In the Shadow of Timna? The Mining Region of Wadi Amram New Analytical and Archaeological Aspects

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Keywords

Archaeometallurgy, Copper Ore, Trace Elements, Lead Isotope Analysis, Provenance Studies, Wadi Arabah

Abstract

The copper mines of Wadi Amram are located only 10 km north of the prehistoric settlements Tall Hujayrat al-Ghuzlan and Tall al-Magass where there is some evidence of copper metallurgy in the Late Chalcolithic / Early Bronze Age I. These two settlements lay near the Gulf of Aqaba and may have been important nodes for the distribution of copper during this period. Earlier lead isotopic studies demonstrated that copper ores from the mines of Timna and Faynan are possible sources for the early copper production, but up until now the much closer copper deposit of Wadi Amram has been ignored. New lead isotope data has shown that the Wadi Amram copper ore can be distinguished from the ore from Timna and closely correlates to the Faynan copper ores. Some of the copper-related objects from Tall Hujayrat al-Ghuzlan and Tall al-Magass have lead isotope ratios more comparable to ore from Wadi Amram than from Timna and warrants a reassessment of the organization mode of the prehistoric mining industry in the southern Levant.

Introduction

Since the Pre-Pottery Neolithic B (8th-7th millennium BC) copper and copper minerals were traded in the Near East (Klimscha, 2013a, p.88). Some of the most important copper ore deposits in this region lay in the Wadi Arabah, which runs from the Dead Sea to the Gulf of Aqaba (Avner, 2002, pp.39-64). The largest ancient copper districts to be found in this region are Faynan (Hauptmann, 2000; 2007) located in Jordan, Timna (Conrad and Rothenberg, 1980) and Wadi Amram in Israel (Figure 1). Wadi Amram is one of the southern branches of the Wadi Arabah and is located c. 10 kilometers north of the Gulf of Aqaba and the Red Sea, at the



Figure 1: Map of the Wadi Arabah in the southern Levant with the most important copper deposits, Chalcolithic / EBI settlements and craft production centers. Image courtesy of U. Avner (Avner, et al., in press).



Figure 2: Topographic map of the mining region of Wadi Amram and the archaeometallurgical features. The area is divided into subareas, in which mining traces of various periods can be found. Image courtesy of U. Avner (Avner, et al., in press). western rim of the Wadi. Timna is about 20 km and Faynan 120 km to the north. In the southwest Wadi Amram directly adjoins the Sinai Peninsula.

The formation of the Wadi Arabah is tied to the Great East African Rift. This rift runs from the east of Africa to southwestern Asia and marks the African and Arabian plate boundary. In the southern Levant the Rift formed the 5-30 km wide Wadi Arabah, which divides region and its ore deposits: Timna and Wadi Amram are located west of the rift and Faynan is east of the rift. The shift of the continental plates is estimated to be 107 km; before the division of the region by the rift the ore deposits of Timna and Faynan were one geological unit. The geology and ore deposits were described in detail by Segev (1986) and Segev, Beyth and Bar-Matthews, (1992).

The copper ore deposit of Wadi Amram is not only overshadowed in size by the nearby Timna deposit, the focus of archaeological research was and is mostly concentrated on Timna. While Timna has been extensively studied for decades leading to numerous publications (e.g., Rothenberg 1972; 1973; 1988; 1990; 1998), Wadi Amram has widely been neglected. Actually, systematic interdisciplinary investigation in the area only began in the last few years.

Archaeological remains in Wadi Amram were first documented in 1949 by Israeli military soldiers. Shortly thereafter, the first archaeological surveys were led by Glueck (1953; 1960). In these reports two mines with shafts and galleries as well as a large mining camp were described. In conjunction with the research of Beno Rothenberg and the Deutsches Bergbau-Museum (henceforth abbreviated DBM) at Timna, further surveys were undertaken in Wadi Amram in the 1960s (Rothenberg, 1963). Mines on the steep slopes of Har Amir and a large mining camp in the south of the area were documented. Around this camp, slag heaps and flat depressions (referred to as "plates") were identified. There are further descriptions of Rothenberg of shafts and mines in the northwest, at the upper end of the Wadi. There was a break in research, which was renewed in 1989 with further archaeological surveys of the mines by Willies (1991). Some of the above-mentioned contexts were documented and studied in detail with particular regard to the chronology, spatiality and geology but also in regard to the mining technology.

Comprehensive interdisciplinary study of the Wadi Amram mining region began in 2011. The archaeological surveys of Uzi Avner carried out between 2002 and 2013 form the foundation for this research (Avner 2002; 2006; Avner, et al., in press). Not only did these surveys reevaluate known archaeological sites, also many mines and mining-related features were discovered (Figure 2, Figure 3 and Figure 4). The interdisciplinary research included, detailed mapping of the inside of 46 mines, geological and geomorphological investigations, subsurface characterization using ground-penetrating radar, LiDAR scans of mine tailings and slag heaps, chemical/mineralogical characterization of copper ore and slag as well as ¹⁴C dating. The results of these investigations are currently being prepared for publication.

This study is a part of this interdisciplinary research. The elemental compositions of copper ore from Wadi Amram and the lead isotope ratios were analyzed, forming the basis for archaeometallurgical provenance studies and enabling the exploration of questions concerning the processing of ore and prehistoric trade and exchange. The basic concepts of these methods and their capabilities and limitations have been the theme of numerous articles and have been thoroughly explained by Gale and Stos-Gale (2000), Pernicka (2014), Pollard (2009) and Wilson and Pollard (2005), to name a few.

Wadi Amram is located only 10 km to the north of the settlement of Tall Hujayrat al-Ghuzlan and Tall al-Magass. Both of these settlements are accessible to the Red Sea via the Gulf of Agaba and already in the transition between the Late Chalcolithic and the Early Bronze Age I (EB I) there is very interesting evidence for copper production (Khalil and Schmidt, 2009). Lead isotope analyses have shown that the copper used for the production of artifacts from Tall al-Magass could most probably have originated in Timna, but perhaps also in Faynan (Hauptmann, Khalil and Schmitt-Strecker, 2009). The fact that Timna and Faynan could not be excluded as possible copper suppliers has considerable repercussions on the dating of the mining, which is often difficult to reconstruct. Based on this information, an argument can be made that the extraction of ore from Timna and Faynan for copper production may have begun in the Chalcolithic.

However, not all the lead isotope ratios from objects from the settlements of Tall Hujayrat al-Ghuzlan and Tall al-Magass could be assigned to already characterized copper ore deposits. The reason for this could be the lack of lead isotope data from mining areas of Wadi Amram (Hauptmann, Khalil and Schmitt-Strecker, 2009, p.303). Up until now, the mining of ore in Wadi Amram could not be clearly dated to the Chalcolithic, however the close geographic proximity to the settlements make this region a good candidate for a potential ore supplier (Avner, et al., in press). The current study explores the questions of whether the copper ore from Wadi Amram could have been used for the production of the copper artifacts from the Chalcolithic settlements or if it can be clearly excluded.



Figure 3: The Amram pillars are one of the landmarks in the southern Wadi Arabah. They were formed by erosion processes in the Red Sandstones of the Cambrian Shehoret formation. Nearby, a number of copper mines are located in the White Sandstone of the exposed Amir- and Avrona formation. They were surveyed by Avner, et al. (in press) and date mostly to the Roman period; however, some of the mines could be older. Photo: I. Löffler, DBM.



Figure 4: Two of the copper mines found in the White Sandstone of Wadi Amram dated to the Roman period. The adits of the galleries are typically very large: the height and the width are more than one meter. Photo: I. Löffler, DBM.

The approach of the study was undertaken in three steps (after Klein, 2007, pp.144-145):

- The ore deposit of Wadi Amram was characterized by elemental and lead isotope analysis in order to create reference data that can be compared with other deposits and artifacts.
- As a result, the Wadi Amram deposit could be compared to the other geographically relevant deposits, chiefly Timna and Faynan. The main question being whether these deposits can be distinguished from one another by lead isotope analysis.
- 3. In the third step, the lead isotope ratios of these deposits were compared with the copper artifacts from Tall Hujayrat al-Ghuzlan and Tall al-Magass. The primary goal was to determine if the copper used could have originated from the Wadi Amram deposit.

Sampling and Methodology

In the framework of the current study, ten ore samples from Wadi Amram were analyzed to obtain elemental compositions and lead isotope ratios. The ten hazelnut-sized ore samples were selected from a larger collection of copper-bearing ore samples taken during the large-scale systematic surveys of Wadi Amram between 2011 and 2013 (example Figure 5 and Figure 6). All samples came from the White Sandstone geologic level in the Lower Cretaceous (Avner, et al., in press). The samples had no archaeological inventory number. The numbers provided in the tables are those given by the laboratory of the DBM. Due to the small number of samples analyzed in this study, the results should not be taken as representative of the entire deposit, but they should provide a reasonable overview of the mineralization of Wadi Amram. Hence, the goal of the present study is to provide a first look at the characteristics of the ore deposit. In the future, the analysis of a larger number of samples is necessary.

The ten samples were processed in the laboratory of the DBM. The samples were crushed with steel tools and ground to powder using agate ball mills (ca. 30-40 minutes). After drying, the pulverized samples were analyzed by X-ray diffraction (XRD) to determine the mineralogical composition (Table 1). The copper minerals are for the most part green malachite and partially (par-) atacamite as well as blue azurite in tiny amounts. Additionally, brochantite and antlerite could be identified, and in two cases the samples were associated with brown-colored goethite and hematite. Cuprite may be present in several of the samples, however, positive identification of this mineral with XRD was hindered



Figure 5: Nodule of a copper ore from Wadi Amram. The nodule is an impregnation of secondary copper minerals in sandstone. The brown colored core consists of a mixture of iron-bearing minerals, and cuprite may be present. It is covered by an incrustation of green copper minerals mainly consisting of malachite and atacamite.



Figure 6: Detail showing the intergrowth of green secondary copper and brown iron hydroxide minerals with the quartz grains of the sandstone and whitish post-depositional calcite. The association of such minerals found in this ore would make it self-fluxing during smelting. Digital microscope image of 50x magnification.

by the presence of quartz peaks near the major cuprite peaks.

Following these preliminary analyses, the samples were prepared for chemical and lead isotope analysis. The elemental analysis was carried out by high-resolution inductively coupled plasma mass spectrometry (HR-ICP-MS) at the DBM and lead isotope analysis was performed by multicollector ICP-MS (MC-ICP-MS) in the Institut für Geowissenschaften at the Goethe-Universität Frankfurt am Main. Regarding the procedures and the instrumentation refer to Klein, et al. (2009, pp.62-64). Table 1: List of minerals determined in the copper ore samples from Wadi Amram as referred in the text.

Mineral	Formula
Antlerite	Cu ₃ (OH) ₄ SO ₄
Azurite	Cu ₃ (OH) ₂ (CO ₃) ₂
Bisbeeite	(Cu,Mg)SiO ₃ ·nH ₂ O
Brochantite	Cu ₄ (OH) ₆ SO ₄
Chrysocolla	Cu_7 -xAlx $(H_2$ -xSi $_2O_5)(OH)_4 \cdot nH_2O$
Chalcocite	Cu ₂ S
Covellite	CuS
Cuprite	Cu ₂ O
Djurleite	Cu ₃₁ S ₁₆
Dolomite	CaMg(CO ₃) ₂
Goethite	FeOOH
Hematite	Fe ₂ O ₃
Limonite	FeOOH
Malachite	Cu ₂ (OH) ₂ CO ₃
(Par-)atacamite	Cu ₂ (OH) ₃ Cl
Quartz	SiO ₂

In addition to the analyses carried out during this study, lead isotope analyses of seventeen samples from Wadi Amram were performed by Asael (2010; Asael, et al., 2012). Since these data stem from study aimed at answering specific geological questions, the sampling strategy was directed towards specific parts of the deposit and cannot be taken as representative of the whole deposit, and for this reason they will be listed separately. The additional analyses taken from the geological literature increase the sample size, but may skew the statistical evaluation of the results.

Comparative lead isotope data for provenance studies can be found for Timna (Segal, et al., 2015), Faynan (Hauptmann, 2000; 2007) and for the objects from the chalcolithic settlements near Aqaba, Tall Hujayrat al-Ghuzlan (Hauptmann & Khalil, in prep.) and Tall al-Magass (Hauptmann, Khalil and Schmitt-Strecker, 2000).

Evaluation of the Data

Several different statistical methods were used in the evaluation of multi-variate datasets produced by chemical and lead isotope analysis. Due to the complexity of obtaining the data and the commonly encountered problem in archaeology, the lack of a truly representative sample size, the use of statistics has had a limited impact. The observations will be based for the most part on simple descriptions of the samples and graphical/visual representations (Stos-Gale and Gale, 2009, pp.202-205; Baxter and Buck, 2000).

The characterization of the chemical composition of copper ore from Wadi Amram was achieved through the calculation of tendencies in the compositions of each element using a box-and-whisker plot. The simplification of the dataset makes it easier to compare with other deposits. The data can be collectively characterized by their quartiles $(Q_1 \text{ and } Q_3)$ and median (Q_2) . To take account of the range and distribution of the data, as well as strong outliers, an additional distinction was made for the lower and upper decile, D_1 and D_0 . The data is graphically represented in a box-and-whisker plot. The data between the lower and the upper quartile are represented by the box, whereas the median is represented by the line in the middle. The lower and upper decile are represented by the whiskers. The crosses outside the box-and-whisker plot field mark the minimum and maximum values.

The graphical representations of lead isotope ratios used in archaeological provenance studies normally consist of two bivariate scatter diagrams. The values are displayed on the following axes : ²⁰⁷Pb / ²⁰⁶Pb vs. ²⁰⁸Pb / ²⁰⁶Pb and ²⁰⁷Pb / ²⁰⁶Pb vs. ²⁰⁴Pb / ²⁰⁶Pb. A further statistical tool is the determination of the Euclidean distance (d) which was used to confirm the conformity of lead isotope ratios of a specific metal artifact to an ore sample, taking into account the 0.1 % analytical error values. Both these scatter diagrams and the box-and-whisker plots were created using the open source software Sci-DAVis version 0.2.4 and are reproducible.

Results

Characterizing the Wadi Amram Ore Deposit

In the following section, the ores from Wadi Amram will be characterized by their chemical composition and their lead isotope ratios. The characteristics of the Wadi Amram copper ore will be described, and at this point, these descriptions will be independent from comparisons to other deposits and archaeological copper objects. The resulting data would not only be a foundation for the provenancing of objects in regard to the Chalcolithic and EB settlements of Tall al-Magass and Tall Hujayrat al-Ghuzlan, it will also serve as be a basis for the investigation of other archaeometallurgically relevant periods and sites, though in other periods, other ore districts Table 2: Chemical compositions of copper ore samples from Wadi Amram. The values are given in weight percent (when noted), otherwise they are in parts per million (ppm). The oxides were determined stoichiometrically. Hydroxide, carbonate and chloride were not measured, and this factors into the analytical totals of under 100 wt. %.

Sample No.	SiO ₂ in %	$\underset{\text{in \%}}{\text{Al}_2\text{O}_3}$	Fe ₂ O ₃ in %	TiO ₂ in %	MnO in %	CaO in %	MgO in %	BaO in %	K ₂ O in %	Na ₂ O in %	Cu in %				
4813-13	72.67	0.76	0.09	0.03	0.003	0.27	0.09	< 0.004	< 0.0005	0.01	8.65				
4814-13	15.76	1.28	0.18	0.14	0.003	2.32	0.62	< 0.004	< 0.0005	0.21	47.30				
4815-13	42.71	0.51	4.48	0.05	0.001	1.59	0.32	< 0.004	< 0.0005	0.03	18.32				
4816-13	72.12	1.32	0.08	0.07	0.001	0.05	0.06	< 0.004	< 0.0005	0.01	6.88				
4817-13	61.49	0.67	0.06	0.06	0.001	0.03	0.05	< 0.004	< 0.0005	0.01	9.79				
4818-13	69.85	1.22	0.10	0.06	0.001	0.24	0.04	< 0.004	< 0.0005	0.03	8.85				
4819-13	66.60	1.25	0.60	0.05	0.005	0.19	0.11	0.08	< 0.0005	0.01	13.83				
4820-13	50.59	2.41	5,95	2.94	0.006	0.20	0.11	< 0.004	< 0.0005	0.02	14.04				
4821-13	30.95	0.14	0.11	0.02	0.002	1.45	0.30	< 0.004	< 0.0005	0.05	41.05				
4822-13	62.01	0.74	0.73	0.32	0.005	0.28	0.25	< 0.004	< 0.0005	0.02	16.4				
Sample No.	S	Zn	0												D
		ZII	Sn	РЬ	As	Sb	Bi	Со	Ni	Ag	Se	Те	Au	U	Р
4813-13	250	3000	8 8	Р Б 70	As < 5	Sb 75	Bi 2	Co 40	Ni 60	Ag 3	Se < 4	Te 40	Au < 2	U 50	Р 180
4813-13 4814-13	250 510	3000 24000	8 9	РБ 70 200	As < 5 < 5	Sb 75 9	Bi 2 < 0.2	Co 40 270	Ni 60 360	Ag 3 7	Se < 4 20	Te 40 10	Au < 2 < 2	U 50 9	P 180 200
4813-13 4814-13 4815-13	250 510 340	211 3000 24000 12000	8 9 3	РЬ 70 200 210	As < 5 < 5 35	Sb 75 9 6	Bi 2 < 0.2 < 0.2	Co 40 270 170	Ni 60 360 270	Ag 3 7 4	Se < 4 20 65	Te 40 10 3	Au < 2 < 2 < 2	U 50 9 15	P 180 200 210
4813-13 4814-13 4815-13 4816-13	250 510 340 130	211 3000 24000 12000 120	8 9 3 < 2	РЬ 70 200 210 10	As < 5 < 5 35 < 5	Sb 75 9 6 < 2	Bi 2 < 0.2 < 0.2 < 0.2	Co 40 270 170 7	Ni 60 360 270 < 15	Ag 3 7 4 < 2	Se < 4 20 65 < 4	Te 40 10 3 <1	Au < 2 < 2 < 2 < 2 < 2	U 50 9 15 2	P 180 200 210 35
4813-13 4814-13 4815-13 4816-13 4817-13	250 510 340 130 130	211 3000 24000 12000 120 6600	Sn 8 9 3 < 2 < 2 < 2	РЬ 70 200 210 10 9	As < 5 < 5 35 < 5 < 5 < 5	75 9 6 < 2 < 2	Bi 2 < 0.2 < 0.2 < 0.2 < 0.2 < 0.2	Co 40 270 170 7 110	Ni 60 360 270 < 15 180	Ag 3 7 4 < 2 < 2 < 2	Se < 4 20 65 < 4 < 4 < 4	Te 40 10 3 <1 <1	Au < 2 < 2 < 2 < 2 < 2 < 2 < 2	U 50 9 15 2 4	P 180 200 210 35 110
4813-13 4814-13 4815-13 4816-13 4817-13 4818-13	250 510 340 130 130 610	24000 12000 120 6600 70	Sn 8 9 3 < 2 < 2 < 2 3	РЬ 70 200 210 10 9 9	As < 5 < 5 35 < 5 < 5 < 5 < 5 < 5	Sb 75 9 6 < 2	Bi 2 < 0.2 < 0.2 < 0.2 < 0.2 < 0.2 < 0.2	Co 40 270 170 7 110 1	Ni 60 360 270 < 15 180 < 15	Ag 3 7 4 < 2 < 2 < 2 < 2	Se < 4 20 65 < 4 < 4 < 4 25	Te 40 10 3 <1 <1 <1 <1	Au < 2 < 2 < 2 < 2 < 2 < 2 < 2 < 2	U 50 9 15 2 4 2	P 180 200 210 35 110 130
4813-13 4814-13 4815-13 4816-13 4817-13 4818-13 4819-13	250 510 340 130 130 610 5400	2000 24000 12000 120 6600 70 940	Sn 8 9 3 < 2	Pb 70 200 210 10 9 9 280	As < 5 < 5 35 < 5 < 5 < 5 < 5 < 5 < 5	Sb 75 9 6 < 2	Bi 2 < 0.2 < 0.2 < 0.2 < 0.2 < 0.2 < 0.2 < 0.2 < 7	Co 40 270 170 7 110 1 50	Ni 60 360 270 < 15 180 < 15 15	Ag 3 7 4 < 2 < 2 < 2 < 2 20	Se < 4 20 65 < 4 < 4 25 < 4	Te 40 10 3 <1 <1 <1 <1 <1	Au < 2 < 2 < 2 < 2 < 2 < 2 < 2 < 2 < 2 < 2	U 50 9 15 2 4 2 5	P 180 200 210 35 110 130 90
4813-13 4814-13 4815-13 4816-13 4817-13 4818-13 4819-13 4820-13	250 510 340 130 130 610 5400 2600	23000 24000 12000 120 66000 70 940 1500	Sn 8 9 3 < 2	Pb 70 200 210 10 9 280 140	As < 5 < 5 < 5 < 5 < 5 < 5 < 5 < 5 < 5 < 5	Sb 75 9 6 < 2	Bi 2 < 0.2 < 0.2 < 0.2 < 0.2 < 0.2 < 0.2 7 1	Co 40 270 170 7 110 1 50 50	Ni 60 360 270 < 15 180 < 15 15 40	Ag 3 7 4 < 2 < 2 < 2 20 25	Se < 4 20 65 < 4 < 4 25 < 4 < 4 < 4	Te 40 10 3 <1 <1 <1 <1 <1 <1 <1 <1	Au < 2 < 2 < 2 < 2 < 2 < 2 < 2 < 2 < 2 < 2	U 50 9 15 2 4 2 5 5 15	P 180 200 210 35 110 130 90 320
4813-13 4814-13 4815-13 4816-13 4817-13 4818-13 4819-13 4820-13 4821-13	250 510 340 130 130 610 5400 2600 32000	23000 24000 12000 120 6600 70 70 940 1500 990	Sn 8 9 3 <2	Pb 70 200 210 10 9 280 140 170	As < 5 < 5 < 5 < 5 < 5 < 5 < 5 < 5 < 5 < 5	Sb 75 9 6 < 2	Bi 2 < 0.2 < 0.2 < 0.2 < 0.2 < 0.2 7 1 1	Co 40 270 170 7 110 1 1 50 50 45	Ni 60 360 270 < 15 180 < 15 15 40 15	Ag 3 7 4 < 2 < 2 < 2 < 2 20 25 4	Se < 4 20 65 < 4 < 4 25 < 4 < 4 < 4 < 4	Te 40 10 3 <1 <1 <1 <1 <1 <1 <1 <1 <1 <1	Au < 2 < 2 < 2 < 2 < 2 < 2 < 2 < 2 < 2 < 2	U 50 9 15 2 4 2 5 15 15 5	P 180 200 210 35 110 130 90 320 30

which may overlap with Wadi Amram must also be taken into consideration. Especially important are the new lead isotope data.

Chemical compositions

Since the mineralizations of copper ores from Wadi Amram are found in the geological formation of the White Sandstone, the main gangue mineral of all samples is quartz. With a median (Q_2) = 61.8 wt. %, and the lower and upper quartiles, Q_1 =42.7 and Q_3 =69.8 respectively, the samples usually consist of more than 50 wt. % silica. However, the silica contents of samples 4813-13 (15.8 wt. %), 4814-13 (42.7 wt. %) and 4821-13 (31.0 wt. %) are lower, but this is reflected in the comparatively higher copper contents of these samples, (47.3 wt. %, 18.3 wt.

%, and 41.1 wt. %, respectively). The copper concentration in the samples has a median of Q_2 =13.9 wt. %, and the lower and upper quartiles are at Q_1 =8.9 wt. % and Q_3 =18.3 wt. %. The samples are, therefore, not particularly rich in copper. The amounts of other compounds like iron oxide (Q_2 =0.15 wt. %) and lime (Q_2 =0.26) are notably small. Other elements are only found in levels under 5 wt. % and are often under 0.1 wt. % (Table 2).

The partitioning of trace elements during smelting of copper, particularly those that could be interesting for provenance studies, have been examined in the past decades (Merkel, 1990; Pernicka, 1990). For the present study, the concentrations of Ag, As, Au, Bi, Co, Ni, Pb, Sb, Sn, and Zn may be meaningful. The concentrations of As, Sb, and Bi are in the lower level of parts per million. Gold (Au) was continuously below the detection limit



Figure 7: Box-and-whisker plots of major and trace elemental compositions mentioned in the text. The graphic representation uses a logarithmic scale, which is particularly useful in comparing trace elements for provenance studies. On the occasion that there is a value under the detection limit, the value used is ten times below the detection limit.

of 2 ppm of the HR-ICP-MS. The use of this ore for the smelting of arsenic or antimony-bearing copper, which is commonly found in ancient metallurgy and particularly during the Chalcolithic and Early Bronze Age periods, can be excluded.

The portions of Ag (Q₂=4 ppm) and Sn (Q₂=3 ppm) are also very little; however, Pb, Co, Ni and Zn can be found in significant amounts. Especially, the zinc contents can be quite high and have a wide distribution. The zinc distribution follows: D₁=95 ppm, Q₁=940 ppm, Q₂=2250 ppm, Q₃=6600 ppm and D₉=18000 ppm. The values for Pb (Q₁=10 ppm, Q₂=105, Q₃=200ppm), Co (Q₁=40 ppm, Q₂=50 ppm, Q₃=110ppm) and Ni (Q₁=15 ppm, Q₂=50 ppm, Q₃=180 ppm) are notably lesser and have a more constrained distribution (Figure 7).

Lead Isotope Ratios

As mentioned, the lead isotope characterization of the Wadi Amram copper deposit is based on the analyses of this study and a previous study (Asael, 2010; Asael, et al., 2012). The samples from the present study were taken from different spots distributed throughout the entire mining region (Table 3). In the study of Asael, the samples analyzed come from specific parts of the deposit, and thus probably do not represent the whole range of lead isotope ratios. For this reason, the expansion of the

lead isotope dataset for Wadi Amram is significant (Figure 8). Together, and particularly through the help of the new data, the characterization of the ore deposit is much better represented; nevertheless, the dataset is skewed because of the significantly larger amount of samples (n=17) of Asaels investigation which tend to plot in a tight field. For this reason, the following descriptions of each sample series will be given separately.

Table 3: The lead isotope analyses of copper ore from Wadi Amram measured by multicollector ICP-MS. The standard deviations (2 σ) are 0.001 for ²⁰⁸Pb / ²⁰⁶Pb, 0.0004 for ²⁰⁷Pb / ²⁰⁶Pb and 0.0001 for ²⁰⁴Pb / ²⁰⁶Pb.

Sample No.	²⁰⁸ Pb/ ²⁰⁶ Pb	²⁰⁷ Pb/ ²⁰⁶ Pb	²⁰⁴ Pb/ ²⁰⁶ Pb
4813-13	2.068	0.8251	0.0526
4814-13	2.077	0.8410	0.0537
4815-13	2.064	0.8346	0.0533
4816-13	2.064	0.8328	0.0531
4817-13	2.058	0.8264	0.0527
4818-13	2.060	0.8312	0.0530
4819-13	2.115	0.8669	0.0554
4820-13	2.061	0.8283	0.0527
4821-13	2.036	0.8235	0.0524
4822-13	2.046	0.8227	0.0523



Figure 8: The lead isotope ratios of copper ore from Wadi Amram. The black points represent the new analyses presented in this study; the gray points represent the published analyses of Asael, et al. (2012). The analytical error is smaller than the size of the symbol.

The data collected for this study characterize the deposit as follows: for the lead isotope ratios ²⁰⁸Pb / ²⁰⁶Pb, the median is $Q_2 = 2.062$, and the lower and upper quartile are $Q_1 = 2.058$ and $Q_3 = 2.068$. The outer deciles are $D_1 = 2.041$ and $D_9 = 2.096$. For ²⁰⁷Pb / ²⁰⁶Pb, the tendencies are $D_1 = 0.8231$, $Q_1 = 0.8251$, $Q_2 = 0.8298$, $Q_3 = 0.8346$ and $D_9 = 0.8540$. The ratio ²⁰⁴Pb / ²⁰⁶Pb can be represent-

ed as follows: D₁ = 0.0523, Q₁ = 0.0526, Q₂ = 0.0528, Q₃ = 0.0533 and D₉ = 0.0552. It is striking that sample 4819-13 in all values is very different from the other lead isotope ratios from Wadi Amram. The lead isotope data are much higher. They are for ²⁰⁸Pb / ²⁰⁶Pb = 2.115, for ²⁰⁷Pb / ²⁰⁶Pb = 0.8669 and for ²⁰⁴Pb / ²⁰⁶Pb = 0.0554 (Table 3). This sample is identical in its isotopic composition with

a cluster of data from Timna and Faynan which at best reflect the original Precambrian copper mineralization in the basement of the crystalline rocks.

In comparison with the data published by Asael, all three lead isotope ratios analyzed in this study tend to be slightly higher. The variation of the lead isotope ratios can be characterized as followed: the ²⁰⁸Pb / ²⁰⁶Pb ratios are D₁ = 2.080, Q₁ = 2.084, Q₂ = 2.087, Q₃ = 2.087 and D₉ = 2.088. The results of the ²⁰⁷Pb / ²⁰⁶Pb are D₁ = 0.8400, Q₁ = 0.8432, Q₂ = 0.8452, Q₃ = 0.8453 and D₉ = 0.8454. For ²⁰⁶Pb / ²⁰⁴Pb, they are D₁ = 0.0535, Q₁ = 0.0538, Q₂ = 0.0539, Q₃ = 0.0539 and D₉ = 0.0539.

The datasets diverge slightly in their distribution and their lead isotope ratio values, but certain points do overlap and show a strong correlation. Together, the two datasets form a relatively cohesive point cloud (with exception of the two outliers). This can be seen particularly when compared to other copper ore deposits in the southern Levant.

Discussion

Comparison to the Lead Isotope Ratios of Ores from Timna and Faynan

For the copper production of the two Chalcolithic settlements of Tall al-Magass and Tall Hujayrat al-Ghuzlan, the ore from the local deposits of Timna, Faynan and Wadi Amram should be considered first. It is problematic in provenance studies when the geologic time scale and the geochemistry of ore formation are similar, and hence result in similar lead isotope ratios and elemental patterns. Therefore, it is important to discuss whether the lead isotope compositions of the copper ore deposits can be differentiated and if it is possible to produce clear results to be used for provenance studies. Through new highly precise analyses of copper ore from Timna, a partial separation of the lead isotope fields from Timna and Faynan have been achieved (Segal, et al., 2015). These new results now illuminates certain differences. In this section, the lead isotope ratios of Timna and Faynan will be presented, and afterwards they will be compared to the new data from Wadi Amram in order to determine to what extent they are distinct.

While the copper ore from the mining region of Wadi Amram lies in rock from the Lower Cretaceous (LC), the ore from Timna comes from layers from both the Lower Cretaceous and Cambrian. The copper minerals found in the LC sandstones of Timna are chalcocite, covelline, malachite and (par-) atacamite intergrown in parts with limonite (so called Ziegelerz in German language). Most characteristically the copper ores substitute plant remains (Bartura, Hauptmann and Schöne-Warnefeld, 1980). This is a unique feature of the copper ores from Timna and was not observed in any other mineralizations discussed in this study. In the sandstone, shales and dolomitic rocks of the Cambrian, minerals like chrysocolla, bisbeeite, malachite, chalcocite, djurleite and paratacamite can be found. The goal of the study of Segal, et al. (2015) was to characterize each of the mineralizations, and therefore the entire deposit of Timna, using lead isotope analysis.

The copper ore from the deposit of Faynan are found in rocks of the Cambrian age. There are two main types of copper mineralizations. In the impregnated Massive Brown Sandstone (MBS) zone, there is malachite, chalcocite and covellite. The other mineralization type is in the form of copper silicates, malachite and paratacamite in the Dolomite-Limestone-Shale Unit (DLS) zone. The lead isotope ratios of the two copper ore types have been discussed by Hauptmann (2000; 2007). In the recent past, the lead isotope data from Timna was considerably smaller making it difficult to distinguish the two deposits.

The new investigations have shown that there is a partial differentiation in the lead isotope ratios from Timna and Faynan. The ore from the MBS and the DLS zones at Faynan form two different groups that lay on the same trend line. The Timna ore from the Cambrian shale and the Cretaceous sandstones as well as the ore of Precambrian age form two groups with the same trend line as the ore from Faynan. Ore from the Faynan DLS zone and the Timna LC ore strongly overlap and are thereby indistinguishable by lead isotope analysis. Another lead isotope group is formed at Timna by the Cambrian dolomite and sandstones. Together with ore from the Sinai Peninsula, in the ²⁰⁸Pb / ²⁰⁶Pb diagram, they form a trend line that is different from the others. Conversely, this means that they can be differentiated from ore from the Faynan MBS zone. For the same ²⁰⁷Pb $/^{206}$ Pb ratio, there are different 208 Pb / 206 Pb values. The ore from the Faynan MBS of the Cambrian can be clearly distinguished from all types of ore from Timna (Segal, et al., 2015, pp.212-216).

When compared to the lead isotope ratios of ore from Timna and Faynan, the ore from Wadi Amram, with one exception (4819-13), fits directly in the isotopic field of the Faynan MBS zone (Figure 9). In other words, the ore from the Cretaceous White Sandstone of Wadi Amram has the same lead isotope ratios as the ore from the Cambrian MBS of Faynan. Like the Cambrian MBS ore of Faynan, the lead isotope ratios of ore from the Cretaceous White Sandstone of Wadi Amram can be



Figure 9: Lead isotope ratios of copper ore from deposits in Wadi Amram, Faynan and Timna compared to copper artifacts from the Chalcolithic settlements of Tall al-Magass and Tall Hujayrat al-Ghuzlan. The symbols used are larger than the analytical error.

unequivocally differentiated from Timna using the $^{207}\mathrm{Pb}$ / $^{206}\mathrm{Pb}$ to $^{208}\mathrm{Pb}$ to $^{206}\mathrm{Pb}$ diagram.

While the point cloud of analyses of ore from Wadi Amram (without the two outliers) represents 25 samples, the Faynan MBS type ore is represented by only five samples. These five from Faynan correlate to the Wadi Amram samples. The variation of ²⁰⁸Pb / ²⁰⁶Pb from these Faynan MBS samples are Q1 = 2.047, Q2 = 2.076 and Q3 = 2.076, for ²⁰⁷Pb / 206Pb Q1 = 0.8215, Q2 = 0.8387 and Q3 = 08393 and for ²⁰⁶Pb / ²⁰⁴Pb Q1 = 0.0523, Q2 = 0.0535 and Q3 = 0.0536. Due to the low number of samples, there are large gaps between the points, whereas the more numerous samples from Wadi Amram better fill the space. Though the two isotope ratio fields overlap, there is still potential for the differentiation of copper artifacts by source.

The Provenance of Copper Artifacts from Tall al-Magass and Tall Hujayrat al-Ghuzlan

The Chalcolithic settlements of Tall al-Magass and Tall Hujayrat al-Ghuzlan are located near the city of Aqaba in now environmentally inhospitable areas. With connections to the Red Sea via the Gulf of Agaba, the location of the settlements makes them predestined for importance in long distance trade. Since the large scale excavations of the settlements had begun in 1998, a number of archaeometallurgical finds have been uncovered (Khalil and Schmidt, 2009). Objects like copper minerals, slag, ingots, casting moulds and casting debris are evidence that the complete chaîne opératoire for a metalworking industry was performed in these settlements - from the smelting of ore to the preparation of metal for trade. Both of the Chalcolithic settlements, more precisely the inhabitants, seem to have been specialists in metalworking (Klimscha, 2013b, pp.45-51). The dating of the metalworking at the settlements can be confined to the period of 4200 to 3600 BC (14C dating) and thus spans the transition period between the Chalcolithic and the Early Bronze I (Hauptmann, Khalil and Schmitt-Strecker, 2009, p.295). For the chronology of the southern Levant refer to Klimscha (2012).

These periods are particularly important for the history of economic development. The growth of copper metallurgy not only brought about new and better tools, but also reflects advances in the organization of labor. Complex tasks like copper mining and smelting need large numbers of specialized workers tied to these activities. Among other industries, the development of copper metallurgy was a catalyst for the division of labor in ancient societies, which, like other industries of the period, had to be tightly organized through control mechanisms and power. Towards the end of the 4th

millennium BC, the first cities and states were developing in the Near East, particularly in Mesopotamia and Egypt. This process can be called urbanization, or in contrast to the Neolithic Revolution, the Urban Revolution (Childe, 1950). This brought with it the expansion of long-distance trade networks, which could ensure the steady supply of raw materials to these cultures, especially in regard to rare and prestigious goods. This raises further questions about the organization of these trade networks, or for example, were Tall Hujayrat al-Ghuzlan and Tall al-Magass trade posts controlled by foreign powers or were they indigenously controlled (Klimscha, 2012, p.190).

In the reconstruction of complex organizations of labor and the resulting trade and exchange of goods, the use of science-based provenance studies can be an important factor. On one hand, provenance studies can be used to link the mining activities to the locations where ore was processed and smelted to metal, and, on the other, it can link the traded metal back to the production sites and the ore deposits. For this reason, lead isotope analyses of copper-related artifacts from Tall Hujayrat al-Ghuzlan (Abdel-Motelib, et al., 2012; Hauptmann and Khalil, in prep.) and Tall al-Magass (Hauptmann, Khalil and Schmitt-Strecker, 2009) were performed. The samples analyzed are composed primarily of objects that can be tied directly to copper production such as copper smelting slag and casting waste. The working hypothesis was that the copper came from Timna, which is approximately 30 km distance. Additionally, the deposit of Faynan, more than 120 km away, and several copper mines in the Sinai were considered (Hauptmann, Begemann and Schmitt-Strecker, 1999).

Some of the artifacts could come from Timna, or more precisely from the LC mineralizations found there. Furthermore, mineralizations from the Faynan DLS and MBS zones are also possible sources. For the transportation of raw materials and supplies between the mines and the settlements, Klimscha proposes small caravans of domesticated donkeys. With regard to proximity, the deposits of Timna seem to be much more likely as a source of raw materials for the two settlements laying near the Gulf Aqaba rather than Faynan, which is significantly further away. The transit from Faynan would have taken days and would have been highly arduous and dangerous, if not unnecessarily so (Klimscha, 2013b, p.46). There is yet still a discussion about why there are no settlements of this period close to Timna. This would have made the transport of copper ore over such distance unnecessary and would have simplified the control over the copper source. A possible reason behind this is that the two settlements truly functioned as trade

posts. From there, other groups such as the Egyptians or mobile social groups may have been responsible for the transport of raw/semi-processed materials (Klimscha, 2013b, p.52).

However, not all of the copper-related artifacts are consistent with these deposits (Hauptmann, Khalil and Schmitt-Strecker, 2009, pp.301-303). The copper ore deposit of Wadi Amram has only recently been studied and, therefore, has not been played any role in the discussion of the provenance or as a potential source of raw materials. Concerning the results of the lead isotope analysis, particularly the ²⁰⁷Pb / ²⁰⁶Pb to ²⁰⁸Pb / ²⁰⁶Pb diagram (Figure 9), it is clear that the ore deposit of Timna was probably not the only source of copper. With the assumption that the ore deposit of Timna has almost totally been characterized (Segal, et al., 2015), all artifacts that do not fit within the Timna isotope field must have another origin. If copper ore from more than one type of deposit were being processed, it is certainly possible that mixing occurred.

Four artifacts from Tall al-Magass and two from Hujayrat al-Ghuzlan are consistent with the field of ore from Wadi Amram and the Faynan MBS zone. Due to the low density of these point clouds, based on the small sample sizes, it is not possible to link every artifact with a certain piece of ore. Particularly in the case of sample JD-35/4 (a bar/ingot-like object from al-Magass), it is relatively far from all other ore samples. Two further samples from Tall al-Magass plot directly near ore samples from the Faynan MBS. One sample from Hujayrat al-Ghuzlan and one from Tall al-Magass are highly consistent with ore from Wadi Amram. Based on the current data pool of lead isotope ratios the copper mines of Wadi Amram were the most likely raw material source for these two artifacts.

Conclusions

The lead isotope analysis of ore and copper artifacts has shown that, next to Timna, Wadi Amram could have been a supplier of copper ore for the metallurgical activities of Tall Hujayrat al-Ghuzlan and Tall al-Magass. Wadi Amram is only 10 km from Tall al-Magass and Tall Hujayrat al-Ghuzlan and it is the closest source of copper. If copper ore from mines in Wadi Amram was processed at the two settlements, the most conclusive evidence would be if traces of Chalcolithic mining could be found in the future. Further archaeological investigations around Wadi Amram and prehistoric Aqaba may help to confirm the results of this study and may reveal further evidence for Chalcolithic copper metallurgy. Furthermore, an increase in the number of ore and copper-related samples from this region for lead isotope analysis would significantly improve the clarity of the results.

With the present results, there might be an indication that the mining of copper ore from Wadi Amram dates back to the Late Chalcolithic. As a result, models on the organization of mining in the region are in need of reevaluation. Additionally, the reconstruction of the organization of labor and the transfer of resources must be reconsidered. The geographically close proximity of the settlements to the copper source would have made direct control of the resources much easier. The location of Tall al-Magass and Tall Hujayrat al-Ghuzlan would make much more sense. The indigenous population as owners of the mines would be then more likely.

Acknowledgements

We would like to thank Uzi Avner and his team for providing us with unpublished results of their archaeological works at Wadi Amram in recent years. Without these preliminaries, this study would not have been possible. We would like to thank Prof. Dr. Lutfi Khalil, Amman-University, Jordan, who permitted us to use the Hujayrat al-Ghuzlan data. Furthermore, we are much obliged to Michael Bode, Regina Kutz and Dirk Kirchner, Materialkundliches Labor, Deutsches Bergbau-Museum, for their support in sample preparation, chemical analyses and XRD. The lead isotope analyses were thankfully conducted by Sabine Klein, Goethe-Universität Frankfurt. Last but not least, Stephen Merkel, Deutsches Bergbau-Museum, is acknowledged for his extensive editorial assistance. We would like to thank the two anonymous reviewers for their comments that helped improve the quality of this paper.

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