Where Does the Gold from the Cemetery of Ur Come From? – Provenancing Gold Sources Using Analytical Methods

Moritz Jansen, Andreas Hauptmann and Sabine Klein

Introduction

The following work is part of a PhD project on the archaeometallurgy of Bronze Age gold in the Middle East. In the current study, the origin of the raw materials for the gold artifacts from Ur will be discussed. The hundreds of golden artifacts from the Early Dynastic (ED) period are unique in the third millennium Mesopotamia. The fact that there are no natural sources of gold in Mesopotamia prompts questions about the origin of the raw material used to create these artifacts. Gold was likely imported from other regions such as Anatolia, Iran, Afghanistan, Egypt or India. The rich corpus of cuneiform tablets could help to identify possible trade relations, as they give an idea of possible origins due to named trading posts. However, these tablets date slightly later than the ED period in Ur and nothing is known about the provenance of the earliest gold artifacts at the site.

Therefore this project seeks to define the geochemical fingerprint of these objects in order to identify the origin of the gold through its chemical and isotopic composition. Ideally, this method could trace the gold back to the location of its natural deposit(s). For our analytical work, we used two different methods: first, the chemical compositions with main, minor and trace elements were determined by electron probe microanalysis (EPMA) and laser ablation inductively coupled plasma mass spectrometry (LA-ICP-MS); second, the isotopic compositions of selected elements (Pb, Os, Cu) were measured by multicollector plasma mass spectrometry (MC-ICP-MS).

Golden alloys – natural or intentional?

The artifacts of Ur present different kinds of alloys (see Hauptmann and Klein, this volume). For the geochemical fingerprint of the gold itself, unalloyed gold artifacts need to be characterized. In alloyed artifacts the other components could obscure the distinct signature of the gold source. In this regard, it seems to be useful to analyze the artifacts of the headdresses with hair ribbons, flowers, diadems and necklaces instead of definitely alloyed artifacts like the chisel, adze and dagger.

The focus of our analysis is on the richly decorated Early Dynastic Royal Tombs. Forty-five samples were taken from artifacts of the Great Death Pit (PG 1237), the King's Grave (PG 789) and other tombs. Nine samples came from objects within the graves, which excavator Woolley attributed to the Second Dynasty and the Sargonid Period. These graves are mixed in date and grouped here as Akkadian to Ur III period. Twelve additional objects are without stratified contextual information and are discussed separately. The main components of the gold artifacts are gold, silver and copper. Between 15 and 45 percent of silver and between 1 and 6 percent of copper was typical for the samples. The jewelry of the Early Dynastic period has the same composition as the later Akkadian and Third Dynasty. Based on analysis of their main components, they could come from the same sources.

It needs to be clarified whether the gold composition is natural or is an intentional alloy. In ancient times silver was exploited by the use of silver-bearing lead minerals such as galena or cerussite. One would expect a positive correlation between silver and lead for the gold artifacts if the silver content of the gold was an intentional alloy (Hauptmann, et al., 2010). However, there is no correlation: the silver of the golden objects does not seem to originate from lead ores.

For comparison, the composition of gold, both primary and secondary, of various occurrences in the Lesser Caucasus is shown in Figure 1. These deposits (unrelated to Ur) form a broad database because they were systematically investigated in recent years by the authors and a second group from Mannheim and Halle (Wolf, et al., 2011; Wolf, et al., 2013). There is an overlap between the silver contents of the artifacts with silver in natural gold from this region. The content of silver of the artifacts is within the range of natural composition. But it is also evident that natural gold does not contain copper in the range of the gold artifacts from Ur. So is this an intentional alloy? This question will be discussed again later.

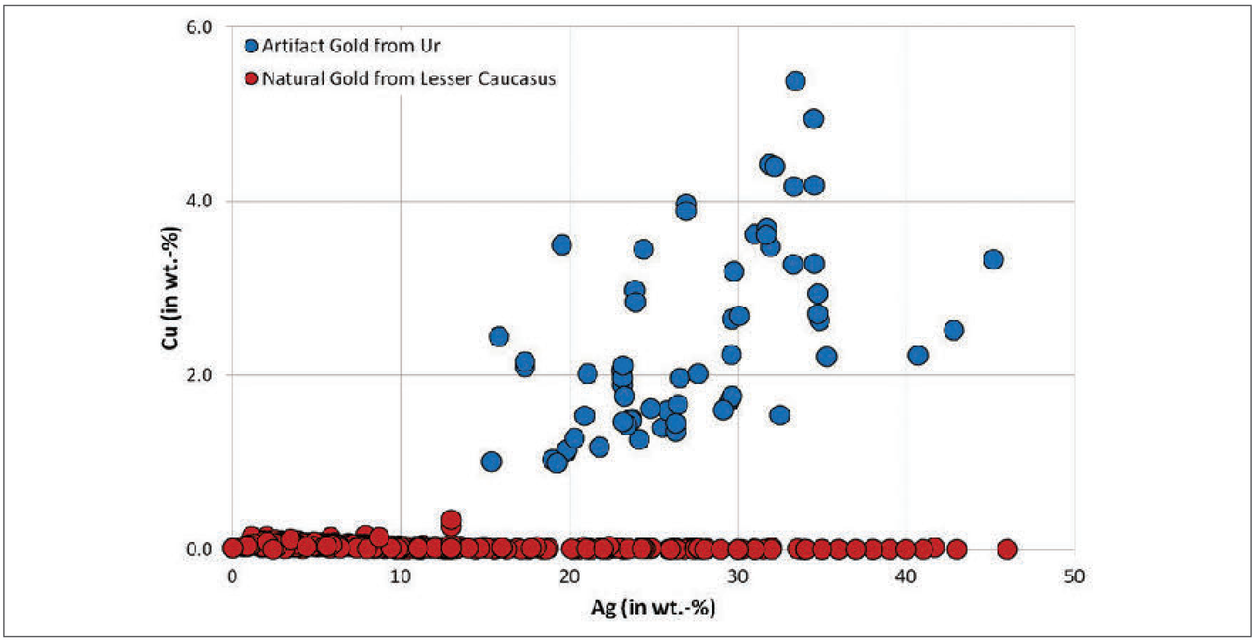

Figure 1. The alloy components of the gold artifacts from Ur are compared to natural gold of the Lesser Caucasus. The variability of the silver content is comparable, but the natural gold does not show significant contents of copper.

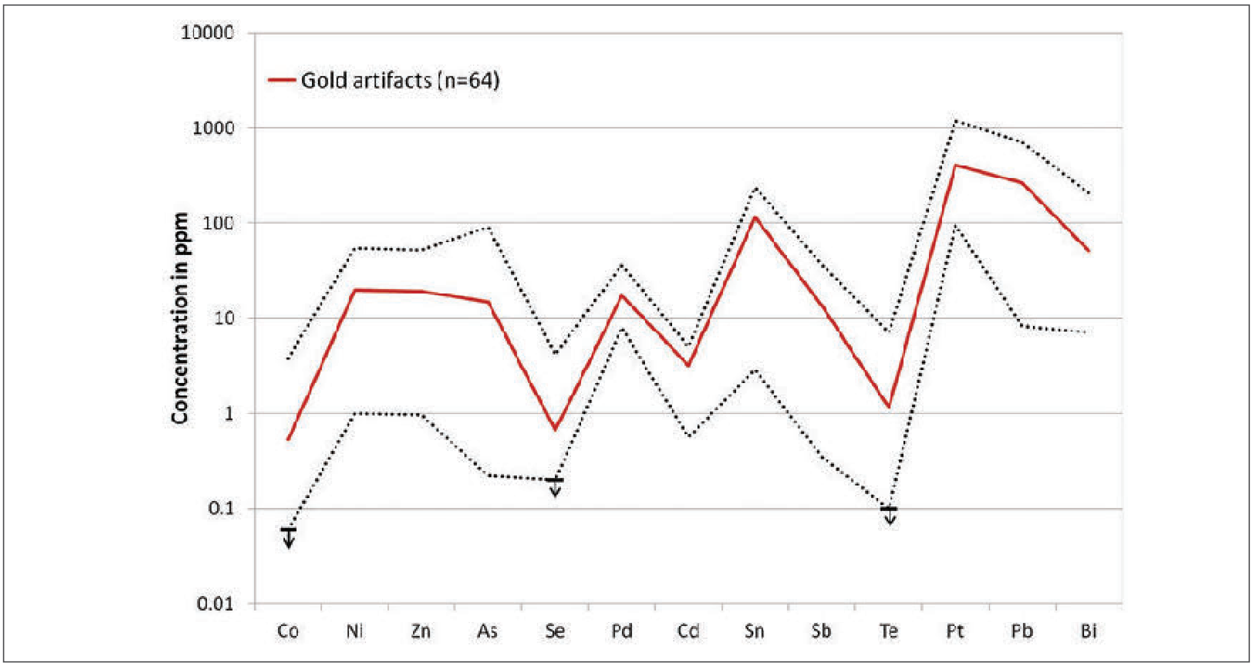

Figure 2. Trace element pattern of 64 objects from Ur. The red line shows the mean of all objects, the black lines show minimum and maximum. Arrow symbols showing detection limits.

Trace Elements as fingerprint

Trace element patterns of gold artifacts used for provenance studies have been discussed in recent years through the introduction of sensitive laser ablation plasma mass spectrometry (Ehser, et al., 2011; Hauptmann, et al., 2010; Kovacs, et al., 2009; Schlosser, et al., 2010). In the following section, the trace element patterns of the gold artifacts from Ur are presented. Following the definition in geochemistry, trace elements are elements with concentrations below 0.1 percent (given as ppm – parts per million) and which do not form their own phases. In Figure 2 the trace element pattern of the gold samples is plotted. There are significant occurrences of tin, platinum and lead above 100 ppm.

Because of the combination of tin and platinum, it is evident that the resources came from a placer deposit. Tin minerals like cassiterite are enriched in primary occurrences in granitic rocks. Platinum minerals are typically formed in basic rocks. After erosion they can

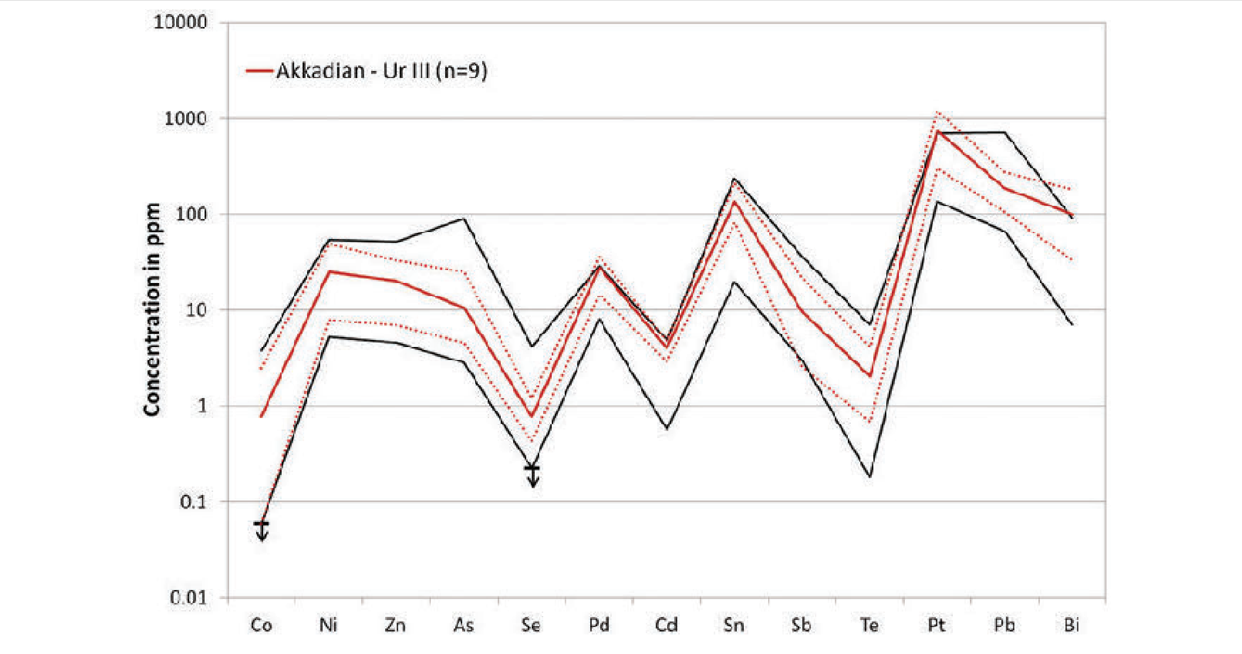

Figure 3. Comparison of the trace elements between the Early Dynastic (given minimum and maximum as black lines) gold artifacts and the later Akkadian / Ur III artifacts (red line: mean; dashed red lines: minimum and maximum). The patterns are the same. Arrow symbols showing detection limits.

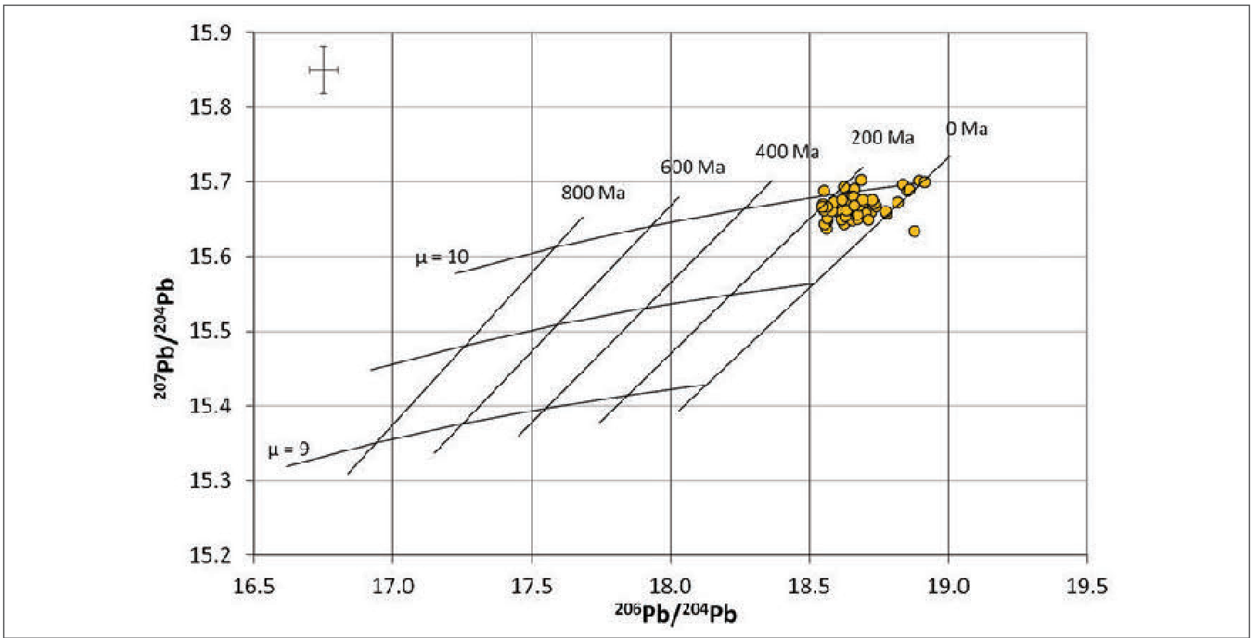

Figure 4. The lead isotope ratios of the gold artifacts from Ur plotted with the Stacey and Kramers lead evolution model (Stacey and Kramers, 1975). The sources of the lead traces in the gold artifacts were formed in the last 200 Ma.

be co-deposited in placers. Compared to the composition of analysis from reef and also alluvial gold, elements like nickel, arsenic and bismuth are also enriched. Only if there are inclusions of other minerals, the analysis of natural gold shows significant traces of other elements. The gold of the artifacts seems to have been contaminated during metallurgical processing. Washing gold never leads to pure gold concentrate. Different heavy minerals change the trace element pattern of the gold flakes / dust during the melting of the concentrate.

By focusing on the dated artifacts, a very small variation in the trace element patterns of all the Early Dynastic gold contents is visible. If we compare the later artifacts of the Akkadian period and Third Dynasty as red lines, these objects have the same pattern (Figure 3). The contents of palladium, platinum and bismuth are slightly higher. That does not mean that other resources were used. Maybe other parts of the same deposits were exploited. A stable use of the same occurrences over hundreds of years might be possible.

Ratios of Isotopes

Isotope ratios of different elements were determined in order to define a geochemical fingerprint. The lead isotope composition provides information about the geological age of the deposits used in ancient times for metal exploitation. For the golden artifacts of Ur a lead evolution model age of 0 to 200 million years can be determined (Figure 4). The source of the lead in the gold artifacts could not have been formed more than 200 Ma. If the groups of different dates are compared, no significant differences are visible. That means that the sources of the gold from Ur may be continuous from Early Dynastic and Akkadian to Ur III times. Both the chemical composition and lead isotope ratios support this conclusion.

A number of copper-based objects from Ur dating between Early Dynastic and the Akkadian period were analyzed for their lead isotopic composition by Begemann and Schmitt-Strecker (2009). The dataset forms two clusters. The gold artifacts have the same composition as one of these clusters (Figure 5). The question arises whether this is due to the alloying of gold with copper from the same source as where the copper-based artifacts originated. Meaning: could it be that the copper content of 1 to 6 percent is an expression of intentional alloying? The lead traces in the gold could originate from the copper due to the low lead content of natural gold.

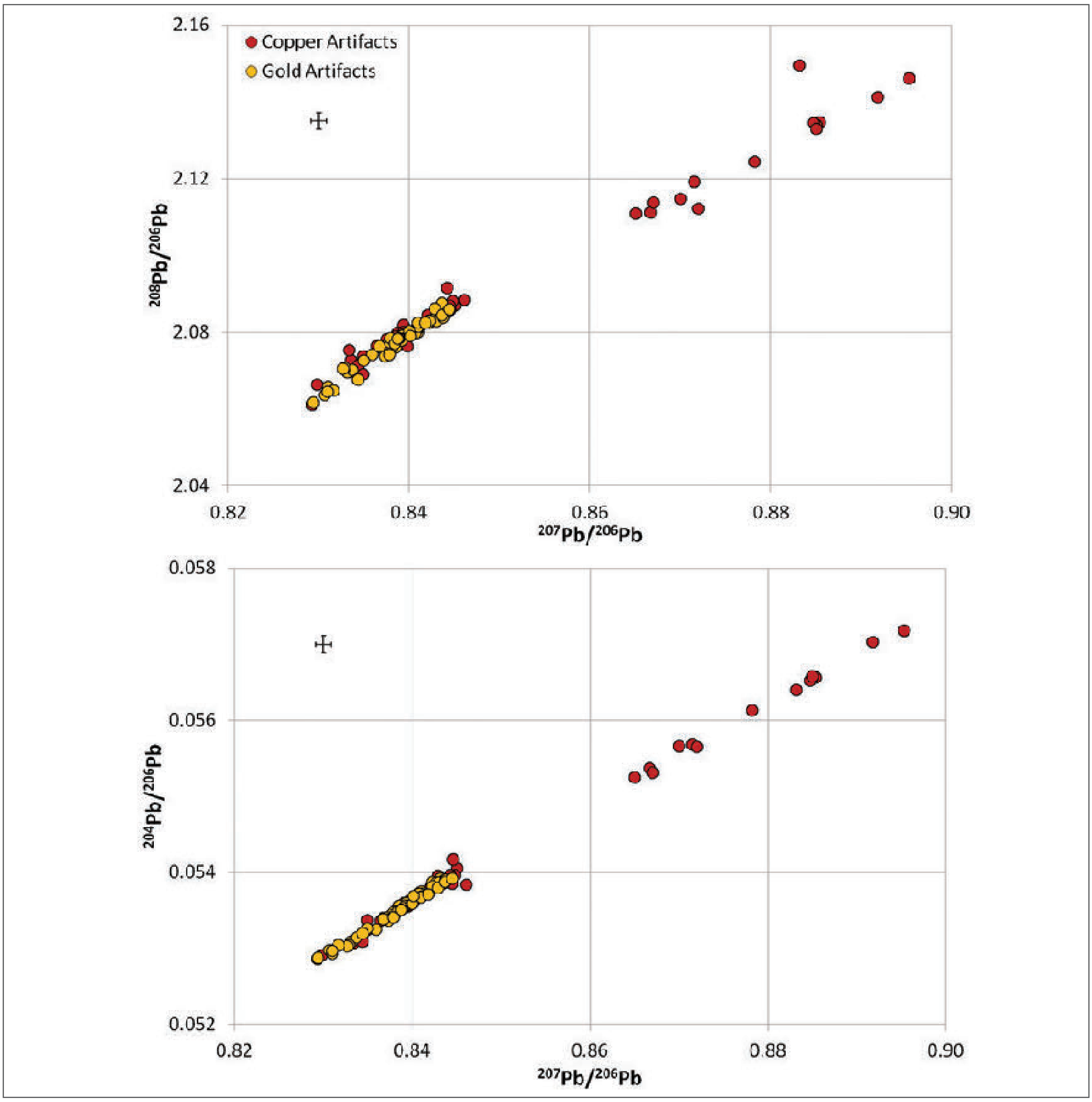

Figure 5. Lead isotope ratios of gold artifacts as well as copper-based artifacts from Ur analyzed by Begemann and Schmitt-Strecker (2009). The gold artifacts are compatible with one of the two clusters.

Most of the analyzed copper objects in addition were characterized for their chemical composition by Lutz and Pernicka (2004). We are able to calculate from these analyses an expected lead content due to alloying with this specific copper (Figure 6a). The expected lead content is much lower than the determined lead content in the objects and there is no positive correlation with increasing copper content. The lead does not originate from an alloyed copper. In addition, a high arsenic content is typical for the copper artifacts of Ur. Because of this we would also expect a much higher arsenic content in the gold artifacts and a positive correlation with copper (Figure 6b). Instead of an intentional alloy, a contamination by the other heavy minerals in the gold concentrate is more realistic.

As the artifact gold can be characterized as originating from alluvial gold, the lead isotopes reflect the sources of a region, eroded and deposited in placers, and not of a single ore occurrence. These sources have the same lead isotope signature as the group of copper artifacts. The gold occurrences could be in proximity to the copper sources. So what is the "metallurgical" origin of the copper content itself in the gold artifacts?

The identification of copper isotope composition allows a characterization of the types of minerals that were used to produce the ancient copper. The copper isotope

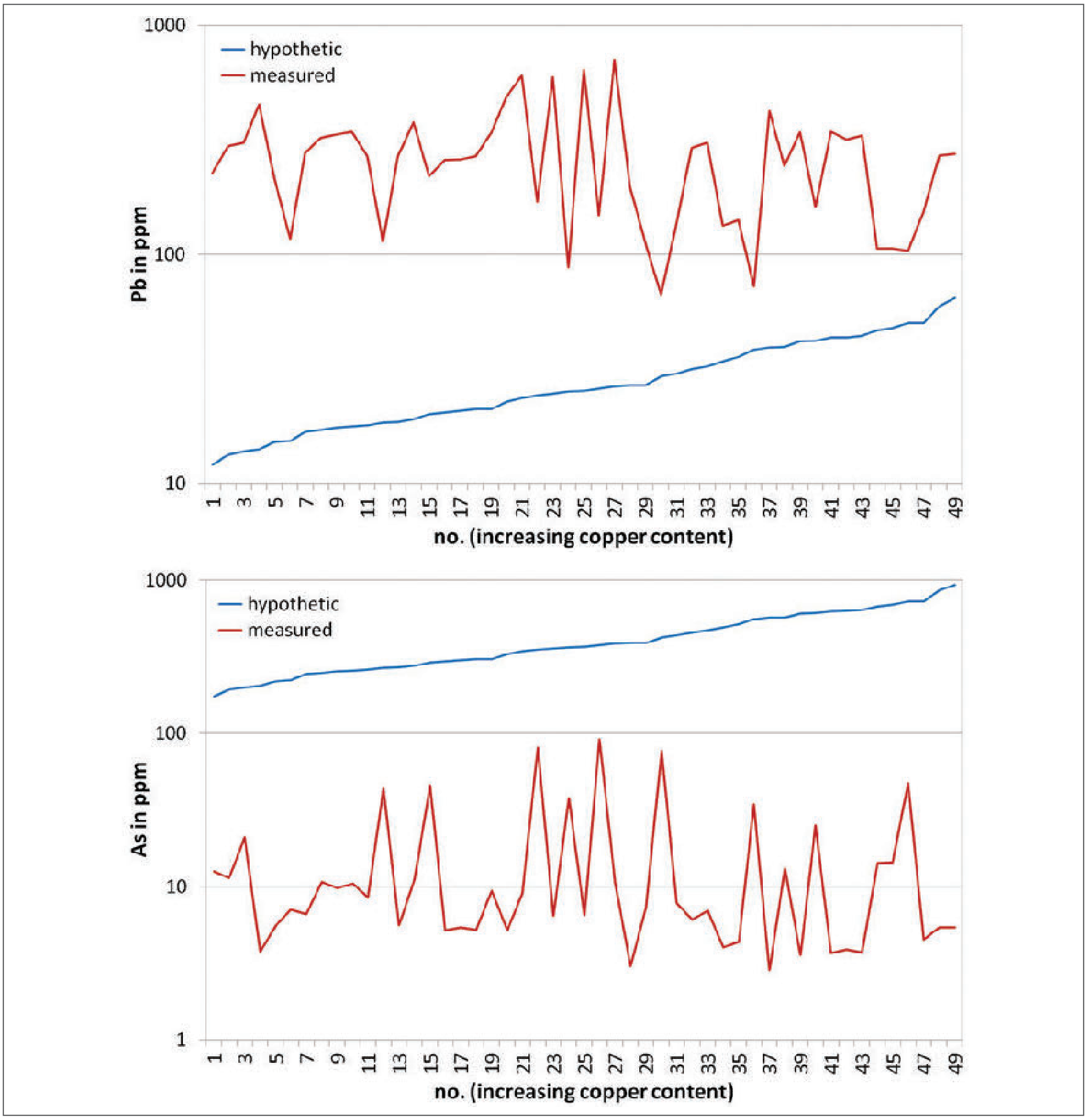

Figure 6. Hypothetic and measured lead and arsenic contents in the gold artifacts. Calculation based on the analyses by Lutz and Pernicka (2004).

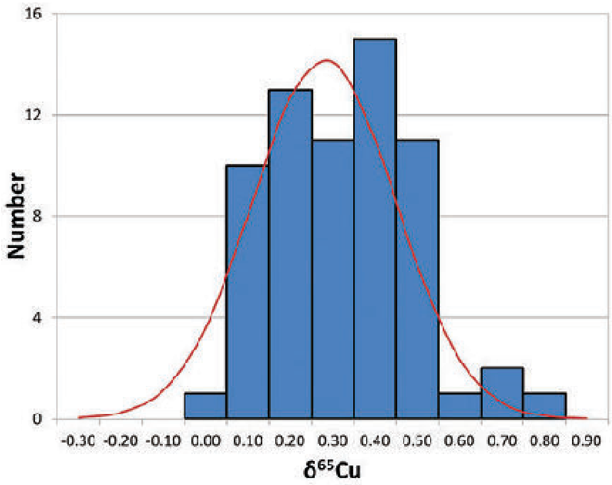

Figure 7. Copper isotopic composition of the gold artifacts from Ur. The positive δ^{65}Cu-values indicate oxidized copper minerals in the gold-bearing heavy mineral concentrates.

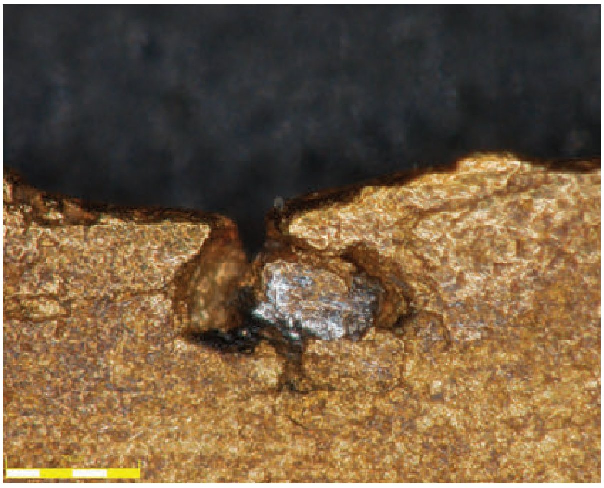

Figure 8. PGE-inclusion in a gold artifact from Ur. The scale is 200 µm.

signature of the gold from Ur shows a range of positive delta values (Figure 7) which is typical for a source of oxidized minerals (Klein, et al., 2010). Heavy mineral concentrates can also contain oxidized copper minerals like malachite (Hauptmann, et al., 2010). The copper in the gold seems to originate from the heavy mineral concentrate containing oxidized copper minerals. The copper source of the gold is also different from the copper-based objects in the copper isotopic composition (Salzmann, et al., this volume), indicating that the copper in the gold does not originate from intentional alloying with this copper.

Geochemistry of PGE inclusions

A significant characteristic is the numerous inclusions of natural alloys of elements of the so-called Platinum Group (PGE). In more than the half of all samples PGE inclusions were found (Figure 8). For comparison, we could identify such inclusions only in two samples of 120 artifacts from Georgia. The PGE inclusions are topic of another article (Jansen, et al., 2016). These Os-Ir-Ru alloys are not soluble in gold. The high contents of Pt and Pd in the trace element pattern originate from a second PGE source: Pt-Pd-minerals which are soluble in gold. The results of the investigation of PGE show that the chemistry of these inclusions is compatible to PGE alloys originating from ophiolite complexes where they formed in chromites of ultrabasic rocks. By calculating different models, a possible age of 290 – 610 Ma can be estimated for the crystallization of the alloys using their most frequent Os isotopic composition.

Discussion

After the geochemical characterization of the artifacts, possible candidates for the origin of the gold from Ur can be discussed. The calculated formation date of PGE inclusions in the gold from Ur is higher than the age of the Mesozoic ophiolites of the Tethyan Eurasian Metallogenetic Belt (TEMB) which hosts most of the ophiolites ranging from the Alps, Balkan, Anatolia, Iran, Afghanistan and further East to the Himalayas.

Older ophiolite complexes can be found in Egypt and Arabia. They were formed 700 to 900 Ma ago. There are many gold occurrences in the Eastern Desert next to these ophiolithe complexes. They were exploited since the third millennium BC (Klemm and Klemm, 2013). The gold-silver ratio of the gold deposits of the eastern desert is the similar to the objects of Ur. The occurrences in Nubia contain less silver. However, the deposits of Egypt and Saudi Arabia can be excluded as sources for the gold from Ur: if these deposits were used for the exploitation they should have the same lead isotopic composition as local ore resources (data published by Doe and Rohrbough, 1977; Stos-Gale and Gale, 1981). They are not compatible with the lead isotope ratios of the golden artifacts of Ur. In addition we can exclude the Egyptian deposits by comparison with Os isotopes. The Os isotope composition of the inclusions of Ur is different from that of the Os-Ir-minerals of the ophiolites from the Eastern Desert with lower ratios. The Eastern Desert ophiolites are older than the calculated model age.

The Omani ophiolite is one of the Mesozoic examples of the TEMB. It also shows a different Os isotopic

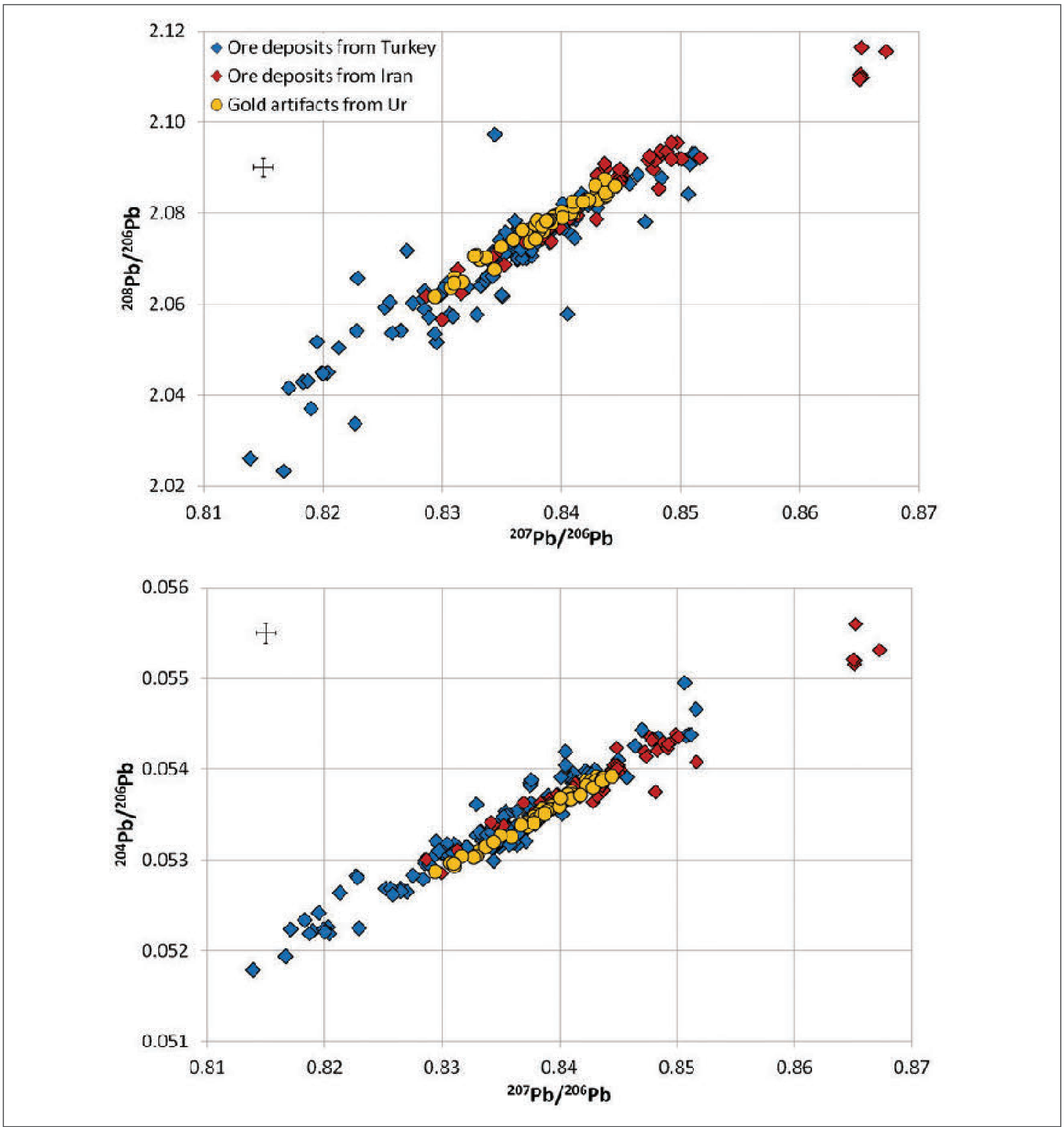

Figure 9. Lead isotopic composition of the gold artifacts compared to deposits from Turkey (Wagner, et al., 1986) and Iran (Mirnejad, et al., 2011; Pernicka, et al., 2011). The gold artifacts are compatible with this composition.

composition with higher ratios. We have to search ophiolites which are older than Mesozoic and younger than Proterozoic ones. In the Middle East there are some rare remnants of Paleozoic ophiolites. The model age of the PGE inclusions fits their age. We have to compare the lead isotope ratios of deposits within the TEMB, where remnants of these ophiolites occur.

The lead isotope composition of ore deposits from Anatolia and Iran fits with the golden artifacts from Ur (Figure 9). Little is known about the lead isotopic composition of deposits from Afghanistan. The rare datasets show some highly radiogenic lead isotope compositions like the deposits of Mes Aynak (Begemann and Schmitt-Strecker, 2009). There is an overlap with a few ore specimens, but additional samples are needed. The general conclusion is that the gold artifacts from Ur fit well to the ore deposits of the TEMB.

The gold occurrences should be next to Paleozoic ophiolites due to the Os isotopic composition of the PGE inclusions. For Anatolia and the Caucasus, there are no gold occurrences in proximity to the Palaeozoic ophiolites. In Iran the largest gold reserve is close to Paleozoic ophiolites next to Takab. There are very rich gold placers which were also worked in premodern times.

Another hint is given through the artifacts artistic style. The leaves of the headdresses of Ur were interpreted as the Indian Rosewood (Dalbergia sissoo Roxb.) which indicate an association to the east (Tengberg, Potts and Francfort, 2008). The present day distribution of this tree is in the Indus Valley, Baluchistan and the Asian coastal area of the Gulf of Oman. The later cuneiform tablets also described gold from *Meluḫḫa*.

Typical for the gold of Ur is the association with lapis lazuli and carnelian. There are some analogies between the inventories of gold from the Harappan culture of the Indus Valley and the gold from Ur, including an association with gemstones, the shape of diadems, and the discoid beads. Unfortunately, only a few analyses were done for the Harappan gold (Kenoyer and Miller, 1999). Five artifacts from Harappa itself contain less silver (6 – 9 wt. %) than the Ur artifacts, while analyses of two artifacts from Lothal (34 & 42 wt. %) have the same silver content. In the Indus Valley gold deposits are not present. The gold sources of the Harappan culture therefore are expected to be to the north or northwest of the Indus Valley, possibly in Afghanistan or Iran.

In Afghanistan some relics of Paleozoic ophiolites exist in Badakhshan. In proximity to the ophiolites, the richest alluvial gold deposits of Afghanistan can be found at the Amu Darya River near Samti in Northern Takhar. This area is also of interest because the Harappan trading post Shortugai was found here. It is hundreds of kilometers away from the Indus Valley. Shortugai has the typical inventory of a Harappan city with its architecture, ceramics, seals, copper-related metallurgy, beads of gemstones, but also some of gold (Francfort, 1989). The lapis lazuli deposit of Sar-e Sang is in its vicinity. Shortugai indicates a direct connection between the Indus Valley and a region of interest for gold.

Conclusions

The golden jewelry from Ur is a natural alloy of gold and silver originating from placer gold. In addition, it contains some copper, presumably from an oxidized mineral source. There are high contents of tin, platinum, lead and other trace elements. In contrast, there are low trace elements in natural gold. For artifact gold, there is a contamination during metallurgical processing due to the presence of other heavy minerals. There are no changes in the amount of alloy components as well as the trace element pattern between Early Dynastic III and Akkadian / Ur III period. The sources of the trace element lead are heavy minerals which are not older than 200 Ma old. There are no changes in the lead isotope composition between EDIIIA and Akkadian to Ur III period. This is explained by the use of the same sources of gold over hundreds of years.

The PGE inclusions are Os-Ir-Ru alloys which are not soluble in gold. In addition, Pt-Pd minerals were dissolved during metallurgical melt. The PGE minerals originate from ophiolites which were formed 290-610 Ma ago.

The provenance of the gold can thus be stated: the source of the gold from Ur is likely located within the Tethyan Euarasian Metallogenetic Belt. The PGE inclusions most probably trace back to older ophiolite complexes. Possible origins of these are rare remnants of Paleozoic ophiolites. There are two locations that are potential candidates for the origin of the gold found at Ur. These two sources are the placer deposits found at Takab in Iran and at Samti in Afghanistan. These sources should be investigated in the future to compare the lead isotopes of gold-bearing heavy minerals and Os isotope ratios of PGE minerals in local chromite occurrences.

References

Begemann, F., and Schmitt-Strecker, S., 2009. Über das frühe Kupfer Mesopotamiens. *Extrait d'Iranica Antiqua*, 44, pp.1-45.

Doe, B. R., and Rohrbough, R., 1977. Lead isotope data bank: 3,459 samples and analyses cited. *USGS Numbered Series, 79-661 open-report*. Washington, D. C.: U.S. Geological Survey.

Ehser, A., Borg, G., and Pernicka, E., 2011. Provenance of the gold of the Early Bronze Age Nebra Disk, central Germany. Geochemical characterization of natural gold from Cornwall. *European Journal of Mineralogy*, 23, pp. 895-910.

Francfort, H.-P., 1989. *Fouilles de Shortugai. Recherches sur l'Asie Centrale Protohistorique, Memoires de la Mission Archeologique Francaise en Asie Centrale*. Paris : *Diffusion de Boccard*.

Stos-Gale, Z. A., Gale, N. H., 1981. Sources of galena, lead and silver in Predynastic Egypt. *Actes du XXme Symposium International d'Archéometrie, Revue d'Archéometrie, Supplement*, 1, pp.285-295.

Hauptmann, A., and Klein, S., 2016. Golden Artifacts from the Royal Tombs of Ur, Mesopotamia. *Metalla*, 22(1), Supplement, this volume.

Hauptmann, A., Bendall, C., Brey, G., Japarize, I., Gambashize, I., Klein, S., Prange, M., Stöllner, T., 2010. Gold in Georgien. Analytische Untersuchungen an Goldartefakten und an Naturgold aus dem Kaukasus und dem Transkaukasus. In: S. Hansen, A. Hauptmann, I. Motzenbäcker and E. Pernicka, eds. 2010. *Von Majkop bis Trialeti. Gewinnung und Verbreitung von Metallen und Obsidian in Kaukasien im 4.-2. Jt. v. Chr.* Bonn: Habelt. pp.139-160.

Jansen, M., Aulbach, S., Hauptmann, A., Höfer, H. E., Klein, S., Krüger, M., and Zettler, R. L., 2016. Platinum Group Placer Minerals in Ancient Gold Artifacts – Geochemistry and Osmium Isotopes of Inclusions in Early Bronze Age Gold from Ur/Mesopotamia. *Journal of Archaeological Science*, 68, pp.12-23.

Kenoyer, J. M., and Miller, H.-L., 1999. Metal Technologies of the Indus Valley Tradition in Pakistan. In: V. C. Pigott, ed. 1999. *The Archaeometallurgy of the Asian Old World, MASCA Research Papers in Science and Archaeology*, 16. Philadelphia: University Museum University of Pennsylvania. pp.107-151.

Klein, S., Brey, G. P., Durali-Müller, S., and Lahaye, Y., 2010. Characterisation of the raw metal sources used for the production of copper and copper based objects with copper isotopes. *Archaeological and Anthropological Sciences*, 2, pp.45-56.

Klemm, R., and Klemm, D., 2013. *Gold and Gold Mining in Ancient Egypt and Nubia. Geoarchaeology of the Ancient Gold Mining Sites in the Egyptian and Sudanese Eastern Deserts*. Heidelberg: Springer.

Kovacs, R., Schlosser, S., Staub, S. P., Schmiderer, A., Pernicka, E., and Günther, D., 2009. Characterization of Calibration Materials for Trace Element Analysis and Fingerprint Studies of Gold using LA-ICP-MS. *Journal of Analytical Atomic Spectrometry*, 24, pp.476-483.

Lutz, J. and Pernicka, E., 2004. Neutronenaktivierungsanalysen. In: H. Hauptmann and E. Pernicka, eds. 2004. *Die Metallindustrie in Mesopotamien von den Anfängen bis zum 2. Jahrtausend v. Chr. Orient-Archäologie*, 3. Rahden / Westfalen: Verlag Marie Leidorf. pp.104-108.

Mirnejad, H., Simonetti, A., and Molasalehi, F., 2011. Pb isotopic compositions of some Zn-Pb deposits and occurrences from Urumieh-Dokhtar and Sanadaj-Sirjan Zones in Iran. *Ore Geology Reviews*, 39, pp.181-187.

Pernicka, E., Adam, K., Böhme, M., Hezarkhani, Z., Nezafati, N., Schreiner, M., Winterholler, B., Momenzadeh, M., and Vatandoust, A., 2011. Archaeometallurgical researches at Arisman in central Iran. In: A. Vatandoust, H. Parzinger, and B. Helwing, eds. 2011. *Early mining and metallurgy on the Central Iranian Plateau. Report on the first five years of research of the Joint Iranian-German Research Project. Archäologie in Iran und Turan*, 9. Mainz am Rhein: Zabern. pp.633-705.

Schlosser, S., Reinecke, A., Schwab, R., Pernicka, E., Sonetra, S., and Laychour, V., 2012. Early Cambodian gold and silver from Prohear. Composition, trace elements and gilding. *Journal of Archaeological Science*, 39, pp.2877-2887.

Stacey, J. S., and Kramers, J. D., 1975. Approximation of Terrestrial Lead Isotope Evolution by a Two-Stage Model. *Earth and Planetary Science Letters*, 26, pp.207-221.

Tengberg, M., Potts, D. T., and Francfort, H.-P., The golden leaves of Ur. *Antiquity*, 82, pp.925-936.

Wagner, G. A., Pernicka, E., Seeliger, T. G., Lorenz, I. B., Begemann, F., Schmitt-Strecker, S., Eibner, C., and Öztunali, Ö., 1986. Geochemische und isotopische Charakteristika früher Rohstoffquellen für Kupfer, Blei, Silber und Gold in der Türkei. *Jahrbuch des Römisch-Germanischen Zentralmuseums*, 33, pp.723-752.

Wolf, D., Borg, G., Pernicka, E., Meliksetian, K., Kunze, R., and Bobokhyan, A., 2011. Geoarchäologische Untersuchungen der Goldvorkommen von Sotk und Fioletovo, Armenien. In: H. Meller and P. Avetisyan, eds. 2011. *Archäologie in Armenien. Veröffentlichungen des Landesamtes für Denkmalpflege und Archäologie Sachsen-Anhalt-Landesmuseum für Vorgeschichte*, 64. Halle: Sachsen-Anhalt-Landesmuseum. pp.51-68.

Wolf, D., Borg, G., Meliksetian, K., Allenberg, A., Pernicka, E., Hovanissyan, A., and Kunze, R., 2013. Neue Quellen für altes Gold? In: H. Meller and P. Avetisyan, eds. 2013. *Archäologie in Armenien II. Veröffentlichungen des Landesamtes für Denkmalpflege und Archäologie Sachsen-Anhalt-Landesmuseum für Vorgeschichte*, 67. Halle: Sachsen-Anhalt-Landesmuseum. pp.27-48.