Copper for the afterlife in Predynastic to Old Kingdom Egypt: provenance characterization by chemical and lead isotope analysis (RMAH collection, Belgium)

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1 - Introduction

Following the first detailed work on ancient Egyptian metallurgy by Garland and Bannister (1927), large-scale characterisations of Egyptian copper alloy compositions have been undertaken (e.g., Cowell 1986, 1987, Farag 1981, Kallfass and Hörz 1989, Lucas 1962, McKerrell 1971, 1973, Pelleg *et al*. 1979, Riederer 1978, 1981, 1982, 1983, 1984, 1988 and more recently Kmošek *et al*. 2016, Philip 2006). These studies have outlined an "evolution" of copper alloy use in Egypt (see Table 1 for periodization used), from copper and arsenical copper in the Predynastic, Old Kingdom and 1st Intermediate Period, to the introduction of tin bronzes in the Middle Kingdom. By the New Kingdom, tin bronze generally replaced arsenical copper, while leaded tin bronze made its appearance during the late New Kingdom, to remain the dominant alloy throughout the Late, Ptolemaic and Roman Periods. Such broad trends must be read critically, however, as they were often based on analytical programs focusing on particular object typologies, sometimes lacking (discussion of) archaeological context. As such, these trends stand to be evaluated to improve our understanding of Egyptian alloy selection for particular purposes in specific contexts. Such an overview should take into account the deep diachronic dimension in which Egyptian copper metallurgy is situated: over three thousand years of technological history are only slowly being illuminated through the recent study of production remains (e.g., Abd el-Raziq *et al*. 2011, Fitzenreiter *et al*. 2014, Rademakers *et al*. 2018b), with many open questions remaining. For example, the method of arsenical copper production remains a moot point, as it does in so many other early contexts across the ancient world (e.g., Charles 1967, Rehren *et al*. 2012). While this paper does not directly address issues of production technology, it touches on these obliquely by considering the provenance of the copper ores used to produce early Egyptian (arsenical) copper.

Very little provenance research of Egyptian copper integrating lead isotope (LI) analyses, has been performed so far. Predynastic copper from Maadi and Tell el-Farkha has been analysed (Abdel-Motelib *et al*. 2012, Rehren and Pernicka 2014), but no published LI data currently exists for the Protodynastic, Old Kingdom, First Intermediate Period, Middle Kingdom or Second Intermediate Period. LI data for New Kingdom copper alloys from Amarna and Pi-Ramesse have been published by Stos-Gale *et al*. (1995) and Rademakers *et al*. (2017), respectively. For the Third Intermediate Period no LI data is available, while Fleming (1982) and Schulze and Lehmann (2014) have published data on Late Period leaded bronzes. Others have measured LI ratios of other Egyptian materials, such as lead, silver, *kohl*, glass, faience, Egyptian blue and frit (e.g., Brill *et al*. 1974, 1993, El Goresy *et al*. 1998, Fleming 1982, Shortland 2006), which – as for leaded bronze – generally relate to lead rather than copper deposits (with the exception of faience and Egyptian blue, cfr. Rademakers *et al*. 2017).

Altogether, there currently exists an enormous gap in our understanding of copper provenance and its production technology in ancient Egypt, despite its importance to the ancient Egyptian economy. Indeed, from its early discovery and use as a precious material for ritual and secular display, its alloys soon grew into the most important functional metals in Egypt, valued for their mechanical properties. Moreover, copper played an important role in the production of various other materials, such as Egyptian blue, faience and coloured glass (Rehren *et al*. 2001), and was often used to alloy (debase) silver and gold (Rehren *et al*. 1996). Its versatile use in ancient Egypt can thus not be underestimated, and implies an intricate system of production, trade and consumption.

Some of this production clearly took place within ancient Egypt's borders throughout pharaonic history, through the smelting of copper ores mined in the Eastern Desert and Sinai Peninsula (Abdel-Motelib *et al*. 2012). From pharaonic to Roman times, the Eastern Desert (extending into Nubia) was equally exploited for gold (Klemm and Klemm 2013, Troalen *et al*. 2014), amethyst (Shaw 1998, 2007) and probably tin (Rademakers *et al*. 2018b and references therein), while other Sinai expeditions focused on turquoise mining (Shaw 1998). During the New Kingdom, the state's zone of influence further extended to the Timna valley, where Egyptian initiative led to a revival in mining activities (Ben-Yosef 2016). However, it is clear that the import of metal from abroad and the recycling of circulating copper must have played an important role in Egypt's layered copper economy – as has been established for the New Kingdom (Rehren and Pusch 2012, Rademakers *et al*. 2017). The importance of these various provisioning systems through time stands to be evaluated.

This paper presents new chemical and LI data on copper alloys from the Predynastic, Protodynastic and Old Kingdom periods, as a first step towards establishing a more continuous history of copper provisioning in ancient Egypt. Though this represents only a first few steps in the dark, these results nonetheless outline important new avenues towards considering early Egyptian copper provisioning. The data are obtained through the analysis of artefacts from the Egyptian collection at the Royal Museums of Art and History (RMAH) in Brussels, Belgium. This collection was acquired at the start of the 20th century, from early archaeological missions in Egypt led by Petrie, Garstang, Amélineau and others. A broader overview of this collection's history, including full discussion of each artefact's excavation context and acquisition, is the subject of a forthcoming publication. Interestingly, finds from many of these early excavations were dispersed to various European collections (e.g. Ny Carlsberg Glyptotek: Bagh 2011). The study of copper alloy artefacts within these collections (e.g., Odler 2016, Kmošek *et al*. in preparation) promises to add further dimension to the results presented here, and welcome discussion on this under-studied subject.

With three exceptions, all analysed artefacts derive from funerary context, and thus represent a particular copper consumption sphere: some may have been purposefully made for the afterlife, while others may have served the owners during their lifetime or were perhaps taken out of broader circulation to accompany Pharaoh and his courtiers for eternity. They may thus differ from alloys commonly employed in, e.g., military or craft environments, and are perhaps more representative for consumption in a wealthy domestic sphere. Figure 1 presents the different ancient sites from which the artefacts were excavated and highlights important mining areas mentioned in this paper.

2 – Materials and methods

Prior to sampling, qualitative surface analysis by handheld XRF (X-ray Fluorescence) was conducted for the entire metal collection at the RMAH (over 500 artefacts), to ascertain the alloy types present (results to be integrated in the collection database). For each artefact, a detailed investigation of its post-excavation history was conducted to achieve the finest possible contextualisation. From the combined results of this contextual and qualitative compositional analysis, a selection of 47 samples was made, listed in Table 2. This Table includes essential contextual data, while further bibliographic details for each artefact are provided in the Online Supplementary Materials (OSM), along with photographs of each artefact. Figure 2 illustrates a selection of sampled artefacts.

Metal samples were either clipped using steel cutting pliers or drilled using a clean TiN-coated steel 1 mm drill bit to obtain core material. Prior to sampling, all surface corrosion was mechanically removed (Dremel rotary tool (Bosch®), steel brush) to ensure a metallic sample. In four objects, important corrosion was present, as noted in *OSM Table 2*. Ore fragments were ground down to a fine powder in an agate mortar, from which homogenised samples were taken.
All samples were completely dissolved following a high-temperature acid digestion procedure. One aliquot was retained for chemical analysis by ICP-OES (Inductively-Coupled-Plasma Optical Emission Spectroscopy), while the remainder was used for lead isolation and LI analysis by MC-ICP-MS (Multi-Collector Inductively-Coupled-Plasma Mass Spectrometry). Full details of sample preparation and laboratory procedures are provided in the OSM (cfr. Rademakers *et al*. 2018a).

When comparing ancient metals to ores or other metals, it is essential to consider the variable sensitivity of these elements to process-related changes in the course of their (usually unknown) technological trajectory (oxidation, volatilisation, metal-slag affinity … during metallurgical operations) which may obscure their initial relation with respect to (often heterogeneous) ore composition (Pernicka 1999). For this reason, we consider the order of magnitude of minor and trace elements, and the ratios between them, more instructive than their absolute contents. This encourages a careful approach to compositional data, in particular for corroded samples, which nonetheless offers important supporting evidence to LI evidence when discussing provenance and insight into metallurgical technology (Pernicka 2014).

3 – Results

LI and chemical analysis have been performed for a total of 40 metal samples and 7 ore samples. Complete LI data are provided in *OSM Table 1* and presented graphically in Figures 3 and 4. Full compositional data are provided in *OSM Table 2*, with selected elements presented in Figures 5, 6 and 7. Discussion of these results is organised along the artefacts' different find contexts, in a chronological order (sections 3.1-3.3). Specific compositional characteristics are discussed for each artefact, while a broader overview of compositional trends across the assemblage is presented in section 3.4.

Previous research has identified the Sinai Peninsula and Eastern Desert as the most likely source for early Egyptian copper. Copper most likely smelted from these sources has been attested already in Predynastic Maadi and Tell el-Farkha. No information on copper provenance in Protodynastic or Old Kingdom Egypt is currently available but continued use of these ores appears the best working hypothesis to test here, which is why comparisons to this source (data from Abdel-Motelib *et al*. 2012, Hauptmann *et al*. 1999, Pfeiffer 2013, Shortland 2006) are discussed in detail in the first instance. While this dataset is an extremely important resource for considering Sinai and Eastern Desert copper exploitation, it has some limitations. A large number of sites are presented, for which exploitation evidence spans several millennia: only few samples are thus usually available for each deposit or region, and not all pertain to the period under study. Furthermore, the data scatter widely along the geochemically expected trend lines, especially in the ratios normalized to ^{204}Pb (Rademakers *et al*. 2017). It thus offers an incomplete picture of Sinai and Eastern Desert ores, which cover a very large geographical area and vary widely in their LI ratios – including strongly radiogenic LI ratios in certain (Southern) Sinai ores.

Comparisons to Arabah valley ores (Faynan: Hauptmann *et al*. 1992, 2015; Timna: Gale *et al*. 1990) are included, as these nearby deposits were equally mined in the Early Bronze Age (EBA: ca. 3300–2100 BCE, roughly equivalent with Protodynastic to Old Kingdom), and likely traded across the Sinai desert (Ben-Yosef *et al*. 2016, Hauptmann *et al*. 1999). Contemporaneous artefacts are considered to give the most secure reference points, though ores discussed here all offer evidence of EBA mining and/or smelting.

As Figure 3 shows, LI ratios of the early Egyptian copper alloys and minerals at the RMAH are generally similar to the Eastern Desert and Sinai ores. This is explored in further detail in the sections below. Figure 4 presents a closer view of Figure 3, with all analysed samples labelled (more radiogenic samples labelled in Figure 3). Additional LI plots in the OSM provide a detailed comparison of the specific deposits within the Sinai and Eastern Desert to the analysed artefacts.

Comparisons to metals in use or circulating the EBA eastern Mediterranean and various regional ore sources (Arabian Shield, Oman, Lavrion, Anatolia) have been made as well (references and selected LI plots of these comparisons in OSM). Archaeological evidence for the exploitation of copper in these regions and the trade of goods between them and Egypt more generally is not always clear for this period (e.g., Diego Espinel 2011, Tallet and Marouard 2016). The hypothesis of "domestic" (geographical proximity of deposits) copper/ore use, for which mining and smelting evidence is abundant in the Sinai and Eastern Desert, is thus favoured although the import of copper from further away is not excluded *a priori*. Where LI and geochemical overlap between the studied samples and multiple ore sources exists, these Sinai and Eastern Desert ores are considered the more likely alternative unless compelling archaeological evidence argues otherwise. It is important to note that not all abovementioned ore deposits offer strong evidence for EBA exploitation, and that some LI data was measured for ore-hosting formations rather than the actual (possibly exploited) copper ores (e.g., Arabian Shield data). As such, these may be representative of available ore sources, but better field evidence is necessary to validate their possible exploitation and circulation in the EBA eastern Mediterranean.

3.1 – Predynastic period

The oldest artefact in the presented assemblage is a needle from a Nagada II tomb in Fayoum. It has LI ratios consistent with Southern Sinai (Wadi el-Regeita) and Eastern Desert (Wadi Semna) ores – both of which were exploited during the EBA. Based on antimony content and higher LI ratio similarity, a Wadi el-Regeita origin is most likely. It has almost 1% arsenic (not typical for Wadi el-Regeita) and overall low trace elements content. Extremely low iron content points to poorly reducing smelting conditions in the production of this copper, from relatively pure copper ore (Craddock and Meeks 1987).

3.2 – Protodynastic period

Tarkhan

This group of ore minerals from Petrie's excavations at Tarkhan may derive either from grave 81 (Dynasty 0 – "Predynastic") or grave 1061 (Dynasty 1 – "Protodynastic"). The copper carbonate has relatively low trace elements content, except for notable barium and zinc and 0.12% lead, and LI ratios compatible with an Eastern Desert provenance. The sulphide mineral is a mixed galena/sphalerite ore, with LI ratios again matching an Eastern Desert provenance.

Faras (Nubia)

Two "piercers" from different female graves at cemetery 3 are made of relatively pure copper (0.1-0.2% As). Differences in iron (very low for E03491), lead and antimony content indicate that they were produced during distinct smelting operations. Their LI ratios indeed verify that different ores were used, though possibly from relatively nearby (Southern?) Sinai deposits. They are isotopically similar to two sheet fragments from Khasekhemwy's tomb (E04825a-b), which have more elevated arsenic content.

Elkab

This chisel is corroded but appears to have had an important arsenic content. It isotopically matches Southern Sinai ores and production waste (and two Old Kingdom chisels from Bêt Khallaf discussed below).

Abydos

The copper and lead minerals from the first Dynasty Tomb M.12 in Abydos are compatible with an Eastern Desert provenance. In comparison to the earlier ore finds from Tarkhan, the copper ore has highly similar LI ratios, though lower barium, lead and zinc and higher iron contents. The LI ratios of the lead sulphide are near identical to those of the mixed sulphide mineral from Tarkhan and are consistent with those of Predynastic/Protodynastic galena/kohl previously attributed to Eastern Desert deposits (Shortland 2006). This includes two "burial galena" from Tarkhan and Abydos (Stos-Gale and Gale 1981). Though Stos-Gale and Gale (1981) did not attribute an Eastern Desert provenance to these galena, Shortland (2006) assumes the Gebel Zeit ore body may well extend to a $^{207}Pb/^{206}Pb$ value of 0.785 up to 0.808.

The adze from contemporary male Tomb 510 (different area of the site) is made of (ca. 1.1%) arsenic copper with remarkably high iron content (ca. 1.6%), indicating the use of poorly refined raw copper. Its LI ratios fall within the range of Eastern Desert and (Southern) Sinai copper ores, and other Protodynastic artefacts discussed in this paper – but differ from the first Dynasty minerals of Tomb M.12. No better match to known contemporaneously exploited copper (ore) can be currently suggested.

A later first Dynasty knife (Tomb 429) is marked by overall low minor (0.4% As) and trace elements content and LI ratios consistent with Southern Sinai and (central) Eastern Desert copper ores.

Gizeh

This first Dynasty adze (contemporary to the knife from Abydos) is strongly corroded but appears to have been another arsenical copper (arsenic content cannot be accurately estimated). Its LI ratios are similar to copper (ore) fragments recovered at Predynastic Maadi and a Nagada IIIc bowl fragment from Kafr Hassan Dawood (Hassan *et al*. 2015), and consistent with (Southern) Sinai ores.

From other graves around Mastaba V, ore minerals were recovered. The copper ore has more radiogenic LI ratios than those noted in earlier analysed fragments, with remarkable cobalt (and manganese) content.

The two lead sulphides are chemically similar but differ in their LI ratios. One is similar to the Tarkhan and Abydos sulphide minerals and Predynastic/Protodynastic galena/kohl (Shortland 2006) and compatible with an Eastern Desert origin. The other has more radiogenic LI ratios which may suggest a related provenance to the copper ore fragment. Such radiogenic LI ratios were equally noted by Stos-Gale and Gale (1981) in galena from Abydos and Nagada (and haematite from Nagada), which they could not match to any known ore source at that time. More recent data, however, allow for a possible Southern Sinai origin, though only copper ores with such LI ratios were described by Abdel-Motelib *et al*. (2012) at Wadi Samra[1]. Furthermore, the newly available data for (central) Eastern Desert copper ores forms an isochron with Eastern Desert lead ore data, implying Eastern Desert copper ores with radiogenic LI ratios may exist (Shortland 2006, Weeks *et al*. 2009). This remains speculative of course, but this finding may suggest the exploitation of geologically associated lead and copper minerals in Protodynastic Egypt.

Tomb of Khasekhemwy

The remaining Protodynastic artefacts all derive from the Tomb of Khasekhemwy (2nd Dynasty) but were obtained during separate excavation campaigns by Amélineau and Petrie: they were found in different rooms of the royal tomb (cfr. OSM).

The large vessel's surface colour – though influenced by unknown earlier conservation treatment – reveals it to be made of alloyed copper, and it indeed has a high (ca. 3.1%) arsenic (and notable zinc: ca. 0.2%) content. Its LI ratios are consistent with ores from Southern Sinai and in particular Wadi Tar, the only known arsenic-rich (associated with high zinc) copper ore deposit in the region (Hauptmann *et al*. 1999), though its lead content is relatively low in comparison. Its LI ratios are identical to those of a 3rd Dynasty arsenic copper chisel (E00916e, cfr. below) which has lower zinc and slightly higher nickel and antimony content.

From a different context in the same grave, over 1200 copper "objects" were discovered by Amélineau. Six copper sheet and strip fragments from this assemblage, now at the RMAH, have been analysed. They vary widely in chemical composition (e.g., 75 μg/g to 2.8% arsenic, 8-1600 μg/g lead) and LI ratios (from "old" to strongly radiogenic). It is therefore clear that they were not made from the same copper batch, let alone fragments from a single large sheet. Interestingly, some reveal marks of being cut, as if by scissors, while others are bent. No direct correlation between these marks and arsenic content (which would influence work-properties) could be noted to suggest intentional selection here, but this sample is of course quite small.

[1] More radiogenic ores still are noted at Wadi el-Regeita and Sheikh Muhsen, sometimes associated with copper sulphides. No high cobalt content was noted in these copper ores.

Their LI ratios are not consistent with variation in a single (known) deposit either, and probably represent different deposits. E04825b (ca. 1.2% As, note higher Bi content) falls between the two first Dynasty artefacts from Abydos in terms of its LI ratios, and is quite similar to one of the Faras piercers. Overall, E04825a-c fall in the broad range attested for the earlier Elkab, Faras and Fayoum samples discussed above and the other Khasekhemwy samples discussed below, for which a Sinai provenance is suggested. Fragments E04825d-f have more radiogenic LI ratios, with E04825e-f falling within the Eastern Desert lead ore as well as Southern Sinai ore range (particularly Wadi el Regeita/Sheikh Muhsen) and E04825d roughly within the (radiogenic) Southern Sinai ore range (Wadi el Regeita/Sheikh Muhsen/Wadi Samra, cfr. Gizeh mineral fragments).

A chisel (E00161) and three needles (E00167-169) from Petrie's excavation of Khasekhemwy's tomb (from seven (model) tools kept at the museum) were analysed. Their arsenic contents vary between 0.5 and 1% (roughly correlated to antimony content), while 0.1-0.2% iron positively correlates to nickel and trace cobalt, sulphur, tin and zinc contents (all roughly inversely correlated to arsenic content). Lead contents vary between 0.01 and 0.1% (no correlation).
Increased iron contents are reflective of a reducing atmosphere during primary smelting (Craddock and Meeks, 1987), whereby elements such as arsenic, nickel and cobalt may be equally co-reduced to some extent, when present in the ore. If all objects were thus smelted from similar minerals, one would expect arsenic and iron content to correlate too. The inverse, witnessed here, may rather be suggestive of a secondary (alloying) process, where arsenic is added to refined copper. However, the LI ratios of these objects suggest they were smelted from different ores (as do their elemental ratios, cfr. Figure 7). As neither lead or arsenic content correlates to the LI ratios, these chemical trends (which are very limited in any case) are insufficient to strongly suggest an active alloying practice.
The LI ratios are compatible with ores (and slag) from Sinai and fall within the same range as those of copper (ores and artefacts) found at Predynastic Maadi in Lower Egypt (Hauptmann *et al*. 2011 – believed to have been smelted using Sinai ores: Abdel-Motelib *et al*. 2012). Available Eastern Desert ore data does not offer good parallels. E00161 is isotopically less similar to the abovementioned Sinai ores, and falls on the border of LI ratios representative of Timna (outside those of Faynan). A Sinai origin is thus most likely for the copper in these four artefacts, but they were smelted from different ores. More specifically Um Bogma (for E00168) or perhaps Wadi el Regeita (E00167/169) could have served as mining areas, all active during this period. E00161, with the highest arsenic content, lies closest to the Wadi Tar range.

3.3 – Old Kingdom

Bêt Khallaf

The largest group of seventeen samples consists of a range of model tools from three 3rd Dynasty tombs in Bêt Khallaf. Eleven samples (E00915a-e, E00916a-e, E00918) belong to Tomb K.1, three (E00917a-c) to Tomb K.2 and three (E00914a,b,d) to Tomb K.4. In terms of LI ratios, this group contains the most radiogenic samples (most extreme in E00914a).

From Tomb K.1, a group of copper lamellae (E00915a-e), small adzes and chisels (E00916a-e) and a thin (model) adze (E00918) were analysed. In terms of LI ratios and chemical composition, three of the lamellae (E00915b,c,d) are indistinguishable (only sulphur content varies slightly). E00915a has higher arsenic, cobalt, nickel, lead, sulphur, antimony, selenium and zinc and lower iron content, and slightly less radiogenic lead (same Th/Pb, slightly lower U/Pb). E00915e has more elevated arsenic, bismuth, nickel and tin content, and less elevated lead, sulphur and antimony content, with more strongly radiogenic lead (slightly higher Th/Pb and U/Pb). This suggests that the former three lamellae were made from the same batch of copper, while the other two were separately produced. Though different,

their similarities are close enough to suggest all copper was originally smelted from the same Southern Sinai ore deposit: variable arsenic content (0.5-1%) and LI ratios may be due to natural ore variations or differential alloying.
The adzes and chisels vary in composition. Three (E00916a,c,d) have strongly radiogenic lead, consistent with Southern Sinai (particularly Sheikh Muhsen) copper ores. E00916b is more similar to E00915a (not exactly the same: differing arsenic and iron content) and consistent with Southern Sinai ores. E00916e shows close LI similarity to the arsenical copper vessel from Abydos (E00561), though without important zinc content. A Southern Sinai (Wadi Tar) ore provenance may be suggested. This small (E00916) tool set appears not to have been cast from the same metal batch, though E00916b and E00916d are chemically very similar (except for iron content). Altogether, important variations exist in terms of arsenic content, from 0.3-0.6% in E00916a,c to 1.1-1.3% in E00916b,d,e, but these appear not to be related to typology.
The thin adze's (E00918) LI ratios are again consistent with Southern Sinai (on trend-line through available Wadi Tar ore samples).

The adze and chisels from tomb K.2 are highly homogeneous in their composition. Particularly E00917a and E00917b are indistinguishable, while E00917c is only slightly different. The trace element patterns are comparable (similar ratios of cobalt, nickel, antimony and lead: Figure 7), with the exception of arsenic (which correlates to sulphur). This suggests that the former two objects were cast from a single metal batch, only slightly different from that used to cast the third – although metal from the same ore source was probably used in both cases. This may be from Wadi Tar (Southern Sinai), or possibly Wadi Hamama in the Eastern Desert. The abovementioned elements may vary naturally within the deposit, or the difference may be explained by the intentional alloying of the same copper with different amounts of arsenic (mineral). In the latter scenario, part of the final (low absolute) lead content may derive from this arsenic mineral, with the resulting LI ratios representing a mixture of the copper and arsenic minerals – which may or may not have been different. Interestingly, a (mixing) line running from E00917c through one of the (As-rich, Cu-poor) Wadi Tar ores (ET-10/1, Hauptmann *et al*., 1999) runs through E00917a/b. The limited available samples from Sinai, however, do not allow us to push this interpretation much further.

The axe from Tomb K.4 (E00914a) is consistent with Southern Sinai ores (Sheikh Muhsen/Watiyah Pass) and a Roman (?) Eastern Desert smelting slag. It has the most radiogenic LI ratios within the assemblage (more pronounced uranogenic component[2]: $^{206}Pb/^{204}Pb$ and $^{208}Pb/^{204}Pb$). The chisels (E00914b-d) are less radiogenic, and indistinguishable in terms of LI ratios as well as chemical composition, strongly suggesting they were both cast from the same metal batch. They are isotopically relatively similar to the Protodynastic chisel from Elkab (which is corroded and chemically different), and a Southern Sinai ore provenance appears likely. Similar iron-cobalt-nickel ratios for the axe and chisels from Tomb K.4 may imply that the axe's radiogenic LI ratios could fall within the natural variation of the same Southern Sinai deposit, although lead, sulphur, antimony and zinc are much lower in the axe. In contrast, their arsenic contents are nearly identical, suggesting a different origin.

The samples from Bêt Khallaf show a large spread in LI ratios, ranging from the oldest geological age in the assemblage to highly radiogenic ratios along a roughly linear trend. This may indicate that these metals were derived from different deposits within a single ore region. The best-known ore match appears to be the Southern Sinai in general, and more specifically the Wadi Tar ores ("older range") and Watiyah Pass, Wadi El-Regeita and Sheikh Muhsen ores (more radiogenic range). These sites offer evidence for the production of copper during the "Early Bronze Age II" (Abdel-Motelib *et al*. 2012,

[2] Overall, there is a tendency for more radiogenic lead in the Protodynastic samples with respect to the Old Kingdom samples, particularly in terms of thorogenic lead (cfr. section 3.4).

Beit-Arieh 2003), coinciding with the Protodynastic to 3rd Dynasty in Egypt. Arsenic contents suggest Wadi Tar ores may have played a role in the production of several of these objects.
Similarity to central Eastern Desert ores (Abu Greida, Wadi Semna III, Wadi Abu Mureiwat) may equally be noted, though the overlaps are not as good (particularly in the more precisely measured ^{206}Pb ratios) and exploitation evidence for this period is not as clear in the Eastern Desert (with better evidence for the Ptolemaic and Roman periods).
The Arabian Shield deposits cover a large range of LI ratios similar to those of the Bêt Khallaf artefacts. No solid evidence is currently available, however, to suggest the exploitation of copper ores in that region during the studied period. Nonetheless, they remain an important source to re-evaluate when further evidence becomes available. Overall, the Southern Sinai ores present the most likely source used to smelt copper for these 3rd Dynasty objects.

The lead content of the Bêt Khallaf samples is consistently below 1000 μg/g and two clusters around 100 μg/g and 300 μg/g may be noted, which weakly correlate to antimony content (but not to iron or arsenic). Cobalt and nickel correlate quite well within the assemblage (except for E00915a and E00918). Two broad arsenic clusters may be noted around 0.5-0.6% and 1-1.3% (cfr. overall pattern in Old Kingdom samples: section 3.4). No clear correlation exists between arsenic content and typology, with similar objects from the same context being made with different alloys. This may suggest the difference between 0.5% and 1% was perhaps not noticed, controlled, or considered relevant in this context.

Qau el Kebir (5th-6th Dynasty)

The 5th Dynasty hasp's strong corrosion implies that its original chemical composition may appear heavily distorted in the reported measurement. Nonetheless, relatively high lead and low arsenic content (with respect to the presented assemblage) may be noted. Its LI ratios reflect a relatively high geological age (older than the E00917 artefacts, and on a different trend-line), suggesting a Central Eastern Desert (Wadi Hamama) or Southern Sinai origin – in terms of dated mining activity, the latter presents a more likely interpretation.

The 6th Dynasty mirror has the highest arsenic content (ca. 4.8%, though possibly over-estimated due to corrosion) in the assemblage, probably related to its reflective functionality, and slightly elevated antimony content. Its LI ratios are consistent with Central Eastern Desert (Wadi Semna III[3]) copper ores.

el Mahâsna (5th-6th Dynasty)

The two hasps from Tomb M.349 at el Mahâsna are made of arsenical copper, with different iron and lead content. Their LI ratios are consistent with chisels from Sheik Muhsen (Southern Sinai) and only poorly consistent with ores from Northern Sinai (sites B-10 and B-50). The (EB II) Sheik Muhsen chisels have a very similar trace element pattern (including lead content), but lack significant arsenic[4] and antimony (Hauptmann *et al*. 1999). This could suggest the use of metal from a similar ore source, with the addition of arsenic (1.2-1.7%) to the metal used in these hasps. Both hasps fall within the Timna ore range and one within the Faynan range as well. The latter is isotopically indistinguishable from Sheik Muhsen chisel 6678 and two awls from Arad (0590/61, 4757/61) for which Hauptmann *et al*. (1999) suggested the use of Arabah copper – they are indeed very different from Sheikh Muhsen (and most Wadi el-Regeita) ores (Abdel-Motelib *et al*. 2012). Strikingly, these are three of only five arsenic-rich (EBA II) artefacts in Hauptmann *et al*.'s (1999) Arad/Sinai sample. The (slightly later dated) hasps from el Mahâsna may thus derive from Arabah ores, with LI ratios and an increased lead content (ca. 0.5%) in E01961c perhaps favouring a Faynan interpretation. A Southern Sinai origin is equally likely, however, and good discriminating characteristics are currently not available. In fact, the notable arsenic

[3] Evidence of copper smelting is present at Wadi Semna III, most likely datable to the Old Kingdom (Abdel-Motelib *et al*. 2012).
[4] Elevated arsenic content is noted in an ore sample from site B-50, but without elevated antimony content (Abdel-Motelib *et al*. 2012).

content may strengthen such a Sinai interpretation, as the hasps' LI ratios are intermediate to those of one of the older Wadi el-Regeita copper ores (ET-58/1, with high Pb) and the arsenic-rich Wadi Tar ores. E01961c and E01961d were in any case smelted from different deposits, based on their differing LI ratios and trace element pattern (Figure 7).

Abydos (6th Dynasty)

A metallic core sample (with intergranular corrosion) was taken from a 6th Dynasty (reign of Pepi I) Abydos vase. It is characterised by generally low minor and trace element content (note minor arsenic and lead). Its LI ratios are indistinguishable from those of an earlier needle from the tomb of Khasekhemwy, and its trace element pattern is broadly similar (except for lower arsenic content). It may thus represent the use of similar Southern Sinai copper ores, or perhaps even the recycling of copper from earlier periods circulating in Abydos or Egypt more broadly.

3.4 – Broad compositional trends

The foregoing sections have highlighting the most important compositional characteristics for individual artefacts from each specific archaeological context. In this section, a broader overview of this compositional data is provided.

Typical elements used to discuss copper provenance are silver, gold, bismuth, iridium (not measured) and nickel, while arsenic, cobalt, lead, antimony, tin, selenium, tellurium and zinc may be related to both provenance and production technology (Pernicka 2014). Out of these, the most important variation within this assemblage is found in arsenic, bismuth, cobalt, nickel, lead and antimony, as illustrated in Figures 5-7. Additional histograms and scatter plots of abovementioned elements are provided in the OSM.

All artefacts in the presented sample consist of strikingly pure copper: apart from arsenic and iron, few contain over 0.1% of these typical provenance elements and often less than 100 μg/g (see OSM Figure 52).

Cobalt content is extremely low in the Protodynastic artefacts, mostly below 1-10 μg/g, with a few outliers containing up to 90 μg/g. In the Old Kingdom artefacts, cobalt ranges from 1 to 135 μg/g (following a roughly normal distribution) and correlates roughly to iron content. This may be indicative of more reducing smelting conditions in Old Kingdom copper production, though particularly cobalt-poor ores appear to have been used in the production of most Protodynastic artefacts (some have less than 1 μg/g "despite" 0.1-0.2% iron).

Nickel contents are on average higher in Old Kingdom artefacts, though the Protodynastic artefacts contain relatively more nickel than cobalt. The ratio of nickel to cobalt quite distinctly shows a normal distribution for Old Kingdom artefacts (around 19 Ni/Co), while a similar distribution in the Protodynastic artefacts is complemented by a longer tail of higher Ni/Co ratios (Ni/Co > 55 ~ low Co).

Lead and bismuth correlate weakly to each other and are on average comparable for Protodynastic and Old Kingdom samples, with slightly higher lead contents in the Old Kingdom. Lead increases slightly with nickel content, though high lead contents (>500 μg/g) are not associated with high nickel content. No direct correlation between lead content and LI ratios can be discerned (see further OSM Figures).

Arsenic does not correlate well to nickel, cobalt or iron (and poorly to lead) but rather varies independently between 3000 and 15000 μg/g for the entire range of nickel, cobalt and iron contents throughout the Protodynastic and Old Kingdom samples. Within this range, arsenic shows a normal distribution around 0.8%, though upon closer inspection two peaks (around 6000 and 12000 μg/g) stand out, particularly for the Old Kingdom samples (Figure 5).

Antimony behaves similar to arsenic with respect to iron and nickel but correlates better to lead. Antimony further increases along with arsenic as well.
While arsenic content thus appears to be unrelated to nickel, cobalt, iron or lead (provenance and smelting conditions), antimony relates to lead and arsenic, bridging the spheres of interaction for these three elements. If arsenic is thus considered to originate from other (copper) minerals than nickel, cobalt and lead, antimony probably derives from both these arsenic-rich and nickel/cobalt-bearing minerals, while lead appears more strongly related to the nickel/cobalt-bearing minerals[5]. A fraction of lead may derive from the arsenic-rich minerals, however, and LI ratios may thus in part reflect these minerals as well as the dominant copper-bearing minerals[6].

Comparing the ratios of nickel to cobalt and lead to antimony (Figure 7), the majority of samples cluster around 19 Ni/Co and below 2 Pb/Sb (mainly due to low Pb contents). A tail of Protodynastic artefacts with high Ni/Co (low cobalt) stands out, while one Old Kingdom artefact (E00494) with similar Ni/Co has relatively elevated Pb/Sb. The LI ratios of this group generally reflect a higher thorogenic lead contribution. With a few exceptions, the Protodynastic artefacts have slightly more radiogenic (especially thorogenic) lead than the Old Kingdom artefacts and their lead content is slightly lower, though important overlap exists.
Looking at the major cluster (Figure 7, bottom left), samples with indistinguishable LI ratios generally match very well in Ni/Co and Pb/Sb ratios (e.g., E00915b,c,d), confirming their orogenetic relation. Tight chemical clustering of objects from a single tomb context may suggest an orogenetic relation even though LI ratios differ slighty (e.g., E00914b,d or E00917a,b-E00917c) or importantly (e.g., E00916b and E00916d, and even the more radiogenic E00916c). These LI variations may thus exemplify variability within particular Sinai ore deposits, as expected from the ranging values presented by Abdel-Motelib *et al*. (2012).
Taken together, this reveals a trend-line in the LI ratios for much of the Bêt Khallaf assemblage, sitting below the abovementioned trend-line for Protodynastic artefacts, indicative of a different ore range being exploited. Other Protodynastic artefacts (e.g., E04825c,e) may equally belong to this Old Kingdom LI trend-line, and match it in terms of trace element pattern (shared normal distribution in Ni/Co). Two distinct ore deposits/regions can thus be inferred to have been exploited: one particularly in the Protodynastic periods and the other in both Protodynastic and Old Kingdom periods. Important overlap exists, however, and changing smelting processes may have introduced significant variations in these trace element patterns, while strong variations (both chemically and in terms of LI ratios) exist within the ore deposits – these generalisations are thus to be treated with caution.

[5] This particular relation may be exemplified by E00161, E00167 and E00168: their cobalt, nickel, iron and lead contents correlate roughly (∼ copper ore?) while arsenic and antimony do not (∼ arsenic minerals?). On a mixing line through their LI ratios, they are ordered along their arsenic and lead content (higher arsenic ∼ lower lead ∼ more radiogenic LI ratios).
[6] E.g., lead and arsenic correlate for E00914b,d E00915a and E00918, with progressively increasing arsenic and lead content (cfr. OSM). In terms of LI ratios, a rough mixing line could be drawn through these samples, with E00915a and E00918 lying closest to the arsenic-rich Wadi Tar deposits.

4 – Discussion

The majority of Predynastic to Old Kingdom copper artefacts presented here can be interpreted as being smelted from Eastern Desert or Sinai ores. Abdel-Motelib *et al*. (2012) indeed note continuous exploitation of this region, with important mining evidence of first the nomadic population during the 4th millennium BCE in both Eastern Desert and Sinai, followed by Nagada pottery at mining sites and pharaonic expeditions during the Old Kingdom. In the presented sample, prevalent LI consistency with Southern Sinai ores is noted through time, with particular ore deposits such as Watiyah Pass, Wadi El-Regeita, Sheikh Muhsen (metallurgical site), Wadi Semna and Wadi Tar most strongly represented. The available exploitation evidence strengthens these tentative identifications. This does not imply that copper from other regions, such as Anatolia, the Arabah Valley or the Arabian Peninsula, did not reach Protodynastic or Old Kingdom Egypt. There, however, is no strong evidence to suggest this from the presented data.

It is thus possible to establish Sinai as the most important source of copper to the early Egyptians, but the wide spread in its LI ratios (both within and between individual deposits) makes it very difficult to consistently pinpoint specific deposits within Sinai with true confidence. A better characterisation of these ores is essential to gain further insight in the possible overlap between different deposits, and their internal variability. The data presented here, if taken to be evidence of Sinai copper ore use, may in this sense further illustrate the variability that is to be expected.

Provenance and production technology: arsenic in early Egypt?

The artefacts' chemical compositions provide further information on provenance as well as production technology. Overall, remarkably pure copper was used in early Egypt, with the exception of arsenic and iron only. Iron content steadily increases in the Old Kingdom objects (750-7500 μg/g) compared to the Predynastic and 1st Dynasty objects (50-2500 μg/g, with one exception of 1.6%), which might be explained by changing ores sources and/or smelting practices. The latter has particularly been noted by Craddock and Meeks (1987) to explain the rise in iron content[7], starting at the 2nd Dynasty in Egypt (they note an average 0.33% iron as opposed to 0.03% up to the 1st Dynasty).

The group of artefacts with extremely low cobalt content (cfr. section 3.4) all have iron contents below 500 μg/g and may represent the early smelting of a particularly cobalt-poor ore type, accompanied by limited iron reduction. The detection limit for cobalt in Cowell's (1987) AAS data is too high to verify if their pre-2nd Dynasty copper is similarly cobalt-poor.

However, though an average increase is observed in the 2nd Dynasty (Khasekhemwy) and later finds discussed here, 1st Dynasty finds from Faras, Elkab and Gizeh already have iron contents in this "higher range". A shift in smelting technology, perhaps as a result of experimentation with different ore types, may thus already be witnessed earlier than previously assumed. Nonetheless, iron contents remain well below percentage levels, indicative of only mildly reduction smelting conditions and/or the use of very pure ores even during the Old Kingdom.

Direct evidence in support of changing technology should, however, be sought in production waste of primary (furnaces) and secondary (crucibles) metallurgy. No analytical evidence is currently available of early Egyptian smelting technology, except at Middle Kingdom Ayn Soukhna (Abd el-Raziq *et al*. 2011, and ongoing research by the authors: Verly *et al*. in preparation). Remains of secondary metallurgy may equally illuminate different types of raw copper in circulation but have only been analysed in detail

[7] This rise was noted in analytical data obtained by Cowell (1987). More reducing primary smelting conditions are considered to have resulted in increased iron content – though ore quality and beneficiation processes would of course affect this. This may have necessitated a refining step to remove excess iron prior to casting or alloying.

for a single New Kingdom site (Rademakers *et al*. 2017, 2018b). More extensive research on metallurgical technology is thus essential to improve provenance research on Egypt's copper.

Overall, it appears likely that most of the copper presented here was smelted from highly pure copper oxides/carbonates. As Abdel-Motelib *et al*. (2012) note: "*copper ores of the Sinai Peninsula and the northern part of the Eastern Desert, with but one exception of the As-rich ores at Wadi Tar, reveal very low minor- and trace-element concentrations. They consist of pure copper ores [with very low concentrations of all those elements which would end up in copper after smelting (arsenic, nickel, cobalt, bismuth, lead, silver, gold)], and, hence, pure copper was produced from the sites we investigated.*". The only elements to deviate from this pattern are iron and arsenic. Unlike iron, arsenic is influenced by redox-conditions but moreover highly reflective of the smelted ore compositions. As noted in section 3.4, arsenic does not correlate to other elements (except antimony to some extent) in the presented samples, indicating it is unlikely to have the same mineralogical origin. Rather, its distribution would suggest its addition to the charge as a separate constituent of the metallurgical batch – either at a primary (smelting) or secondary (alloying) stage. While the volatility of arsenic may skew its relation to other copper ore trace elements, particularly under oxidising metallurgical conditions (e.g., Mödlinger *et al*. 2017b), the relatively high concentrations witnessed here suggest a different explanation.

Although arsenic is present up to a few 100 μg/g in different Sinai and Eastern Desert copper ores (Abdel-Motelib *et al*. 2012), even extremely reducing smelting conditions could barely account for copper with the percentage-level concentrations of arsenic witnessed here[8]. The copper's iron contents do not favour such a hypothesis[9].
Indeed, arsenic contents in all investigated artefacts are one or more orders of magnitude higher than those of nearly all ores from Sinai and the Eastern Desert. The only known exceptions are a mineral (ET-65/6: 1% As) from settlement site B-50 in Northern Sinai (Abdel-Motelib 2012, Pfeiffer 2013) and arsenic-rich (copper) minerals (ET-10/1-2: Abdel-Motelib *et al*. 2012, Hauptmann *et al*. 1999) from the Wadi Tar deposit in Southern Sinai (close to Wadi Samra and Wadi Kid: Pfeiffer 2013). The former's LI similarity to the Wadi Tar minerals suggests it may in fact derive from there, making Wadi Tar the only known deposit with such high arsenic contents. Could arsenic-rich minerals from Wadi Tar have been used to obtain the ranging arsenic contents in early Egyptian copper? Is the arsenic content in these early Egyptian artefacts intentional?

The intentionality of arsenical copper production in the Early Bronze Age more generally is a highly debated and challenging issue. For example, early SE Iberian metals often contained percentage-levels of arsenic but are commonly considered to be the result of complex and variable ore mineralogy (e.g., Murillo-Barroso *et al*. 2017, and references therein). Indeed, arsenic levels vary strongly between sites, and appear reflective of the local geology. They are the result of directly smelting arsenic-rich ores and their distribution shows a majority of artefacts with low (1-2%) arsenic concentrations, with falling numbers at higher concentrations (Rovira 2011).
In Italy, complex poly-metallic (fahl)ores were smelted to produce arsenic-rich copper, with often important antimony-contents as well (e.g., Dolfini 2014, and references therein). Here too, the alloy is a result of smelting particular ore types, with regional alloy differences reflective of varying ore compositions in northern, central and southern Italy. Early Bronze Age smelting of poly-metallic ore in

[8] Murillo-Barroso *et al*. (2017) note limited loss of arsenic during Late Chalcolithic smelting in SE Iberia (from ores with average arsenic contents of 18%), but high losses during melting and casting (even down to 1%). Under similar conditions, most Sinai and Eastern Desert ores could not have produced the witnessed arsenic contents.

[9] The oldest artefact is an interesting case in point: with only 75 μg/g iron and overall low trace elements content, its 0.8% arsenic content stands out. This implies the smelting of a very pure copper ore, without the reduction of significant iron during smelting. To obtain 0.8% arsenic in this setting, an arsenic-rich ore must have been (co-)smelted, or arsenic added during secondary (crucible) metallurgy.

the Alps equally resulted in copper with high arsenic and antimony and sometimes nickel and bismuth (e.g., Höppner *et al*. 2005).
In Early Bronze Age Iran, in contrast, arsenical copper (often 2-5% arsenic in final artefacts) was produced in a two-step process involving speiss (iron arsenide) as an intermediate product to be alloyed with copper (Rehren *et al*. 2012, Thornton *et al*. 2009).
In both Iberia and Italy, however, there is an apparent lack of selectivity for arsenic content with respect to object typology, particularly when arsenic is below ca. 3%. This random use supports the general interpretation that production of these alloys was not strictly controlled, or even perceived by the early metallurgists (Dolfini 2014, Rovira 2011). Recent research further supports this threshold of ca. 3% arsenic as a perceptive category (physical and mechanical properties) likely to have been relevant to early metalworkers (Kuijpers 2017, Mödlinger *et al*. 2017a). Lechtman (1996 – in the context of Andean metallurgy), in contrast, defined copper-arsenic alloys and arsenical bronze at 0.1-0.5% and >0.5% arsenic respectively, based on appreciably changed mechanical properties upwards of 0.5%, while a threshold of 1% was suggested by Craddock (1976) for Early Bronze Age Aegean deliberate alloying.

Such thresholds should take into account the relevant geological context, which is indeed particular for the case of Egypt. As discussed above, there is no context of poly-metallic ores here, with all but one deposit supporting the "accidental" production of arsenic-rich copper. Cowell (1987), building on earlier work by Lucas (1962), similarly came to the conclusion of intentional selection for arsenical copper in Egypt on the basis of chemical analysis alone (noting only one 2nd Dynasty exception, where antimony and bismuth are particularly high).
This intentional selection within Egypt is further supported by the chemical and especially LI data presented here. Indeed, these indicate that Sinai and Eastern Desert ores were probably used to produce most early Egyptian copper, with certain artefacts having LI ratios compatible with the Wadi Tar deposit[10]. The notable lack of arsenic in all other apparently exploited deposits, and its prevalence in the analysed artefacts with respect to other trace elements, provides a strong indirect argument for the intentional selection for arsenical copper in ancient Egypt.
This can be contrasted with evidence from the nearby Southern Levant. EBA ingots discussed by Hauptmann *et al*. (2015) are consistent with a Faynan (Arabah Valley) provenance and have systematically lower (less than 1000 μg/g) arsenic contents. Some artefacts in the EBII Kfar Monash hoard (Hauptmann *et al*. 2011) have 0.4-3.8% arsenic, accompanied by 0.4-2.8% nickel. Nickel contents in the presented assemblage are all below 0.2%, and arsenic-rich Egyptian copper analysed by Cowell (1987) is equally low in nickel (below 0.1-0.2%). The LI ratios (ca. 2.06-2.07 $^{208}Pb/^{206}Pb$ and 0.827-0.83 $^{207}Pb/^{206}Pb$, plotted in OSM) for these high arsenic/nickel alloys, interpreted by Hauptmann *et al*. (2011) as metal of Anatolian provenance, differ significantly from those in the presented assemblage. This strengthens the hypothesis that this early Egyptian arsenical copper is a specific, local phenomenon.

The contingency of available ores, early smelting evidence, and consumption evidence matching that production implies very early metallurgical developments in Egypt. These may have followed independent rationales, the nature of which should be further explored through detailed excavation and analysis of metallurgical sites.
While it appears that copper-arsenic alloys were desired by the early Egyptians, this data is insufficient to resolve whether particular copper ores were selected to obtain a raw alloy, or if arsenic was actively mixed in a secondary process. The variable arsenic content seen between artefacts, even within a tomb assemblage, perhaps argues that ores from Sinai were used for which the arsenic content varied naturally. Though this may imply a "lack of control" over the final alloy, one must also consider that

[10] Abdel-Motelib *et al*. (2012) and Hauptmann (2007) mention native copper, copper arsenides and the possible remains of a prospecting trench at Wadi Tar but could not ascertain if these served for production of copper arsenic alloys during the (Early) Bronze Age.

artefacts from different periods, produced by different workshops/craftspeople are compared here – they are thus reflective of variability in a long Egyptian metallurgical tradition, and do not explicitly suggest variability – let alone (a lack of) "control" – within a single workshop[11].
As such, "alloy types" within a certain range (around 0.6% to 1.2% and even 3% arsenic[12]) may have been intended through careful ore selection. Alternatively, variable quantities of arsenic (mineral) may have been alloyed with "clean copper", which could be obtained from most Eastern Desert and Sinai ores, in different contexts (either during primary or secondary metallurgical processes). A final possibility is that arsenic was more widespread in Sinai and Eastern Desert ores than hitherto shown, but this appears unlikely based on currently available data.

Though no apparent correlation between object typology and absolute arsenic content can be noted in the presented assemblage, there is a tendency for slightly higher arsenic contents (>1% As) in larger objects and slightly lower (<1% As) in smaller objects. A similar pattern was noted by Cowell (1986) for 2nd Dynasty and Old Kingdom full-size axes compared to model axes (see Odler 2016 on such model tools). Clearly, a larger database of Protodynastic and Old Kingdom copper is required to assess this utilitarian selection, but the appearance of arsenical copper from the onset of Egyptian metallurgy (cfr. Hassan *et al*. 2015, Rehren and Pernicka 2014) is remarkable.
Although arsenic contents are relatively low in comparison to early arsenical copper in other regions, they are high with respect to the specific Egyptian geological setting and thus draw attention. While the perception of such low-arsenic alloys to early Egyptians, in terms of mechanical and physical properties, is difficult to reconstruct, the foregoing discussion has aimed to underline its particularity with respect to early arsenical alloys elsewhere.

Returning to provenance

A better understanding of the possible extent of the Wadi Tar deposits is clearly desirable, as currently only four samples are available for comparison (for two of which ^{204}Pb ratios were not measured). Important to note, is the relatively high lead content of (some of) these Wadi Tar ores: up to 100 times (and more) higher than that of many other Sinai copper ores (Hauptmann *et al*. 1999). The addition of Wadi Tar copper/arsenic to other Sinai copper might thus obscure the latter in terms of its LI ratios. For example, E00917a,b,c have near identical trace element patterns (Figure 7), but slightly different LI ratios, lying on a mixing line through the Wadi Tar ores (cfr. section 3.3): E00917a,b have higher arsenic contents, and lie closer to the Wadi Tar ores on this LI mixing line. This may imply a variable shift in LI ratios due to variable addition of Wadi Tar copper/arsenic to an otherwise homogeneous copper batch. Indeed, several of the metals presented in this paper (and Predynastic metal from Maadi, Tell el-Farkha, Kafr Hassan Dawood) have lead contents exceeding that of many ores characterised by Abdel-Motelib *et al*. (2012). Though orders of magnitude should be compared, the importance of considering lead contents in ores and metals for provenance research is essential to avoid unrealistic attributions and recognize mixing effects, e.g., involving lead-rich (Wadi Tar) and lead-poor ores (cfr. Rademakers *et al*. 2017: mixing of lead-poor oxhide ingots).
This issue is further reflected in the LI dissimilarity between many Sinai ores and smelting slag (Abdel-Motelib *et al*., 2012). While this can be partially explained by exploited ore ranges being broader than revealed by currently available ore data, some ore-slag fractionation during smelting processes[13] (Baron *et al*. 2014) or the use of mixed ore charges during primary smelting may play a role.

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[11] Variability is, however, likely based on observations elsewhere (e.g., Iberia: Murillo Barros *et al*. 2017) as well as in later (New Kingdom) Egypt itself (Rademakers *et al*. 2017).
[12] Cowell (1987) notes up to up to 7% arsenic in a 2nd Dynasty axe.
[13] Many ore and slag samples analysed by Abdel-Motelib *et al*. (2012) have only low (often < 10 mg/g) lead contents. Rather than fractionation, lead contamination (during smelting) may thus be an important factor to take into account.

With these considerations in mind, it is difficult to confidently assess any shift in exploitation between different regions, for example the Eastern Desert and Sinai, or Northern and Southern Sinai, through time. Furthermore, the sample presented here is too small to assess in detail such shifting exploitation over several centuries *a priori*, particularly without further studies of smelting evidence.

For now, we may tentatively note exploitation of a broad range of ore sources during the Predynastic and Protodynastic period, followed by an increasing focus on the Southern Sinai during the Old Kingdom (3rd Dynasty). Exploitation of Eastern Desert ores is more notable amongst Protodynastic samples, and shifting exploitation is evident from broad trends in the chemical and LI data (section 3.4). In the 5th-6th Dynasty, the possible use of Arabah copper is noted for the first time, though this may equally have been Sinai ores. This coincides with a "logical" expansion of ore exploitation starting at deposits nearest to the Nile Valley and moving further away through time, with the true Old Kingdom unification of Egypt under Pharaonic rule heralding more large-scale mining campaigns and regional trade (e.g., Tallet 2018). Kmošek *et al*. (in preparation, pers. comm.) arrive at similar conclusions for the provenance of contemporary Egyptian copper alloys, and future combination of our datasets may further delineate these trends, as well as more context-specific variations.

A notable similarity between the Old Kingdom (Bêt Khallaf: section 3.4) copper "trend" and Arabian Shield (base metal) LI ratios (and al-Middamam metals consistent with these ores: Weeks *et al*. 2009) exists. Though the exploitation of these ores is currently mainly attested (indirectly) during later periods (Liu *et al*. 2015, Weeks 1999, 2003, Weeks *et al*. 2009), they are worth considering as a possible copper source for Egypt. Trade along the Red Sea coast with the so-called "land of Punt" (e.g., Fattovich 2012, Meeks 2003, Tallet 2013) is attested already during the Old Kingdom and may have peaked during the Middle Kingdom period. Most likely, the core of this land was situated on the Arabian Peninsula, with trade posts along the Red Sea coast. The "Mine of Punt" (Bia-Punt), a hinterland possibly encompassing parts of Sudan and Eritrea, is believed to have been the source of metals (particularly gold), ebony and other exotic materials imported to Egypt. Trade of copper may have taken place along this route, in both directions. The Arabian Peninsula must thus be considered as a possible ore source for early Egyptian copper, but currently available evidence more compellingly suggests the use of Sinai copper in the period under study.

The Red Sea and Sinai (donkey trail) trade routes may further have allowed copper from Oman to reach Egypt, as suggested for the New Kingdom period by Rademakers *et al*. (2017). Neither chemical, LI nor exploitation evidence currently available would favour such an interpretation over the presented Sinai and Eastern Desert interpretations for the artefacts presented here.

Occasional similarity of artefacts to Anatolian ores (and Troy, Syros and EBA Greek Mainland artefacts) can further be noted. Evidence for trade of Anatolian metals in the eastern Mediterranean existed (e.g., with Cyprus: Webb *et al*. 2006, Troy: Stos-Gale *et al*. 1984, the Levant: Hauptmann *et al*. 2011) and Egyptian (consumer) involvement should thus be considered. However, low trace element contents (particularly nickel[14]) again favour suggested Sinai and Eastern Desert interpretations.

A comparison to the available data on later (New Kingdom) copper alloys reveals little overlap – these reflect the influx of eastern Mediterranean sources such as Cyprus and the Arabah valley, as well as Sinai, Eastern Desert and Omani copper (see Rademakers *et al*. 2017, Stos-Gale *et al*. 1995). Some of the "Intermediate samples" noted in Pi-Ramesse are isotopically similar to several Protodynastic metals, strengthening the interpretation that such "Intermediate samples" may indeed reflect metal already circulating in Egypt from earlier periods, replenished by raw metal import from the Eastern Desert, Sinai

[14] Lightly increasing nickel content through time may indicate an influx of Anatolian metal and its contamination of circulating recycled/mixed metal in Egypt. Proposing such a framework of extensive mixing is premature on the basis of this data, however.

and Oman (Rademakers *et al*. 2017). The range of Sinai and Eastern Desert ores attested in these earlier periods is much wider, however, as has been witnessed in Predynastic Tell el-Farkha and Maadi.

Copper for the afterlife

The artefacts presented in this paper mainly derive from funerary context. Objects deposited with the dead may have been specifically produced for use in the afterlife or have served a prior function outside of this funerary context. In either case – as for all archaeologically recovered copper – they may have been directly produced from raw copper, or by recycling older copper. Ancient recycling of copper is very difficult to detect by modern analytical techniques, as is the mixing of copper from different sources. While the former does not influence LI ratios, the latter can mix these up beyond recognition. Though it is often assumed that mixing of copper occurred relatively infrequently up until Late Bronze Age times (e.g., Pernicka 2014), this practice cannot formally be excluded (Bray *et al*. 2015) and has been shown to be of importance to the New Kingdom copper economy (Rademakers *et al*. 2017, Rehren and Pusch 2012). Furthermore, the mixing of ores from various deposits in a single smelting furnace may have occurred (cfr. infra: Wadi Tar). Smelting evidence for the period under study invariably occurs close to the Eastern Desert and Sinai mines (Abdel-Motelib *et al*. 2012) and no smelting furnaces have hitherto been recovered in the Nile Valley. Middle Kingdom evidence, in contrast, indicates that ores from mining campaigns in the Sinai were collected and smelted in a centralised location at Ayn Soukhna (Abd el-Raziq *et al*. 2011), likely involving mixing at that stage: several different ore types are encountered within individual smelting workshops (Verly *et al*. in preparation). Furthermore, the recycling of used metal tools from these mining and smelting campaigns appears to have been an important part of the (state-controlled) Middle Kingdom metal economy (ongoing research by the authors).

This possible mixing of copper (ores) most likely does not affect provenance interpretations offered here. Mixing would result in the shifting of LI ratios in the resulting copper to a position intermediate between those of the mixed ores/copper. Such mixed LI ratios would mostly still fall in the range of Sinai and Eastern Desert ores discussed here and may thus go undetected. However, the presented data show strong dispersion, not adhering to linear mixing patterns (except for arsenic mixing?). Such disparate LI ratios are indicative of a wide variety of copper sources entering circulation in early Egypt, in contrast to a convergence towards homogenised LI ratios which could be expected if mixing were "total".

Returning to the funerary contexts, different situations may be noted. Copper and lead minerals[15] are encountered in the 1st Dynasty tombs, and their geochemical similarity suggests the exploitation of these different minerals from a similar geological setting in Protodynastic Egypt. Interestingly, minerals in Tarkhan and Abydos are nearly identical in their LI ratios, reflective of Eastern Desert mining. These may not have been directly related to metallurgy, however, as contemporary artefacts have different LI ratios, more likely reflective of Sinai ores. Eastern Desert lead ores are indeed exploited for cosmetics (*kohl*), while lead metal was apparently imported (Shortland 2006) – though this remains to be verified through more exhaustive analysis of lead artefacts. Later finds from Khasekhemwy (E004825f), however, may imply exploitation of these same deposits for metallurgical purposes during the 2nd Dynasty.

In Khasekhemwy's Royal Tomb at Abydos, copper lamellae (E04825a-f) were interred for which the origin and composition varies, suggesting they were not the result of a single "state-controlled production order" with associated mining expedition. Rather, they are more likely reflective of the

[15] The provenance attribution of lead (ores) is often problematic due to limited isotopic constraints on Egyptian lead ore deposits (Shortland 2006, Shortland *et al*. 2000).

variety of metal in circulation at that time. They appear not to have been selected for any obvious aesthetic attribute (highly irregular shapes), but perhaps rather for their intrinsic material value. Their particular find context may even suggest that they are scrap from metalworking, perhaps of other objects within the funerary assemblage – though these were not analytically detected in this study. Such scrap, which could have been remolten elsewhere, may still have been valuable enough to inter as a cache in this royal tomb. Diversity in terms of provenance is the rule for Khasekhemwy's tomb assemblage, though Southern Sinai was probably the most important mining region. It may thus seem that this 2nd Dynasty king relied on the availability of metal from a variety of sources, rather than preparing a funerary assemblage using freshly smelted copper from a single expedition.

In 3rd Dynasty Bêt Khallaf, not all objects in the tombs derive from the same copper source. However, copper from different tombs often has similar provenance. Furthermore, in each of the tombs, several objects appear to have been cast from a single copper batch – at least this is as close as one can expect to come to identifying a nearly 5000-year old crucible batch by current analytical means. This suggests that some of these objects were produced as an assemblage: they may have been "made to order" for direct burial purposes, or perhaps they were previously owned as a group of objects (not separated after production) by the tomb owner or their family. In the later el Mahâsna tomb, the hasps are again typologically similar but their copper from different provenance. Though not royal tombs, different patterns may thus be noted in these Old Kingdom finds. Obviously, future analyses must illuminate whether these are recurrent diachronic patterns, related to social status, or not.

Overall, geochemical (LI) similarities can be noted between specific artefacts from the 1st and 2nd, 2nd and 3rd, and 2nd and 6st Dynasties. This, together with the overall results discussed in this paper, suggests an important continuity in copper provisioning throughout early Egyptian times. Part of this was organised through continued expeditions to the Eastern Desert and Sinai Peninsula, but part of it may already have been fulfilled through the recycling of circulating metals. The importance and evolution of these different production mechanisms must be investigated for a much wider variety of contexts, ideally including metallurgical workshops, to gain deeper insights into the organisation of this particular segment of the ancient Egyptian economy.

6 – Conclusion

This paper has presented the first lead isotope data for Egyptian copper used in the Predynastic, Protodynastic and Old Kingdom periods. Through detailed comparisons of its isotopic and chemical composition to contemporarily mined ore deposits and circulating metals in the wider region, this dataset shows the predominant reliance on "local" ore sources from the Eastern Desert and Sinai Peninsula during this formative period. While the consumption of imported copper from the wider Mediterranean region during this period can certainly not be excluded, this is not apparent from the presented sample. The results indirectly imply significant developments in smelting technology, which may have been adapted to different ore types. Indeed, smelting processes may have diverged earlier than previously assumed, as a response to newly exploited ore deposits. Importantly, arsenic stands out as an integral alloy component in the majority of metal artefacts, incompatible with their Eastern Desert and Sinai ore sources. Though arsenic contents are lower than those observed in other Early Bronze Age settings, the particular geological setting argues for their specific selection. These new chemical and LI data confirm that Southern Sinai ores (particularly those from Wadi Tar) may have provided the opportunity to intentionally produce such alloys – either directly through primary (co-)smelting or by secondary alloying processes. These findings are based on a very particular segment of the ancient Egyptian metal economy (funerary consumption) and thus only reveal a tip of the iceberg. We hope they may nonetheless serve as a background for future studies of early Egyptian copper provisioning, which clearly relied on a variety of mining and production zones. The organisation of these early supply networks and the metallurgical techniques underlying copper production are the subject of ongoing research, which is only slowly revealing their deep and intricate history in ancient Egypt.

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