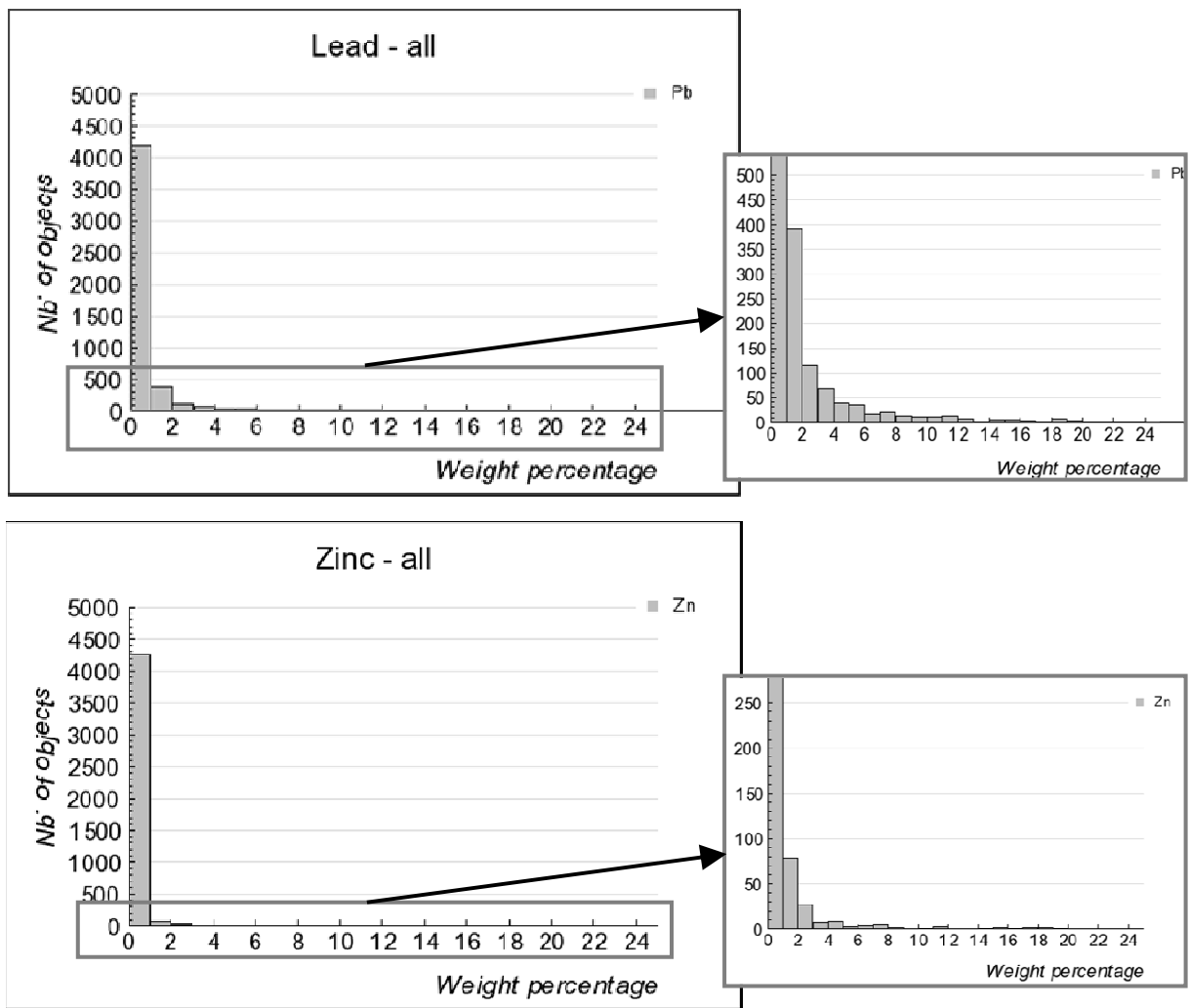


Figure V-12. Distribution of the lead content (top) and zinc content (bottom) for all of the objects of the database

a. Leaded copper

Lead is likely to have been added to copper by ancient metal-workers in order to facilitate casting in moulds of complex shapes (Moorey, 1994, p.293). As seen in Chapter 2, leaded-copper occurs from as early as the 4th millennium BC in Iran. Malfoy and Menu in particular have noted high amounts of lead in copper-based objects from Susa in the late 4th millennium BC (Malfoy & Menu, 1987). Figure V-13 shows the evolution of the ubiquity of leaded copper objects in the assemblage for Iranian regions. We have chosen here the arbitrary limit of 2% Pb to define “leaded copper”. This cut-off value is quite low and it can be argued that a higher value (e.g. 5%) would be a better indicator of intentional

alloying, but it has been chosen to reflect the shape of the distribution seen in Figure V-12. Figure V-13 shows the high ubiquity of leaded copper in Khuzistan objects in the 4th millennium BC mentioned above (around 33% of the assemblage) and also a relatively high proportion in the mid 2nd millennium (18%). Leaded copper is also very common in the mid 1st millennium BC in Luristan where it represents about half of the assemblage. This value is to be taken with caution however as it is only based on the analysis of 13 objects. In all of the other Iranian regions, leaded copper objects never represent more than 15% of the copper-based metal assemblage and of the “leaded copper” objects many have less than 5% Pb (Figure V-12) and could arguably be accidental alloys or originate from the recycling of leaded copper objects. It is interesting to note that, unlike for tin bronzes, there isn't here a clear chronological pattern for the adoption of leaded copper. Going back to the transition between the Bronze Age and the Iron Age in particular, there isn't an obvious change between these two periods in terms of the use of lead as an alloying element: the great increase in the proportion of leaded copper object observed in Luristan occurs only at the end of Period 2.

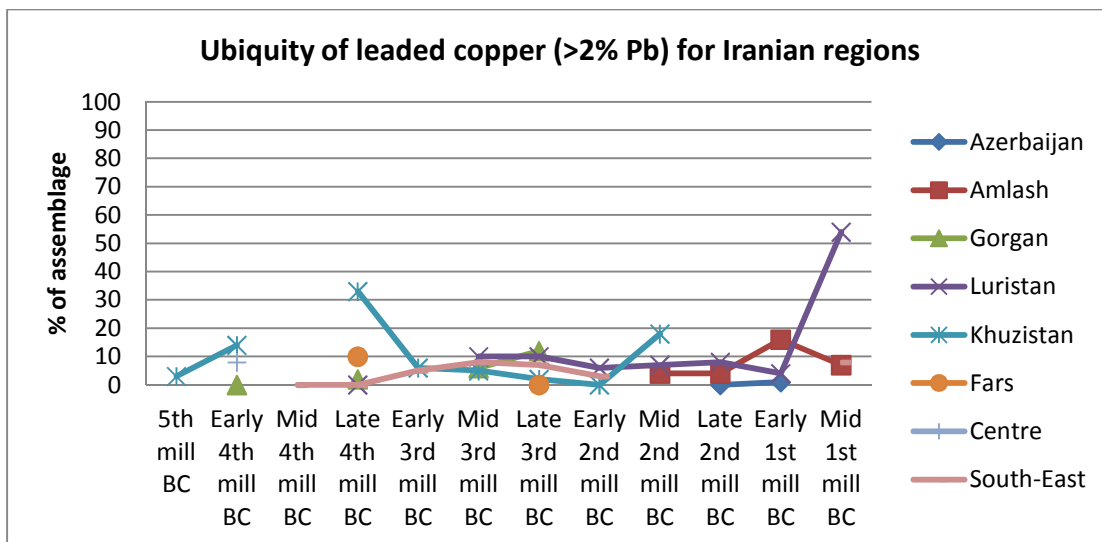


Figure V-13. Chronological evolution of the ubiquity of leaded copper objects in the copper-based metal assemblage for Iranian regions

Figure V-14 shows the evolution of the ubiquity of leaded copper for the regions around Iran. In Mesopotamia, the maximum ubiquity (16% of the assemblage) is reached in the early 3rd millennium BC, and the use of lead seems to decrease thereafter. In Oman on the other hand leaded-copper appears to occur more frequently in the 2nd millennium with a maximum of 24% of the assemblage reached in the early 2nd millennium BC. Again, the chronological pattern observed here doesn't demonstrate a period of experimentation followed by a generalized adoption of the alloy as for tin, but rather a more or less constant use of lead for generally less than a quarter of the objects produced.

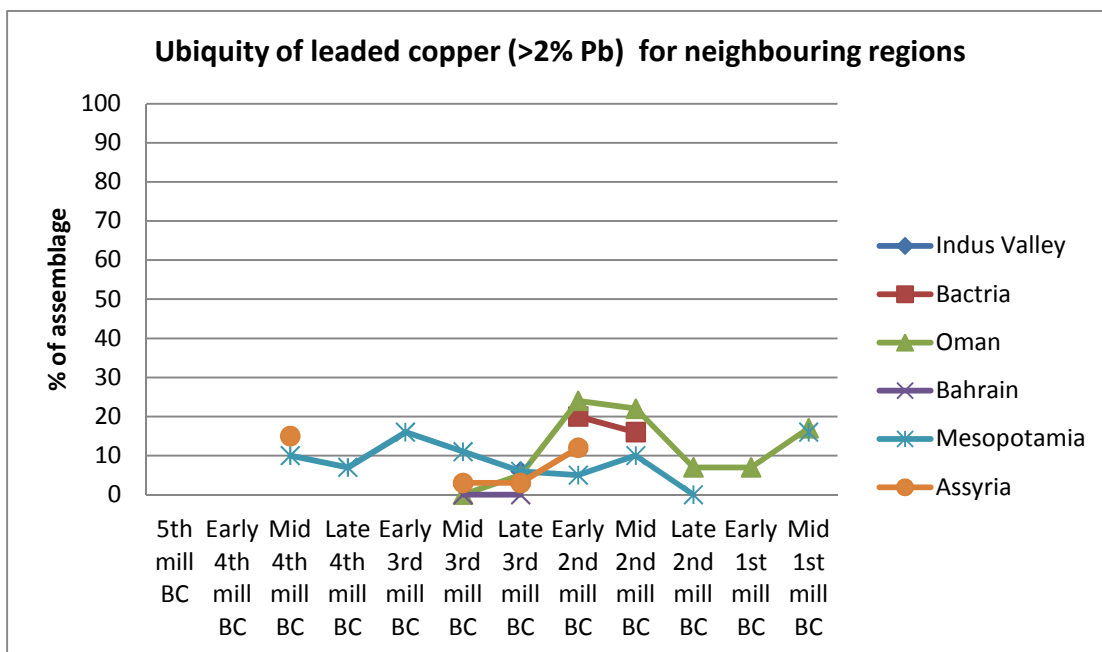


Figure V-14. Chronological evolution of the ubiquity of leaded copper objects in the copper-based metal assemblage for neighbouring regions

Moreover, Figure V-15 shows that even in regions where leaded copper is used relatively frequently as in Khuzistan and Oman, there doesn't seem to be, as observed for tin, a well defined recipe resulting in a clear pattern on the distribution graphs.

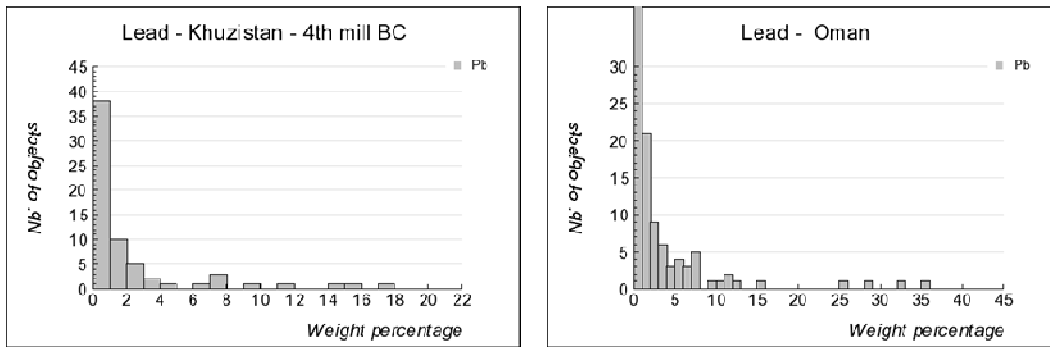


Figure V-15. Distribution of the lead content in copper-based objects for Khuzistan in the 4th millennium BC (left) and Oman for all periods (right)

To investigate a potential relationship between lead and tin, the evolution of the ubiquity of leaded copper in tin-less copper only and in tin-bronze only have been plotted for Iran (Figure V-16) and for the neighbouring regions (Figure V-17).

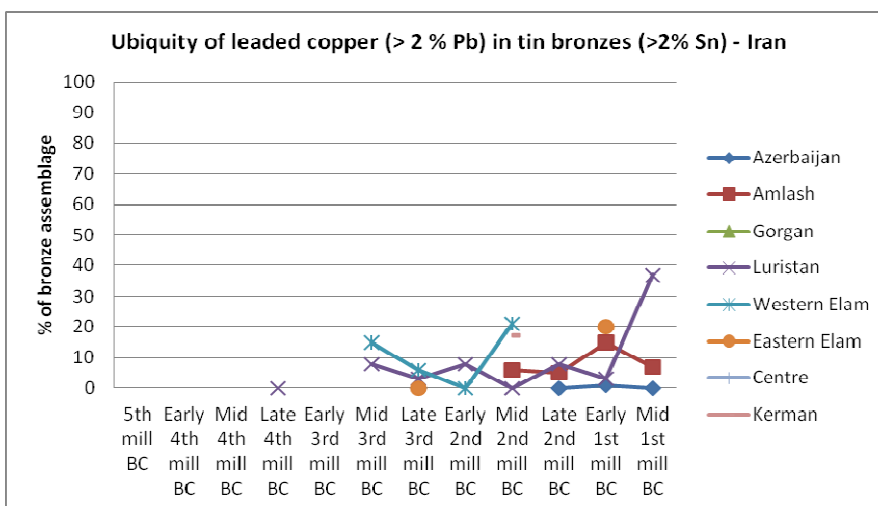
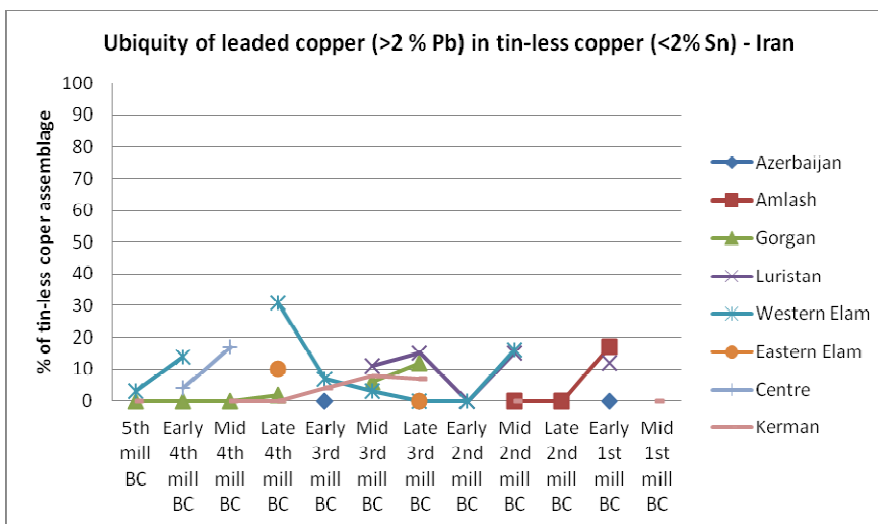
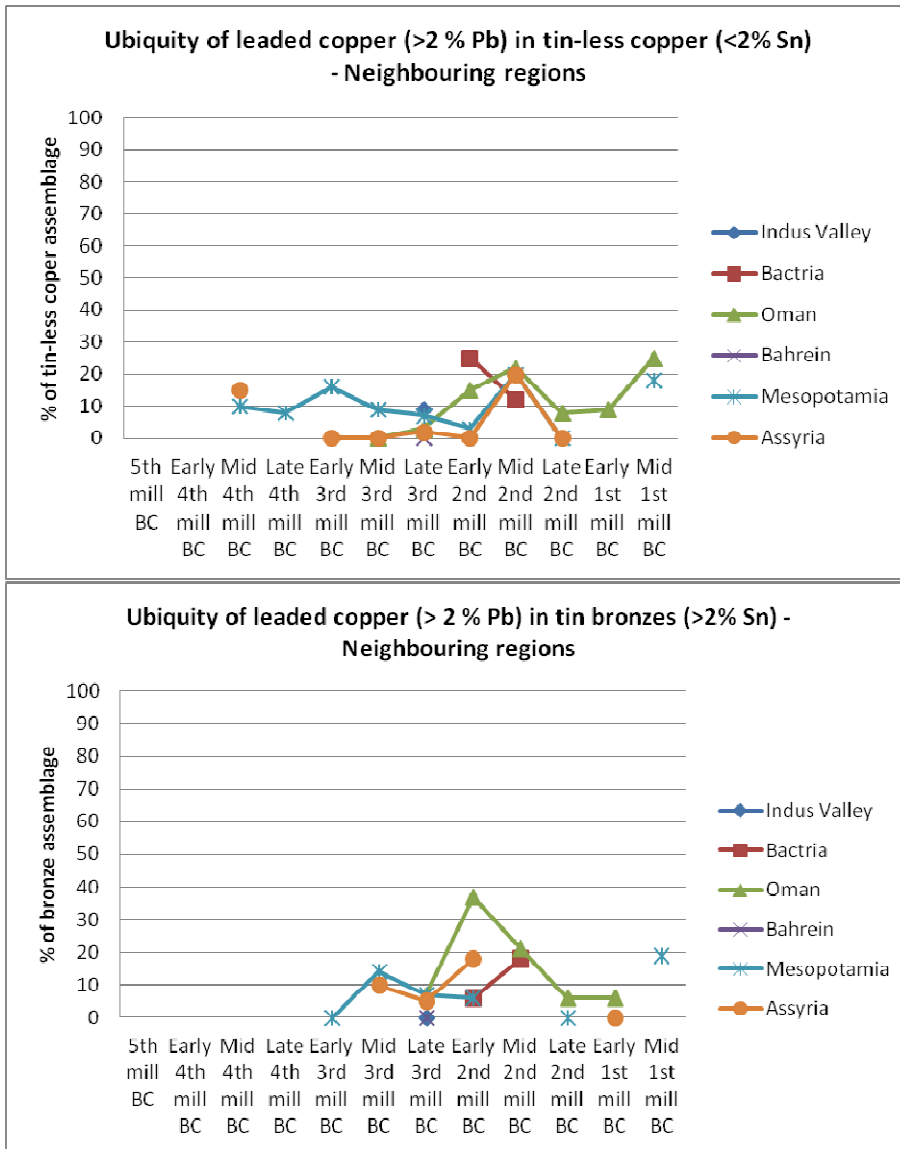


Figure V-16. Chronological evolution of the ubiquity of leaded copper objects in the assemblage of tin-less copper objects (< 2% Sn, top) and of tin bronze objects (>=2% Sn, bottom) in Iranian regions



**Figure V-17. Chronological evolution of the ubiquity of leaded copper objects in the assemblage of tin-less copper objects (< 2% Sn, top) and of tin bronze objects (>=2% Sn, bottom) in neighbouring regions**

These figures show that there isn't an obvious link between the presence of tin and the presence or not of lead. In Oman, in the early 2nd millennium BC, lead is more common in tin-bronzes than in tin-less copper for example, but in Luristan the opposite trend is visible in the mid to late 3rd millennium BC. On a very basic level, we can say that lead was present in a roughly equivalent proportion of tin bronzes than of tin-less copper objects. An interesting further direction of research could be a more detailed study of the relationship between the percentages of tin and lead and what they can tell about production processes. For example if tin was added at a late stage to an already alloyed leaded-copper, we could

expect to see a drop in the lead content due to an oxidative loss of lead. However, since there are no obvious changes in the frequency of alloying with lead between Period 1 and Period 2, and characterising the transition between these two periods is the main aim of this thesis, this subject is not studied here.

#### b. Brass

Brass is a yellow-coloured alloy of copper and zinc. In Chapter 3 we have seen that Thornton et al. discovered elevated levels of zinc (17 to 20%) in objects from the site of Tepe Yahya. These jewellery fragments from the 15th century BC are some of the oldest known brasses in the Near-East (Thornton et al., 2002). As shown in Figure V-18 however, objects with more than 2 % of zinc are very rare everywhere in Iran and never exceed 10% of the assemblage. Once again it must be noted that the cut-off value of 2% chosen here is very low. Thornton for example, in his review of south-west Asian copper-zinc alloys, looks at objects with more than 8% Zn (Thornton, 2007). This limit is, in Thornton's own words, "somewhat arbitrary", but was picked because above about 8% Zn the alloy is recognisable by its golden colour and it is more likely for the production of a zinc-alloy to be intentional. Here however, we choose to include all objects with over 2% Zn as we believe that low zinc contents might occur when recycling or mixing true brasses and can therefore also be indicative of the practice of alloying copper with zinc.

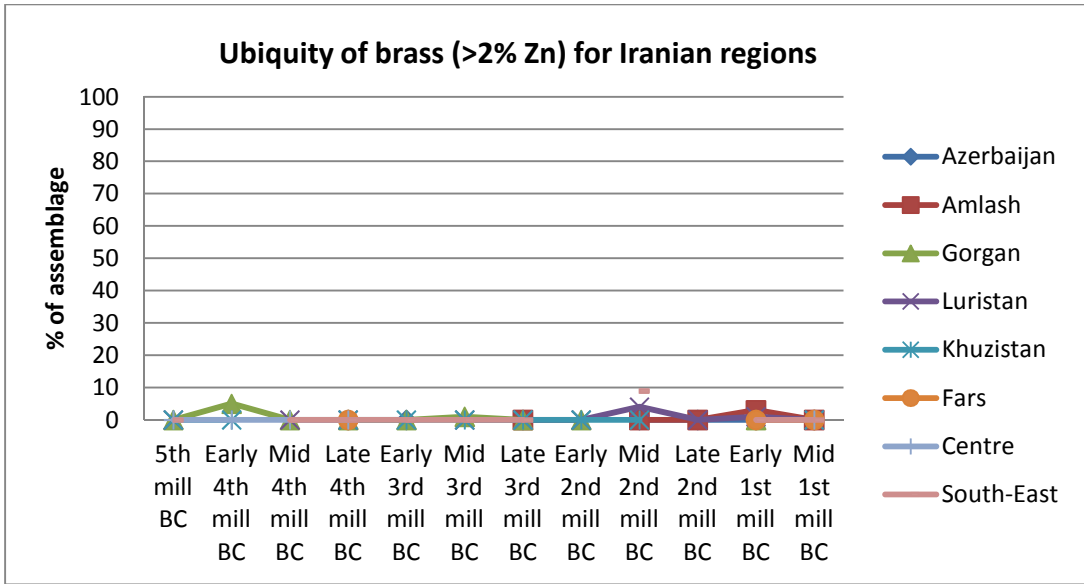


Figure V-18. Chronological evolution of the ubiquity of brass objects in the copper-based metal assemblage for Iranian regions

The situation is similar in the regions neighbouring Iran with the notable exception of Bactria in the early 2nd millennium BC (Figure V-19). There, zinc was sought for in only ten objects, two of which presented 2% Zn or more: a vessel with 8.2% Zn and spatula with 2.0% Zn, for which an accidental occurrence of zinc, or perhaps the recycling of brass with higher zinc content, could probably be argued.

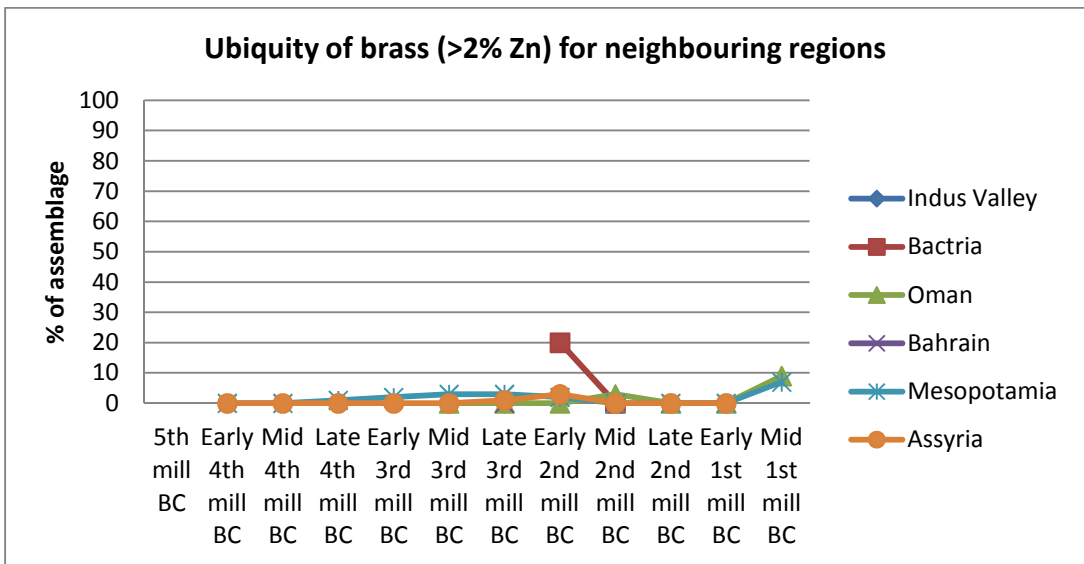


Figure V-19. Chronological evolution of the ubiquity of brass objects in the copper-based metal assemblage for neighbouring regions

The maps of Figure V-20 show the regions where brass objects have been recovered in Period 1 and Period 2. Bactria, Mesopotamia and south-eastern Iran show very early examples of brass objects while elsewhere brass seems to have been very rare. From this geographical pattern it would be possible to speculate that brass came to Iran from Bactria. This hypothesis has indeed already been put forward by Thornton, based on the observation that the Tepe Yahya brasses were found in “an area of Tepe Yahya that shows material links with the BMAC” (Thornton et al., 2002, p.1459). However, much more research and analytical data would be needed to ascertain this possibility.

Figure V-21 shows the relationship between tin and zinc in the brass objects of Bactria, Tepe Yahya and Mesopotamia. The objects from Tepe Yahya are all tin-less copper with high levels of zinc. The Bactrian objects both also have low amounts of tin (1.5% and 2.4%), but the Mesopotamian objects present a wide variety of tin contents, including very high tin bronzes. There appears to be a slight inverse correlation between the two elements: objects with the highest zinc contents are low-tin bronzes, while the very high tin-bronzes have low amounts of zinc. An interesting question for further research, once a greater bulk of data becomes available, would be to investigate whether this inverse correlation can be linked back to recycling processes and for example a drop of the zinc content of a brass caused by a later addition of tin. This, if true, could have a wide range of implications, including the possibility that despite the relative rarity of brasses, it wasn't considered necessary to protect it from recycling.

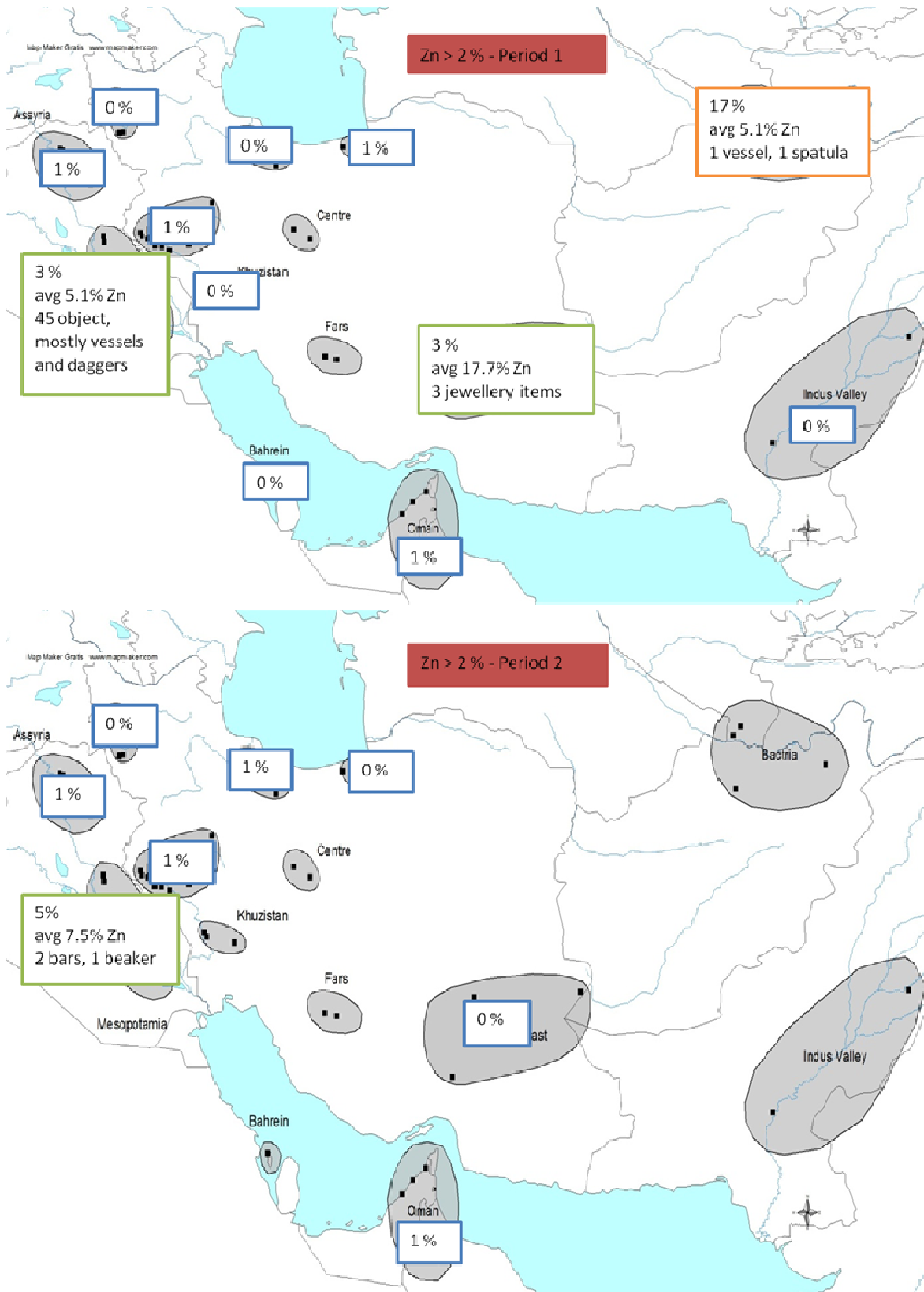


Figure V-20. Maps showing the ubiquity of brass objects (>2% Zn) in Period 1 (early 3rd to mid 2nd millennium BC, top) and Period 2 (late 2nd to mid 1st millennium BC, bottom). For regions where brass represented more than 2% of the assemblage, the average percentage of zinc in the brass objects, as well as the type of the objects that were made of brass are given.

Thornton's comprehensive review of brasses and gun-metal objects in South-western Asia known before 2006 revealed a geographical and regional correlation between the occurrence of tin-bronzes and that of zinc-copper alloys. The tin and zinc contents in objects presenting both alloying elements were, however, rather haphazard. In a very interesting argument, he suggests that this might be the result of confusion or ambivalence on the part of local consumers, for whom only the characteristic golden colour of both alloys was diagnostic, and who might therefore have mixed the two alloys together creating Cu-Sn-Zn alloys with somewhat random compositions (Thornton, 2007).

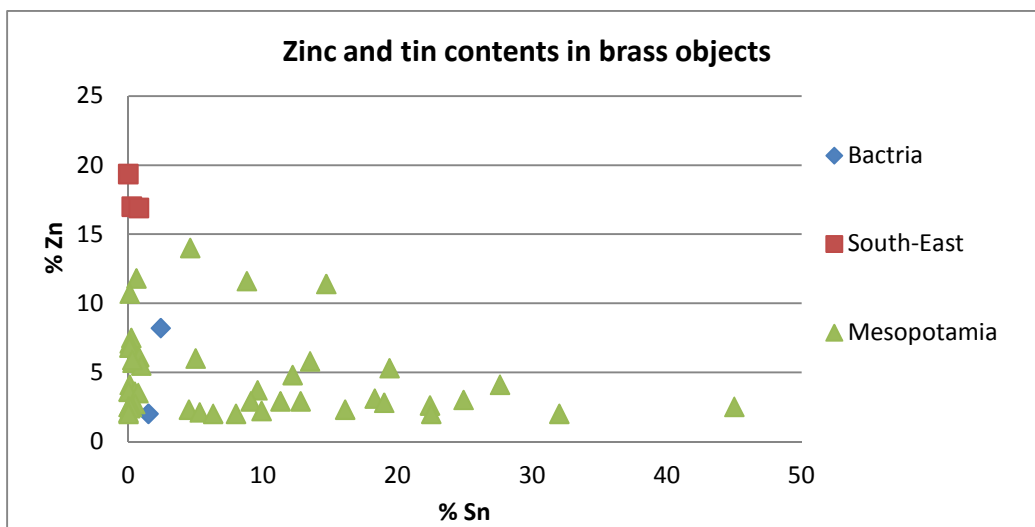


Figure V-21. Zinc content against tin content for brass objects ( $\geq 2\%$  Zn) in Bactria, south-eastern Iran and Mesopotamia)

This section echoes Thornton's work in showing that it is no longer possible to ignore the early occurrence of brass in the Near-East and early brasses could be a very important area of research. They must be studied by considering not only the production of the metal, but rather all the stages of its life-cycle, including possible trade, re-use and re-manufacture, before deposition. However, given the limited amount of data available here, we are unable to push the analysis any further in this thesis.

#### 4. Nickel, antimony, silver and cobalt

The presence in copper-based objects of nickel, antimony, silver and cobalt is generally thought to have been related with the provenance of the ore, rather than being a combination of ore provenance and intentional alloying as for the elements discussed above. Detailed geographical patterns for these four elements and arsenic are discussed in Chapter 6. Here, we limit ourselves to looking at the chronological evolution of their concentrations for Iran, Mesopotamia and Oman, the three regions for which data is available for most periods.

##### a. Nickel

Figure V-22 shows the evolution of the nickel content in Iran, Mesopotamia and Oman. We can see that in Iran and Mesopotamia, the median nickel content stayed fairly constant up to the mid 2nd millennium BC. In Mesopotamia, this is accompanied by a restriction of the range of nickel contents that could be explained by change of emphasis in the importation of copper from sources with higher nickel contents to sources with lower nickel contents. In these two regions, the median and the range of Ni contents dropped in the late 2nd millennium BC and remained low until at least the mid 1st millennium BC. We attempt to explain this drop in relation to copper sources in Chapter 6. In Oman, the nickel levels are higher than in the other two regions and there is also a slight drop at the turn of the Iron Age which as we have seen above could be related to a shift towards the exploitation of the sulphide deposits containing low levels of As and Ni (Weeks, 2003, pp.118-19).

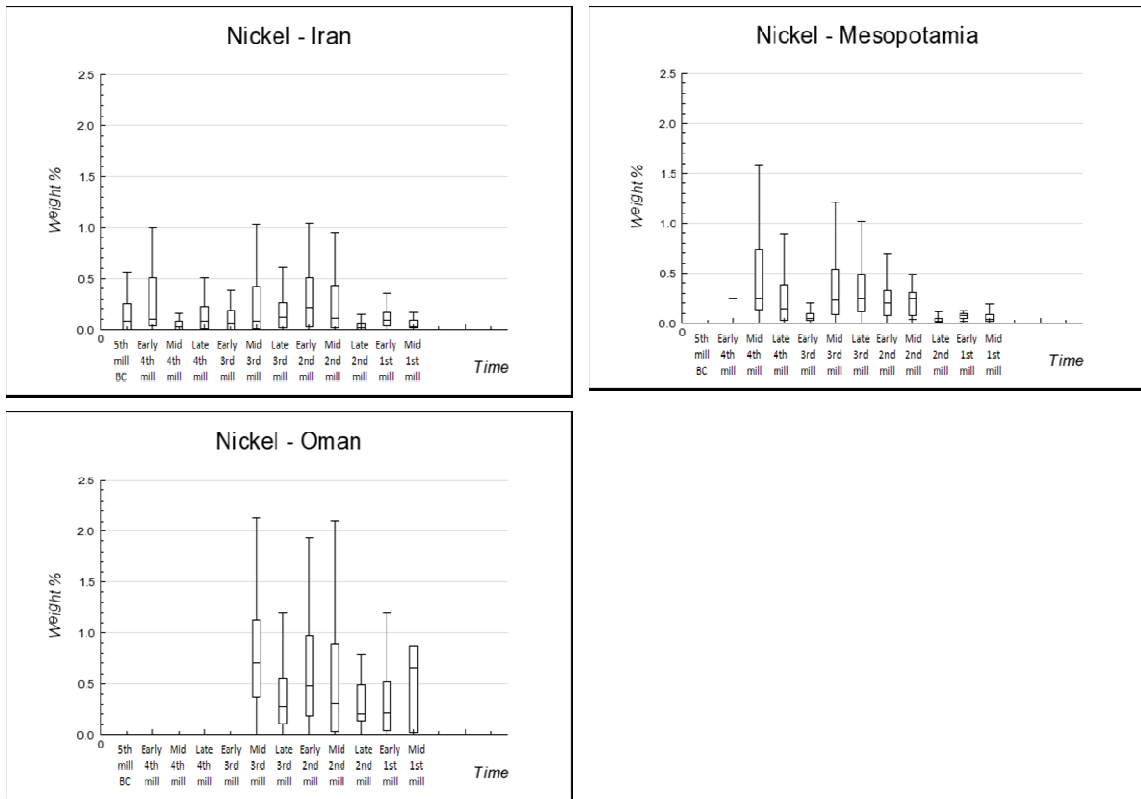


Figure V-22. Evolution of the nickel content in Iran, Mesopotamia and Oman over time.

b. Antimony and silver

Figure V-23 and Figure V-24 show that similar observations can be made of the evolution of the levels of antimony and silver. In Oman, the concentration in these two elements is consistently extremely low with only very few objects showing concentrations of over 0.02% Sb or Ag. In Iran and Mesopotamia the median levels are also very low (consistently under 0.1%) but wider ranges of concentrations are observed. In Iran, the levels of Sb and Ag stay fairly constant throughout the time-frame of this study except for higher levels of Sb in the 4th millennium BC. In Mesopotamia, the concentration in both elements seems to drop at the beginning of the 2nd millennium BC. Overall it would appear that Mesopotamia shifts from using metal with a significant amount of impurities, to using a ‘cleaner’ metal.

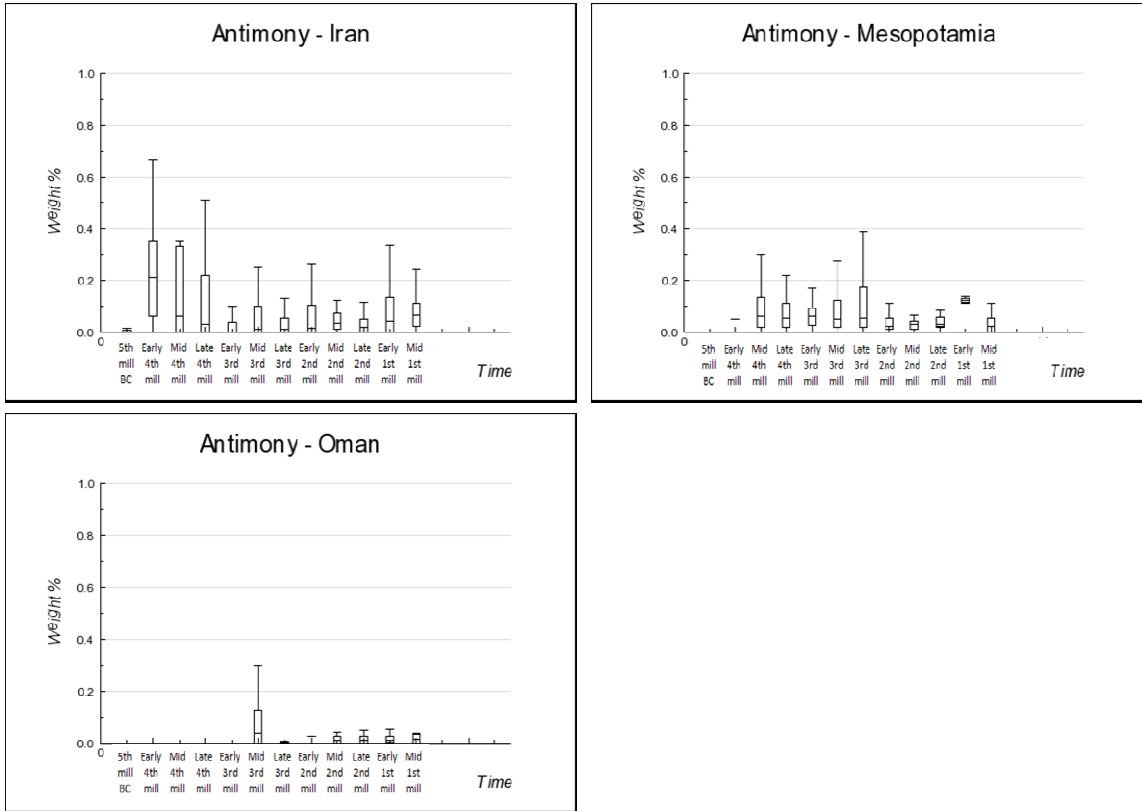


Figure V-23. Evolution of the antimony content in Iran, Mesopotamia and Oman over time.

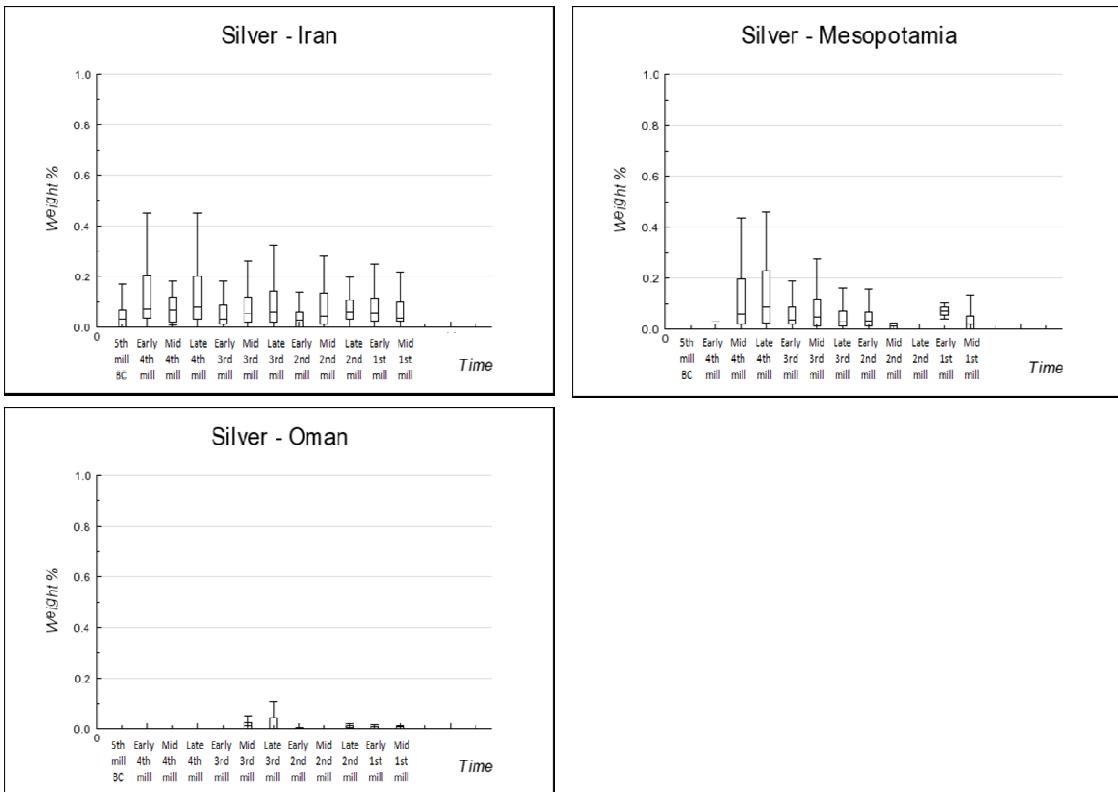


Figure V-24. Evolution of the silver content in Iran, Mesopotamia and Oman over time

### c. Cobalt

Finally, a look at the concentration of cobalt in the objects shows that levels of cobalt of over 0.05% were not uncommon in Omani objects (37% of the objects for which cobalt was measured) but were very rare elsewhere. Moreover, it would appear that objects with high cobalt contents were much more common in Period 1 than in Period 2 (Figure V-25).

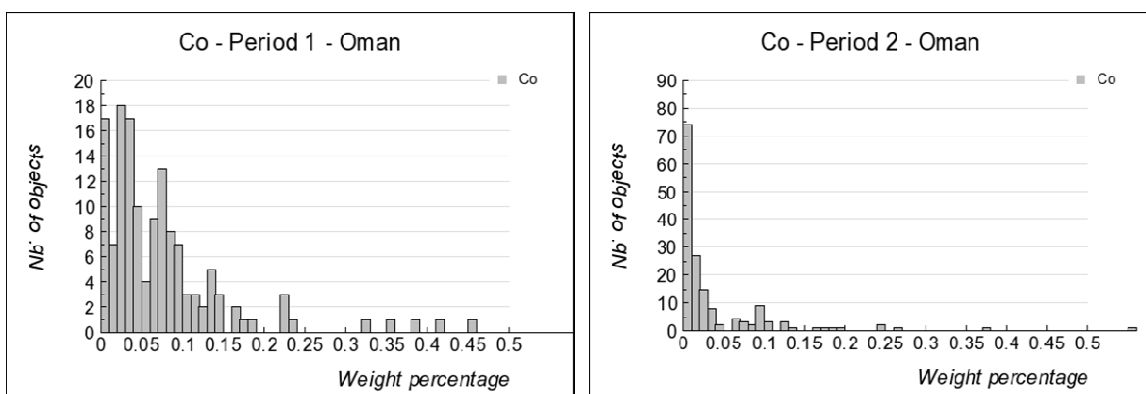


Figure V-25. Distribution of the cobalt content in copper-based objects from Oman in Period 1 (early 3rd to mid 2nd millennium BC, left) and Period 2 (late 2nd to mid first millennium BC, right)

### 5. Lead isotope analysis

Lead isotope analysis has fuelled debates on ore provenance since its first use in archaeology in the 1970s. This technique is based on the fact that the relative abundance of four isotopes of lead ( $^{204}\text{Pb}$ ,  $^{206}\text{Pb}$ ,  $^{207}\text{Pb}$  and  $^{208}\text{Pb}$ ) varies from one ore deposit to another. Indeed, while  $^{204}\text{Pb}$  was present at the formation of the earth, the other isotopes are derived from the decay of uranium and thorium ( $^{238}\text{U}$ ,  $^{235}\text{U}$  and  $^{232}\text{Th}$ ). Therefore, the lead isotope composition of an ore deposit depends on the age of the deposit and the relative concentrations of lead, uranium and thorium that were present at its formation (see Pollard & Heron, 2008, pp.302-345 for example for an overview of the technique). The analysis of the ratio of these isotopes in archaeological objects and the comparison of these to lead

isotope ratios of ores from different deposits has therefore been seen as a tool of great potential to determine the provenance of the ore used to produce the objects.

For copper-base objects in particular several assumptions need to be made in order to use this method for provenance studies (Pollard & Heron, 2008, p.322). The first one is that the isotopic ratios in the copper ore are not affected by production processes such as smelting, casting and working, the main worry being the possibility that lead would fractionate during these processes. Secondly, we must assume that they are also unaffected by the addition of tin or other alloying element. Thirdly, we must assume that the objects were not produced by the mixing of ores or metals from different sources. The first assumption has been proven to be a reasonable working hypothesis: it has been demonstrated that the fractionation of lead during production processes is negligible (Pollard & Heron, 2008, pp.322-25). The second assumption, also generally considered as valid given the low concentration of lead in cassiterite, has however been recently questioned (see summary of discussions in Weeks, 2003, p.138). In the case of alloying with lead, a practise that; as shown above, wasn't uncommon in the Near-East, it is certain that the trace amounts of lead initially contained in the copper would be swamped by the added lead and the results of LIA can not be seen as indicating the provenance of the copper ore. The third assumption (the lack of mixing of different copper ores) is highly questionable. The deposits of high quantities of scrap metal in major sites of the Harappan Period in the Indus Valley for example is likely to indicate that the recycling and mixing of metal for the production of new objects was a common practice in that area (Hoffman & Miller, 2009, p.248). In Chapter 4 we have seen that a significant number of bronze objects present tin contents ranging between approximately 2 and 6% Sn. We have suggested that these were produced by the mixing of metals with different tin contents, or by a loss of tin upon remelting. In Chapter 6 we also present evidence for the recycling of certain metal types. If

mixing was indeed common practice, the likelihood of being able to determine their provenance by using LIA is highly compromised, unless all the metal that is mixed comes from the same ore source. This could be the case if we envisage a production using local ores, with little contact and trade with foreign regions. However we have seen that in the period and region we are interested in, and in particular in Period 1, long distance trade of metals was probably taking place, in particular through the Persian Gulf. Another important limitation of LIA for provenance study is the fact that distinct ore bodies can have overlapping ranges of lead isotope ratios. This has been found to be increasingly problematic as the number of ores analysed grew bigger and the overlap became more significant (Pernicka et al., 1990, p.278). All in all, it seems that LIA, rather than being considered as a tool to determine an absolute provenance, should be seen as a way of characterising the metal and perhaps recognising objects that are likely to have had a similar story, including their provenance, trade, recycling, mixing, etc. In effect, the results of lead isotope analysis are not very powerful when used in isolation but can become very useful when constrained by other sets of data, such as the chemistry and the chronology.

For this thesis, we have collated published LIA analysis of ore samples (mostly copper ores and galena) from the three potential Iranian ore sources mentioned in Chapter 2, and ore from Oman, Afghanistan, Bactria, the Caucasus and the Indus Valley (Nezafati, 2006, p.90; Vatandoust et al., 2011, p.677; Begemann et al., 2010; Begemann & Schmitt-Strecker, 2009; Hegde & Ericson, 1985; Srinivasan, 1999).

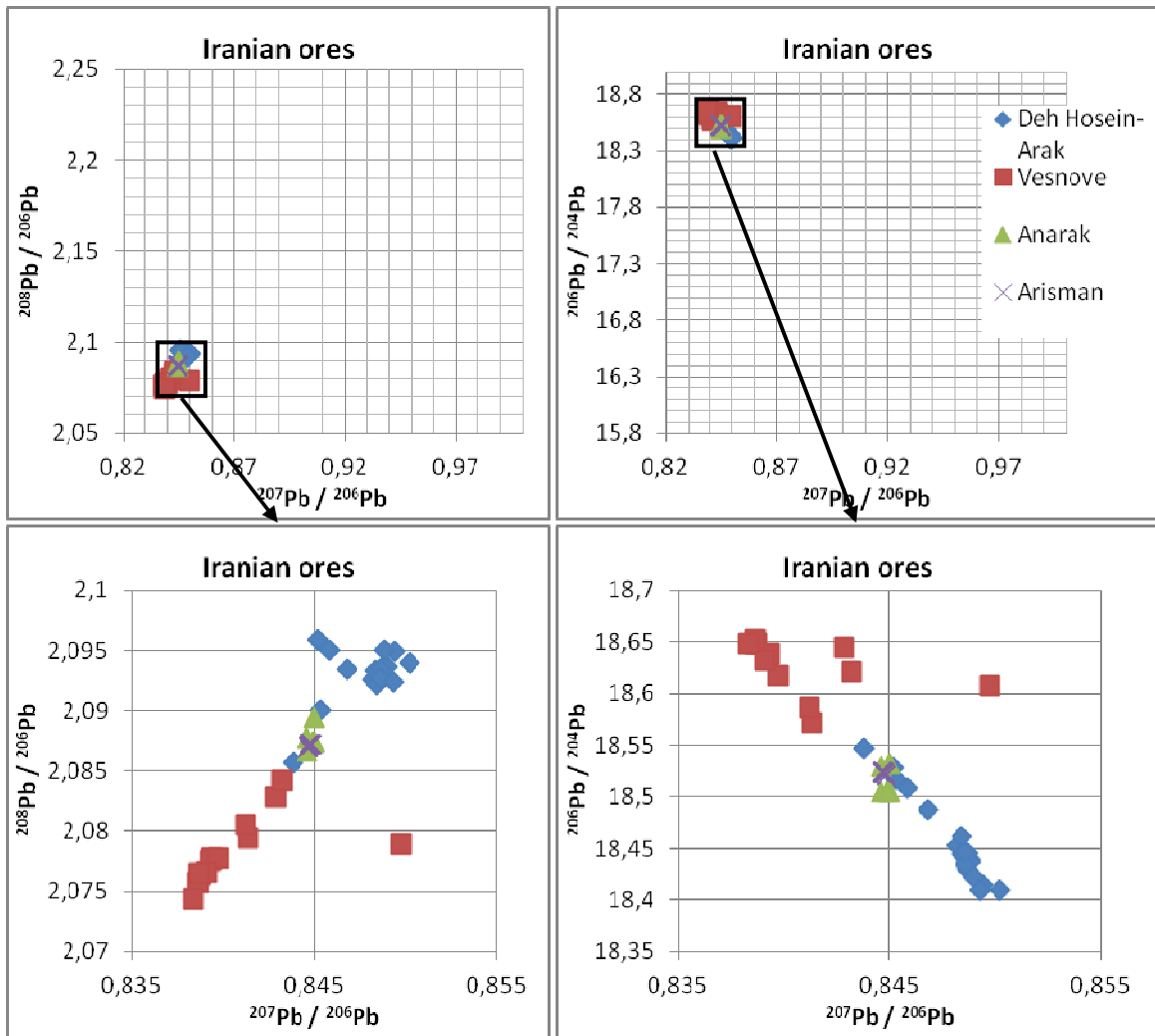


Figure V-26. Lead isotope ratios for ores from Iranian mining areas

As we can see on Figure V-26 the different ore sources in Iran plot in somewhat distinct regions of the graph, however the overlap between these sources and the Omani, Caucasian and Afghan sources in particular is high (Figure V-27 and Figure V-28). The ores from the Aravalli hills and particularly Rajasthan on the other hand are quite distinct in that they have much higher  $^{207}\text{Pb}/^{206}\text{Pb}$  and  $^{208}\text{Pb}/^{206}\text{Pb}$  ratios (Figure V-27).

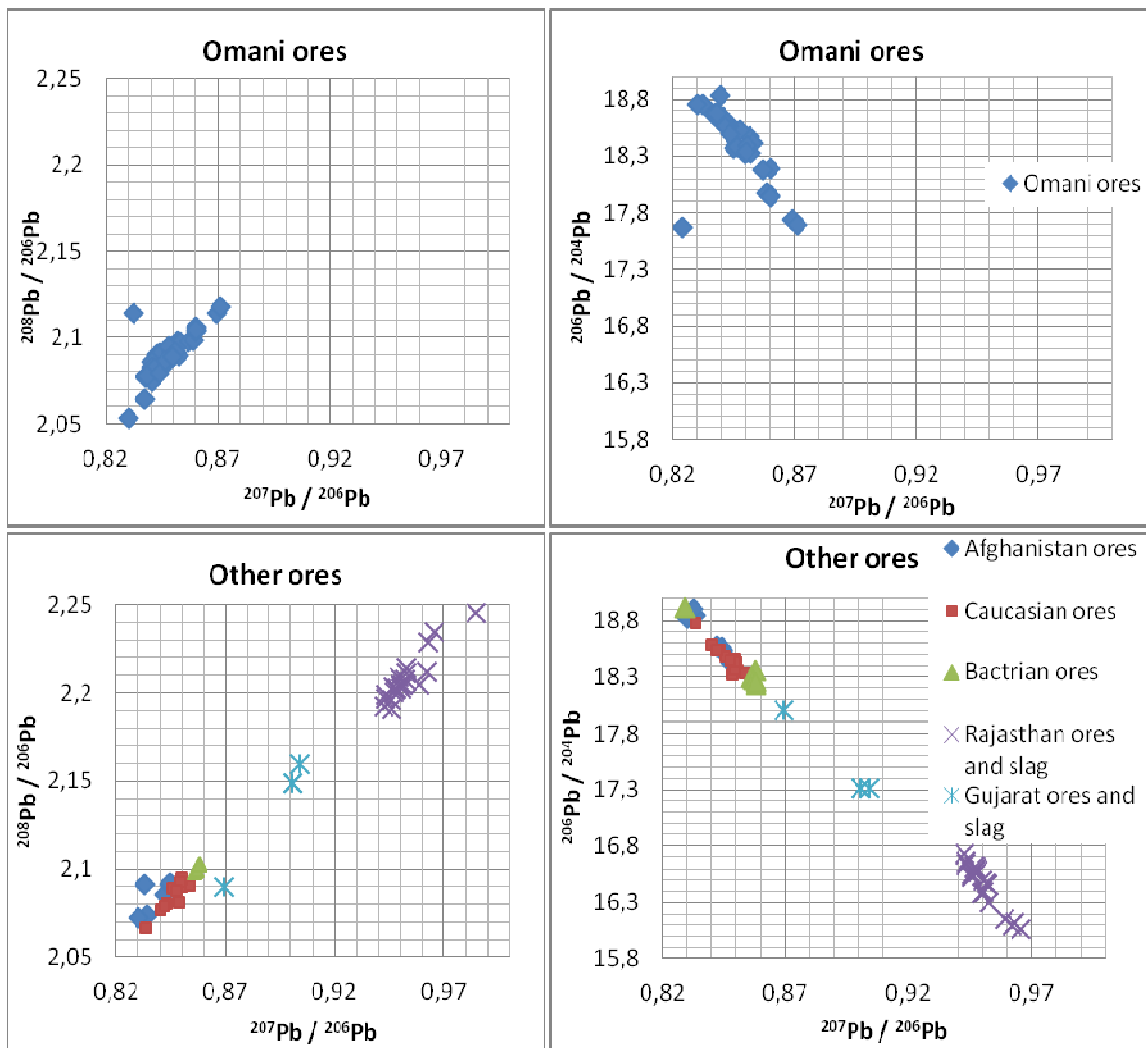


Figure V-27. Lead isotope ratios for ores from Oman, Afghanistan, Bactria, the Caucasus and the Indus Valley

The LIA analysis of 378 objects from Luristan (Begemann et al., 2008; Nezafati, 2006, p.88), Oman (Weeks, 2003, p.73; Begemann et al., 2010) and Mesopotamia (Begemann & Schmitt-Strecker, 2009) and of prills and slag from central Iran (Vatandoust et al., 2011, p.677) have been collated in our database. Interestingly, almost all of these objects have also been subjected to chemical analysis, allowing us to use these two datasets simultaneously. The following section summarises the observations derived from previous studies of LIA in our region of interest and attempts to find patterns from looking at the LIA data in connection with the concentration in As, Ni, Sb and Ag, taking each one of

these elements separately at this stage. It demonstrates the difficulty of finding an archaeological structure within the chemical and lead isotope datasets.

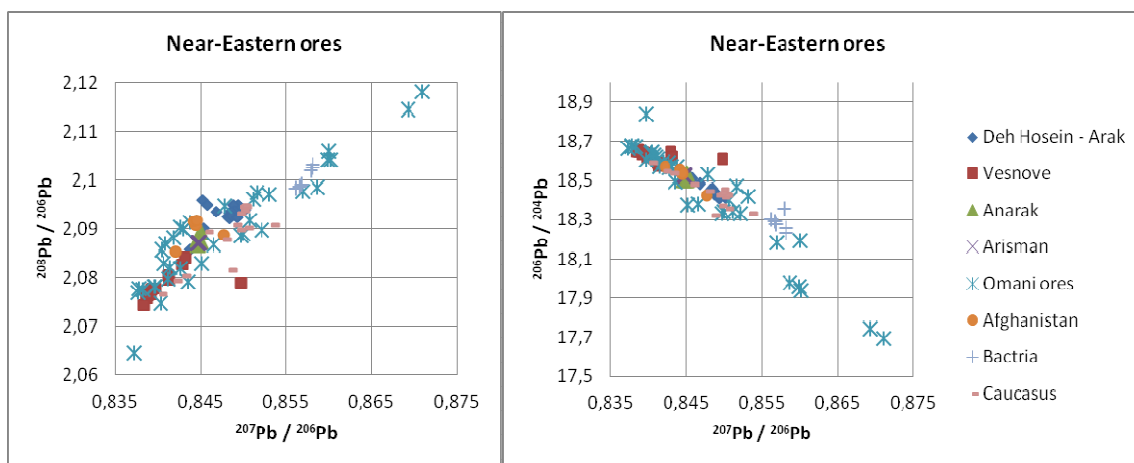


Figure V-28. Overlap between the lead isotope abundance ratios of ores from various Near-Eastern regions.

Unfortunately, almost all of the objects that have been subjected to LIA are from Period 1 (early 3rd millennium BC to mid 2nd millennium BC). Only 32 objects are from Period 2 (late 2nd to mid 1st millennium BC) and 29 of these come from a single site: Tell Abraq in the Peninsula of Oman. This means that LIA is of little use to us in the study of the changes in copper provenance for Mesopotamia and Iran at the transition between the Bronze Age and the Iron Age. It can however help define the metal used in the Bronze Age which is a necessary first step in understanding the transition.

Another important point that needs to be emphasized here is that 68 out of the 78 Iranian objects for which LIA data is available come from Luristan with the other 10 coming from the site of Shahr-i Sokhta in Eastern Iran. Figure V-29 shows that there is a significant overlap between the two sets of data. Objects from Shahr-i Sokhta appear to fall at the lower end of the Luristani range for  $^{207}\text{Pb}/^{206}\text{Pb}$  ratios and possibly have slightly higher  $^{208}\text{Pb}/^{206}\text{Pb}$  ratios probably indicating a different provenance for the copper ore in eastern and western Iran. However the number of eastern Iranian objects analysed at this point is

far too small to make any definite conclusion. Therefore, in this section we only consider the Luristani objects when discussing LIA.

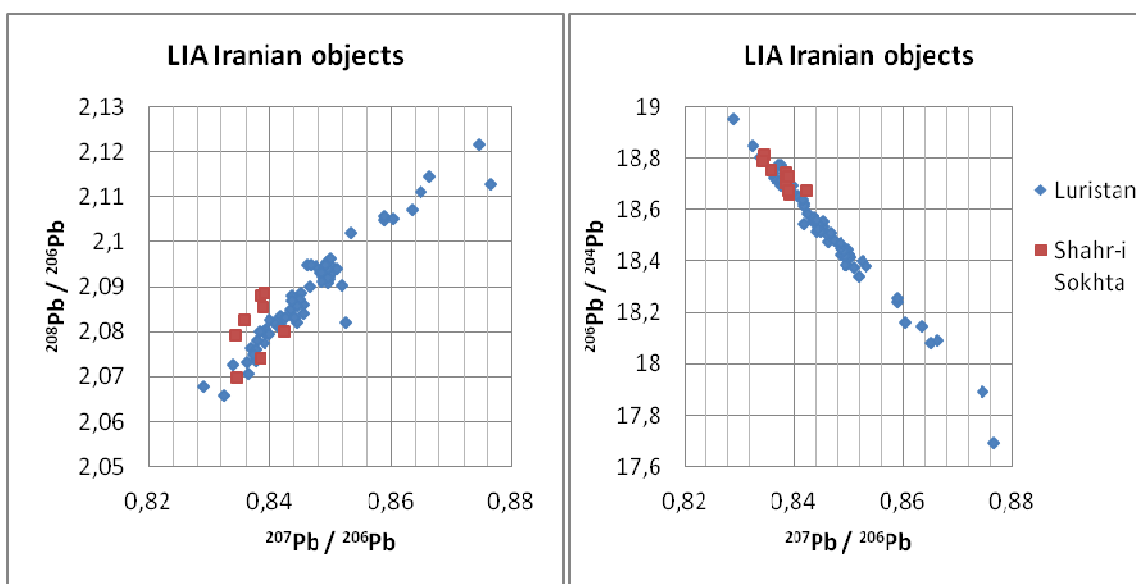


Figure V-29. Lead isotope ratios for Iranian objects

a. Luristan

The LIA of a total of 70 objects from Luristan were published by Begemann et al. (2008) and Nezafati (2006). The artefacts represent all types of objects (weapons, tools, vessels and ornaments) and, as mentioned above, most date from Period 1 with only two objects from Period 2 (from the mid 1st millennium BC). Aside from a small number of outliers with higher  $^{207}\text{Pb}/^{206}\text{Pb}$  and  $^{208}\text{Pb}/^{206}\text{Pb}$  ratios, most of the objects fall in two distinct groups: objects with between approximately 0.835 and 0.847  $^{207}\text{Pb}/^{206}\text{Pb}$ , and a cluster around 0.85  $^{207}\text{Pb}/^{206}\text{Pb}$ , 2.093  $^{208}\text{Pb}/^{206}\text{Pb}$  and 18.45  $^{206}\text{Pb}/^{204}\text{Pb}$  (Figure V-29), as discussed by Begemann et al. (2008). The good overlap between this cluster and the analyses of the ores from Deh Hosein and other sources near the city of Arak have led them to suggest a local provenance in the Eastern Zagros for the ore used in the manufacture of these objects. Unfortunately, ores from Anatolia, western Afghanistan or

the Caucasus also correspond on the basis of their lead isotope ratios, but in the case of Anatolia, the low arsenic content of the ores makes it an unlikely provenance (Begemann et al., 2008, p.31). The objects with  $^{207}\text{Pb}/^{206}\text{Pb}$  ratios lower than this cluster have an isotopy that matches ores from Vesnave, Anarak and Bezarak in eastern Afghanistan (Begemann et al., 2008, p.31). Interestingly, all but one of the outliers date from second half of the 3rd millennium BC, perhaps indicating the use of copper from a source in that period, that was subsequently abandoned in the 2nd millennium. Begemann et al. found that these higher ratios corresponded to the lead isotope ratios of ores from Mushiston in Bactria for some of the objects, Timna in Israel and Horzum in south-eastern Anatolia for some others. However, in the case of Timna they state: “We mention this just for the sake of completeness, not to suggest such a provenance to be a likely possibility” (Begemann et al., 2008, p.35), and they emphasize the fact that our knowledge of the lead isotopy of the Near East is too limited to pin-point the provenance of these objects.

Begemann et al. looked at the arsenic, lead and tin contents of the objects they analysed in connection with the LIA, but gave little notice to other elements. They noticed that all three of the lead isotopes ratio groups described above present tin-less copper objects, tin bronzes, lead rich and lead poor objects. To this we can add that the objects with  $^{207}\text{Pb}/^{206}\text{Pb}$  ratios above the main cluster have a relatively high nickel content ( $>0.1\%$ ) and a relatively low antimony content ( $<0.1\%$ ) (Figure V-30). At present, however, given our limited knowledge of potential sources corresponding with these higher  $^{207}\text{Pb}/^{206}\text{Pb}$  ratios, this information is of little use as such.

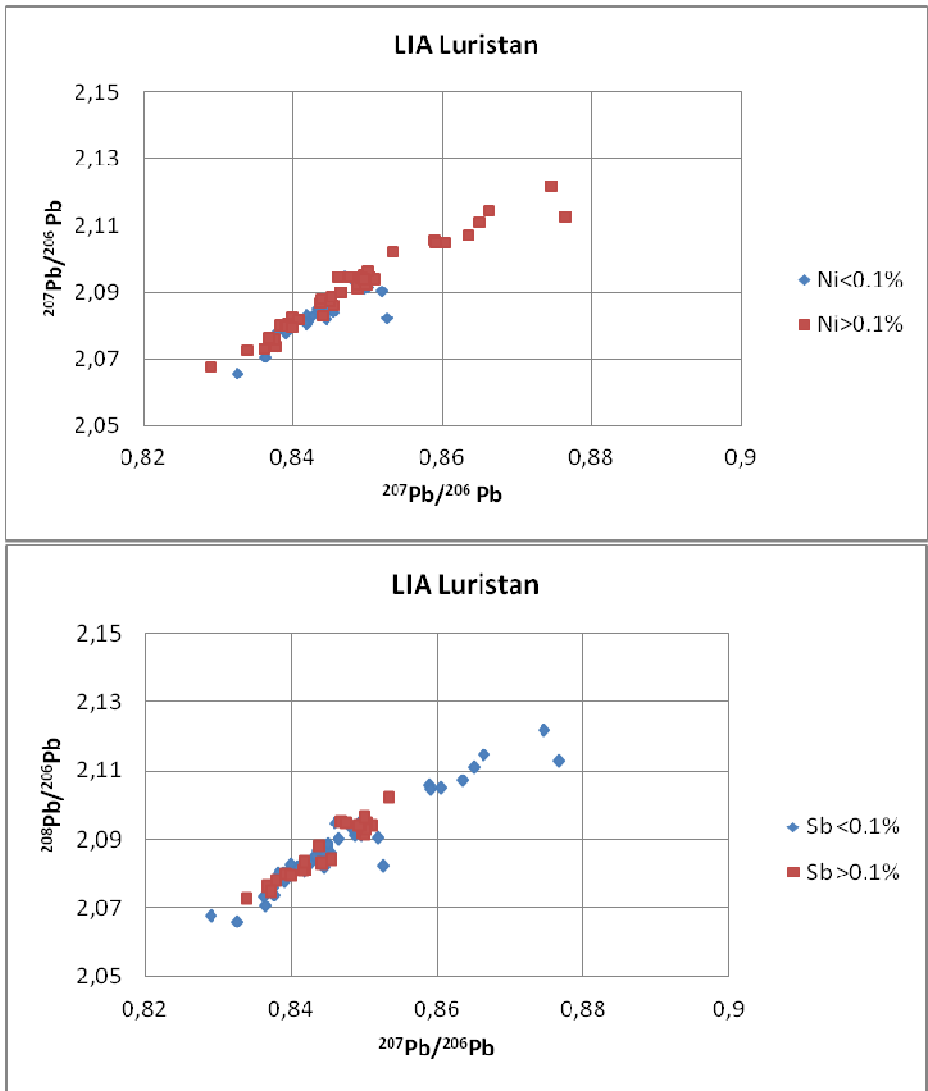


Figure V-30. Lead isotope ratios for low nickel (<0.1% Ni), and high nickel (>0.1% Ni) objects (top), and low antimony (<0.1% Sb) and high antimony (>0.1% Sb) objects (bottom) from Luristan

#### b. Mesopotamia

Begemann and Schmitt-Strecker published the results of the LIA of 96 Mesopotamian objects (Begemann & Schmitt-Strecker, 2009), mostly weapons, but also ornaments and tools dating from the Uruk period (4th millennium BC) to the Akkadian period (late 3rd millennium BC).

The spread of isotopic ratios observed for Mesopotamia is wider than for Luristan (see Figure V-32) which has lead Begemann et al. (2008, pp.37-38) to state that Mesopotamia used copper from a wider variety of sources than Luristan. This, of course, doesn't come as

a surprise given the political importance of Mesopotamia and the lack of mineral resources in that region. Interestingly, the signature of the main cluster observed in Luristan is practically absent in Mesopotamia, suggesting that this ore, arguably of eastern Zagros provenance (see above), was not used in Mesopotamia (Begemann et al., 2008, pp.36-38). From a chronological point of view, the authors observed in the Uruk period low  $^{207}\text{Pb}/^{206}\text{Pb}$  ratios (around 0.83) that could correspond to ores from central and northern Anatolia. This signature then becomes rare (Figure V-31) and the tin-less copper objects from the 3rd millennium BC could correspond to a variety of different ores sources (south-eastern Anatolia, Iran, Oman and Timna) (Begemann & Schmitt-Strecker, 2009).

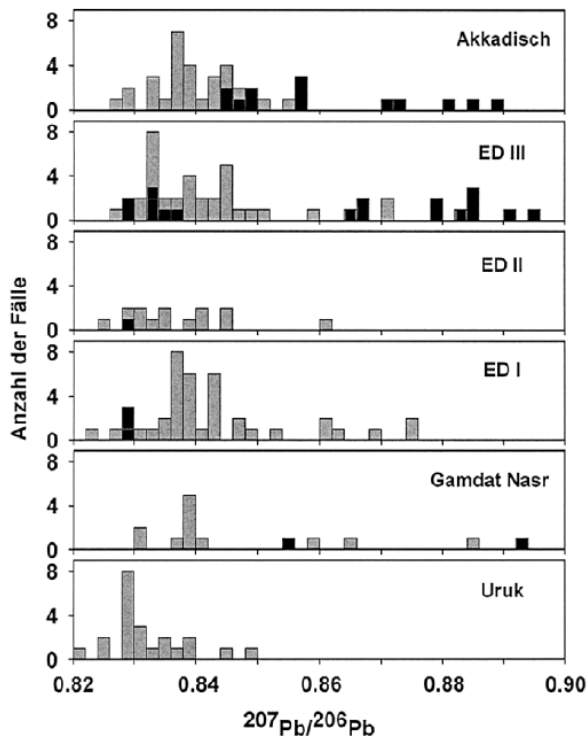


Figure V-31.  $^{207}\text{Pb}/^{206}\text{Pb}$  ratios for Mesopotamian tin-less copper (grey) and tin bronze (black) objects from different periods (from Begemann & Schmitt-Strecker, 2009, p.12, fig.5).

From the Early Dynastic III period (mid 3rd millennium BC) onwards, bronze objects with higher  $^{207}\text{Pb}/^{206}\text{Pb}$  ratios start to appear. The authors suggest for this bronze a provenance in the Indus Valley (Gujarat or Southern Rajasthan). However, the data collected for this

thesis (see Figure V-27) suggests that the Indian ores have even higher  $^{207}\text{Pb}/^{206}\text{Pb}$  ratios and the answer to the provenance of this metal could lie elsewhere. This metal also presents relatively low levels of arsenic (between 0.25 and 1%, see Figure V-32). However, the absence of higher arsenic contents in these objects probably shouldn't be seen as a signature of the ore, but rather as a result of the process of alloying with tin, which would have resulted, as explained above, in a loss of arsenic. For this thesis, the nickel, antimony and silver contents were also examined one by one, in relation to the lead isotope ratios, but no specific patterns were observed.

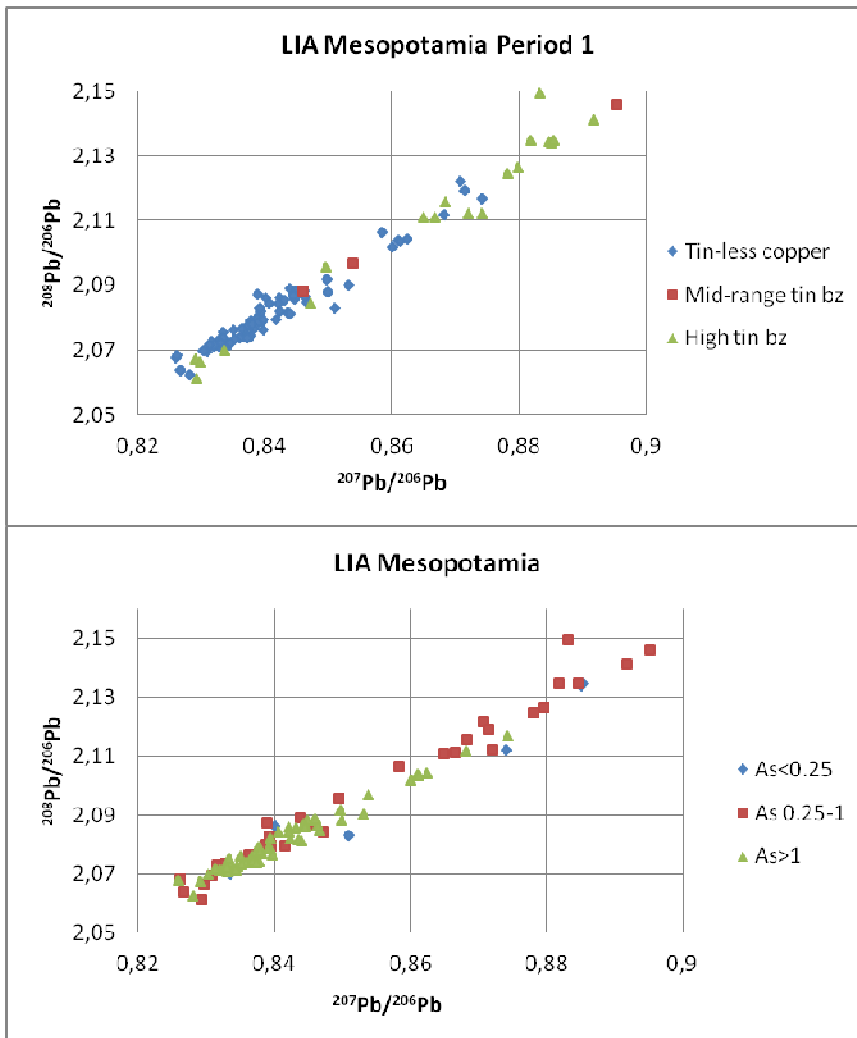


Figure V-32. Lead isotope ratios for tin-less copper (<2% Sn), mid-range bronze (2-6% Sn) and high tin bronze (>6% Sn) objects (top) and low As (<0.25% As) middle As (0.25 to 1%) and high As (>1%) objects (bottom) in Mesopotamia

c. Oman

For Oman, the LIA of 69 objects from Period 1 and 29 objects from Period 2 were added to the database. The Period 1 objects range from the mid 3rd to the mid 2nd millennium BC and come from various different sites of the Oman Peninsula. The objects are mostly pins, rings, vessels and waste but there are also a small number of weapons, tools and ingots. All of the objects from Period 2 on the other hand come from one site: Tell Abraç. They are mostly pins, vessels, arrowheads and waste.

Begemann et al. (2010) have studied both the chemistry and the lead isotopy of artefacts from sites on the eastern side of the peninsula, where the ores are located, while Weeks (2003) has studied the LIA of objects from sites on the western coast of the peninsula, in modern U.A.E. In both cases and in both Period 1 and Period 2, aside from a few outliers, the artefacts have an isotopic signature broadly compatible with Omani ores, however this signature is not unique and other sources could be envisaged (Weeks, 2003, p.145-163; Begemann et al. 2010, pp.153-156; see also Figure V-33). Weeks noted that the isotopic range for tin bronze was wider than for tin-less copper, implying either that the addition of tin shifted the lead isotope ratios of alloyed object, or that different sources were used for copper and bronze in Oman (Weeks, 2003, pp.150-151). The high  $^{207}\text{Pb}/^{206}\text{Pb}$  abundance ratios ( $>0.875$ ) observed in some Mesopotamian tin bronzes with a suggested provenance from the Indus Valley (see above) are rare in the Omani artefacts (only two objects from Al Sufouh and Unar 2 present this signature but they have low tin levels: 0.71 and 2.05 % Sn respectively). For this reason, Begemann et al. have tentatively suggested a scenario of protectionism whereby “on the Arabian side of the Gulf the copper market was heavily protected so that ‘local’ copper did not have to compete with imports even though the latter may well have been of superior quality” (Begemann et al., 2010, p.162).

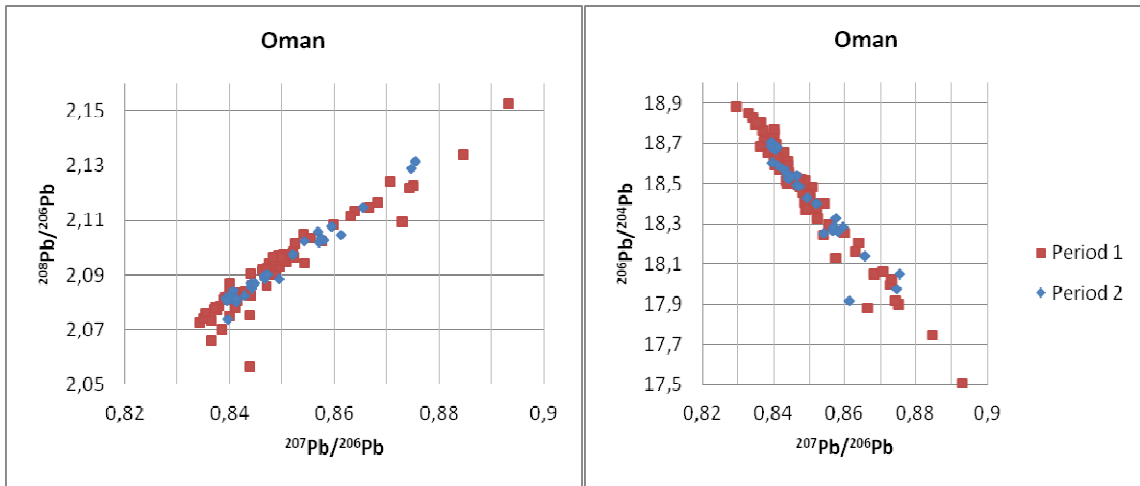


Figure V-33. Lead isotope ratios for artefacts from Oman in Period 1 and Period 2

The study for this thesis of the arsenic, nickel, antimony and silver contents (taken separately) of Omani objects in relation to their lead isotopy has shown no notable trends. In their paper on the LIA and chemical composition of Omani artefacts, Begemann et al. (2010) suggested that looking at the relative proportions of arsenic and nickel could be a way of determining whether or not objects are compatible with Omani copper sources. Indeed, they noticed a positive correlation between arsenic and nickel in the Omani ores and objects (Begemann et al., 2010, p.152) that was not observed in the Mesopotamian objects from the Gamdat Nasr, ED I and ED II periods. Moreover, they noticed that the ratio of arsenic to nickel in Oman was generally smaller than ten, which was very rarely the case for Mesopotamian objects. Based on these considerations, they regarded objects with  $As/Ni < 10$  as being “chemically compatible” with Omani ores. When comparing the lead isotope signature of the compatible objects to those of Omani ores and objects they noticed, for the tin-less copper objects, an important overlap and concluded that these objects “may well have come from Oman” (Begemann et al., 2010, p.157). However, the authors didn’t fully address the limitations of this method and a great deal of caution has to be taken when applying it. Indeed, as explained above, many factors can change the chemistry of an object and in particular its arsenic content. Working and remelting, for

example, result in a loss of arsenic. This phenomenon is clearly visible on the graphs of Figure V-34 for Luristan and Mesopotamia: as discussed above, the arsenic contents of the bronze objects are generally lower than those of the tin-less copper objects. This can be seen, at least in part, as a result of the melting of the copper in the process of alloying with tin. Given that the nickel content is mostly unchanged when copper is melted, the As/Ni ratio will drop, meaning that alloyed or recycled objects are more likely to fall within the range of “chemical compatibility” with an Omani provenance. Assuming that arsenic is not added as an alloying element after trade, which given the new evidence for speiss production (see above and Chapter 2) could be debated, and that limited mixing took place, this method could be used to indicate the incompatibility of objects with As/Ni ratios over 10 with an Oman provenance. The objects that fall in the compatible range however can be made of copper from many different sources, and in particular recycled objects and tin bronzes that are more likely to have low As/Ni ratios. Begemann et al. used LIA to verify whether or not these “chemically compatible” objects were likely to come from Oman, but given the high overlap between the lead isotope signatures of various Near-Eastern ores (Figure V-28), conclusions must be seen as tentative at best.

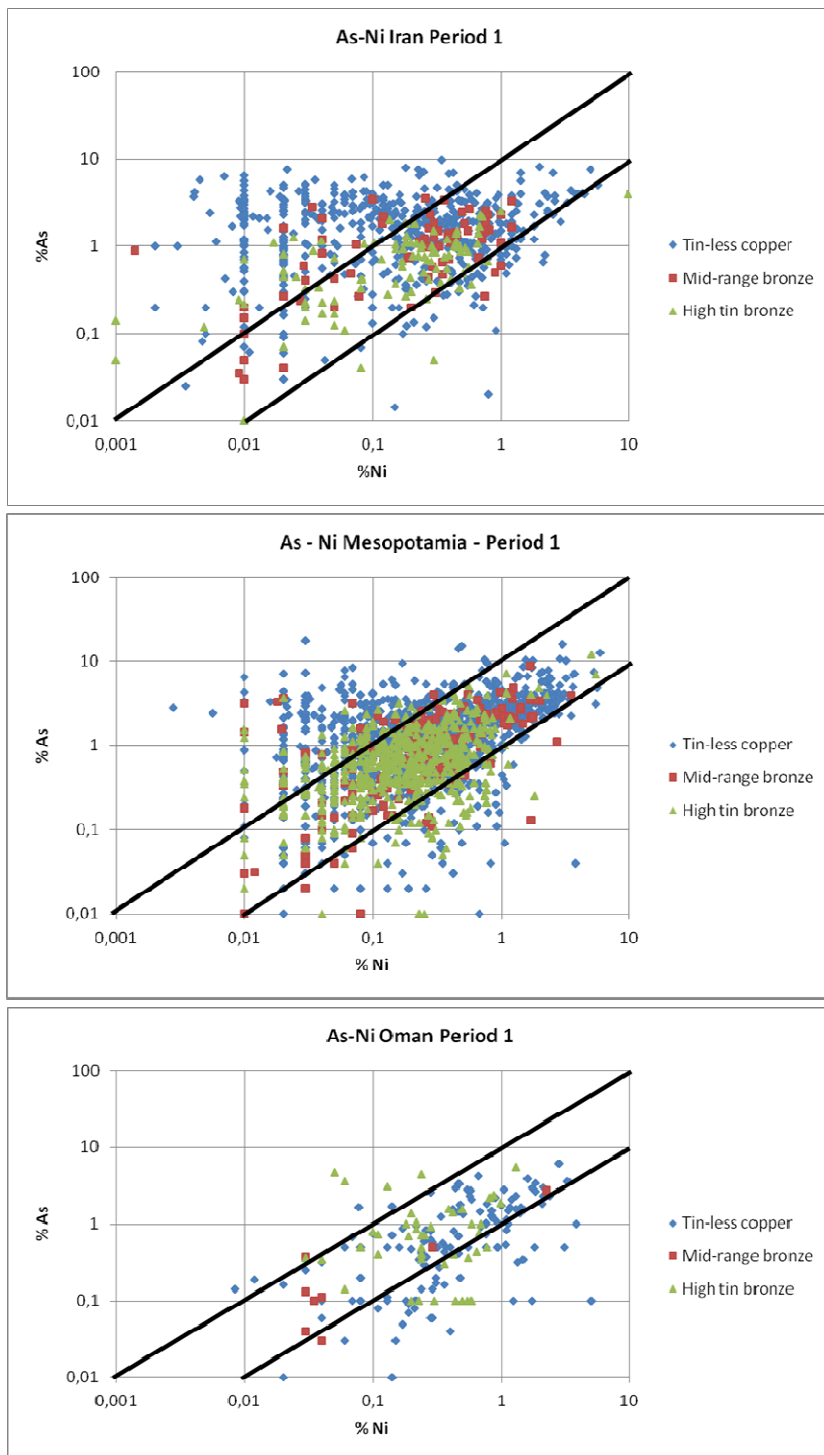


Figure V-34. Plot of the arsenic content against the nickel content of tin-less copper objects (<2% Sn), mid-range bronzes (2-6% Sn) and high tin bronzes (>6% Sn) for Iran (top), Mesopotamia (middle) and Oman (bottom) in Period 1 (early 3rd to mid 2nd millennium BC)

In this section we have seen the difficulties associated with the study of lead isotope abundance ratios, even when these are combined to chemical composition data. All in all,

very few convincing conclusions have derived from LIA studies. These conclusions can be summarized by the following points:

- Association of the cluster of Luristani objects with ratios of around 0.85  $^{207}\text{Pb}/^{206}\text{Pb}$ , 2.093  $^{208}\text{Pb}/^{206}\text{Pb}$  and 18.45  $^{206}\text{Pb}/^{204}\text{Pb}$  with ores from Deh Hosein and the Arak area.
- Import of copper with particularly high  $^{207}\text{Pb}/^{206}\text{Pb}$  ratios to Mesopotamia either in the form of tin-bronze or later alloyed with tin, and absence of this isotopic signature in objects from Oman and Luristan. A provenance in the Indus Valley has been proposed for this copper but more work would be needed to confirm this hypothesis. If it were to be the case, it would be remarkable, as it implies a complex trade system through the Persian Gulf but by-passing Oman.
- General compatibility of copper from both sides of the Oman Peninsula with Omani ores.

The difficulty of finding an archaeological structure within a large amount of chemical and LIA data, as seen here, is, of course, not specific to the Near-East and scholars have tried to develop methods to make sense of their data. An important example is the work of Rohl and Needham on objects from the Early Bronze Age in the British Isles (Rohl & Needham, 1998). They divided the artefacts into groups called IMP-LI (IMPurity and Lead Isotope compositions), such that an IMP-LI is constituted of a number of objects “belonging to a limited phase of activity” and having “both chemical and lead isotope characteristics falling in uninterrupted ranges”. They argued that these groups represented either metal from one source or from a coherent mixture of different metals (Rohl & Needham, 1998, p.84). This approach, however, will not be used in this thesis as it gives too much importance to the typological (and therefore chronological) distinctions and thus makes it impossible to assess continuity or changes in metal movement or recycling patterns (for a

more detailed discussion of the limitations of the IMP-LI system see Bray 2009, Ch.3). Instead, the following chapter uses a method proposed by Bray (2009) that defines metal groups based solely on their chemical composition without taking into account their dating or lead isotopy. Chronological and lead isotope patterns are then discussed for each group. This allows us to get a good idea of the changes that occurred in the Bronze and Iron Ages.

## 6. Conclusion

This chapter has continued to show the changing nature of copper metallurgy throughout the Bronze Age and the Iron Age. In general, it seems that the copper used in the Iron Age was cleaner (had lower levels of impurities) than in the Bronze Age. Alloying with lead, however, seems to have occurred broadly speaking with a similar frequency in both periods. Zinc was also used early on as an alloying element, and much more research would be needed on the subject to fully understand the scale of this practice.

A great deal of this chapter has been focused at trying to explain the remarkably obvious decrease in arsenic levels observed notably in Iran and Mesopotamia from the beginning of the 3rd millennium BC (Figure V-2). We have shown that this decrease was probably due to a combination of different factors, partly, but not exclusively linked the arrival of tin bronzes, including changes in ore sources and production processes but also an accumulation of recycled metal that would have lost some of its arsenic through successive transformations. However, the approach presented in this chapter hasn't enabled us to understand the contribution of each one of these factors.

The overview of the lead isotope data has also emphasized how complicated it is to assess the ore sources used in the Near-East and therefore how difficult it is to observe potential

changes in provenance in different time-periods. As seen above, attempts have been made to combine the lead isotope data and the chemical data but they haven't been very fruitful, mostly because of the overlap in the isotopic signature of ores sources and the changing nature of the chemical data.

In the following chapter we attempt to go further in our understanding of metal movement by using a different approach to the study of the copper composition: by dividing the database into different metal groups and observing, within each group, patterns of depletion in certain elements, we can put forward new hypotheses about the provenance of the ore, patterns of trade and the practise of recycling and how these factors changed between the Bronze Age and the Iron Age.



## Chapter VI. Metal flow and recycling

In the following section we attempt to go further in our understanding of the development of metallurgy in the Near-East by looking at the contribution of different potential ore sources and the movement of the metal. In order to do this, we adapt the two stage method developed by Bray in his thesis on British Isles metals (Bray, 2009) and further expanded in Bray and Pollard (2012): first each object of the database is assigned to a metal group based on its composition, then, for each group, we examine geographical patterns and chemical changes that can give us information on metal flow or recycling. This approach uses the fact that certain elements present in the copper are lost through oxidation when the copper is re-melted or reworked (Bray & Pollard, 2012, p.854). Instead of considering these losses as a hindrance to the study of the metal as has often been the case (Tylecote, 1970; McKerrel & Tylecote, 1972; Merkel, 1982, p.287), Bray turned things around and used them as the basis of his analysis: “if we shift our theoretical focus somewhat, these potential chemical changes under melting could present us with an extremely powerful interpretative tool” (Bray & Pollard, 2012, p.855).

The nature of the data used in this thesis is however quite different from Bray’s dataset. The dynamism of the metal industry in and around Iran, with many ores sources and production centres, makes it a fascinating area to study the beginnings of metallurgy, but also comes with its challenges. The most significant challenge is the typological

variety of the objects. Bray was able to compare hundreds of flat axes of very similar typology from different areas of Great-Britain. Here, comparing objects of similar typology would leave us with too few objects to draw conclusions of any statistical significance and we are therefore unable to go into much typological debate. Another difference between the two regions is the chronology. As explained in Chapter 3, the objects aren't always precisely dated and for this reason, we chose to use broader time scales than in Bray's work: a division into thirds of a millennium and the longer Period 1 and Period 2 defined above.

What these distinctions mean for the present work is that we cannot expect the findings of this thesis to have the same level of clarity and precision than Bray's work. The patterns obtained will be blurred by the signals of many different ore sources, trade systems and technological practices. We can nevertheless use the same broad principles. Indeed whether in Great Britain or in the Near East, the division of the assemblage into a set number of metal groups independent of time and geography, is a simple way of starting to tease out metal movement and chronological changes in metal production. Secondly, the laws of thermodynamics are of course the same in Great Britain and the Near-East, and, if the practise of recycling metal objects through re-working and re-melting was common, we can expect from taking into account losses in arsenic and antimony in particular in the Near-Eastern data to derive some meaningful information on the metallurgy of the region.

Furthermore, contrary to Early Bronze Age Great Britain, in the Near-East we are fortunate to know of texts dealing with the production and trade of metal objects and we can therefore compare the results of our analysis with the texts and through this improve our understanding of both the analytical results and the writings. Finally, we also know of more copper and tin ingots in the Near-East and Eastern Mediterranean than in

Britain, which means that the mechanisms of trade were different in both area, but it also means that we have access to primary fresh metal as a basis for comparison with more processed objects. Unfortunately only few ingots were analysed. They are pointed out throughout this chapter when they exist.

Because of the lack, for Iranian metalwork, of a history of synthetic analysis and because of the need to make an unbiased assessment of the data, we have chosen for this thesis to define entirely new metal groups. As we have seen in Chapter 3, a few authors have defined metal groups for their datasets but these were mostly aimed at summarizing the compositional data rather than for analytical purposes (Moorey, 1971; Mahboubian, 1997; Hopp et al., 1992). An important part of our work has therefore been to define groups based on the chemical composition of the objects. This is presented in the first section of this chapter. The second section examines geographical and chronological variations in the ubiquity of the most important groups, as well as patterns in the lead isotope data within each group and depletion patterns in their chemistry.

#### 1. Defining metal groups

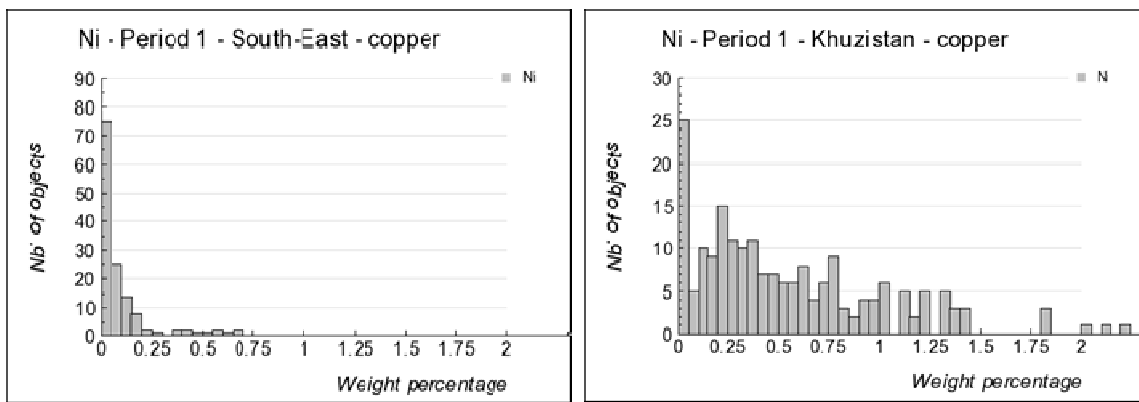
To define the metal groups used in this chapter, we have chosen to stay in line with a long tradition of archaeometallurgical studies going back to Coghlan and Case (1957) and before, and use the following four elements: arsenic, nickel, antimony and silver. Indeed these elements have the benefit of having almost always been recorded (see Chapter 3) and tend to be diagnostic of an ore source (Bray & Pollard 2012, p.854). We have seen above that cobalt is another element that shows a geographical pattern, being

present mostly in Oman. However, since it has not systematically been sought for in all of the studies that constitute our database, using it to define metal groups would limit the range of objects that can be assigned to a category.

Our first step was to consider the 16 possibilities of absence or presence of each one of these four elements. This approach has the advantage of being unbiased and all inclusive: it does not force any prior knowledge of ore sources onto the chemical evidence. It is a useful first filter that is then refined in the second stage of this approach using archaeological evidence, dating, lead isotopes, etc. Each category is designated by a binary number, 0 representing absence of an element, 1 representing its presence. The 16 resulting categories are 0000, 0001, 0010 and so on, up to 1111. The first number represents arsenic, the second one nickel, the third antimony and the fourth silver. 0101 for example would represent objects with high levels (presence) of nickel and silver but low (absent) arsenic and antimony. The objects of all of these groups can have tin, lead, etc. in any concentration. The definition of these groups is meant to be linked to the copper sources and does not take into account the addition of alloying elements.

To define the presence or absence of an element we have had to choose cut-off values. These are to some degree arbitrary as continuous ranges of concentrations from 0 to several percents are observed for each element and depend not only on the composition of the copper ore but on a wide variety of other factors such as the production processes (including the intentional alloying of arsenic in particular), and the subsequent transformations that might have occurred in the life of the metal such as re-working, or re-melting or recycling. As we have seen in Chapter 3, the percentages entered in our database also depend to some degree on the analytical methods used in the original publication. To aid our choice of cut-off values, we have looked at distribution diagrams of each of these elements for each region in Period 1 and Period 2. For arsenic, the

choice of a cut-off value was made difficult by the wide variety of distribution patterns as shown in Figure V-11. 0.25% was chosen as it proved useful in the description of many of the arsenic distributions presented in the Chapter 5. The average levels of nickel being lower than arsenic, we have chosen a lower limit of 0.1% of that element. We can clearly see on Figure VI-1 that, given this limit, some regions such as the South-East have mainly nickel-free objects, while some others like Khuzistan mostly present objects for which nickel is defined as being present.



**Figure VI-1. Distribution of the nickel content of tin-less copper objects (<2% Sn) in Period 1 (early 3rd to mid 2nd millennium) in the South-East (left) and Khuzistan (right)**

For antimony and silver, the average values are even lower than for nickel. It therefore seemed logical to choose a lower cut-off value for these two elements. Indeed Figure VI-2 shows that a cut-off of 0.1% for silver would cut-out half of what looks like a believable almost gaussian distribution centred around 0.1% for regions such as Luristan in Period 1 for tin-less copper or Azerbaijan in Period 2 for bronze objects.

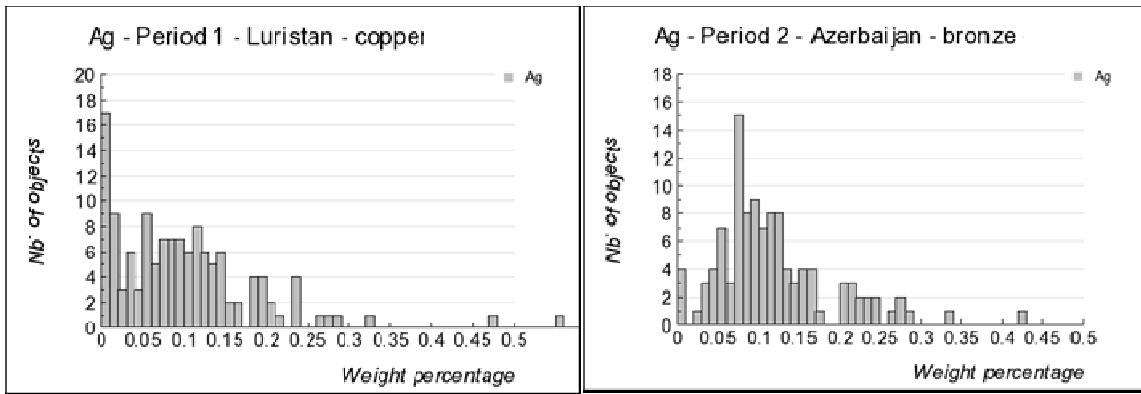


Figure VI-2. Distribution of the silver content of tin-less copper objects (<2% Sn) in Period 1 (early 3rd to mid 2nd millennium) in Luristan (left) and bronze objects (>2% Sn) in Period 2 (late 2nd to mid 1st millennium BC) in Azerbaijan (right)

For antimony, it would have meant that regions like Luristan and Gorgan, which clearly contrasts in Period 1 in terms of antimony levels (Figure VI-3) would have been considered similar in that they both presented almost no objects with a presence of antimony.

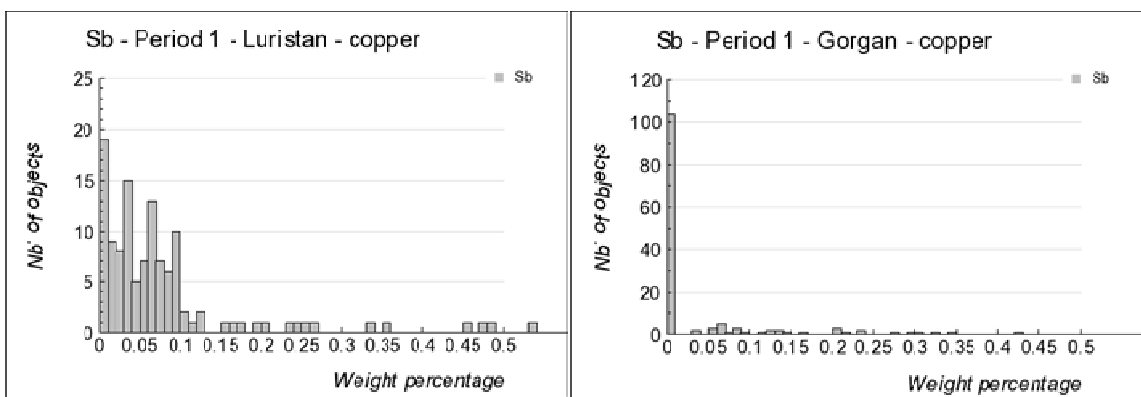


Figure VI-3. Distribution of the antimony content of tin-less copper objects (<2% Sn) in Period 1 (early 3rd to mid 2nd millennium) in Luristan (left) and in Gorgan (right)

However, given the detection limits of some of the instruments used to collect the data, a very low cut-off value would have had no significance. Not many of the publications used for our database gave the detection limits for each element, but as an example we can cite Frame's electron microprobe analysis of Tepe Godin material in which she gives the average detection limits for Sb at 0.043% and Ag at 0.044% (Frame, 2010).

Weeks gave the detection limits for his PIXE analysis of Oman material at 0.070% for Sb and 0.024% for Ag (Weeks, 2003, pp.72-75). Given these restrictions, we have chosen for both elements a cut-off value of 0.05%.

Using these cut-off values of 0.25%, 0.1%, 0.05% and 0.05% for As, Ni, Sb and Ag respectively, we have assigned each object of the database for which all four of these elements were analysed to one of the 16 categories defined above. The values used to assign an object to one of these groups are the corrected values taking into account the addition of tin as explained at the beginning of Chapter 5. The results of this assignment are shown on Figure VI-4. We can see that the three most common groups in our database are groups 1100, 1111 and 1110, i.e. groups representing objects with respectively arsenic and nickel only, all four elements, and arsenic, nickel and antimony with no silver. These three groups all represent more than 12% of the assemblage and together represent more than 40% of it. The other groups represent between 0.9% of the assemblage for the rarest, nickel and antimony, and 8.8% for group 0000. We call the latter “Clean Metal” in this thesis, by which we mean that it contains less than our cut-off values of arsenic, nickel, antimony and silver. This is not to say however that objects assigned to this group cannot contain other elements and in particular tin.

Figure VI-4 shows that all 16 metal groups are populated. However we are not implying that the 16 groups represent 16 different sources within our study area. One metal group can indeed be populated by objects of several different sources as shown below. Some of the groups are also populated by objects that are made of recycled copper or a mixture of copper from different sources and do not represent an initial source. As mentioned above, the division into 16 groups is a useful first step in the analysis that then needs to be refined with other types of evidence.

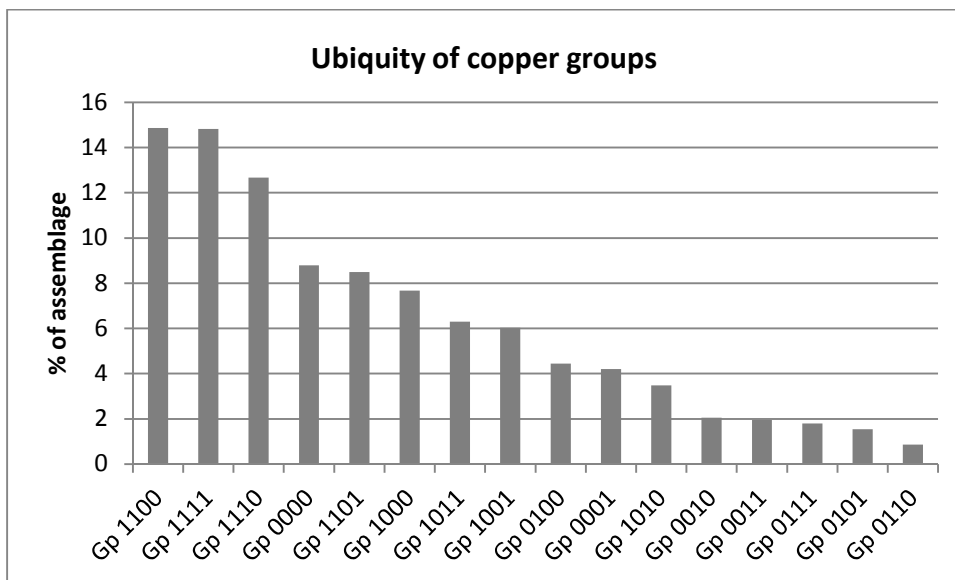


Figure VI-4. Ubiquity of the 16 different copper groups

Figure VI-5 shows the number of objects in each group, and the regions where these objects were found in Period 1 and in Period 2. We can immediately see important differences between the two periods and in particular a bigger preponderance in Period 1 of categories defined by the presence of both arsenic and nickel. We can also see on these graphs that most metal groups are represented by objects from many different regions. A few, however, jump out as being constituted of objects coming almost exclusively from a single region. This is the case for example of the nickel, antimony and silver group, for which, in Period 2, more than 80% of the objects come from Iranian Azerbaijan. This suggests a very localised use of this particular type of copper.

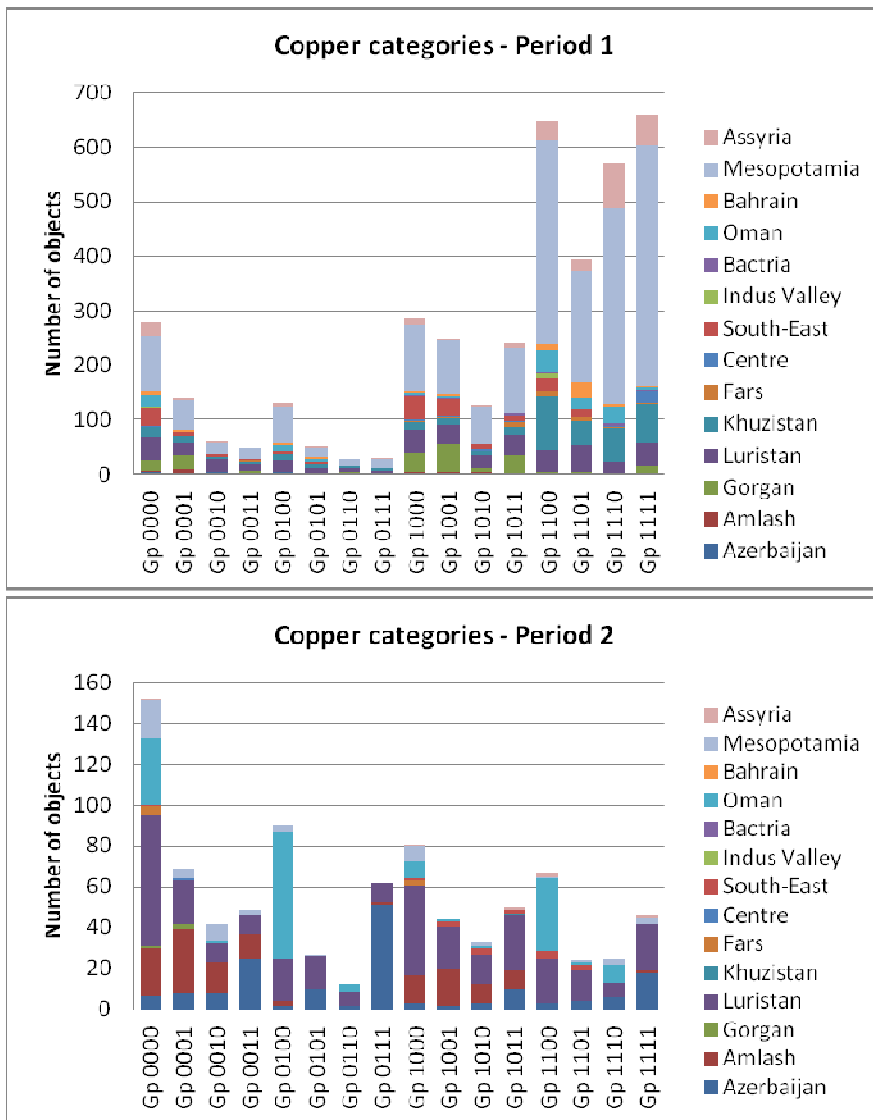


Figure VI-5. Number of objects falling in each copper category in Period 1 (top) and Period 2 (bottom)

The following section is dedicated to the study of the three prevalent groups as seen on Figure VI-4: AsNi, AsNiSbAg and AsNiSb, to which we have added Ni Only metal, As Only metal, and Clean Metal. Indeed, the relationship between group AsNi and group Ni Only, and the relationship between group As Only and Clean Metal can give us some information on the practice of recycling (see below).

Table VI-1 summarizes the composition of the six main groups studied in this thesis. These groups are to be understood as metal types, based on the chemical composition of

the objects. It must be emphasized that the metal from one group or type can come from several different ore sources.

	As	Ni	Sb	Ag
AsNi	$\geq 0.25$	$\geq 0.1$	$< 0.05$	$< 0.05$
AsNiSbAg	$\geq 0.25$	$\geq 0.1$	$\geq 0.05$	$\geq 0.05$
AsNiSb	$\geq 0.25$	$\geq 0.1$	$\geq 0.05$	$< 0.05$
As Only	$\geq 0.25$	$< 0.1$	$< 0.05$	$< 0.05$
Clean	$< 0.25$	$< 0.1$	$< 0.05$	$< 0.05$
Ni Only	$< 0.25$	$\geq 0.1$	$< 0.05$	$< 0.05$

Table VI-1. Definition of the copper groups used in this thesis

Figure VI-6 shows the proportion of the assemblage represented by each group over time. As we can see, there is a clear predominance in the 4th millennium BC of the AsNiSbAg group that represents more than a third of the assemblage in the early 4th millennium BC.

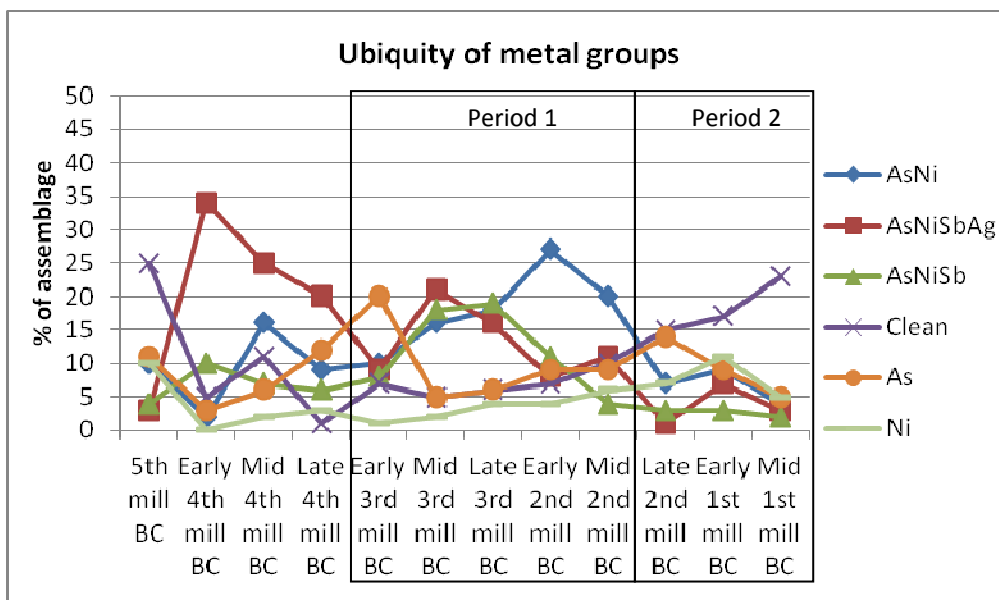


Figure VI-6. Chronological evolution of the percentage of the assemblage represented by each of the studied metal group

In the mid to late 3rd millennium, the assemblage is mostly constituted of AsNi, AsNiSbAg and AsNiSb metal. These three groups all have very similar ubiquities

during this period: between 16 and 21% of the assemblage. In the beginning of the 2nd millennium however AsNiSbAg and AsNiSb become less common, while the ubiquity of AsNi increases up to 27% in the early 2nd millennium before it starts declining as well. As Only and Clean Metal increase steadily from the mid 3rd millennium BC and, at the transition between Period 1 and Period 2, Clean Metal becomes more common than AsNi. Clean Metal continues to increase during Period 2, while As Only starts to decline. Ni Only shows a steady increase throughout both periods and its ubiquity only drops in the mid 1st millennium.

Going back to the question of the transition between the Bronze Age and the Iron Age at the centre of this thesis, it seems that the changes in metallurgy that occur from one period to another do not only have to do with iron or tin as discussed in previous chapters, but also with changes in the copper used by the ancient metalworkers. Figure VI-6 shows that the transition between these two periods is marked by the decline of the common metal groups of the 3rd millennium, while groups with fewer impurities (As Only and Ni Only) continue a gradual increase that started in the 3rd millennium. Many factors would probably have contributed to these changes: trade routes shifting, mines opening and closing, recycling, etc. To try and unravel some of the mechanisms behind these changes, the following section takes each group separately and looks at geographical and chronological patterns as well as the chemical and lead isotope data.

## 2. Type 1100: Arsenic Nickel

Group AsNi is constituted of objects containing over 0.25% As and over 0.1% Ni but under 0.05% of both Sb and Ag. As shown above, this group was one of the three main

groups of Period 1, and was particularly important in the early 2nd millennium when it represented 27% of the assemblage. However this importance then began to decrease and in Period 2 it represented less than 10% of the analysed objects. The number of objects that fall in the AsNi group for each period are summarized in Table VI-2.

	<b>Period 1</b>	<b>Period 2</b>
<b>Number of objects</b>	648	67
<b>Percentage of the assemblage represented by AsNi</b>	16 %	8 %
<b>Ubiquity of tin-bronze (&gt;2% Sn)</b>	35 %	74 %

Table VI-2. Number of objects that fall into Group AsNi in Period 1 (early 3rd to mid 2nd millennium BC) and in Period 2 (late 2nd to mid 1st millennium BC), percentage of the assemblage that this represents for each period and ubiquity of tin-bronze for this group

a. Arsenic Nickel: geographical patterns

The two maps of Figure VI-7 represent the proportion of objects falling into the AsNi group in each one of the regions for which more than 10 objects were analysed. An uneven distribution of this group is immediately obvious: in Period 1 it is almost absent in northern Iran (Azerbaijan, Amlash and Gorgan) but much more common in regions surrounding the Persian Gulf. The regions with the highest percentages are the Indus Valley with 85% and Fars with 60%. AsNi is also well represented Oman and Khuzistan with 28% of the assemblage in both cases. It is worth mentioning that for the Indus Valley this very high percentage is in fact only represented by 11 objects out of the 13 that we were able to assign to a metal group. Very broadly speaking, it seems that the ubiquity of this metal type decreases the further we get from the Indus Valley, with Luristan and Assyria at the very end of the Persian Gulf trade route having only 11 and 12% of AsNi metal respectively. Based solely on this geographical pattern, it is possible to envisage that AsNi metal was traded through the Gulf to western Iran and Mesopotamia from a source in the Indus Valley and perhaps others in Elam or Oman.

Overland trade from the Indus Valley to Bactria and south-eastern Iran could also be a possibility.

It is worth mentioning however that the AsNi objects from the Indus Valley all date from the late 3rd millennium BC and come from the site of Harappa, relatively far upstream, away from the Persian Gulf. Some objects dating from the same period and coming from Mohenjo-Daro, a site further downstream of the Indus, were also analysed (Kenoyer & Miller, 1999), but their silver content was unfortunately not measured. There, only 16% of the objects had arsenic and nickel but no antimony (and could therefore be attributed to either group AsNi or AsNiAg). The most common type (37%) was objects presenting arsenic, nickel and antimony and therefore falling into group AsNiSb or group AsNiSbAg. Although this doesn't undermine the possibility of a Persian Gulf trade originating in Harappa, it clearly shows that AsNi wasn't the only metal type used in the Indus Valley.

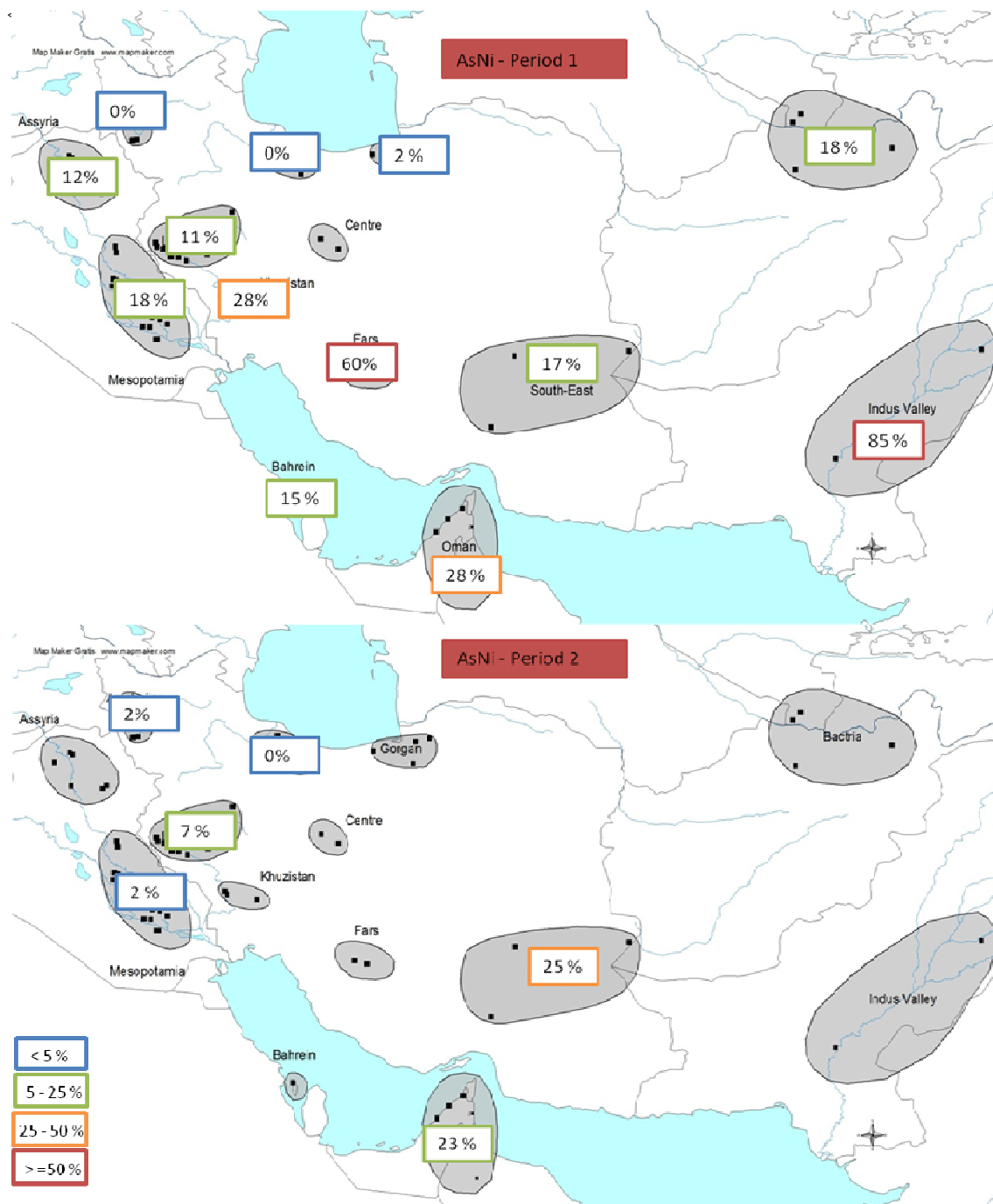


Figure VI-7. Percentage of the assemblage of each region represented by Group AsNi objects for Period 1 (top) and Period 2 (bottom). Percentages are only given for regions for which more than 10 objects were analysed.

In Period 2, group AsNi is still almost entirely absent from northern Iran. In Oman and the South-East, the ubiquity of this group remains quite high, close to the levels of Period 1: respectively 23 and 25%. In Mesopotamia however, AsNi is much less common than in Period 1: it drops from 18% of the assemblage to 2%. From this map it

seems that either the sources or the trade routes that brought AsNi copper to Mesopotamia were discontinued. This isn't surprising given the fall of the Harappan civilisation in the early to mid 2nd millennium BC and the resulting interruption of the Persian Gulf trade as mentioned in Chapters 1 and 2. The continued presence of AsNi copper in Oman and the South-East could suggest the use of local sources, which is possible given the presence of copper deposits in both of the regions (see Chapter 2).

#### b. Arsenic Nickel: chemistry and lead isotope ratios

This section aims to explore in detail the patterns observed on the ubiquity maps by looking at the chemical and lead isotopes contents of the objects. Archaeometallurgical research as well as practises from the modern copper industry agree on the fact that “a bronze object will be depleted in certain of its metals depending on how many times it has been reheated, and to what temperature” (Bray & Pollard, 2012, pp.854-55). The degree to which elements are lost depends on their oxygen affinity but also on a number of other factors (agitation of the metal, solubilities, activities, volatilisation and partial pressures) (Bray & Pollard, 2012, p.854). Bray's comparison between the chemical compositions of the blades and the rivets of a group of halberds has confirmed the fact that arsenic, due to its high volatility, is lost in a much higher percentage than the other elements (Bray, 2009, Ch.3). Antimony is also lost relatively easily while nickel and silver tend to be retained in the metal. Bray has demonstrated that this depletion, rather than only being a hindrance to metallurgical studies, can be used to infer patterns of metal flow. He noted that the levels of arsenic and antimony drop the further away we get from the source of the metal, due to the increased likelihood that the objects would have been subjected to multiple working or melting events. Indeed we can imagine that

new owners would have wanted to rework an object, or recast it to a shape that fitted better with the tradition of its new locality (Bray, 2009; Bray & Pollard, 2012, p.861).

This hypothesis can be backed by the existence of regional differences in the typology of the metal assemblage of Iran and its neighbours. Moorey, in particular, reviewed the different metalworking traditions that existed in Iran during the Bronze and Iron Ages. He showed that, while many parallels indicate strong trans-Iranian contacts, there were also marked typological distinctions between different cultural horizons. The repertory of metal objects from Luristan, for example, shows many parallels to its neighbours to the west and south: it was “part of a common tradition embracing much of central and southern Mesopotamia into the Zagros foothills” (Moorey, 1982, p.89). However, the zoomorphic decorations observed on cast weapons and tools dating from the mid 3rd millennium BC onwards are a distinctive style, not found in Mesopotamia, where only the plain counterparts of these objects occur (Moorey, 1982, p.89). Another example is the repertory of objects from the site of Tepe Hissar in the 3rd millennium BC, described by Moorey as follows: “A standard range of cast copper tools and weapons was mass-produced, as in Western Iran, but within a distinctive typological range with relatives elsewhere in northern Iran. Local fashions are still more evident in such lost-wax castings as the “wands” and zoomorphically decorated maceheads. Animal pendants belong to a universal fashion; but here turtles mark more local tastes” (Moorey, 1982, p.92). A final example from the Iron Age site of Hasanlu can be given. There, the metal objects, and in particular the daggers, arrowheads and spearheads, show very little parallels with objects from Assyria, while parallels in other classes of objects clearly indicate trade between these two regions (Thornton & Pigott, 2011). Given these regional specificities, it is possible to assume that objects were altered

when they “moved away” from their source and therefore that the model of depletion described by Bray is applicable here.

However, as described in Chapter 3, the fragmentation of our database, and in particular the fact objects from different regions have usually been analysed as part of different projects, using different analytical methods, means that caution must be taken when using averages. The necessary division into metal groups, time periods, regions and tin-less copper/tin-bronze means that we are often left with very few objects to study the chemistry of a certain metal type in a certain region. Regions for which less than 10 objects were available were systematically deleted from the graphs showing chemical averages below, but care must still be taken with the remaining regions. That is why small differences in chemistry are overlooked in this section and we only comment on broad trends.

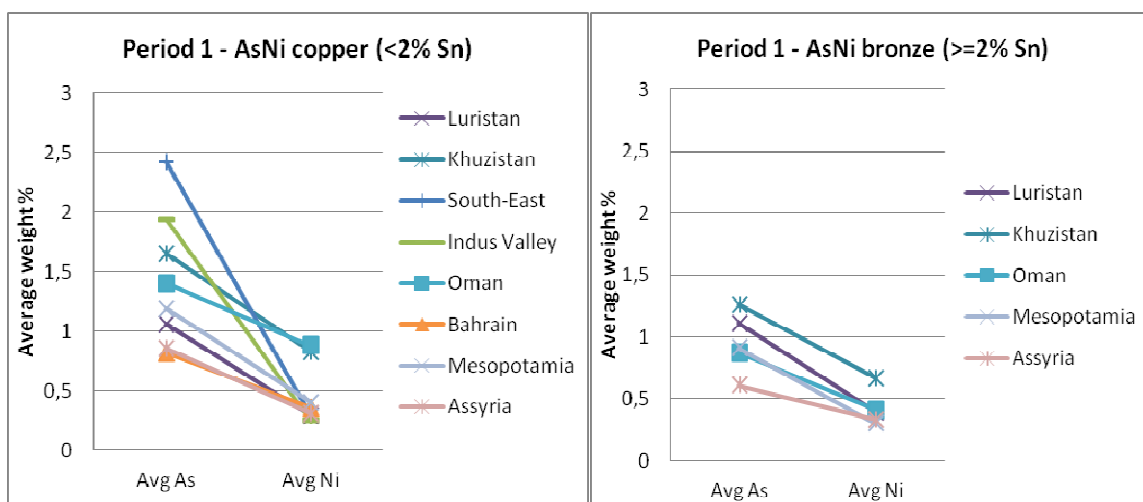


Figure VI-8. Average levels of arsenic and nickel for tin-less copper objects (<2% Sn, left) and tin bronze objects (>=2% Sn, right) of the Arsenic Nickel group in various regions in Period 1 (early 3rd to mid 2nd millennium BC)

Figure VI-8 shows the average levels of As and Ni in tin-less copper objects and bronze objects of the AsNi group. Firstly we can see for most regions (with the exception of Luristan) a lower arsenic content in the bronze objects than in the tin-less copper objects, which is not surprising. Indeed, if bronze is produced by the mixing of molten

tin and molten copper or by putting cassiterite in contact with molten copper (see Chapter 2), the melting of the copper results in a loss of its volatile elements: arsenic for the metal group studied here. Once again, we must emphasize that the averages are calculated on the corrected concentrations as explained in Chapter 5. The drop in arsenic content observed between tin-less copper and bronze is therefore not due to the tin washing out the arsenic signal, but to true losses due to melting and oxidation (or to an altogether different ore source). The fact that this trend is observed here is not unexpected, but it (and the same observation for the other metal groups, as presented below) confirms that simple thermodynamical mechanisms are visible in the archaeological record and can therefore be used to understand more about metal flow.

Secondly, on the graph showing the averages for tin-less copper, we can see two different levels of nickel: one around 0.3-0.4% and the other around 0.85%. The high nickel regions are Oman and Khuzistan, two regions for which high nickel contents have already been commented on. Malfoy and Menu in particular commented on the metal used at Susa in the 4th and 3rd millennium BC and suggested that it came from Anarak in the beginning of this period with a later shift to Omani ores (Malfoy & Menu, 1987). The pattern observed here highlights once again the similarity between the copper used in these two regions but the similarity between the trace element set of copper from the Anarak region and the Omani copper means that it is difficult to tell whether Elam would have imported its copper from Oman. The arsenic level is slightly higher in Elam than in Oman, which could be an argument against the fact that this AsNi copper was imported from Oman. The data available from ingots is also unclear: only five ingots from Khuzistan have been analysed. These are all bun-shaped and dated from the 3rd millennium BC and they all belong to this AsNi group. It has previously been argued that these ingots might be Omani imports where this shape is quite

common (Pigott, 1999b), but in Oman, of the sixteen 3rd millennium BC ingots analysed, only three were of the AsNi group while six were AsNiAg and five NiAg attesting to a relatively wide range of compositions produced in Oman<sup>5</sup>. All in all, it is at present impossible to state whether the copper used in Elam came from Oman, despite the very similar chemical signature of the copper in both regions.

In the regions for which a lower nickel average is observed, we can see a general depletion in arsenic when we go from the eastern regions to the western regions. This confirms the general trend suggested by the geographical pattern of an east to west trade of AsNi metal originating in the Indus Valley, or perhaps in south-eastern Iran. It is reassuring to see that the regions with the lower ubiquity of AsNi metal (Luristan and Assyria) also have the most depleted metal, suggesting once again the validity of this type of analysis.

	Luristan	Khuzistan	South-East	Indus Valley	Oman	Bahrain	Mesopotamia	Assyria
Ubiquity of bronze (%)	63.3	14.1	9.1	18.2	30.0	9.1	42.0	33.3

Table VI-3. Ubiquity of tin bronze (>2% Sn) amongst the AsNi objects in Iranian and neighbouring regions in Period 1 (early 3rd to mid 2nd millennium BC)

Table VI-3 shows the ubiquity of bronze in regions with more than 10 AsNi objects. We can see that the regions where we suggest the AsNi trade might have originated (the Indus Valley and the South-East) are amongst the regions with the lowest ubiquities of bronze. On the contrary, the regions at the receiving end of the Persian Gulf trade route (Luristan, Mesopotamia and Assyria) are the ones where bronze was most common.

<sup>5</sup> Since the submission of this thesis, the publication of 22 more chemical datasets for Omani ingots from the 3rd millennium BC (Prange, 2001) was brought to my attention. They too show the prevalence of the AsNiAg (9 ingots), AsNi (6 ingots) and NiAg (6 ingots) groups. Interestingly the same publication also has 16 datasets for ingots found on Bahrain. Of these, the majority (9) fall in the AsNi group with 4 in the Ni Only group. This confirms that AsNi copper must have been traded through the Gulf but the exact origin of this copper and of that of the AsNiAg group remains uncertain.

This trend would again have to be verified when more data, especially for the Indus Valley, becomes available, but it could indicate that AsNi was traded as copper, only then to be alloyed with tin.

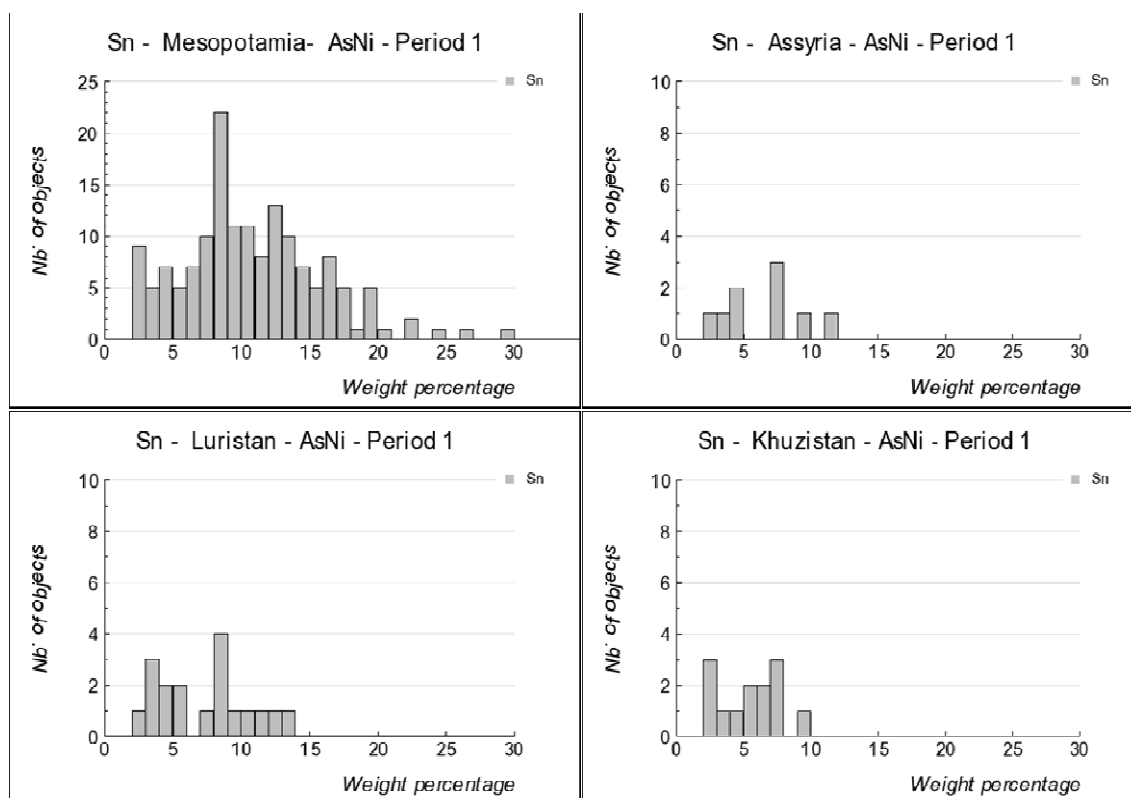
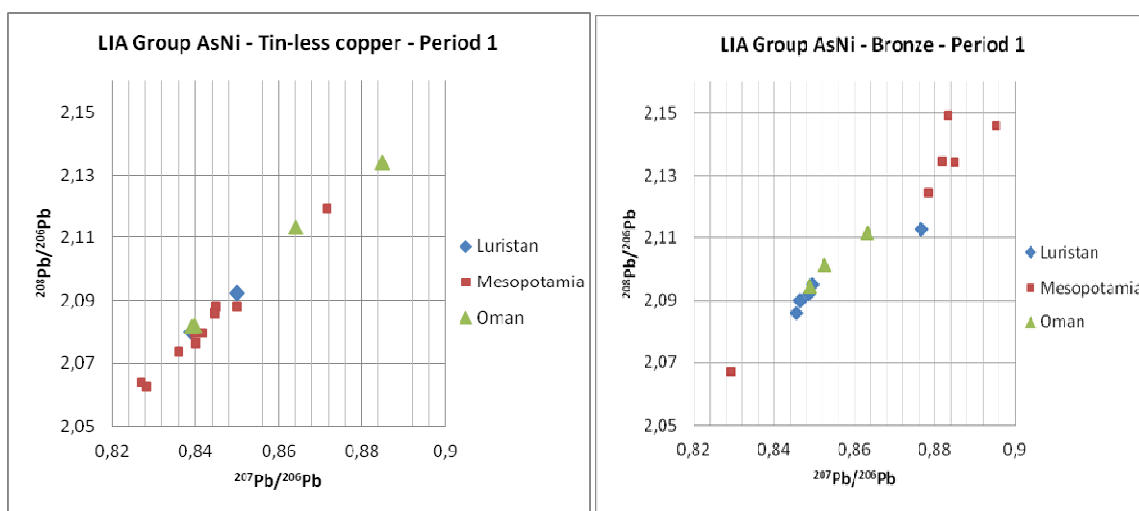


Figure VI-9. Distribution of tin content of AsNi tin bronze objects (>2% Sn) in Mesopotamia, Assyria, Luristan and Khuzistan in Period 1 (early 3rd to mid 2nd millennium BC)

This hypothesis is reinforced by the tin distribution in AsNi bronze objects shown in Figure VI-9: indeed these show, in the regions at the receiving end of the AsNi trade, an important number of objects in the 6-15% Sn range and certainly a higher ratio of high tin bronzes (>6% Sn) to mid-range tin-bronze (2-6% Sn) than for objects of the other two main groups of Period 1 as shown in the next sections. This indicates that a minimal amount of these tin-bronzes have been subjected to recycling or mixing after the addition of tin, making it likely that the tin was added at a late stage, probably after it had travelled from east to west. Perhaps strengthening this argument is the fact that all seven of the analysed ingots from this group were tin-less copper, indicating that they

were probably traded before alloying. However these ingots come from Oman and Elam where the composition of AsNi copper was, as we have seen, slightly different and the analysis of ingots from Mesopotamia would be particularly interesting in this context.

Figure VI-10 shows the lead isotope ratios for objects of the AsNi group. We can see that very different ore sources were used for the copper and the bronze used in Mesopotamia. Both series of objects date from the same time periods (mainly early and mid 3rd millennium BC), but the copper objects almost all have  $^{207}\text{Pb}/^{206}\text{Pb}$  ratios under 0.850, while for the bronze objects this ratio is almost always over 0.875.



**Figure VI-10. Lead isotope ratios for tin-less copper (<2% Sn) and bronze (>=2% Sn) objects of the AsNi group dating from Period 1**

The ratios for the copper fall in a region of the graph that could correspond to many different ore sources, notably Oman, the Caucasus or Afghanistan (see Figure V-28). However, given the low nickel average for the AsNi Mesopotamian copper, Oman can probably be excluded as its source. The ratios for the bronze objects are closest to the signature of ores from Gujarat but as seen in Chapter 5 do not quite overlap and much more data on both ore sources and objects would be necessary to pin-point the source of the metal used for both copper and bronze. It would also be very useful to extend the

area of study to the west and the north to investigate whether Mesopotamia could have received its metal from Anatolia the Levant or the Caucasus for example, but this is beyond the scope of this thesis.

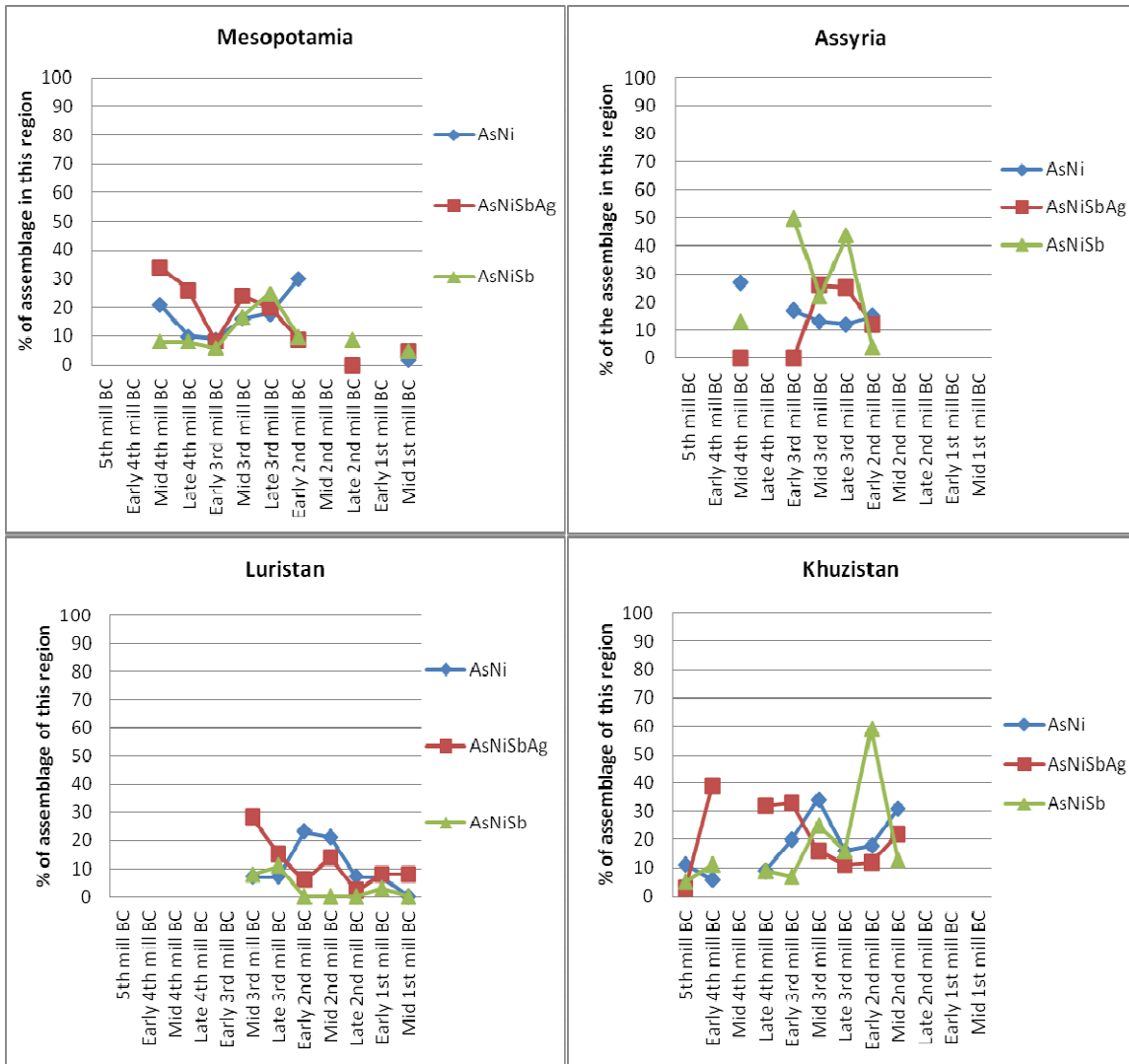


Figure VI-11. Chronological evolution of the percentage of the assemblage represented by the AsNi, AsNiSbAg and AsNiSb groups in Mesopotamia, Assyria, Luristan and Khuzistan

Moreover, it would be interesting to have some LIA data for Mesopotamian objects from the beginning of the 2nd millennium BC, as AsNi metal became more common in Mesopotamia at that time, unlike the other two big groups of the 3rd millennium (AsNiSbAg and AsNiSb) that started declining (see Figure VI-11). For now, all we can say concerning Mesopotamia, is that the AsNi copper used for tin-less copper objects

and for bronze objects came from different sources in the early to mid 3rd millennium BC, unless the process used to make the tin bronze, perhaps involving mixing, considerably shifted the LIA signature.

As shown on the ubiquity maps (Figure VI-7) the AsNi type of copper was much less common in Period 2. Only in Oman and the South-East did it remain present in a relatively high proportion with respectively 23 and 25% of the assemblage. Unfortunately, the amount of data available do not allow for the study of chemical averages or of lead isotope ratios. For both regions, the chronology is also very patchy (Figure VI-12). In Oman, it is impossible to say when AsNi metal became important, but it appears that in Period 2 it remained present in about the same proportions as in Period 1. In the South-East, AsNi seems to have gained in importance in the late 3rd millennium and subsequently decreased only slightly throughout the rest of Period 1 and Period 2. The continuity for these two regions between Period 1 and Period 2 possibly indicates the use of local sources rather than a reliance on trade from Harappa for example.

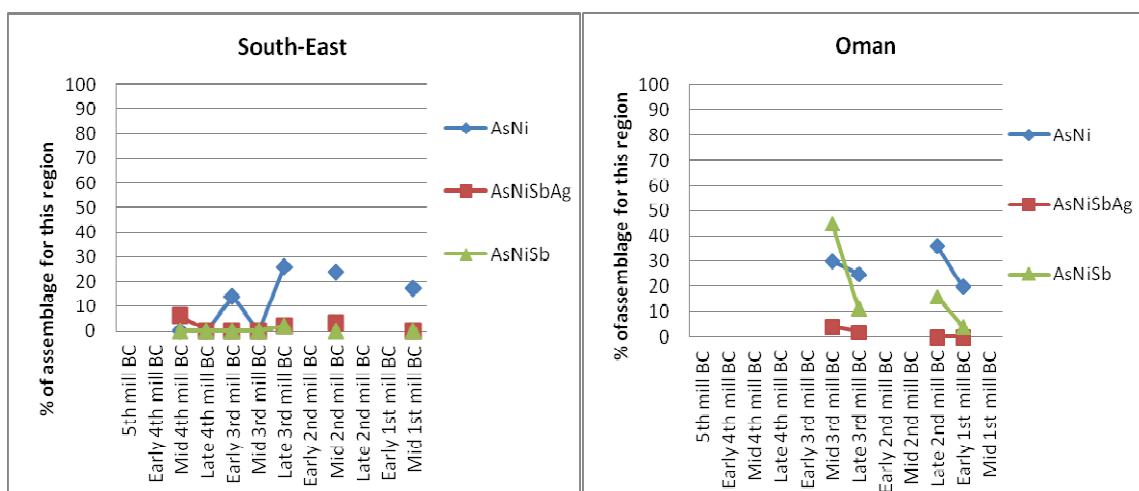


Figure VI-12. Chronological evolution of the ubiquity of AsNi, AsNiSbAg and AsNiSb in south-eastern Iran and Oman

c. Relationship between the Arsenic Nickel group and the Nickel Only group

As explained above, the remelting of copper of AsNi type would have resulted in a copper depleted in arsenic while the nickel content remained more or less unchanged. After successive remelting events, the copper of the AsNi type can therefore transition into the Nickel Only group. Bray has demonstrated that it is possible to observe the progressive accumulation of recycled objects through the steady increase of the ubiquity of “recycled” metal types (Bray, 2009; Bray & Pollard, 2012, p.862). Figure VI-13 shows the ubiquity of AsNi and of Nickel Only for the entire assemblage. We can indeed see a slow increase in the ubiquity of Nickel Only metal probably at least in part as a result of this mechanism (as seen below, Ni Only ore sources can also have played a role in this increase). It must be kept in mind that the degradation of metal of other groups and in particular NiSb (very rare, see Figure VI-4) and AsNiSb (see below) would have also contributed to the accumulation of Ni Only metal.

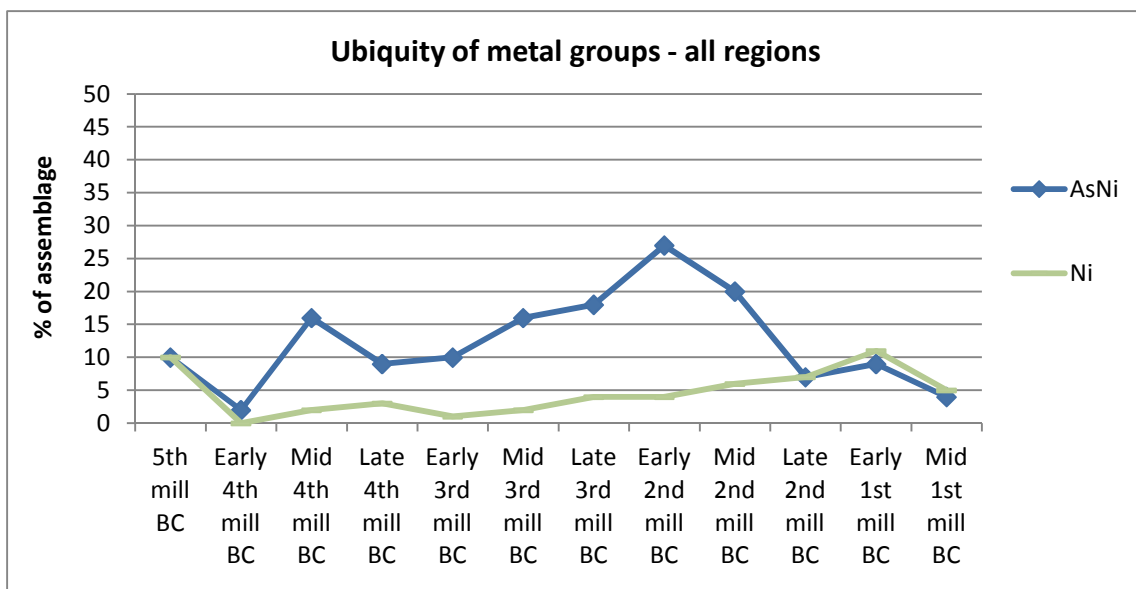
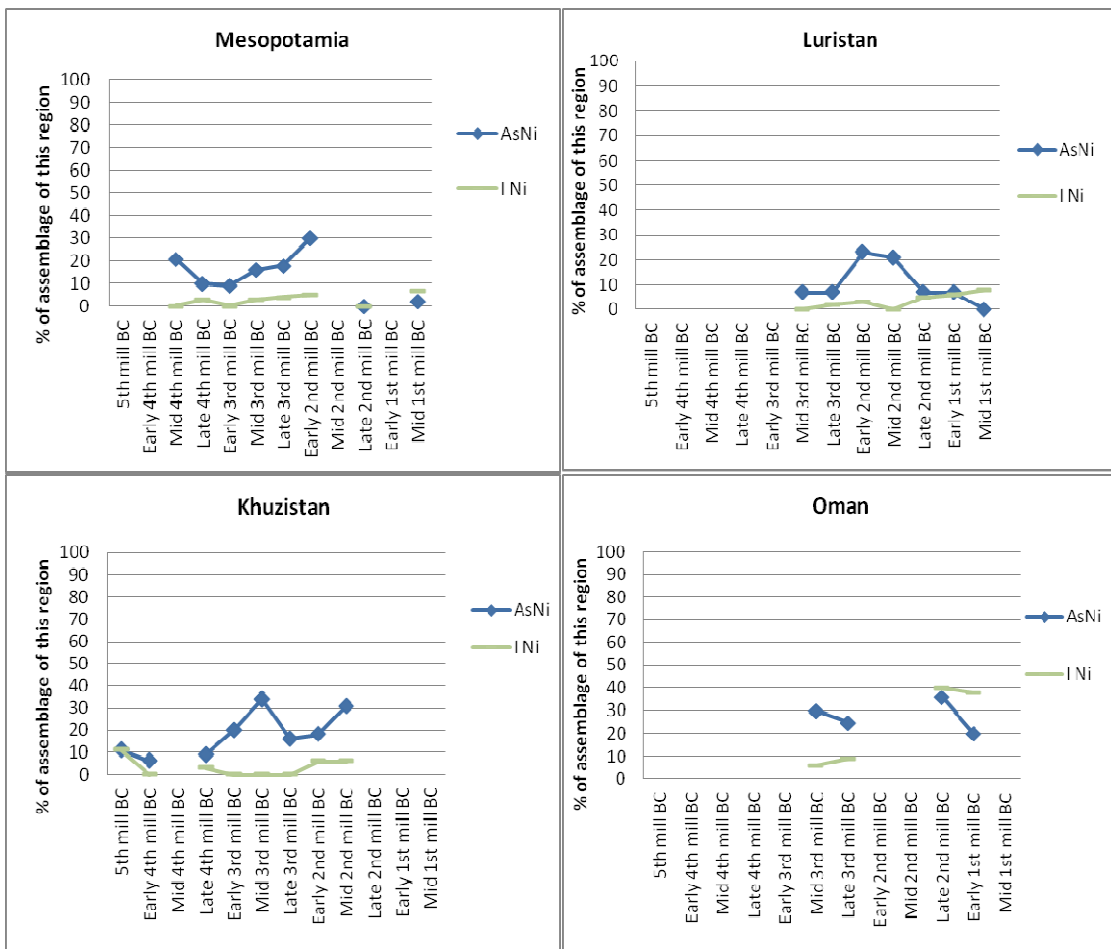


Figure VI-13. Chronological evolution of the ubiquity of AsNi and Nickel Only metal for all regions

This pattern is visible in particular in Mesopotamia and perhaps Khuzistan in Period 1, and Luristan in Period 2 (Figure VI-18). In Oman, the proportion of Nickel Only metal reaches 40% of the assemblage in the late 2nd millennium BC. Although not enough data is available for the early and mid 2nd millennium BC, it is difficult to envisage that this was the result of a slow accumulation of this metal type. A better explanation for Oman is probably the beginning of the exploitation, some time in the early or mid 2nd millennium BC, of a new copper source that was then used along side the source producing AsNi copper<sup>6</sup>. This could fit in with the story proposed by Weeks (2003, pp.118-19) of a shift from the exploitation of the small arsenic rich oxide deposits to the larger sulfide deposits with less arsenic in Oman.



**Figure VI-14. Chronological evolution of the percentage of the assemblage represented by the AsNi and Nickel Only groups in Mesopotamia, Luristan, Khuzistan and Oman**

<sup>6</sup> Since the submission of this thesis, the publication of the analysis of 24 Iron Age ingots from Masafi was brought to my attention. Of these, about half fell in the AsNi metal group and half in the Ni Only group, confirming the exploitation of a source of Ni Only metal alongside AsNi in Period 2 in Oman.

Not much data is available to study the characteristics, and in particular the tin content, of the Ni Only metal type. The evidence available at present shows that the Nickel Only metal from Luristan has a relatively high proportion of objects with between 10 and 15% Sn (Figure VI-15), which is a high percentage for Luristan (see Chapter 4). Much more data would be needed to confirm this trend, but it can perhaps indicate that the accumulation of a depleted form of AsNi can partly be explained by the production of these high tin bronzes. Indeed, as explained in Chapter 2, high tin contents can be achieved by putting cassiterite in contact with molten low tin bronzes, a process during which arsenic and antimony would have been lost. Figure VI-15 also shows a higher proportion of mid range tin-bronzes in the Ni Only group than in the AsNi group. These objects can be interpreted as resulting from the recycling of AsNi or Ni Only tin bronzes, where the loss arsenic is accompanied by a loss of tin by oxidation or mixing with tin-less copper.

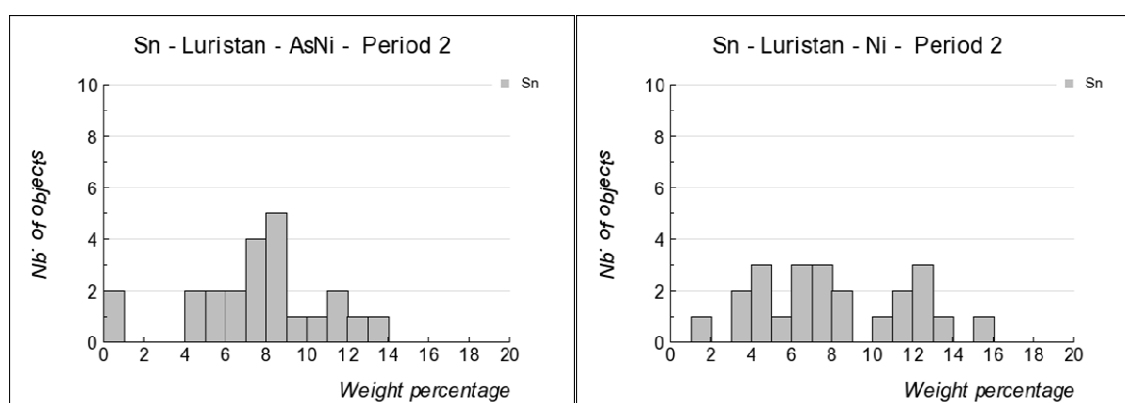


Figure VI-15. Distribution of the tin content in objects of the AsNi type (left) and Ni Only type (right) in Luristan in Period 2 (late 2nd to mid 1st millennium BC)

From the AsNi group to the Nickel Only group, we do not see a significant increase in the overall proportion of tin bronzes (42% for AsNi to 43.9 for Ni Only in Mesopotamia in Period 1 and 91.3% to 92% in Luristan in Period 2), perhaps because one alloying

event isn't enough for the composition of one object to change enough to shift into the Nickel Only group. What we do see on the other hand is, within the tin bronzes, an increase in the proportion of objects falling in the 2-6% Sn range, probably mixed or recycled tin bronzes and of objects falling in the 10-15% Sn range that can be interpreted as secondary alloys, ie. objects for which the copper has undergone more than one addition of tin in order to achieve a higher tin content.

	Mesopotamia Period 1		Luristan Period 2	
	AsNi	Ni Only	AsNi	Ni Only
<b>Recycled/mixed bronze 2-6% Sn</b>	21 %	35 %	19 %	29 %
<b>Primary tin bronze 6-10% Sn</b>	40 %	15 %	57 %	38 %
<b>Secondary tin bronze 10-15% Sn</b>	39 %	50 %	24 %	33 %

Table VI-4. Proportion, within the tin bronzes ( $\geq 2\%$  Sn) of the AsNi and Ni Only groups, of objects falling in the 2-6% Sn, 6-10% Sn and 10-15% Sn ranges for Mesopotamia in Period 1 and Luristan in Period 2.

d. Arsenic Nickel: A Tentative story

From this analysis, we can tentatively derive the following story for metal of the AsNi type. This story will of course need to be refined or altered when more information becomes available and the area of study is extended. In the 3rd millennium BC, AsNi was one of the predominant metal types with AsNiSb and AsNiSbAg and it became the most common type in the early 2nd millennium BC. We can envisage at least two systems of movement of this metal: one originating in the Indus Valley or South-Eastern Iran and being traded to Mesopotamia and then redistributed to Assyria and Luristan, and one operating between Oman and Khuzistan, probably with metal of higher nickel content being traded from Oman to Elam (see Figure VI-16). It seems likely that, as a general rule, copper was traded unalloyed and tin, perhaps from the eastern Zagros or traded from the east along-side copper, was later added to the objects.

Finally, it seems that during Period 1 AsNi copper was recycled, resulting in the accumulation of a depleted form of the metal (presenting only nickel) notably in Mesopotamia and Khuzistan.

In Period 2, AsNi metal became less ubiquitous. South-eastern Iran and Oman might have continued to use the local sources that were already in use in Period 1 (see Figure VI-17). In Oman, this was accompanied by the use in Period 2 of Nickel Only metal, potentially resulting from the opening of one or several new mines in the 2nd millennium BC. In Luristan, it seems that AsNi metal was recycled resulting in the accumulation of a depleted form of this metal type.

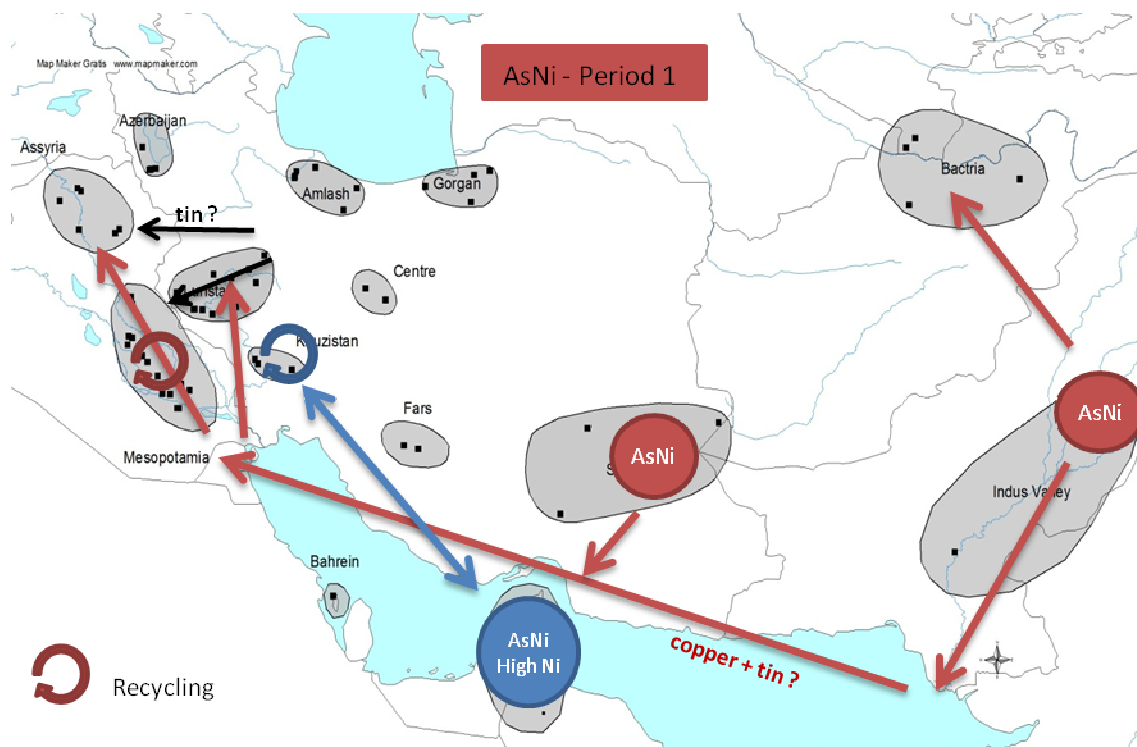


Figure VI-16. Tentative reconstruction of sources and movement of AsNi metal in Period 1 (early 3rd to mid 2nd millennium BC)

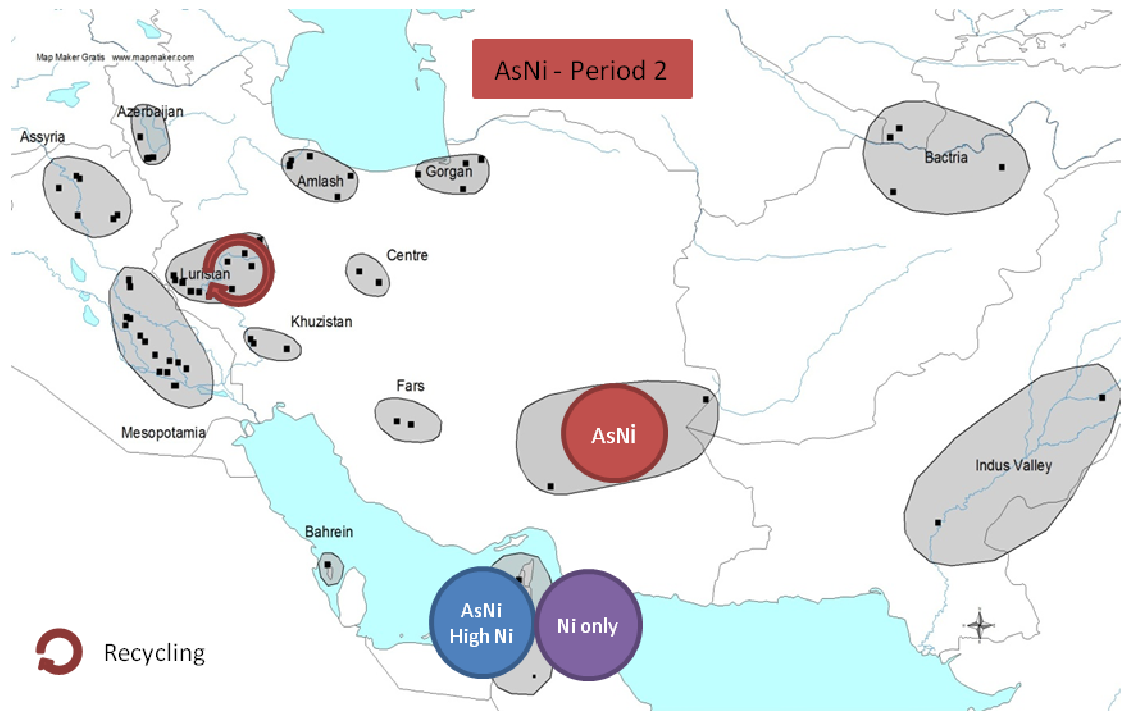


Figure VI-17. Tentative reconstruction of sources and movement of AsNi metal in Period 2 (late 2nd to mid 1st millennium BC)

### 3. Type 1111: Arsenic Nickel Antimony Silver

The AsNiSbAg group is constituted of objects where the levels of arsenic and nickel are respectively above 0.25 % and 0.1 % and the levels of arsenic and silver are both above 0.05%. As shown in Figure VI-6 above, this group was very common in the 4th millennium BC when it constituted a third of the assemblage. It was, with AsNi and AsNiSb, one of the three main groups of the 3rd millennium BC and was particularly important in the mid 3rd millennium BC when it represented 21% of the assemblage. After that, its importance gradually decreased and it represented only a small portion of the assemblage in Period 2.

The numbers of objects that fall in the AsNiSbAg group for each period are summarized in Table VI-5.

	Period 1	Period 2
<b>Number of objects</b>	660	46
<b>Percentage of the assemblage represented by AsNiSbAg</b>	17 %	5 %
<b>Ubiquity of tin-bronze (&gt;2% Sn)</b>	35 %	90 %

Table VI-5. Number of objects that fall into Group AsNiSbAg in Period 1 (early 3rd to mid 2nd millennium BC) and in Period 2 (late 2nd to mid 1st millennium BC), percentage of the assemblage that this represents for each period and ubiquity of tin-bronze

a. Arsenic Nickel Antimony Silver: geographical patterns

Figure VI-18 shows for each period the percentage of AsNiSbAg objects in the assemblages of each region (when more than 10 objects were analysed for the region).

We can see that in both periods, objects of this metal type are most common the western part of our area of interest. In Period 1, the highest ubiquity (23%) is found in Assyria with very slightly lower ubiquities in Mesopotamia, Luristan and Khuzistan. From this pattern, we can suggest that AsNiSbAg metal came from somewhere to the west or the north into Assyria and was traded further down the lowlands in Mesopotamia and Elam and into Luristan. This group also represents 12% of the assemblage in Bactria and it is possible that some of it was traded towards Tepe Hissar where it represents 5% of the assemblage. This group is however virtually absent from the regions connected with the Persian Gulf to the east and the south: Fars, south-eastern Iran, Oman, Bahrain and the Indus Valley.

In Mesopotamia, Assyria and Luristan, the maximum ubiquity of AsNiSbAg is reached in the mid 3rd millennium BC and decreases significantly at around the end of the 3rd and the beginning of the 2nd millennium BC perhaps as a result of the closing of mines or the interruption of trade routes from the west (Figure VI-11). In Khuzistan on the other hand this group is more common in the 4th and early 3rd millennium BC.

In Period 2, this group is still present mostly in the west (Azerbaijan, Luristan and Mesopotamia), but the levels in these regions are generally low with a maximum of 11% of the assemblage in Azerbaijan. Again we can suggest a north-western origin for this metal.

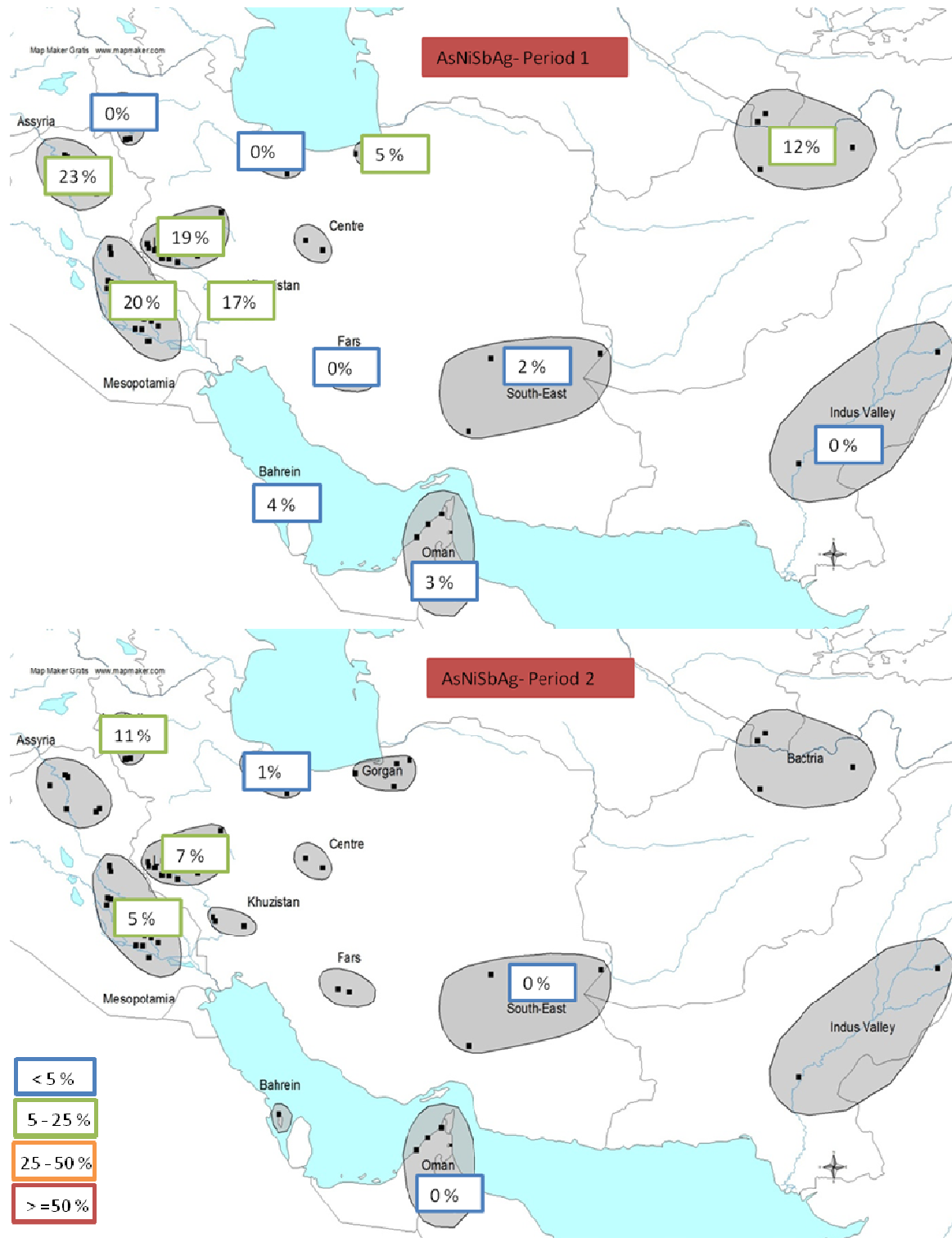


Figure VI-18. Percentage of the assemblage of each region represented by Group AsNiSbAg objects for Period 1 (top) and Period 2 (bottom). Percentages are only given for regions for which more than 10 objects were analysed.

b. Arsenic Nickel Antimony Silver: chemistry and lead isotope ratios

The chemical patterns of Figure VI-19 again show lower levels of arsenic in the bronze objects than in the copper objects. This time we can add that there are also slightly lower levels of antimony in the tin bronze: in the copper the average Sb levels range between 0.19% and 0.31% while in the bronze they range between 0.14% and 0.20%. This can be seen as an indication that the drop in arsenic observed here and for the other metal types is indeed due an oxidative loss in the most volatile elements during the production process of tin-bronze rather than a choice of preferably alloying with tin copper devoid of arsenic.

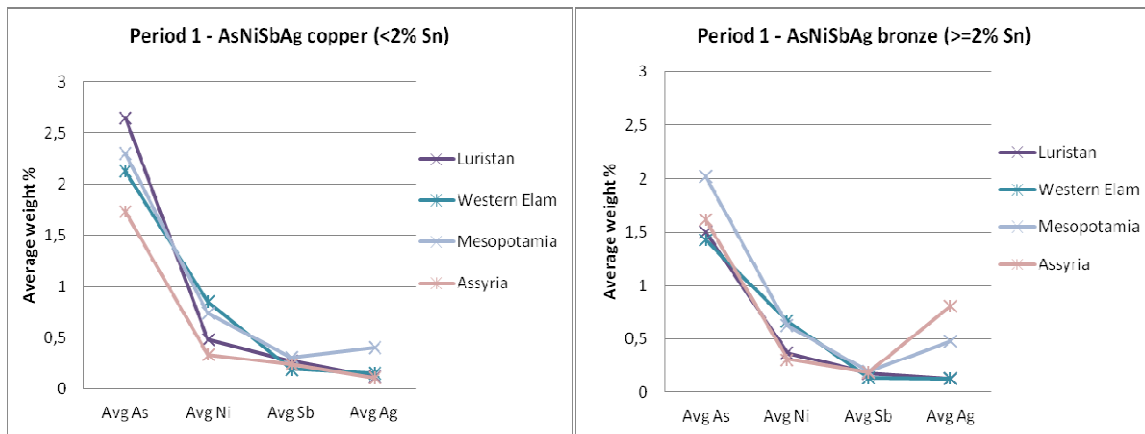


Figure VI-19. Average levels of arsenic, nickel, antimony and silver for tin-less copper objects (<2% Sn, left) and tin bronze objects (>=2% Sn, right) of the AsNiSbAg group in various regions in Period 1 (early 3rd to mid 2nd millennium BC)

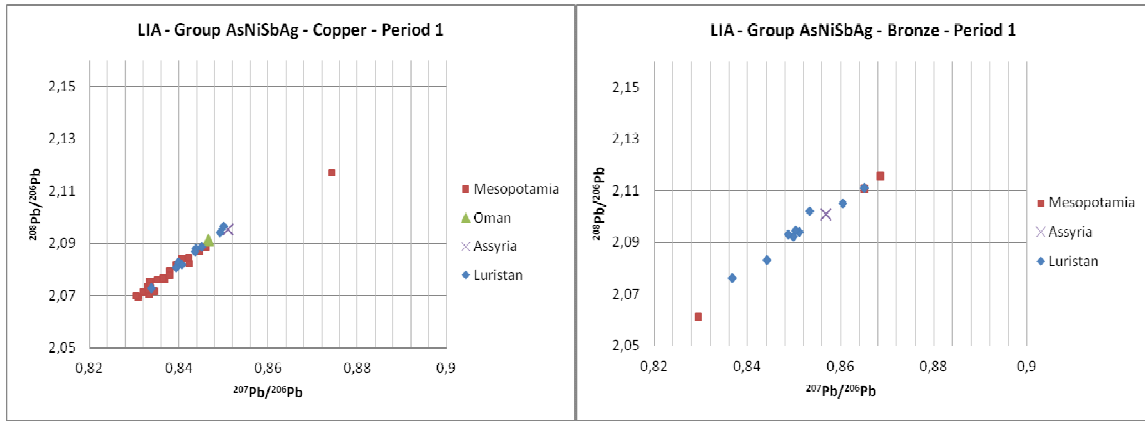
Figure VI-19 shows two different levels of nickel in both copper and bronze: high nickel in Mesopotamia and Khuzistan and low nickel in Assyria and Luristan. We can imagine that one type of metal was being traded between the lowlands of Mesopotamia and Elam and another further north through the passes of the Zagros between Assyria and Luristan. The high levels of silver that are observed in Mesopotamia for copper and in Mesopotamia and Assyria for bronze are due to a small number of pins and vessels

from the mid to late 3rd millennium BC with a very high silver content (between 4.4 and 36.0% Ag). In these objects silver was probably intentionally added and didn't come from the copper ore. Once these outliers are taken out of the calculation of the averages, the silver levels in Mesopotamia and Assyria are very close to those of the other regions. The high levels of arsenic observed in the Luristan tin-less copper, on the other hand, raises the question of a local source being used there, perhaps alongside copper imported from the west. This possibility is reinforced by the lead isotope data shown on Figure VI-20. Indeed, two Luristani and one Assyrian tin-less copper objects and four Luristani tin-bronze objects fall in the cluster that can be matched with ores from the eastern Zagros. For tin-less copper, the other objects from Luristan show a good overlap with the Mesopotamian objects in the 0.830 to 0.847  $^{207}\text{Pb}/^{206}\text{Pb}$  and 2.069 to 2.092  $^{208}\text{Pb}/^{206}\text{Pb}$  ranges (only the  $^{208}\text{Pb}/^{206}\text{Pb}$  against  $^{207}\text{Pb}/^{206}\text{Pb}$  bivariate plot is shown here, but the  $^{206}\text{Pb}/^{204}\text{Pb}$  against  $^{207}\text{Pb}/^{206}\text{Pb}$  plot shows similar results), indicating that the copper used in both regions came from the same source or sources. These ratios match the signature of many different ore sources (see Chapter 5), so from this data, it is impossible to conclude on a provenance for this type of metal.

The study of the nickel level of AsNiSbAg objects in combination with their lead isotopy has shown no obvious correlation between the two. Perhaps then, the two nickel levels observed in Figure VI-19, rather than corresponding to the flow of metal from two different origins, reflect the mixing, in Mesopotamia and Khuzistan, of AsNiSbAg metal with metal of higher nickel content such as the AsNi metal discussed above, that was particularly common notably in Khuzistan.

Finally, very few data points are available for the LIA of AsNiSbAg bronze objects and much more research would be needed to identify their provenance. Interestingly though, none of the data points available fall in the high  $^{207}\text{Pb}/^{206}\text{Pb}$  range ( $> 0.875$ ) that has

been tentatively attributed to ores of Indus Valley (see Chapter 5) and in which fall some of the bronze objects of the AsNi group (Figure VI-10). This reinforces the hypothesis that AsNiSbAg metal did not come from the Indus Valley.



**Figure VI-20. Lead isotope ratios for tin-less copper (<2% Sn) and bronze (>=2% Sn) objects of the AsNiSbAg group dating from Period 1**

Table VI-6 shows the ubiquity of tin-bronze amongst the AsNiSbAg assemblage of the regions where this metal was most common. Unlike for AsNi metal, bronze is most common in the regions we believe were closer to the sources of the metal (Assyria and Mesopotamia). What's more, the proportion of mid-range tin-bronzes (2-6% Sn) as opposed to high tin-bronzes (>6% Sn) is much higher than for the AsNi group for example (Figure VI-21), indicating that a fair amount of recycling or mixing probably took place after the copper was alloyed. It therefore seems possible that this type of metal was traded as ready alloyed tin bronze as well as in the unalloyed form. The source of tin for this bronze is then an interesting question. Given that the period when this metal type was most common is the mid to late 3rd millennium BC, the mines of Kestel/Göltepe thought to have operated in the 3rd millennium (see Chapter 2) could be an option.

	Luristan	Khuzistan	Mesopotamia	Assyria
Ubiquity of bronze (%)	25.5	28.8	37.8	39.0

Table VI-6. Ubiquity of tin bronze (>2% Sn) amongst the AsNiSbAg objects in Iranian and neighbouring regions in Period 1 (early 3rd to mid 2nd millennium BC)

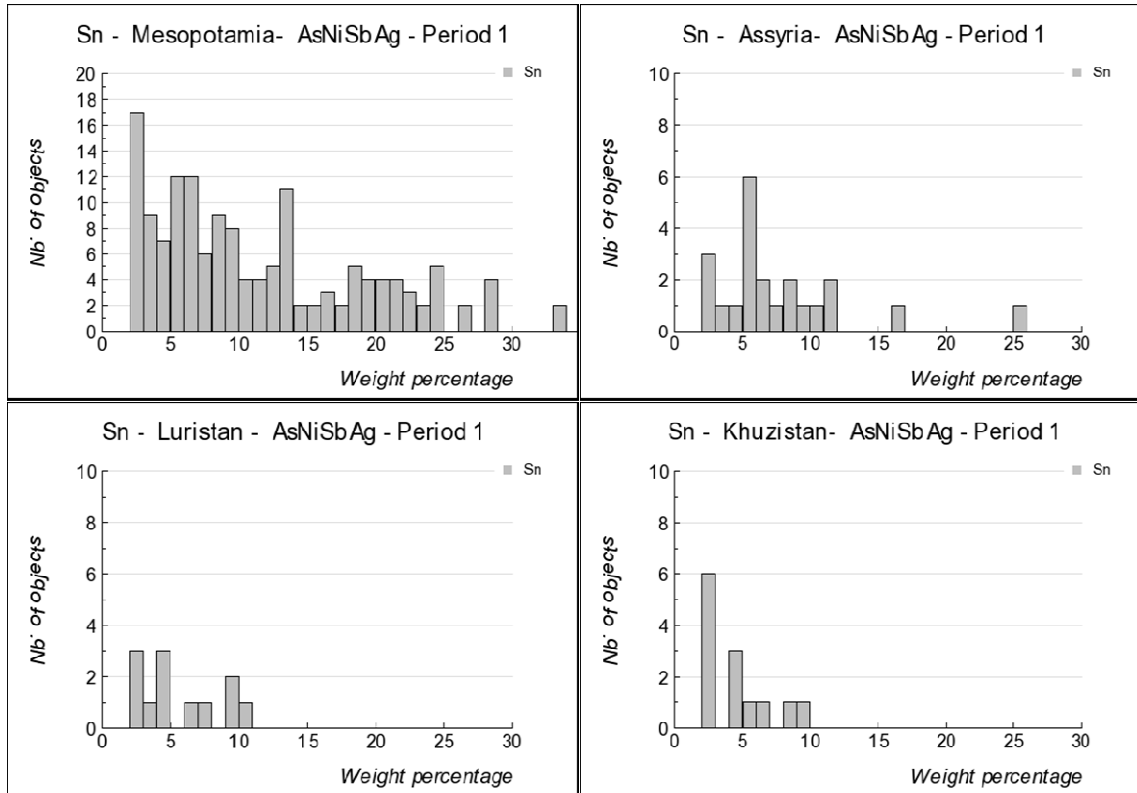


Figure VI-21. Distribution of tin content of AsNiSbAg tin bronze objects (>2% Sn) in Mesopotamia, Assyria, Luristan and Khuzistan in Period 1 (early 3rd to mid 2nd millennium BC)

In Period 2, the AsNiSbAg group is again present only in the north and the west of our area of interest; suggesting once more a north-western provenance, probably outside of that area. The ubiquity of tin-bronze in Luristan and Azerbaijan, the only two regions to have more than 10 AsNiSbAg in Period 2 (respectively 21 and 19) are both very high (see Table VI-7), meaning that this type of copper was either traded as bronze, or that tin from the Zagros was added to it at a later stage. However, too little chemical and lead isotope data are available for Period 2 at this stage, to discuss the movement of this metal type any further.

	Azerbaijan	Luristan
Ubiquity of bronze (%)	100	87.5

Table VI-7. Ubiquity of tin bronze (>2% Sn) amongst the AsNiSbAg objects Azerbaijan and Luristan in Period 2 (late 2nd to mid 1st millennium BC)

Metal of the AsNiSbAg type, upon remelting will lose preferentially its arsenic and antimony. Several groups can then be seen as depleted forms of this metal type: NiSbAg, AsNiAg and NiAg. This complex mechanism is not treated here.

c. Arsenic Nickel Antimony Silver: A Tentative story

From the analysis above we can derive a tentative picture of the use of the AsNiSbAg type of metal, shown on Figure VI-22. Again this is only intended as a basis for further research, as too little information is available for this type of metal especially in Period 2. Metal of the AsNiSbAg type was most common in the north and the west particularly in the mid and late 3rd millennium BC and we have suggested that the source or sources for this metal lied outside of the area of study of this thesis, perhaps in Anatolia or further away. These sources seem to have lost of their importance by the end of the 3rd millennium BC. This copper was traded in both tin-less copper and bronze form and we have tentatively suggested Anatolia and the mines of Göltepe as a source for the tin. We have also suggested that Luristan produced some metal of this type perhaps for use on a local scale.

In Period 2, this metal was generally very rare, but there were again perhaps sources in the north-west from which a small quantity of metal ended up in Azerbaijan and Luristan.

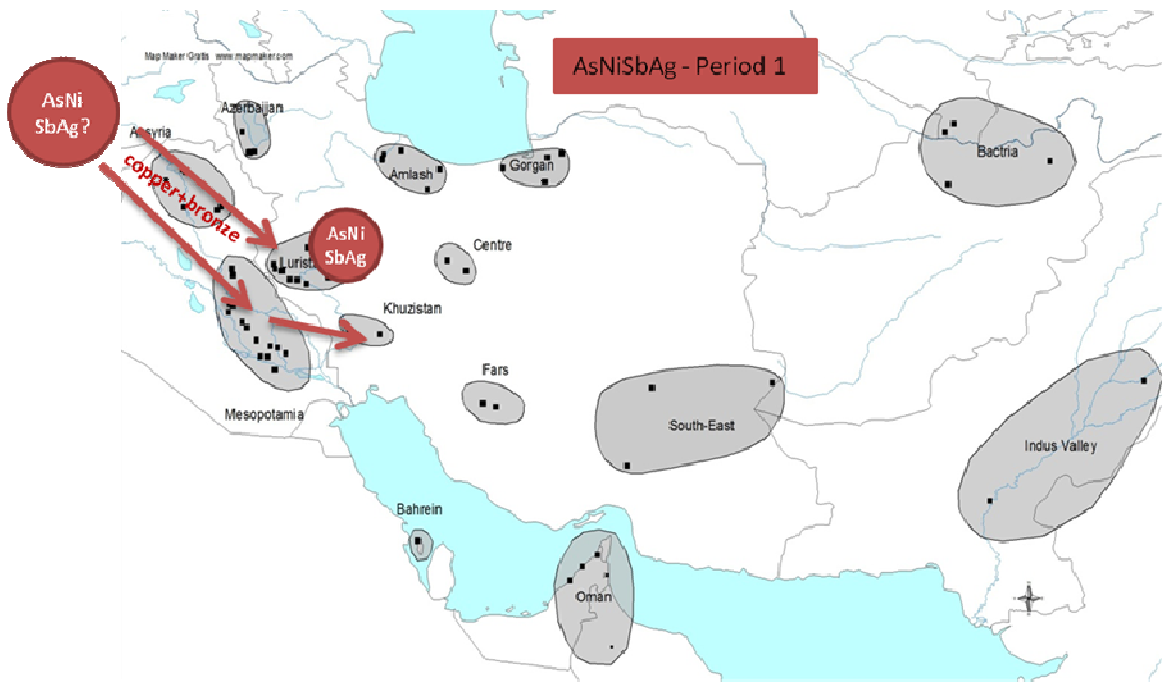


Figure VI-22. Tentative reconstruction of sources and movement of AsNiSbAg metal in Period 1 (early 3rd to mid 2nd millennium BC)

4. Type: 1110: Arsenic Nickel Antimony

The third most important group of our database is the AsNiSb group, constituted of objects with over 0.25% As, over 0.1% Ni, over 0.05% Sb but under 0.05% Ag. This group was most common in the mid and late 3rd millennium BC: it reached a maximum ubiquity of 19% of the overall assemblage in the late 3rd millennium BC (Figure VI-6). Its ubiquity subsequently declined and from the mid 2nd millennium BC onwards it was consistently under 4%. The number of objects that fall into the definition of this metal type in both time periods and the ubiquity of tin bronze for each period are given in Table VI-8. We can see that the ubiquity of tin bronze for this group in Period 1 (19%) is much smaller than for the AsNi and AsNiSbAg groups described above (both 35%).

	Period 1	Period 2
<b>Number of objects</b>	572	25
<b>Percentage of the assemblage represented by AsNiSb</b>	14 %	3 %
<b>Ubiquity of tin-bronze (&gt;2% Sn)</b>	19 %	73 %

Table VI-8. Number of objects that fall into Group AsNiSb in Period 1 (early 3rd to mid 2nd millennium BC) and in Period 2 (late 2nd to mid 1st millennium BC), percentage of the assemblage that this represents for each period and ubiquity of tin-bronze

a. Arsenic Nickel Antimony: geographical patterns

In Period 1, the AsNiSb metal type was common in many places, but virtually absent along the Caspian Sea, in the South-East of Iran and in the Indus Valley (Figure VI-23). It was most common in Assyria, where it represented 35% of the assemblage, and in Bactria, where it represented 24% of the assemblage. It was also an important metal type in Oman and Khuzistan where it represented respectively 21 and 23% of the assemblages. In Period 2, the AsNiSb group was very uncommon: the highest ubiquity is reached in Oman with 6% of the assemblage.

From this geographical pattern, we can imagine again a source in the north-west, from which Assyria got about a third of its metal. This metal was perhaps then traded down the Tigris and the Euphrates to Mesopotamia and further east to Khuzistan. We can envisage another source in the north-east that supplied Bactria and perhaps a source producing the copper used in Oman and Khuzistan. It is worth mentioning that only two of the 38 3rd millennium ingots analysed so far were of the AsNiSb group. One came from Maysar in Oman and the other from Tall Asmar in Mesopotamia.

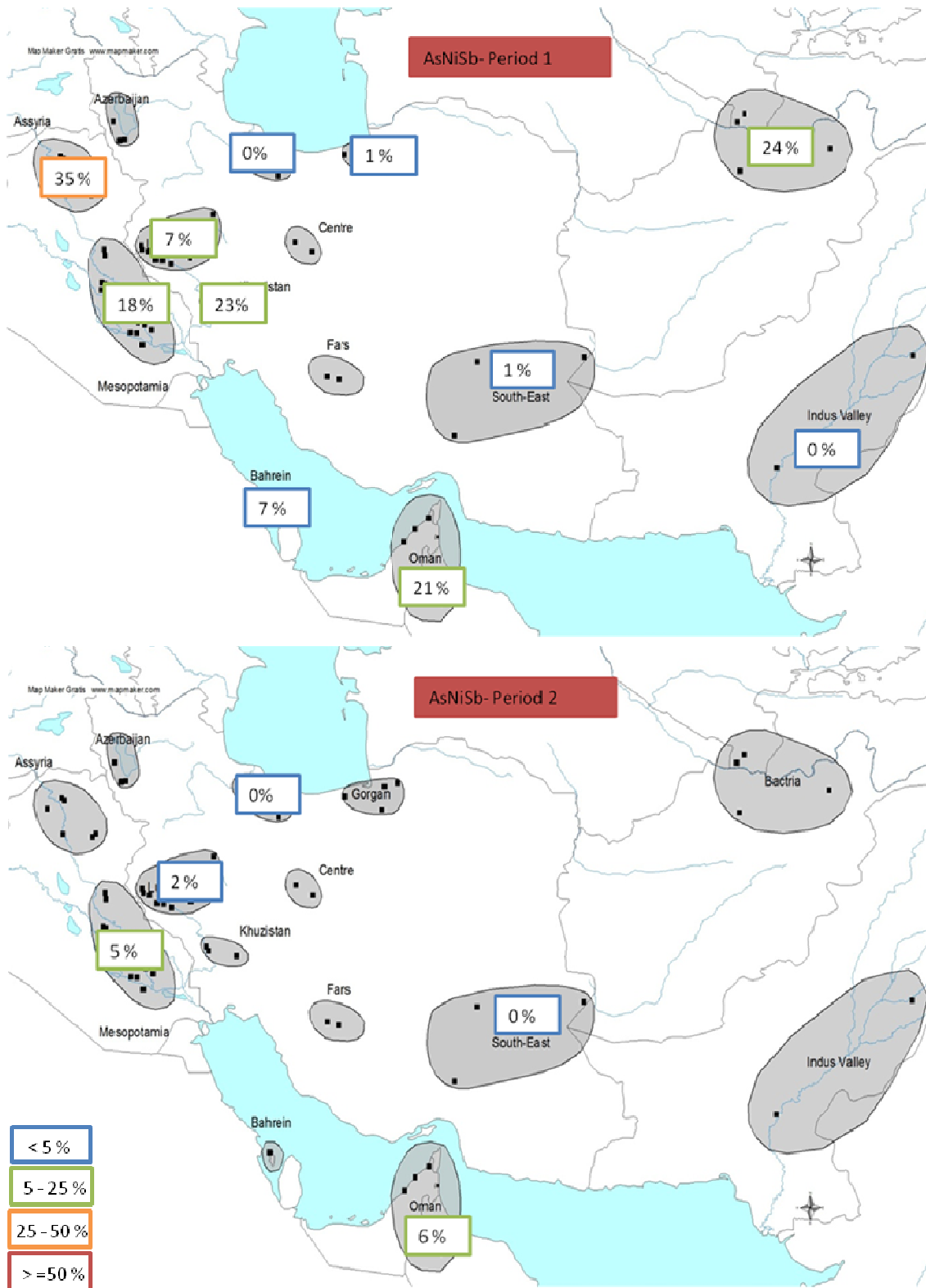


Figure VI-23. Percentage of the assemblage of each region represented by AsNiSb objects for Period 1 (top) and Period 2 (bottom). Percentages are only given for regions for which more than 10 objects were analysed

All of these sources, or the routes used to trade AsNiSb metal (with the exception perhaps of the north-eastern source near Bactria for which we have no information for Period 2), seem to have shut down in Period 2. Figure VI-11 above, shows that the decrease in the ubiquity of AsNiSb metal happened around the same time in Assyria, Mesopotamia and Luristan: between the late 3rd and the early 2nd millennium BC. A north-western source or trade route must then have closed around the turn of the millennium. In Khuzistan on the other hand, this metal represented almost 60% of the assemblage in the early 2nd millennium. We can therefore suggest that either Elam kept access to a north-western source while Assyria, Mesopotamia and Luristan lost it, but this seems unlikely given the geographical situation of Elam, or that the metal used at that time in Elam originated from somewhere different to that used in the 3rd millennium in the west. For Oman, unfortunately, we have no information for the early and mid 2nd millennium BC and so far, we only know that AsNiSb was very common in the mid 3rd millennium BC (Figure VI-12).

b. Arsenic Nickel Antimony: chemistry and lead isotope ratios

Figure VI-24 shows the averages in As, Ni and Sb in metal of the AsNiSb type in regions where more than 10 objects were available to calculate averages. Once again, the levels of arsenic are lower for the tin bronze than for the tin-less copper.

As for the AsNi group, the levels of nickel are significantly higher in Khuzistan and Oman than in the other regions. This suggests that the copper used in this area was different from the copper used further north-west and probably came from the ore deposits of the Oman Peninsula, known for their high nickel content (see Chapter 2). This fits in well with the chronological pattern described above that indicated that

Khuzistan maintained access to an AsNiSb source in the 2nd millennium BC, while Assyria, Mesopotamia and Luristan lost it.

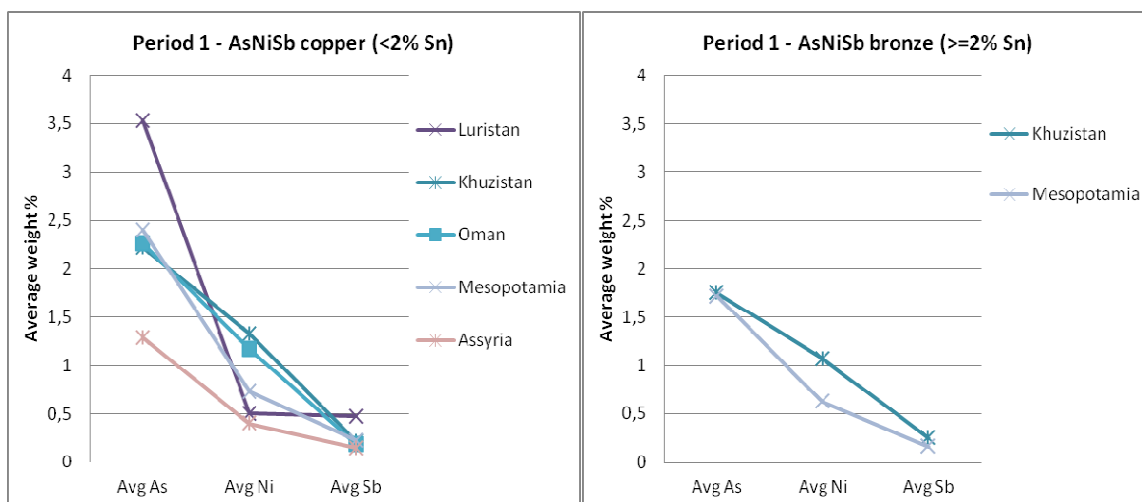
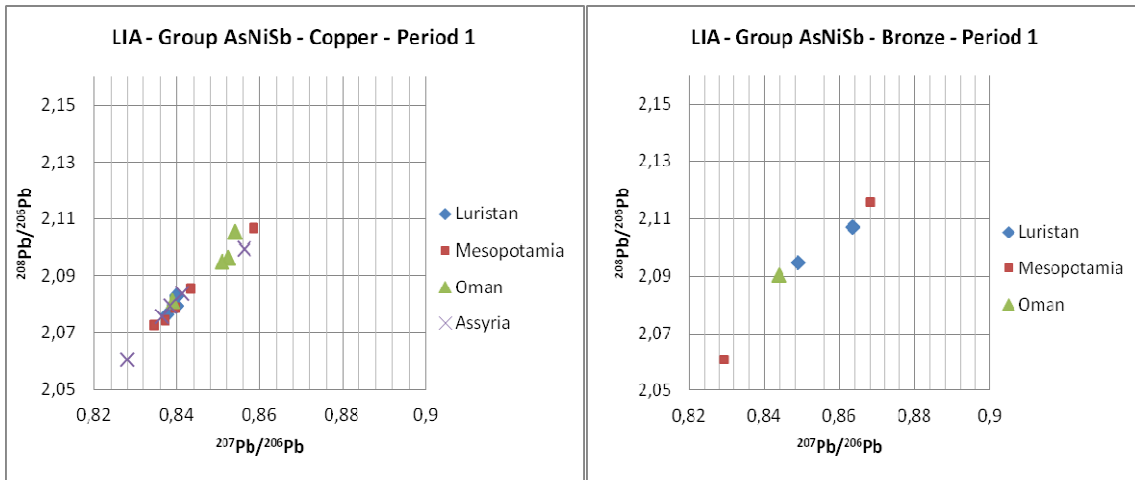


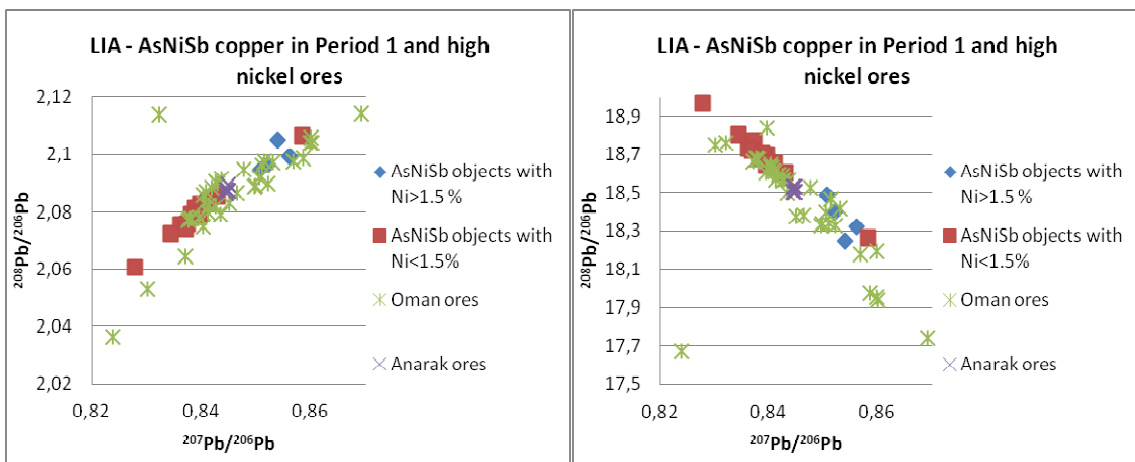
Figure VI-24. Average levels of arsenic, nickel and antimony for tin-less copper objects (<2% Sn, left) and tin bronze objects (>=2% Sn, right) of the AsNiSb group in various regions in Period 1 (early 3rd to mid 2nd millennium BC)

Again Luristan shows an unexpectedly high As average, given the low ubiquity of this type of metal there and we can once more suggest a local source operating on a small scale. Very few AsNiSb objects from Luristan have been analysed for their lead isotope ratios and none of the tin-less copper objects show an isotopy that match the eastern Zagros sources, one of the tin-bronzes however does (Figure VI-25), but much more objects would have to be analysed to assess whether or not Luristan indeed produced AsNiSb metal.



**Figure VI-25.** Lead isotope ratios for tin-less copper (<2% Sn) and bronze (>=2% Sn) objects of the AsNiSb group dating from Period 1

Figure VI-25 shows an important overlap between the lead isotope ratios for tin-less copper objects from Assyria, Mesopotamia and Luristan while the Omani objects mostly fall in a separate part of the graph. LIA of objects from Elam would be crucial to assess whether, as suggested by the chemistry, Khuzistan also used ore of Omani provenance. Indeed, at this stage other Ni rich ore sources, like the Anarak-Talmessi complex (see Chapter 2) cannot be excluded. At the moment though, the objects with the highest Ni contents (in blue on Figure VI-26) cluster together and match the signature of some of the ores from Oman but not those of Anarak.



**Figure VI-26.** Lead isotope ratios for high Ni (>1.5% Ni) and mid-range nickel (0.1 to 1.5% Ni) tin-less copper (<2% Sn) objects of the AsNiSb group dating from Period 1 and ores from Oman and Anarak

As mentioned above, the overall ubiquity of tin-bronze in the AsNiSb group is low compared to the previous two groups, which means that both chemical and lead isotope data for this AsNiSb bronze are scarce and little can be said on its movement and trade. As described for the AsNi group, the regions we think were closest to the sources of AsNiSb metal based on the ubiquity maps (Assyria and Oman) are also those where tin-bronze was most uncommon (Table VI-9), suggesting that this metal was mostly traded as unalloyed copper. However, many of the tin-bronzes of Mesopotamia and Khuzistan (the two regions for which enough data was available) have a relatively low tin-content (Figure VI-27), in sharp contrast with objects of the AsNi group (Figure VI-9). The origin of the tin in objects of the AsNiSb group might then come from mixing with other high tin bronze objects rather than an addition of tin after trade.

	Luristan	Khuzistan	Oman	Mesopotamia	Assyria
Ubiquity of bronze (%)	35.3	32.8	6.7	19.8	5.6

Table VI-9. Ubiquity of tin bronze (>2% Sn) amongst the AsNiSb objects in Iranian and neighbouring regions in Period 1 (early 3rd to mid 2nd millennium BC)

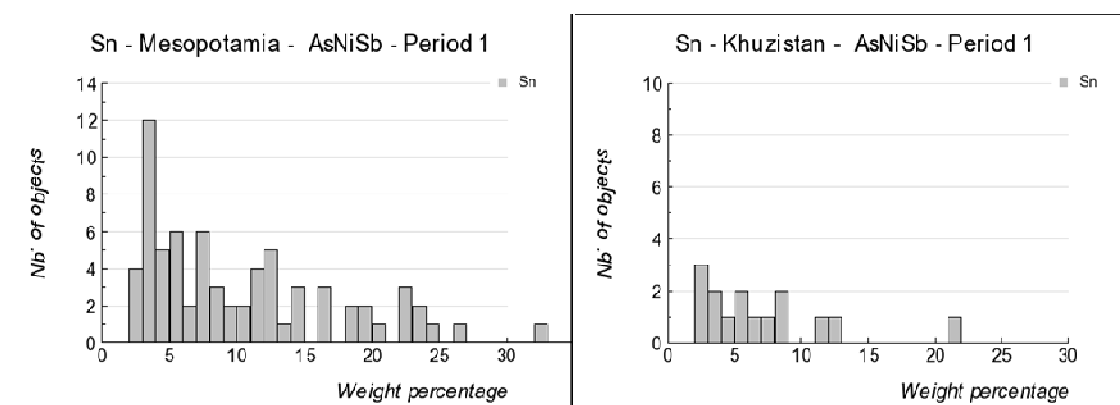


Figure VI-27. Distribution of the tin content in AsNiSb bronze objects (>2% Sn) in Mesopotamia and Khuzistan in Period 1 (early 3rd to mid 2nd millennium BC)

Objects of the AsNiSb group, when remelted, would have preferentially lost their arsenic and antimony. AsNi, NiSb and Ni Only can therefore theoretically all be seen as depleted forms of this metal type. Depletion of AsNiSb is of course not the only pathway that leads to these three compositions. Each group can also contain objects made of prime metal from an ore source. For example, the depleted AsNiSb objects falling in the AsNi type will be lost in the overwhelming signal of the AsNi metal sources described above. AsNi, NiSb and Ni Only can also be populated by objects coming from the depletion of other groups: depleted AsNiSb objects falling into the Ni Only type would have added to the slow accumulation of this recycled type described above, but it is impossible based on the information available to date to set apart objects initially coming from the AsNi type and from the AsNiSb type. Finally, objects falling in the NiSb group are too few to show any trends (44 objects over the entire database).

c. Arsenic Nickel Antimony: A Tentative story

The following story can tentatively be derived of the analysis presented above: in the mid and late 3rd millennium BC, AsNiSb metal was traded to Assyria and further south and east to Mesopotamia and Luristan mostly in the form of tin-less copper. This source (or sources), or the trade routes used to bring the metal to these regions lost their importance at the turn of the millennium. One or several other sources, with higher nickel contents operated in Oman, probably trading their copper to Elam. These sources remained active at least at the beginning of the second millennium, but also lost all of their importance in the Iron Age.

We can envisage that a third source of AsNiSb copper was located in or close to Bactria and operated in the early 2nd millennium BC and possibly earlier or later, and that

Luristan also produced locally a small amount of AsNiSb metal, but too little information is available at this stage to say more about these two potential sources.

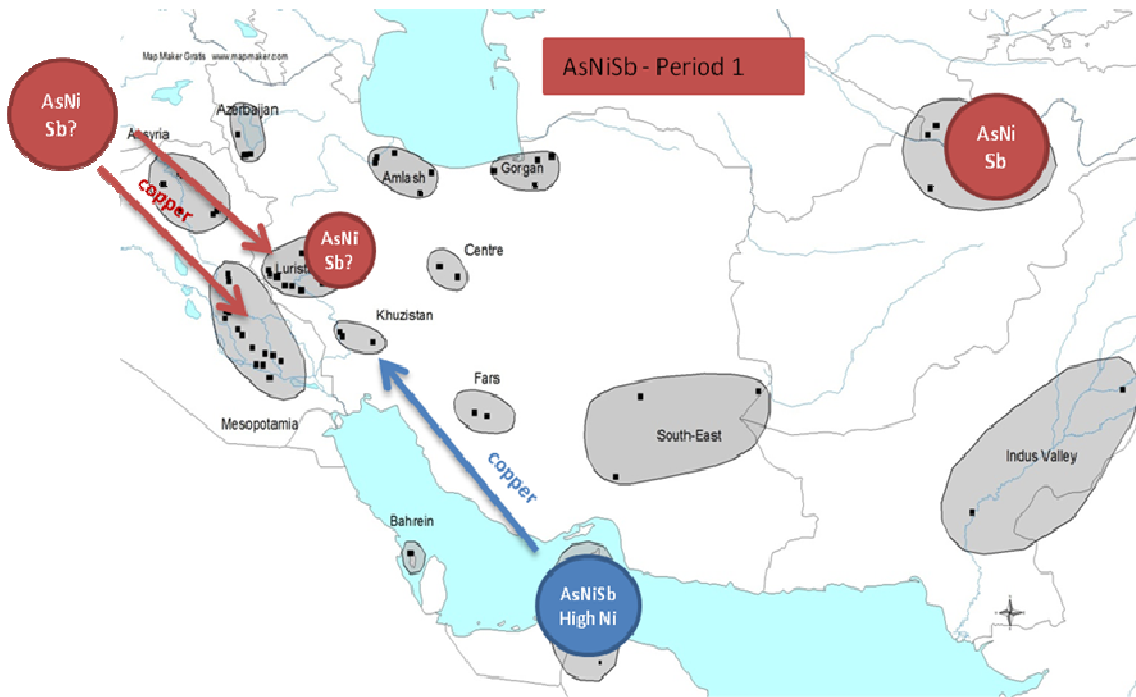


Figure VI-28. Tentative reconstruction of sources and movement of AsNiSb metal in Period 1 (early 3rd to mid 2nd millennium BC)

##### 5. Type 1000: Arsenic Only metal

The final group studied for this thesis is the “Arsenic Only” group, represented by objects with over 0.25% As, under 0.1% Ni and under 0.05% of Sb and Ag. Although it is only the 6th most common group overall in our database (Figure VI-4), it is added here, over the 5th most common (AsNiAg) as the study of As Only metal in relation to Clean Metal can tell us about recycling as demonstrated below.

Figure VI-6 shows that As Only metal was most common in the early 3rd millennium BC when it reached 20% of the overall assemblage, and peaked again in the late 2nd millennium BC at 14% of the assemblage.

The number of objects representing this metal type and the ubiquity amongst them of tin-bronzes are shown in Table VI-10.

	Period 1	Period 2
<b>Number of objects</b>	286	81
<b>Percentage of the assemblage represented by As Only</b>	7 %	9 %
<b>Ubiquity of tin-bronze (&gt;2% Sn)</b>	36 %	81 %

Table VI-10. Number of objects that fall into Group As Only in Period 1 (early 3rd to mid 2nd millennium BC) and in Period 2 (late 2nd to mid 1st millennium BC) percentage of the assemblage that this represents for each period and ubiquity of tin-bronze

a. Arsenic Only: geographical patterns

The maps of Figure VI-29 show that, in Period 1, As Only metal was most common in south-eastern Iran, where it represented almost a third of the assemblage. It was also used in the north: in Bactria and along the Caspian Sea in Amlash and Gorgan (12, 12 and 14% of the assemblages respectively) but was less common in the west and along the Persian Gulf.

In Period 2, the ubiquity of As Only metal in south-eastern Iran was much lower than in Period 1 (6%) but had increased in Luristan and Mesopotamia where it represented 14% of both assemblages.

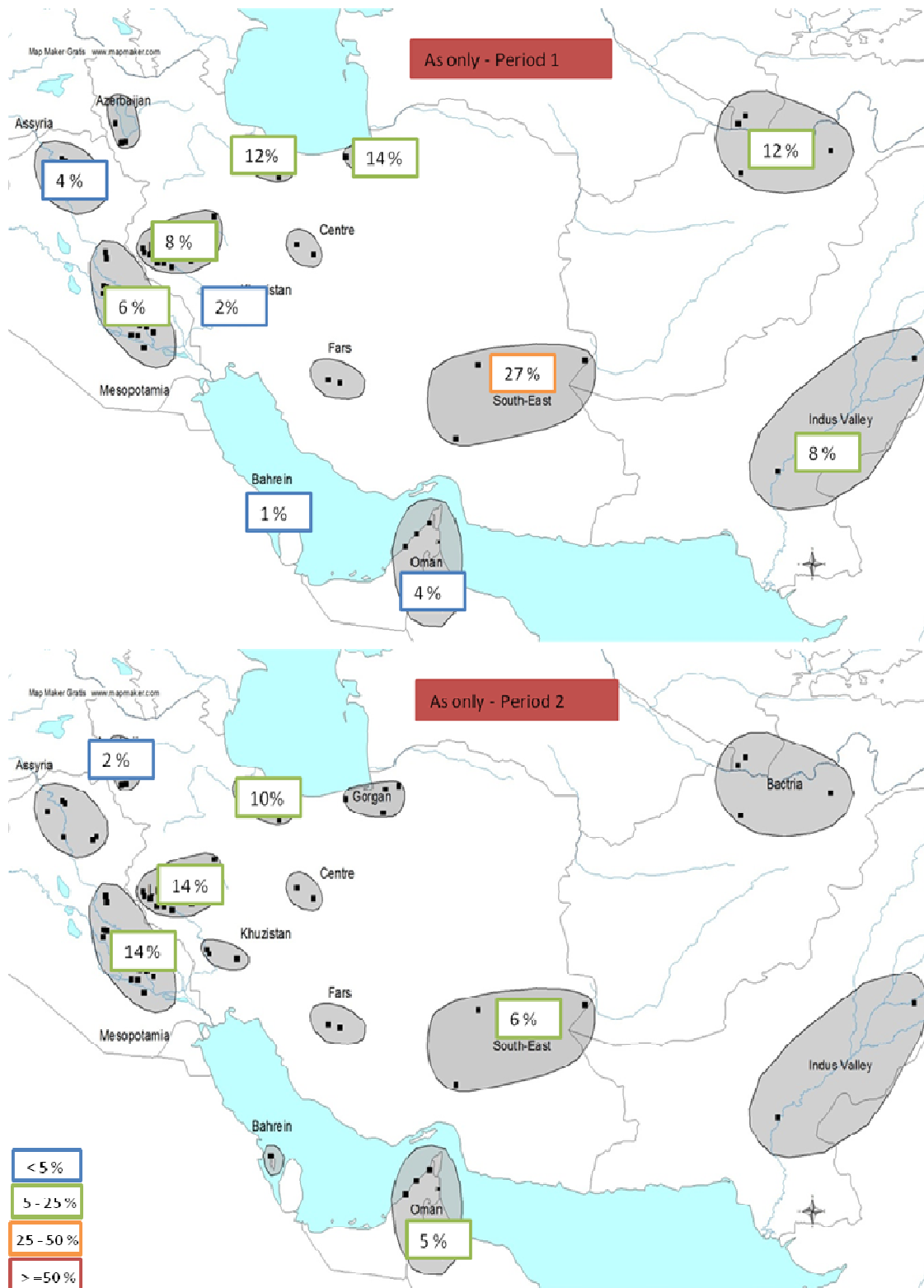


Figure VI-29. Percentage of the assemblage of each region represented by As Only objects for Period 1 (top) and Period 2 (bottom). Percentages are only given for regions for which more than 10 objects were analysed

Figure VI-30 shows the chronological evolution of the ubiquity of As Only metal for the regions where it was most common (the Indus Valley and Bactria are not shown

since for both of these regions we only have data for one time period, respectively the late 3rd millennium BC and the early 2nd millennium BC). It seems that, in the east (south-eastern Iran and Gorgan), As Only metal was very common around the end of the 4th millennium BC and the beginning of the 3rd millennium BC. The importance of this metal subsequently decreased, but the data available doesn't give us much information on this decrease. In Luristan and Amlash on the other hand, it seems that this group was most important around the mid to late 2nd millennium BC. Its importance then slowly decreased throughout the Iron Age.

From these geographical and chronological patterns, we can suggest that there was at least one source of As Only metal in south-eastern Iran exploited in the 4th and 3rd millennium BC. The metal from this source was perhaps being traded north towards Gorgan and west all the way to Mesopotamia. One or several other sources must have been located somewhere in the north-west, providing As Only copper to Luristan and Amlash from the late 3rd millennium BC onward, but peaking in the second half of the 2nd millennium BC. Of course as explained in Chapter 5 it is incredibly difficult to assess whether the arsenic in the metal came directly from the ore source or was intentionally added. Therefore by "As Only" source one must understand one or several mineralisations capable of producing "As Only" type metal whether intentionally or not. The addition of arsenic at a late stage, as for example after copper was traded, is also a possibility that is likely to blur the results obtained here but at this stage the recovery of speiss at Arisman, Tepe Hissar and Shahr-i Sokhta are the only clues as to where this practice might have taken place. It is therefore possible that it was Clean Metal that was mined in south-eastern Iran but that this metal was subsequently alloyed with arsenic in the different regions where it was traded and in particular at Tepe Hissar. Indeed the two

objects that have been tentatively identified as ingots and analysed from Shahr-i Sokhta are both made of Clean Metal.

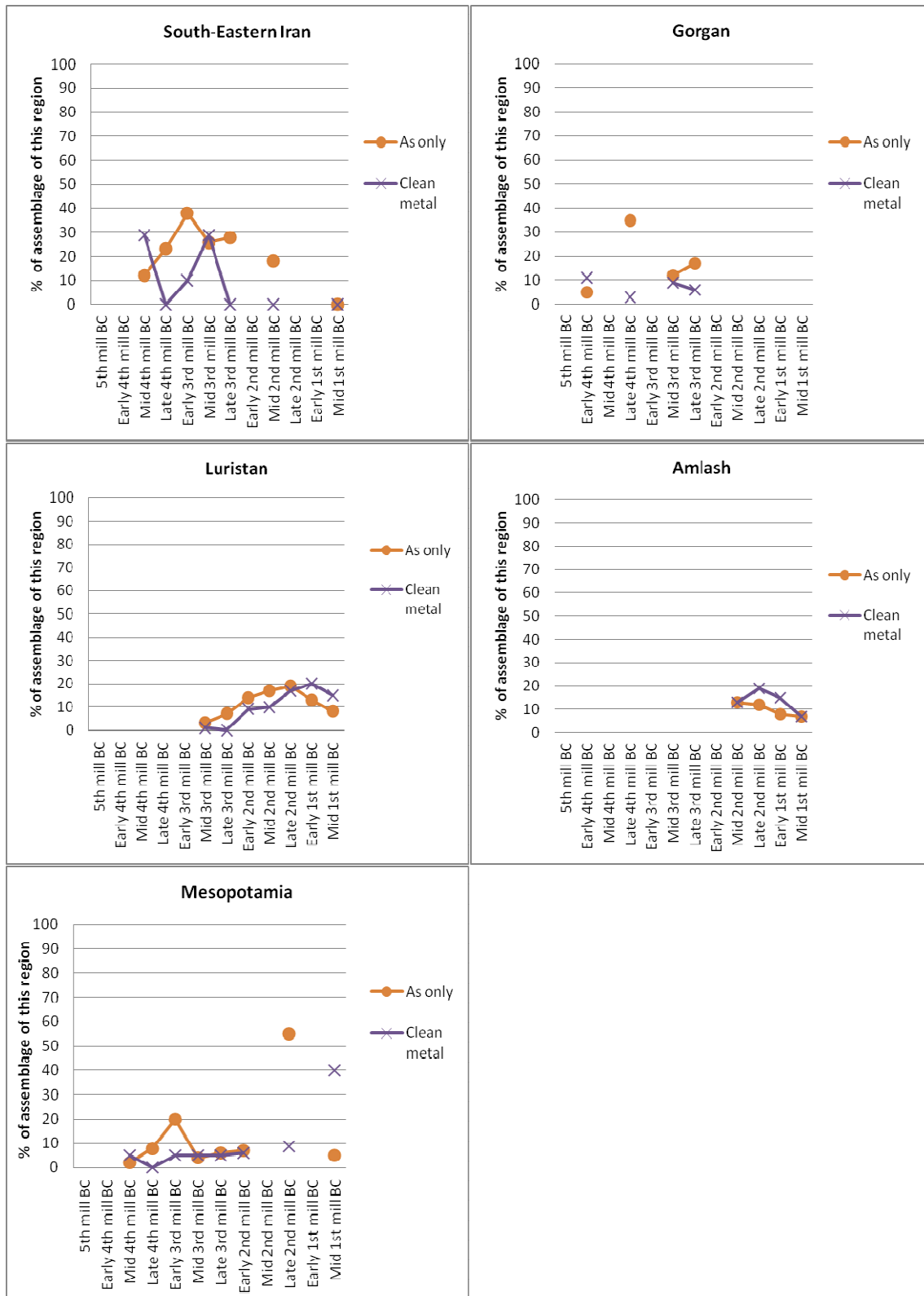


Figure VI-30. Chronological evolution of the percentage of the assemblage represented by the As Only and Clean Metal groups in the South-East, Gorgan, Luristan, Amlash and Mesopotamia

b. Arsenic Only: chemistry and lead isotope ratios

Figure VI-31 shows the average arsenic content within the As Only group in tin-less copper objects of Period 1 and tin bronze objects of Period 1 and 2, calculated after correction to take into account the addition of tin (see Chapter 5). As shown for the other metal types, the arsenic content is lower in tin-bronzes than in tin-less copper in Period 1. The fact that this trend has been observed consistently, across four independent sets of data confirms that patterns based on thermodynamic principles can be observed in the archaeological data.

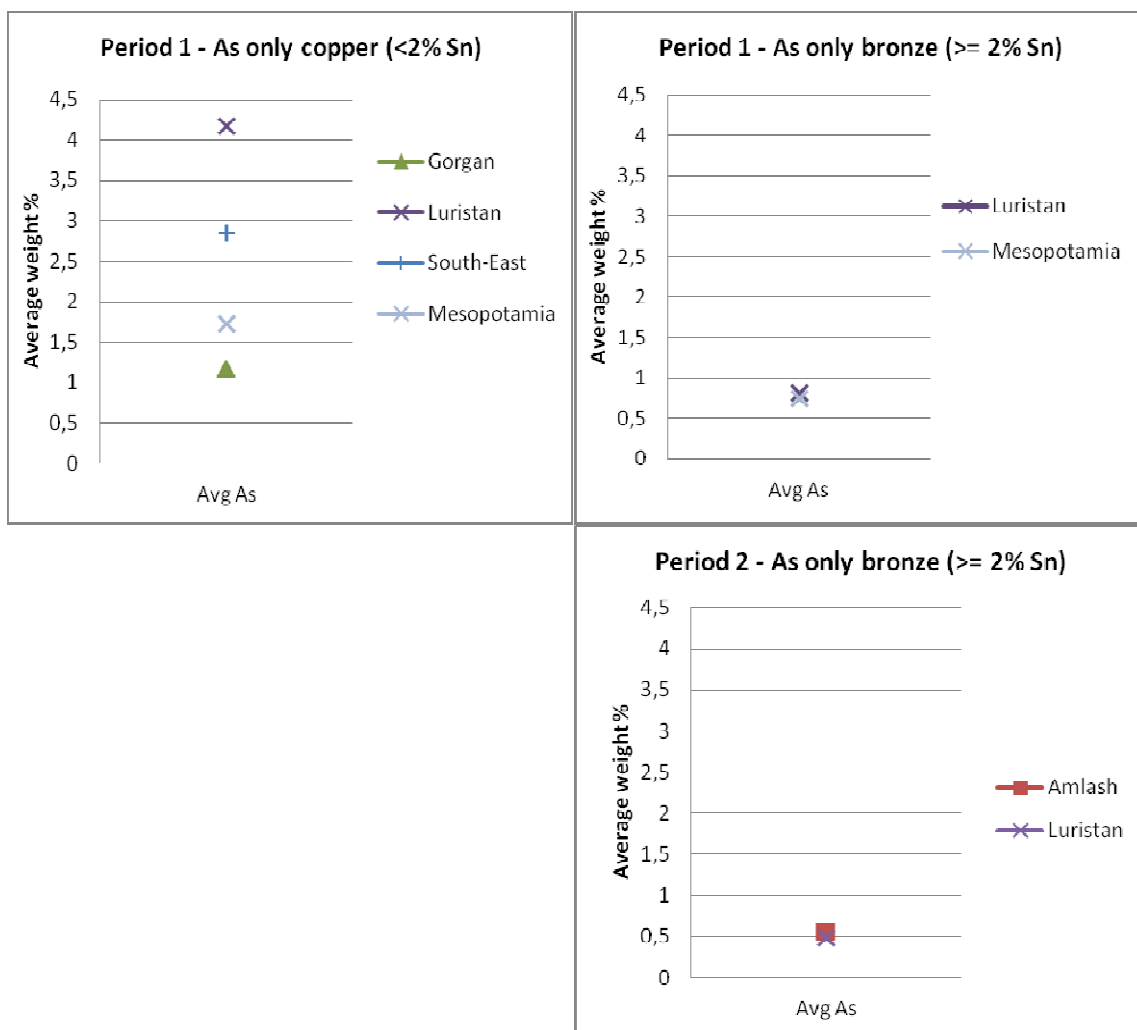


Figure VI-31. Average levels of arsenic for tin-less copper objects (<2% Sn, top left) and tin bronze objects (>=2% Sn, top right) of the As Only group in various regions in Period 1 (early 3rd to mid 2nd millennium BC) and of tin bronze objects in Period 2 (late 2nd to mid 1st millennium BC, bottom right)

In Period 1, the South-East shows a high As content while Gorgan and Mesopotamia, that were probably further away from the source of metal based on the ubiquity maps of Figure VI-29, have a lower arsenic content. This pattern, of course, is not enough to prove that the metal used in these three regions came from the same place and was simply more degraded the further away it got from its source, however it certainly does not go against this hypothesis.

Once again, Luristan shows a surprisingly high arsenic content in the copper of Period 1 given the relatively low ubiquity of As Only metal there (8% in Period 1) and it is again tempting to suggest that this metal came from a local source. The results of LIA analysis of As Only metal (Figure VI-32) show one Luristani tin-less copper object and two bronze objects (one from Luristan and one from Mesopotamia) that fall in the range of lead isotope ratios compatible with the signature of the ores from the eastern Zagros, probably confirming the production of As Only copper in Luristan although more data would of course be desirable.

Only one As Only object from south-eastern Iran (from the site of Shahr-i Sokhta) was subjected to lead isotope analysis. It falls in the same range as the Mesopotamian tin-less copper objects, perhaps suggesting that the same source indeed provided copper to both regions, but again much more analyses are needed to confirm or disprove this hypothesis. Interestingly, the tin-bronze objects from Mesopotamia show the same high  $^{207}\text{Pb}/^{206}\text{Pb}$  and  $^{208}\text{Pb}/^{206}\text{Pb}$  ratios as those observed for the AsNi group (Figure VI-10) and tentatively attributed to the Indus Valley. This trend suggests the same origin for Mesopotamian bronze of the AsNi and As Only groups. Given the low ubiquity of As Only metal in the Indus Valley in Period 1 (8%), it is tempting to suggest a source in south-eastern Iran rather than the Indus Valley for objects with this lead-isotope

signature. The analysis of ores from the southern part of the Iranian plateau would be crucial to further research in that direction.

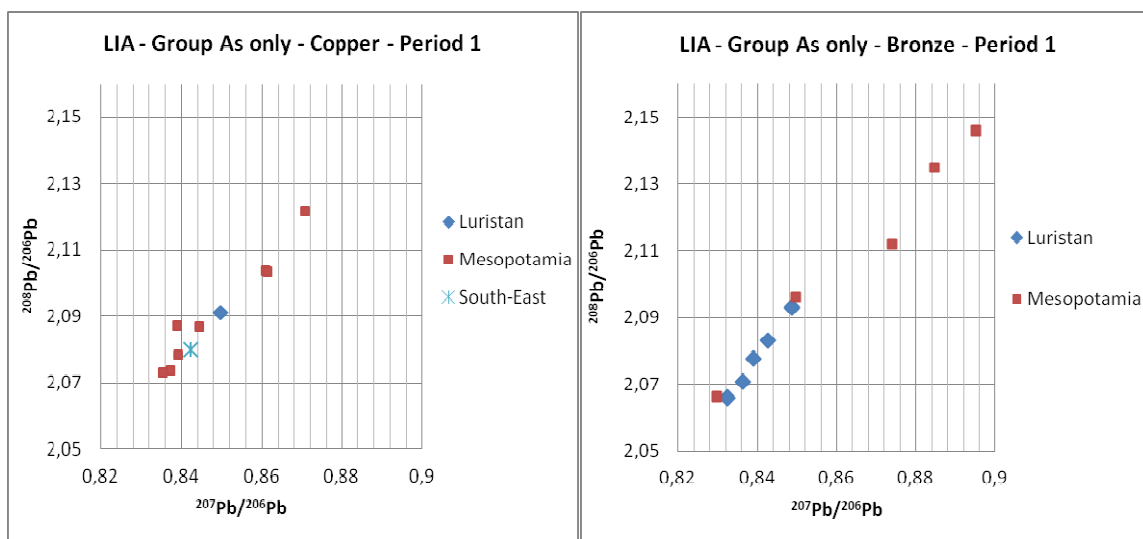
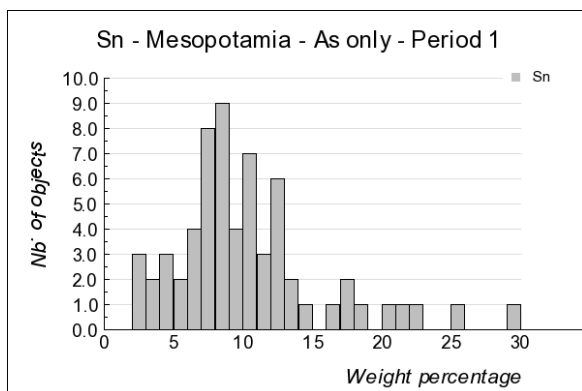


Figure VI-32. Lead isotope ratios for tin-less copper (<2% Sn) and bronze (>=2% Sn) objects of the As Only group dating from Period 1

If the source of AsNi and As Only bronze in Mesopotamia was south-eastern Iran, the virtual absence of tin-bronze in that area as shown in Chapter 4 and for As Only metal in particular as shown in Table VI-11 below is as baffling as ever. The tin distribution plot for Mesopotamian As Only objects of Period 1 (Figure VI-33) shows, as did the plot for AsNi Mesopotamia tin-bronzes (Figure VI-9) a well defined distribution centred on about 8-10% Sn suggesting that the tin was added at a late stage and in a controlled fashion and hasn't been lost by recycling or mixing.

	Amlash	Gorgan	Luristan	South-East	Oman	Mesopotamia
Ubiquity of bronze Period 1 (%)		0.0	47.4	5.0		47.4
Ubiquity of bronze Period 2 (%)	71.4		95.7		80.0	

Table VI-11. Ubiquity of tin bronze (>2% Sn) amongst the As Only objects in Iranian and neighbouring regions in Period 1 (early 3rd to mid 2nd millennium BC) and Period 2 (late 2nd to mid 1st millennium BC)



**Figure VI-33. Distribution of tin content for As Only tin bronze objects (>2% Sn) in Mesopotamia in Period 1 (early 3rd to mid 2nd millennium BC)**

c. Relationship between the Arsenic Only group and the Clean Metal group

Clean Metal (defined here as metal with under 0.25% As, 0.1% Ni and 0.05% Sb and Ag) can be seen as a depleted form of several metal groups and especially those that do not present any nickel or silver as these two elements are difficult to lose when copper is brought to a molten state: As Only, Sb Only and AsSb. Sb Only and AsSb are not very common in our database (Figure VI-4) and here we focus only on the contribution of the degradation of As Only metal to the accumulation of Clean Metal, but it must not be forgotten that this contribution is not unique.

Figure VI-34 shows how the overall ubiquity of As Only metal and Clean Metal evolved over time. From the beginning of the 3rd millennium BC, we can see a steady increase of the proportion of Clean Metal similar to what we saw for Ni Only metal on Figure VI-13. This can be interpreted as the progressive accumulation of depleted copper as a result of recycling activities.

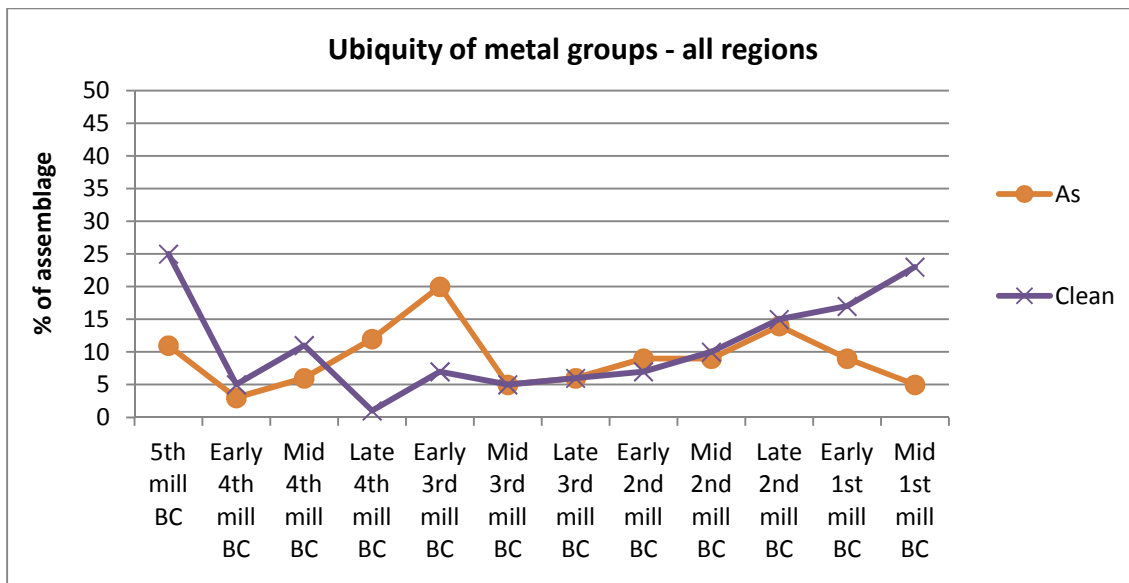


Figure VI-34. Chronological evolution of the ubiquity of As Only and Clean Metal for all regions

On a regional scale (see Figure VI-30), this increase can be seen perhaps in Mesopotamia from the beginning of the 3rd to the beginning of the 2nd millennium BC and in Luristan from the late 3rd millennium BC. In Luristan it occurs with a delay of about a third of a millennium after the appearance of As Only metal. Amlash perhaps follows the same pattern as Luristan, but data from earlier periods would be needed to confirm this. In south-eastern Iran on the other hand, Clean Metal increases sharply between the late 4th and the mid 3rd millennium BC, decreases sharply between the mid and late 3rd millennium and then remains practically absent from this region. This pattern is probably to be seen as a relatively short exploitation of a source of Clean Metal in the south-east and most likely has nothing to do with recycling. Indeed the two ingots from mid 3rd millennium Shahr-i Soktha were made of Clean Metal.

Once again, the proportion of tin bronze in the Clean Metal assemblage of the regions that appeared to have recycled As Only metal is not significantly higher than in the As Only metal (47.4% in As Only to 50.8% in Clean Metal in Mesopotamia in Period 1 and

95.7% to 92.5% in Luristan in Period 2), except in Amlash where it represents 71.4% of the As Only metal and 91.7% of the Clean Metal.

The breakdown into different ranges of tin contents for Mesopotamia and Luristan (the Amlash material currently does not provide enough data), presented in Table VI-12, shows that in both cases, the proportion of objects in the 2-6% Sn range is significantly higher in the Clean Metal group than in the As Only group. This is to be expected as the recycling of a tin bronze would have resulted in a loss of arsenic, in some cases making the object switch into the Clean Metal group, but also in a loss of tin. Unlike for AsNi and Ni Only metal however, there is no visible increase of the proportion of high tin-bronzes meaning that secondary alloying didn't play a big role in the depletion of As Only metal and the accumulation of Clean Metal.

	Mesopotamia Period 1		Luristan Period 2	
	As Only	Clean Metal	As Only	Clean Metal
<b>Recycled/mixed bronze 2-6% Sn</b>	19 %	48 %	15 %	25 %
<b>Primary tin bronze 6-10% Sn</b>	46 %	25 %	49 %	50 %
<b>Secondary tin bronze 10-15% Sn</b>	35 %	27 %	37 %	25 %

Table VI-12. Proportion, within the tin bronzes ( $\geq 2\%$  Sn) of the As Only and Clean Metal groups, of objects falling in the 2-6% Sn, 6-10% Sn and 10-15% Sn ranges for Mesopotamia in Period 1 and Luristan in Period 2.

d. Arsenic Only: A Tentative story

The analysis presented above allows us to draw the following picture concerning the movement of Arsenic Only metal in and around Iran (Figure VI-35 and Figure VI-36):

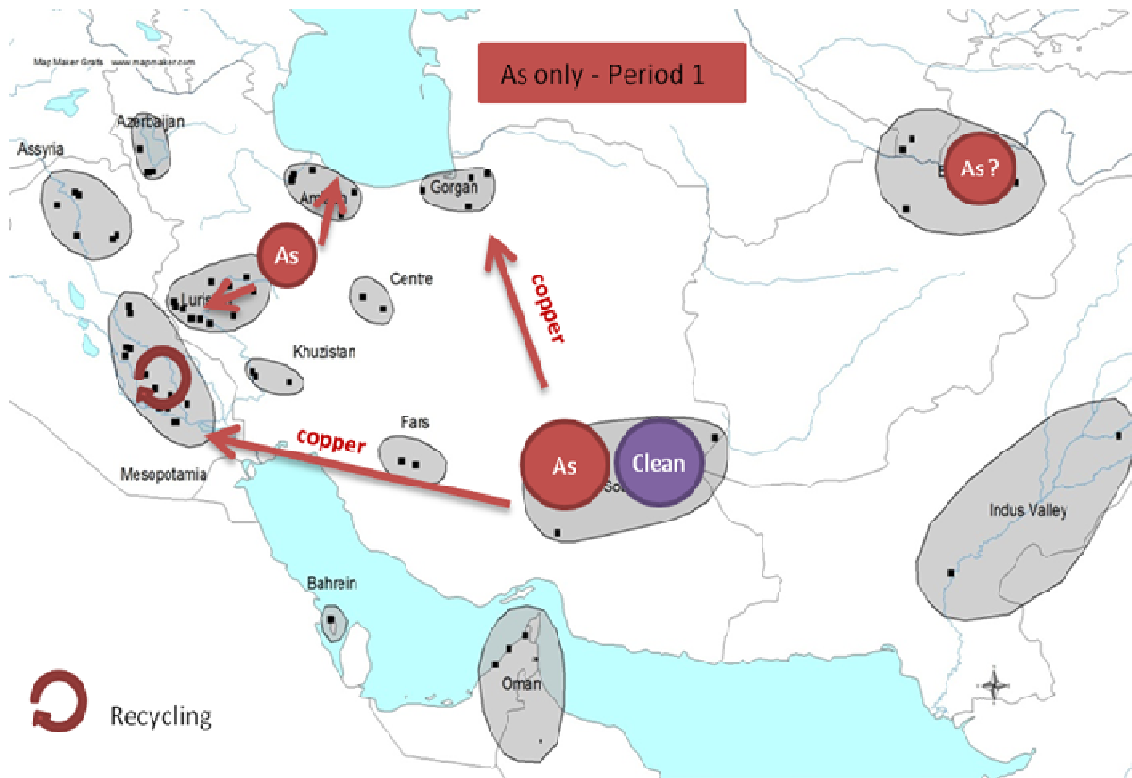


Figure VI-35. Tentative reconstruction of sources and movement of As Only metal in Period 1 (early 3rd to mid 2nd millennium BC)

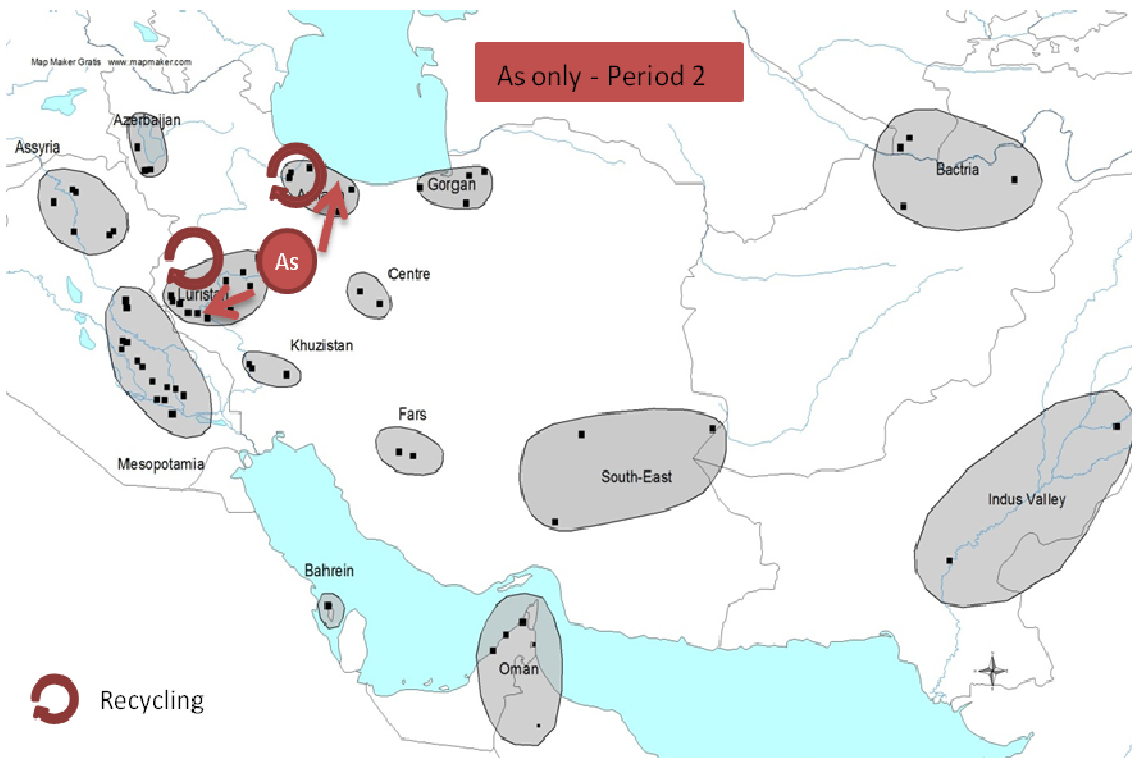


Figure VI-36. Tentative reconstruction of sources and movement of As Only metal in Period 2 (late 2nd to mid 1st millennium BC)

It appears likely that a source situated in south-eastern Iran was exploited at least during the second half of the 4th and in the 3rd millennium BC. This could also have been the exploitation of a source of Clean Metal followed by intentional alloying with arsenic. In the first half of the 3rd millennium BC, this was accompanied by the exploitation, also in the South-East of a source of Clean Metal for which the metal was left unalloyed with arsenic. Copper of the As Only type might have been exported to the north and the west and reached Mesopotamia where it was recycled from the 3rd millennium BC onward.

One or several other sources must have provided As Only copper to Luristan and Amlash. Their exploitation probably started around the middle of the 3rd millennium BC, but peaked in around the end of the 2nd millennium BC. Luristan and Amlash probably recycled this metal, resulting in the accumulation, with a slight delay, of Clean Metal.

## 6. Other significant groups

The analysis above has focused on the main three metal groups and on groups that could give us some information on recycling patterns. A focus on major groups is necessary, as more evidence is available for these, giving a better statistical weight to the analysis. These groups are also more likely to be related to the primary ore sources and the major technological patterns of recycling, alloying, mixing, reuse etc. However, as shown on Figure VI-4, all 16 of the metal groups are populated. Some of the groups that we have not yet mentioned are likely to also be indicative of primary ore sources in or near our study area, as shown below. Some others, on the other hand, are probably mostly populated by objects made of mixed or recycled copper. Indeed, these two practices can

result in strange combinations of trace metals. A small number of objects may also have been traded from a great distance and therefore have a trace element content related to a remote ore source.

One of the less common groups worth mentioning is the AsNiAg group. This group, much like AsNi, AsNiSbAg and AsNiSb, was most common in Period 1 but quite rare in Period 2. It reached a maximum ubiquity of 13% of the assemblage in the late 3rd millennium BC. In Luristan, Khuzistan and Oman in particular its ubiquity was relatively low before and after the late 3rd millennium BC, but in the late 3rd it peaked at respectively 40%, 36% and 20% of the assemblage. It however remained relatively rare in Mesopotamia and Assyria. For this group, there isn't a significant difference between the Ni levels of Khuzistan and Oman and those of the other regions. Once again Luristan has the highest As levels for tin-less copper and we can tentatively suggest a short exploitation of a mine in the Zagros and an export of this metal towards Elam and Oman. However, many of the ingots recovered in Oman fell into the AsNiAg group and it is therefore also possible to envisage the exploitation in the Umm an Nar period of an Omani mine producing this type of copper.

Another notable group is the NiSbAg metal type. This group is practically absent in Period 1 (<2% of the assemblage in all regions), but in Period 2 it represents 31% of the assemblage in Azerbaijan and 3% or less elsewhere. Most of the objects from this group are tin bronzes that have been recovered in the destruction levels of Hasanlu and date from ca. 800 BC. The site of Hasanlu presents evidence of copper production in the form of crucibles and mould (Pigott, 1999b, p.91). The metalworkers of Hasanlu must have had an easy access to a source of NiSbAg metal and used it to meet the needs of the local population and equip the warriors of the fortress. This is reinforced by the fact that four of the five tin bronze ingots analysed from Hasanlu were made of NiSbAg

metal. This metal could perhaps have come from the northern Zagros or the west. At this stage though, it seems that this metal wasn't exported into any of the other regions studied here. A study of bronze objects from outside of our focus area, notably from Urartian sites, would be necessary to understand both the provenance and trade of this metal.

## 7. Conclusion

This chapter has shown that the analysis of the concentrations of a limited number of elements is a powerful tool for the study of the origin and movement of the metal. It has provided a rigorous test for a series of new ideas regarding the use of composition analysis in copper alloy technology studies, and has identified a number of new interesting patterns, conclusions and ways forward.

Figure VI-37 summarises the findings of this chapter and draws a picture of the movement of copper and bronze in the Bronze Age and the Iron Age, allowing us to go back to the question of the transition between these two periods.

The most striking difference between Period 1 and Period 2 is the discontinuation of the trade of the three main metal groups discussed in this thesis: AsNi, AsNiSb and AsNiSbAg. The first one was probably traded from the Indus Valley or south-eastern Iran via the Persian Gulf and stopped around the end of the Bronze Age, unsurprisingly given the fall of Harappa as explained above (see in particular Muhly, 1973). The other two must have come from somewhere in the north-west and travelled to Mesopotamia and western Iran in the form of both tin-less copper and alloyed bronze. Their importance started to decrease a little bit earlier than the first group: around the end of

the 3rd millennium BC. Too little information is available at this stage for Elam in the Iron Age, but it would be interesting to see if it still traded metal with Oman in that period. Indeed we have observed in the Bronze Age a close relationship between the two regions: they both presented AsNi and AsNiSb metal with high Ni content and we have suggested that Elam imported some Omani metal, as had already been envisaged by Berthoud and Françaix (1980).

With the decline of these trade routes, it seems that the metal-workers came to rely on more local sources of metal. In Period 2, we can see the use of copper of new types, probably following the opening of new mines: Ni Only metal in Oman and NiSbAg metal in Azerbaijan.

In this chapter, we have also been able to suggest that recycling was a common practice at least in Luristan, Mesopotamia, Khuzistan and Amlash. The “recycled” metal groups, here Ni Only and Clean Metal are characterised by their depleted chemistry, their slow accumulation, with a delay on the use of the metal type from which they derive and their tin content: they present more objects with either low amounts of tin (2-6%), as a result of a loss of tin during remelting, or a higher tin content (10-15%) possibly as a result of secondary alloying. Recycling however is not a characteristic of the transition between the Bronze Age and the Iron Age as we have seen that, in Mesopotamia in particular, it appears to have started in the 3rd millennium BC.

In the next chapter, we conclude this thesis by reflecting on how the advent of iron fits into this new picture. We also discuss the strengths and limitations of our approach and the direction to give to future research.

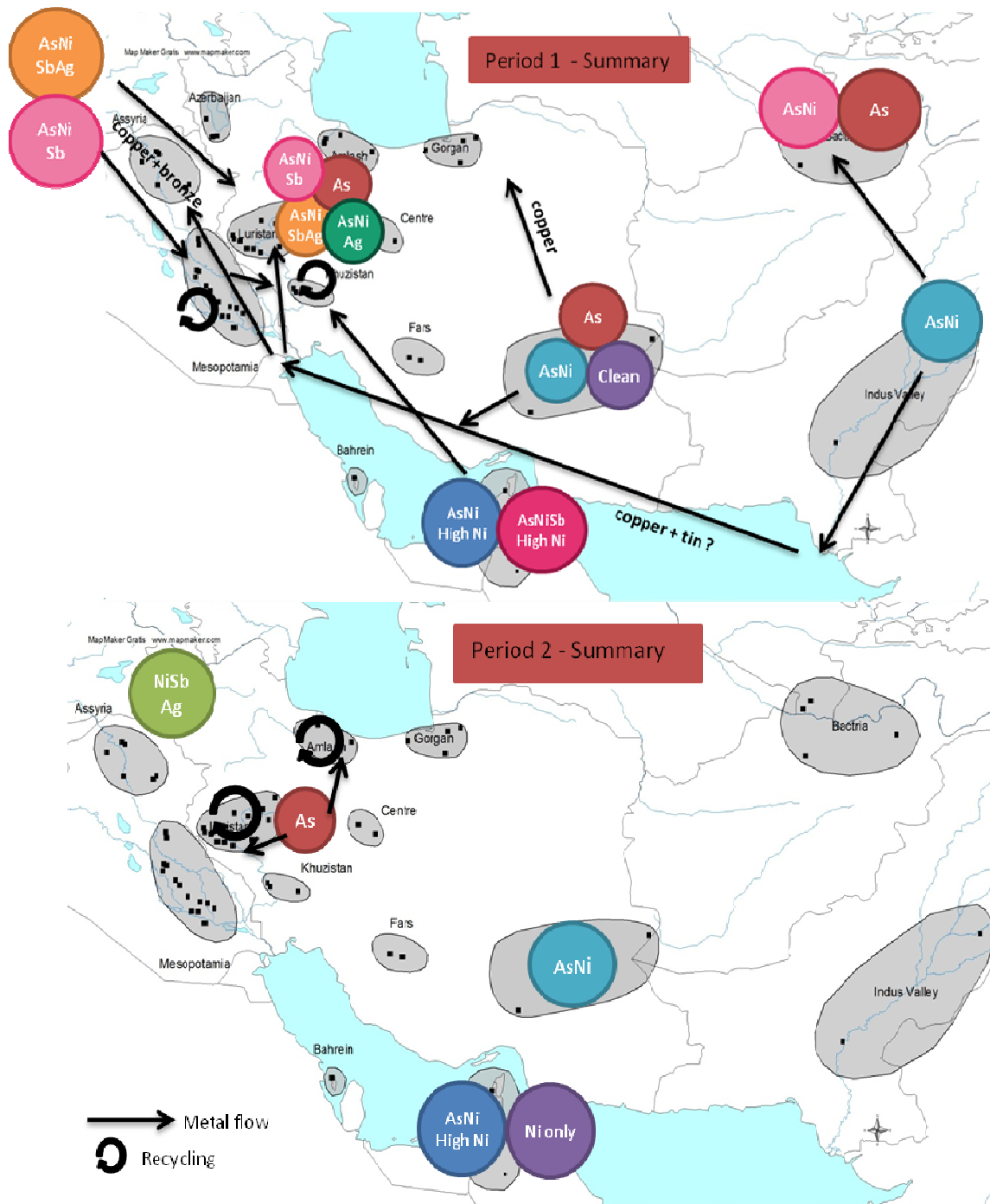


Figure VI-37. Tentative reconstruction of the movement of copper-based metal during Period 1 (early 3rd to mid 2nd millennium BC, top) and Period 2 (late 2nd to mid 1st millennium BC, bottom). The circles are not intended to represent the exact geographical location of a copper source, but the general area where metal of a certain type might have come from.



## Chapter VII. Discussions and conclusion

The primary aim of this thesis as defined in the introduction was to provide a new perspective on the nature of Iranian metallurgy in the transition between the Bronze Age and the Iron Age. To do so, we have reassessed the development of copper-based metallurgy and in particular the changes in alloying practises and metal-flow that occurred between ca. 3000 BC and 500 BC. In this chapter, we discuss these changes in relation to the appearance of iron and we question whether the new perspective emerging from this thesis can help better understand why the new metal was first adopted.

The secondary aim of this thesis was to test a recent method for the analysis of early metallurgy based on the synthesis of old datasets. Here, we discuss the strengths and limitations of this approach in the context of the data that was available to us, and we propose ways of improving it.

Finally, we examine how the synthesis of large databases can contribute to the discussion on technological innovation and in particular on the adoption of new technologies.

1. The appearance of iron in the context of bronze metallurgy

- a. Hypothesis of a response to economic pressure

From the middle of the 2nd millennium BC onwards, iron started to appear in the Near-East. Present only in small quantities at first, it had replaced bronze for most uses by the 7th century BC. A recurring question among scholars in the field is the cause for this transition. Why did people start using a metal that was admittedly more widely available, but also more difficult to smelt and work than bronze? Several hypotheses have been put forward, including the ubiquity of the ore and the mechanical superiority of iron.

In the 1970s Anthony Snodgrass and Jane Waldbaum suggested that the appearance of iron in the eastern Mediterranean might have been an indirect consequence of the collapse of several major civilizations towards the end of the 2nd millennium BC (Snodgrass, 1971, pp.231-39; Waldbaum, 1978, pp.12-73). They argued that these political events caused major disruptions in trade routes and in particular in the trade of tin, which in turn led to difficulties in producing bronze. This would have encouraged metal workers to use iron as an alternative material. Although their region of interest was located to the west of ours, the question of the appearance of iron as a response to a crisis in bronze supply remains interesting. Waldbaum herself questioned the validity of this hypothesis (Waldbaum, 1989), but so far, no alternative explanation is entirely satisfying. Having reviewed the changes in social setting and political organisation that occurred in and around Iran in the second half of the 2nd millennium BC (Chapter 1) and the changes in bronze technology and metal flow during this period (Chapter 4, 5 and 6), we can reconsider the validity of this hypothesis in a new light.

In Chapter 1, we have seen that in our area of study, a period of urbanisation and international exchanges gave way around 1600 BC to a period of hiatus, marked by a decrease in settlement density especially in the east and probably a shift to a more nomadic lifestyle until the beginning of the 1st millennium when the density of settlements increased again. The repercussions of these disturbances, whatever their causes, are visible in the metallurgical data. In Chapter 3 we have seen that it has had an impact on the availability of data. Indeed, very few objects from Iran or the neighbouring regions and dating from the mid and late 2nd millennium BC have been subjected to chemical analysis (Figure III-7 and Figure III-8). This has made the understanding of the exact nature of the transition into the Iron Age difficult, however we have been able to compare the chemical data before and after the hiatus and clear differences have become apparent. In Chapter 4, we argued, based on textual and archaeological evidence and on a study of the tin content of the objects in our database, that an organised international trade of tin gave way to a reliance on more local sources. We suggest that in the Iron Age, the Persian Gulf trade lost of its importance and tin from the east was potentially traded into Oman, but not much further. On the other hand, sources in the Zagros Mountains, such as Deh Hosein and possibly others yet undiscovered, seem to have supplied the intensive bronze production in north-western Iran and Luristan and Azerbaijan in particular. The results from Chapter 6 mirror these conclusions: a great predominance of AsNi, metal in the regions implicated with the Persian Gulf trade suggests an organised trade of this metal. In Mesopotamia and in Luristan in particular, the decline of this metal group in the Iron Age is evident, suggesting the loss of access to copper from the east. AsNiSbAg and AsNiSb metal, possibly traded to Iran from the west also decline at the end of the 3rd millennium BC showing that western sources or trade routes were also disrupted. Other metal groups,

on the other hand, gain more importance in the Iron Age. This is the case of As only metal in Luristan and Amlash, Ni only metal in Oman and NiSbAg metal in Azerbaijan. Although the source of the metal used in north-western Iran is still unknown, we can postulate a reliance on sources from the Zagros or the Iranian plateau.

These changes are undoubtedly related to some degree to the disturbances of the end of the 2nd millennium BC. The fall of Harappa in particular must have been one of the causes for the decline of the metal trade through the Gulf. The changes we have observed show that the disruptions led to a reorganisation of the metal trade, but was the pressure on metal supply enough to have also triggered the beginning of the use of an entirely new metal? To answer this question, we look at two technological aspects that could indicate a real difficulty in getting hold of copper or tin: alloying and recycling.

We suggest that if it truly became difficult to obtain tin towards the end of the 2nd millennium, this may show in the evolution of the ubiquity of tin-bronzes in the assemblage or in the tin contents of the bronzes. In Chapter 4, we have demonstrated that, in Iran, the mid and late 2nd millennium BC were marked by a continued increase in the ubiquity of tin-bronze, and little variation in the tin content of these bronzes. In other regions and in particular Mesopotamia and Oman however, this period sees a stagnation of the ubiquity of tin-bronze. The decline of the Persian Gulf trade may well have affected these two regions, but it seems that north-western Iran's metallurgy remained mostly untouched by the changes and getting hold of tin wasn't too challenging.

The second indicator examined to have an idea of the pressure on metal supply is recycling. We can imagine that if importing tin became difficult, the metalworkers might have resorted to recycling the bronze they already had. In Chapter 6 we have

indeed demonstrated that the practise of recycling was used in the Near-East. But this practise appears to have originated around the beginning of the 3rd millennium BC, not at the end of the 2nd. Indeed, certain groups of metal that can be seen as a degraded form of more common groups show a gradual accumulation throughout the Bronze Age and the Iron Age. Recycling in the Iron Age appears to merely be a continuation of practises already known and used in the Bronze Age. Moreover, we have seen that the degradation of certain metal groups can sometimes be tentatively associated to secondary alloying of objects of this group. This suggests a ready access to tin, rather than a shortage. Rather than being an indicator of a pressure on resources, it seems that recycling is a better indicator of the level of familiarity with copper and its alloys and of the understanding of the properties of copper and its ability to be re-melted, re-alloyed, recast and mixed. Interestingly, the appearance of “recycled” metal groups occurs in the first half of the 3rd millennium which coincides with the appearance of tin-bronze objects. In Chapter 2 we have seen that the end of the 4th and the 3rd millennium BC are also marked by a diversification of the repertoire of objects and the developments of new metal-working techniques (lost-wax casting and production of sheet-metal objects for example etc.). At that time, the arsenic content of the objects also reaches a maximum and metal-workers no doubt experimented with different ways of producing arsenical copper. All in all, it seems that recycling, just like alloying with tin, appeared about 2000 years after the beginning of a widespread use of copper in the Near-East. During these two millennia metal-workers would have had the time to familiarise themselves with all of the possibilities offered by copper. In his thesis Bray came to the same conclusions and summarised this idea as follows:

*“Rather than being a pseudo-economic response to dwindling metal stocks, recasting and mixing appear to be a consequence of familiarity with metal and therefore a*

*comfort with regularly changing artefact shape and alloy composition.*” (Bray, 2009, Ch.7)

Going back to the question of the appearance of iron, we cannot conclude that the use of recycling in Iron Age Iran and Mesopotamia indicates a difficulty to acquire copper and tin and therefore the need for the use of a new metal. However, we can be quite confident that iron first appeared within a well developed and well understood tradition of copper making. In Chapter 2 we have seen that the oldest iron artefacts recovered in Iran come from the north-west of the country, a region with particularly active copper industries in the Iron Age. Of course, part of the reason for this is that the south and the east are almost archaeologically silent in the Iron Age. Nevertheless, one cannot help but wonder if this dynamic copper-making tradition played a role on the appearance of iron. Another interesting question is that of the influence of Iran’s north-western neighbours on the introduction and development of iron-working in Iran.

b. North-western influence on the appearance of iron

Chapter 4 and 6 in particular have demonstrated through the study of tin profiles and As, Sb, Ni and Ag contents that Luristan, Iranian Azerbaijan and potentially Amlash were regions of primary bronze production in the Iron Age. The source of the metal used in these three regions is still uncertain. We have seen that the mine of Deh Hosein is a good candidate for both copper and tin used in Luristan, but the metal used in Azerbaijan is slightly different (NiSbAg type) and the source of the copper and tin used there is unknown. We have postulated the existence of other, yet undiscovered, sources in the northern Zagros that could have had a major role in supplying copper and perhaps tin to Azerbaijan. Muhly, before the discovery of the Deh Hosein source, had already

suggested this possibility and our analysis seems to reinforce his hypothesis (Muhly, 1973). From his study of Old Assyrian and Babylonian texts and of the tablets found in Kanesh he suggested a trade route bringing tin from the northern Zagros to Assyria, via passes at the level of Hasanlu and Dinkha Tepe and then being redistributed from Assur to Anatolia and down the Tigris to Babylon. These texts for the most part date from the beginning of the 2nd millennium BC, but given that north-western Iran appears to have been active in producing bronze it seems possible that such a trade pattern would have survived into the Iron Age. The question of the trade of metal between Azerbaijan and Assyria in particular has been recently discussed in Thornton and Pigott's study of Hasanlu's weaponry (Thornton & Pigott, 2011). They noticed that unlike other categories of objects, the metal weapons of Hasanlu had few parallels in Assyria but had similarities with types found in Transcaucasia and Gilan (see Chapter 1). However, this doesn't mean that raw materials couldn't have been traded through the Zagros and towards Assyria. It is also conceivable that bronze objects were traded from Azerbaijan to Assyria but then recast to suit the Assyrian style. Unfortunately, our database only holds six objects for Assyria in the Iron Age making it impossible to test this hypothesis.

All in all, it seems likely that, at the beginning of the Iron Age, the regions of north-western Iran interacted closely with their neighbours, particularly through the trade of metals. How did iron fit into this picture? Was it brought into Iran from these neighbours, perhaps in exchange for copper or tin? Was the idea of working iron introduced by them to north-western Iran, only to then flourish in a region of skilled metalworkers?

In 2004, McConchie studied the development of the Urartian iron industry. This kingdom located to the north-west of Iran and centred on Lake Van has yielded an

important number of iron objects, notably during the excavation of the capital Tushpa and the fortress of Toprakkale. The fortress of Teishebaini (modern Karmir-Blur in Armenia) is thought to have been an important iron-working centre when it was under Urartian influence in the 7th century BC (Pleiner, 1969, pp.17-18). The presence of an iron working civilization in Urartu is also attested by written evidence: the cuneiform ideogram for iron AN.BAR appears in the annals of king Sarduri II (764-735 BC). It states that the king ordered a seal of iron to be made during a campaign against Kulkha. Assyrian annals also mention iron in the form of spoils taken from Urartu after it had been defeated by king Sargon II (Pleiner, 1969, pp.16-17). McConchie however highlighted the fact that very few Urartian iron objects predate 800 BC, which is about the time when the Urartian kingdom expanded its borders all the way to Lake Urmia in north-western Iran (McConchie, 2004, p.76). As we have seen in Chapter 2, in Iran the development of iron was well under way by that time, and although it was still mostly limited to weapons and ornaments, an important number of objects were recovered in Azerbaijan, Luristan and Amlash. McConchie concluded that the contact between Urartu and its Iranian neighbours might have triggered the development of iron in Urartu and that “iron was adopted thereon comparatively swiftly and enthusiastically” (McConchie, 2004, p.81). This may be further proof of the influential position of north-western Iran in terms of metallurgy, but it shows that Urartu is unlikely to have been the source of the impetus for iron-working in Iran.

Another region that might have played a role in the appearance of iron in Iran is Assyria. There, texts mention iron from an earlier date: it is first mentioned in the inscriptions of king Shalmaneser I (1274-1245) (Grayson, 1972, p.84; Postgate, 1973). In the 12th century BC iron-working seems to have been important enough to require the presence of a blacksmith at the court of Ninurta-Tukulti-Assur (Curtis et al., 1979).

At that time, king Tiglath-Pileser also reports using iron arrow-heads for hunting purposes (Grayson, 1976, p.16). However the texts only start mentioning iron in quantity from the 9th century BC onwards. In this period it is regularly mentioned as part of tributes given to Assyrian kings after a military success. The archaeological evidence comprises of a large number of iron artefacts dating from the 8th and 7th century BC discovered at Assyrian sites and in particular at Khorsabad, Nineveh, Ashur and Nimrud.

In 1974 Pleiner and Bjorkman summarized both textual and archaeological evidence in a comprehensive review of Assyrian iron (Pleiner & Bjorkman, 1974). They found that from the mid 13th century BC, when iron first appears in texts, to the end of the 10th century, it seems to only be mentioned in relation with the court and the palace. From the 9th century onwards there is evidence of the use of iron for military purposes through weapons and sapping implements. Other types of tools (knives, agricultural implements, etc.) appear in the middle of the 8th century BC. It is also from that period that we have the first mentions of iron used for fetters, handcuffs and bonds, objects that appear very frequently in Neo-Assyrian texts and even became a symbol of the king's power. From the 7th century there seems to have been a well established iron civilization in Assyria, where iron was used for all kinds of tools and weapons and also as a structural material for building.

Overall, when comparing this picture to the one presented in Chapter 2 for Iran for which we sadly do not have the luxury of textual evidence to fine-tune the picture provided by archaeological remains, it seems that the development of iron in Assyria and Luristan occurred quasi-simultaneously. With no clear anteriority of one over the other, it is difficult to establish the influence that Assyria could have had on north-western Iran and vice-versa. Moreover, as mentioned above, the weaponry from

Hasanlu shows little parallels to the Assyrian material. So, even if the two regions were clearly in contact, as attested by the presence of parallels for other types of objects, and material resources (probably including copper and tin) and ideas undoubtedly travelled from one to another, the development of an iron-working tradition in Iran is likely to have had an important indigenous component.

c. Indigenous origin for Iranian iron-working

In their 2009 article on the production of speiss at Tepe Hissar Thornton *et. al* suggested that Iran might have been “one of the ‘heartlands’ of iron metallurgy” (Thornton et al., 2009, p.315). The discovery of speiss at Tepe Hissar other Iranian sites (Shahr-i Sokhta and Arisman – see Chapter 2) and in Anatolia led them to suggest that the production of ferrous speiss could have been “widespread and formalized” from as early as the 3rd millennium BC. Although, this ferrous alloy was probably related to an operation designed to provide a more reliable source of arsenic, they considered that the metal-workers were likely to have been familiar enough with iron-like metals not to discard it. Indeed, there is ample evidence for the sporadic occurrence of iron objects in the Near-East in the 3rd millennium BC (Waldbaum, 1999, pp.28-31).

This familiarity with the metal and the presence of workable iron deposits in Iran might have been the precursors for an independent development of iron-metallurgy within a community of skilled metal-workers. Indeed, although iron is not excessively abundant the Iran, there are still a number of deposits in north-western Iran as well as eastern Iran that could support a local production of iron (Figure II-6).

We have seen that, by the second half of the 2nd millennium, when iron first came about, copper was fairly well understood: copper-based metal objects were recycled, tin-bronze had already been used for more than a millennium, and although its ubiquity was still increasing, the levels of tin added had reached a “normalised” level. It wouldn’t be surprising for the bronze-smiths, especially in “aesthetically sensitive” regions like Luristan, to have been looking for new things to do with the metal, to change the appearance or properties of the objects (Pigott, 1999b, p.95). In other words the first steps in the development of iron-working, rather than following a separate path to copper metallurgy, were simply yet another innovation of this industry. The development of iron as a result of the experimentations of bronze-smiths is attested notably by the fact that many of the earlier occurrences of iron objects are in fact bi-metallic objects made partly of copper-based metal and partly of iron. But bronze and iron have very different properties (see Chapter 2). How then was iron perceived and understood by the bronze-smiths?

d. Early understanding of the properties of iron – the question of craft specialization

We have seen that the metal-workers understood the “changing” nature of copper alloys relatively well. They knew how to cast it, cold work it, alloy it, alter it and recast it into entirely new objects. What then would they have made of a metal that doesn’t allow for such flexibility? Iron can admittedly be forged and slightly altered multiple times, but it wasn’t cast until much later in history in this part of the world and doesn’t allow for alloying or complete remodelling as bronze does. Would this have made it seem more “eternal” in the eyes of the metal-workers (rather ironically given its rapid corrosion

rate), in the sense that the processes that lead to a finished iron objects are on the whole irreversible?

What is certain is that iron was a luxury when it first appeared. As mentioned in Chapter 2, the first objects made of iron were mostly small ornamental objects. This is not surprising given the difficulties that the metal-workers would have no doubt experienced to produce them. Towards the end of the 2nd and the beginning of the 1st millennium, iron was becoming more common, but we suggest that it was still worked by the same people that made bronze objects and not fully understood. Two points support this hypothesis:

- The existence of two different types of “transitional” objects, both described in details by Moorey (1991): bi-metallic objects (such as swords with an iron blade and a bronze grip or pins with an iron shaft and a bronze head) and the iron bracelets and swords mentioned in Chapter 2 that were made of many different pieces riveted together.
- The findings of metallographic analysis of early iron objects presented in Chapter 2. These have shown that the structure of most early iron objects was quite inhomogeneous and that the use of processes aimed at “improving” the iron was very limited (France-Lanord, 1969; Smith, 1971; Tylecote, 1972; Pigott, 1981).

The highly decorated Luristan short-swords in particular have split scholars between those arguing that the lack of welding, the use of iron for such intricate shapes and the overall bad quality of the iron demonstrated a poor understanding of the metal (Maxwell-Hyslop & Hodges, 1966, p.169) and those who argued that such a virtuous

display of skill could only be the product of a well established industry (Ternbach, 1964, p.51). It seems that both sides of the story are in fact not incompatible. The objects were indeed made by expert metal-workers who had developed good forging skills, and they were representative of a well-developed and active metal industry, but this industry was still mostly one of unspecialised metal-smiths that were mostly used to copper-alloys and therefore did not yet understand the very different properties of the new metal. However, interest in iron is evident at this stage, and as mentioned by Rehder, these swords suggest an “intentional display of virtuosity” (Rehder, 1991, p.16). They are likely to have been a mark of prestige, intended for rituals or perhaps a way of showing off newly acquired forge-skills.

The bi-metallic objects are an obvious indication that both metals were worked in a single workshop. They may however be the first indicators of a better understanding of the metal. Indeed, in a great majority of cases iron is used for the working part of the object and contrary to the bracelets and swords mentioned above, no attempt is made to decorate the iron parts (Moorey, 1994, p.285). It is unclear whether this indicates an understanding of the potential of iron to be harder and stronger than bronze or whether it indicates a realisation that it was not adapted to intricate designs. Either way, bi-metallic objects seem to represent the beginning of a differentiation of the two metals.

If the production of finished objects of bronze and iron were carried out in the same workshops resulting in a delayed specialisation of skills, the story was possibly different for an earlier stage of the production phase: the smelting. Indeed, the two metals require different smelting conditions and although the same type of furnaces can be used, the control of the conditions necessary to obtain a workable metal is much more difficult for iron (Moorey, 1994, p.282). More significantly, it is likely that these activities would have been carried out close to the mines (Moorey, 1994, p.243), i.e. in different places

and by different people for bronze and iron. This separation of tasks might be responsible for an earlier understanding of the properties of iron by the people who smelted it than by the people who forged it to its final shape. In particular, Rehder noticed the presence of internal welds in the Luristan shorts-swords that were probably the result of the consolidation of several small blooms into a larger one after the smelting, but he noted that welding was never used to join the different parts of the swords together (Rehder, 1991, p.18). However, the level of knowledge of iron-smelters must not be overstated: regarding the welding in particular, Pigott argues that “the process of coalescing a bloom or blooms through a sequence of heatings and hammerings does not necessarily imply a true understanding of forge welding” (Pigott, 1999b, p.93). On the subject of the smelting itself, given the inhomogeneous carbon content and variable slag content of Iranian iron until relatively late (France-Lanord, 1969; Smith, 1971), it would appear that control of smelting conditions in the furnaces was a skill that, just like “true blacksmithing”, took several centuries to develop.

In their thorough review of Assyrian iron metallurgy, Pleiner and Bjorkman remarked that although the texts differentiate blacksmiths and copper-smiths from as early as the 12th century BC, there is textual evidence of both blacksmiths working with copper alloys and copper-smiths working with iron. On the subject of specialization they note: “So the process of specialization, caused by the different properties of the material and begun early (probably among palace smiths), was slow, and most smiths had to master a broader field of operations.” And also: “Other kinds of work, bi-pointed bars, and large chains, and heavy tools apparently manufactured in sets, are indicative of a true blacksmith. It is not impossible that a sort of specialization took place on another level, viz., the production of different types of goods.” (Pleiner & Bjorkman, 1974, p.303). It seems possible that a similar situation would have also occurred in Iran: the production

of iron objects remained intimately associated with that of bronze ones until, probably triggered by the need for bigger, stronger objects, a branch of “true blacksmithing” developed. By Iron Age III in Iran, iron objects are representative of this shift to a better understanding of iron. They are for the most part made entirely of iron and present simpler shapes, better adapted to forge work.

e. The status and symbolism of early iron

So for the metal-smiths, it seems that until relatively late iron would have been considered as a metal different to bronze, of course, but used as an extension of the same craft. But how did the people who used the objects consider this new metal as compared to bronze? Did they accept the new material easily despite its greyish and easily corroded appearance?

In Iran, the limited number of iron objects recovered and the fact that most of the sites where iron was unearthed are burials means that assessing the status of iron in the society, beyond the simple view of iron being first considered as a precious material before it became utilitarian, is difficult. In other Near-Eastern civilisations, the presence of texts relating to early iron can at least inform us on its status within the highest levels of the society. In Assyria, iron and in particular iron daggers were a symbol of military strength. However this symbolism was not exclusively reserved to iron and in certain texts bronze is found in the same contexts (Pleiner & Bjorkman, 1974, p.305). In some Hittite and Neo-Babylonian texts iron appears as symbol of “permanency, indestructibility” and “deathlessness” (Pleiner & Bjorkman, 1974, p.305). This perhaps echoes what the metal-smiths would have rapidly noticed: that iron does not have the ability that bronze has to be almost infinitely remodelled. In any case, in the early

centuries of the mention of iron in texts, it appears that it is very much a metal of the kings (Pleiner & Bjorkman, 1974, p.286) who probably had an interest in its novelty and rarity. It is perhaps under their influence that the metal-smiths developed their intricate forging abilities for the production of spectacular display items. The development of the more practical side of blacksmithing on the other hand could have been driven forward by the use of iron in the military (see Pleiner & Bjorkman 1974, p.296). Indeed, both in Assyria and in Iran weaponry is the best represented category of objects in the beginning of the Iron Age.

## 2. Strengths and limitations of the synthesis of old datasets

As exposed in Chapter 3, the reassessment of bronze metallurgy undertaken in this thesis relies on data collected from a total of 51 different publications that add up to over 5000 objects. In this section we discuss the strengths and limitations of this approach and the direction to give to further work.

### a. The synthesis of old datasets: a promising approach

Traditionally, it is considered that advances in the understanding of ancient metallurgy will be made by analysing new collections of objects if possible with new analytical methods (Pernicka, 1998). However, a more recent approach pioneered in particular by Bray in his doctoral thesis (Bray, 2009) has been to try and find some archaeological meaning in old datasets. Indeed, the compilation of published analyses can produce surprisingly large datasets. For Iran only, we have seen that an unexpectedly large total of over 2000 analyses of copper-based objects have been published. Not only is the

amount of readily available data larger than probably suspected, but Bray has demonstrated that despite a number of problems related to the use of different and sometimes old techniques, the data is richer in archaeological information than is often believed. His main contribution has been to consider that given the knowledge of the behaviour of a limited number of chemical elements during processes of transformation of copper-based metals, we can observe, in a large enough dataset, broad geographical and chronological patterns that can be linked to the movement or alteration of the metal. Eventually this information can allow us to sketch a picture of the metallurgy of this region, even if the individual stories of the objects are all mostly lost.

The application of this method to a dataset of Near Eastern objects has enabled us not only to better understand the transition between the Bronze Age and the Iron Age in this region, but has also been a way to test Bray's approach with a whole different set of data, in a very different archaeological setting. The starting point of this undertaking was that although the archaeology is different, the underlying chemistry is universal: the fact that arsenic is lost in higher quantities than nickel when copper is melted should be true in Iron Age Iran just as it is in Bronze Age Britain. And indeed the results obtained here are promising: we have been able to identify believable trade and recycling patterns. Notably encouraging is the fact that for a given metal group, the regions with the lowest ubiquity of this group are often also the regions where the metal is most degraded (lower levels of As and Sb) indicating that they are far removed from the production area. Also encouraging is the observation of different distributions of tin contents in "recycled" metal groups (a higher proportion of objects in the 2-6% Sn and 10-15% Sn ranges) that could indicate the practises of remelting and secondary alloying.

One of the strengths of this approach is that it works from the bottom-up: letting the chemical data speak for themselves before it is reconciled with the current understanding of the archaeological setting. This means that there is a much smaller chance of repeating old misconceptions. In particular in this thesis we have been able for Mesopotamia to challenge the vision that tin-bronze was first used for ornaments, then for weapons and finally for implements (see Chapter 4). But this bottom-up approach importantly also means that we can infer the presence, in a particular area, of a significant metallurgical centre, even if it has not yet been discovered. In our case for example, we can perhaps suggest the presence of one or several copper mines and conceivably also tin mines in the north-eastern Zagros or Azerbaijan able to fuel the important industry of Azerbaijan in the Iron Age and perhaps even earlier. Indeed, Azerbaijan is known to have important copper deposits, but at present our knowledge of mining there is limited to the presence of a grooved hammer and “traces of ancient mining” reported from the Mazraeh area (Nezafati et al., 2008). Similarly mineralisations containing tin are now known to occur in the Sanandaj-Sirjan geological zone of Iran, but Deh Hosein is the only confirmed mine with Iron Age working (Nezafati et al., 2008). This aspect is particularly interesting for areas of the world where very little is known about potential metal sources and metal-working centres and could be used to guide prospection work in the field.

b. Limitations to this approach and further work

In Chapter 3, we have mentioned a certain number of limitations and difficulties due to the nature of our dataset. In particular we have discussed problems in the chronology of Iranian sites, the geographical patchiness and fragmentation of the data and the

difficulty of integrating studies using different analytical methods. We will not go through these again but instead assess what the true limiting points were in our attempt to better understand Iranian metallurgy.

Although a better chronological definition would be desirable, we have seen that much can be said already with a division in roughly thirds of a millennium. Plots showing chronological evolutions of certain quantities (the ubiquity of tin bronzes in Iran for example) have shown surprisingly smooth variations and enabled us to describe the evolution of copper-based metallurgy with more precision than before despite a relatively rough scale.

More limiting on the other hand is the patchiness of the data in some geographical areas. For our study of the provenance of tin in particular, we have been greatly restricted by the small number of analyses of metal from Afghanistan, the Indus Valley and Bactria. Although the synthesis of old datasets seems to have a promising future, in some areas new analyses would also be welcome. One of the benefits of assembling old publications is that it makes the identification of regions and time periods for which more data is required very straightforward. In our case, for the tin story, data from objects of sites from the Helmand tradition for both the Bronze Age and the Iron Age would be particularly needed. For the understanding of the transition of copper-based metallurgy into the Iron Age, we have seen that data from Iranian Azerbaijan would be greatly helpful as we suspect that this region played a major role in the production and trade of tin bronze in the Iron Age, but also potentially in the Bronze Age, which is a period for which we have almost no data. For the same reasons, a compilation of previously published analyses of Caucasian, Syrian, and Anatolian copper-based metal would also be greatly beneficial as it would potentially help understand the source of the

AsNiSbAg and AsNiSb metal that appears to have been traded to Assyria, Mesopotamia and western Iran.

We realize that for some of our metal groups, the “tentative stories” proposed are based on the analysis of very small datasets. However these stories are only intended as a working hypothesis that can be a basis of reflection for future work. A way to assess the significance of our observations would be to use a statistical test such as the Mann-Whitney U-test (Bray & Pollard, 2012). Obviously, a greater bulk of chemical analysis would help make our stories stronger, but other approaches could also help verify them. In particular there is a need for a bigger LIA database as we have seen that, at the moment, the data available can only provide very limited information. Prospection and excavation work in the field will also help refine or modify our conclusions. The recent discovery of the mine of Deh Hosein has been instrumental in better understanding the Luristan metallurgy and the study of Iranian metalwork would greatly benefit from other such discoveries.

### 3. Conclusion: the study of large datasets as a basis of reflection on technological innovation

Through the question of the transition between the use of bronze and the use of iron, this thesis has touched on the much wider subject of technological innovation. This subject encompasses a number fascinating questions, many of which still have a place in our modern world. Amongst these are: How are new technologies “discovered”? What are the forces that drive their adoption? How do they spread? Why are they adopted faster in some areas than others?

Our study of the bronze dataset has enabled us to discuss the adoption of several “new technologies”: the alloying of copper with tin, the recycling of copper and the adoption of iron. We have demonstrated that phenomena that are usually intuited by discoveries made during excavations can be shown with more precision and even quantified with the study of large datasets. For example, the graph showing the evolution with time of the ubiquity of tin-bronze in Iran (Figure IV-2) clearly shows a period of experimentation with tin-bronze in the 4th millennium BC, followed by the beginning of the adoption of this new technology in the 3rd millennium and the technology finally reaching maturity in the late 2nd millennium BC. Quantifying the rate of adoption of a new technology can be interesting in that it allows for more precise comparative studies of different technologies or different regions for example.

Of course, these numbers and graphs then have to be reconciled and explained with our knowledge of the social setting in which the innovations took place. The present study has only skimmed the surface of this complex question and in particular presents very little discussion on the outlook that the people who used the metallic objects had on the different metals. However, it does reflect on the technological innovations from the perspective of those who made the metal objects. In particular we argue that the adoption of iron metallurgy in Iran is to be seen as the extension of the craft of the bronze-workers rather than the adoption of an entirely separate technology. This discussion is based not only on the nature of the early iron artefacts, but also on the realisation of the level of development of the bronze industry, made possible by our study of a large dataset of analysis. Indeed, we have demonstrated that in the beginning of the Iron Age in Iran, there wasn't, economically speaking, the need for a new metal. Although the Persian Gulf trade was greatly diminished, it seems that the use of local sources made for a flourishing bronze industry and we have argued that in Iran the Iron

Age is to be seen as a “mature Bronze Age”. This thesis has made obvious the fact that tin-bronze had been around for about a millennium and a half (although only becoming ubiquitous in the Iron Age) and that the recycling of copper-based objects had been in practise for almost as long, meaning that the bronze-smiths had a very good understanding of the “fluid” nature of this metal. What perhaps pushed the bronze-smiths to expand their repertoire and experiment with iron, beyond simply curiosity or enthusiasm for the different aspect of the metal, might have been the more durable, inalterable nature of iron and the symbolism ensuing from the realisation of this property.

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## Appendix A: Chronology

Date BCE	Mesopotamia		Elam		Luristan	Western Iran	
	Assyria	Babylonia	Khuzistan	Fars			
3000		Jemdet Nasr	<i>Susa III</i>	<i>Anshan</i> <i>Banesh</i>	Early Bronze I	<i>Godin IV</i>	
		Early Dynastic I-II		"Transitional"			
2500		II-III	<i>Susa IV</i>		Early Bronze II	<i>Godin III</i>	
	Akkadian				Early Bronze III		
	Ur III		Old Elamite	Kaftari			
2000		Isin Larsa					
	Old Assyrian	Old Babylonian	<i>Susa V</i>				Middle Bronze
1500	Mitanni				Late Bronze	<i>Hiatus</i>	
	Middle Assyrian	Kassite period	Middle Elamite <i>"peak of Elam"</i>		Iron I A		
		2nd Isin Dynasty			Iron I B		
1000		<i>Instability</i>	<i>Decline</i>		Iron II A		
	Neo-Assyrian		Neo-Elamite	<i>Hiatus?</i>	Iron II B		
					Iron III	<i>Godin II</i> Medes	
500		Neo-Babylonian					

Date BCE	Iranian Azerbaijan	Urartu	North-Central Iran	North-Eastern Iran	Bactria Turkmenistan		
3000	Early Bronze <i>Hasanlu VII</i>		<i>Ghabristan / Arisman</i> <i>Sialk IV</i>	<i>Hissar II</i>	<i>Namazga III</i>		
			"Kura Araxes"	<i>Abandonment</i>	<i>Namazga IV</i>		
2500							
2000					Central Grey Wares		<i>Namazga V</i> BMAC
			Middle Bronze <i>Hasanlu VI</i>				<i>Namazga VI</i>
1500					<i>Yaz I</i>		
	Iron I <i>Hasanlu V</i>						
1000	Iron II <i>Hasanlu IV</i>	Pre-Urartian	<i>Sialk V</i>		<i>Yaz II</i>		
			<i>Sialk VI</i>				
	Iron III <i>Hasanlu III</i>	Urartian		<i>Tureng IV</i>			
500					<i>Yaz III</i>		
		Post-Urartian					

Date BCE	South Eastern Iran			Oman	Indus Valley		
	Jiroft	Kerman	Sistan				
3000	Proto-Elamite expansion	<i>Tal-i Iblis IV</i>	<i>Shahr-i Sokhta I</i>	Hafit	Early Harappan		
	Abandonment	<i>Shahdad Takab IV</i>	<i>Shahr-i Sokhta II</i>				
2500			Yahya IV	III	<i>Shahr-i Sokhta III</i>	Umm An Nar	Harappan
	<i>Shahr-i Sokhta IV</i>						
2000	II					Wadi Suq	Late Harappan
1500	Hiatus			Late Bronze Age			
				Iron I	Black and Red Ware		
1000				Iron II <i>Lizq</i>		Painted Grey Ware	
				Yahya III			
500	Yahya II			Iron III			

