Journal of Archaeological Science: Reports 52 (2023) 104267

Contents lists available at ScienceDirect

Journal of Archaeological Science: Reports

journal homepage: www.elsevier.com/locate/jasrep

Prehistoric gold from Lake Sevan Basin? New research on Armenian gold deposits and objects

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A B S T R A C T

The article gives an overview of the results of a joint Armenian-German project in the area of today's open pit gold mine of Sotk in Eastern Armenia. Special attention is given to the preliminary results of the gold analysis. Interdisciplinary studies on regional natural gold deposits - especially the Sotk and Tsarasar deposits - and their archaeological context are presented together with the analytical results of 43 gold and four silver objects from five different sites in Armenia. The LA-ICP-MS data are discussed in terms of their alloy and trace element composition. They provide information on the use of raw material sources, intentional alloying, and the preferred use of these alloys. The archaeometric investigations show that the gold objects were mostly made of secondary placer gold. However, for one object, the oldest object examined in this study, the use of primary rock gold could not be excluded. Furthermore, the data provide information on the serial production of typologically identical types found at different sites, indicating that these objects were probably traded over long distances.

1. Introduction

Due to its geographical and cultural proximity to the Near Eastern world, the area south of the Great Caucasus Mountain Range is a little noticed but all the more important region for early metallurgy and the related material and immaterial exchange between the Caucasus and the Near East (Bobokhyan et al., 2017; Kavtaradze, 2013; Meliksetian et al., 2011).

Until now, the research focus has been mostly on selective, less large-scale and mostly purely archaeological investigations. Only in recent decades have efforts been made to structure and contextualise research in the region, on the one hand, and to include the geological and geochemical framework in the research, on the other hand, in order to address the resulting archaeometallurgical questions (most recently Jansen, 2019). In particular, the South Caucasus represents one of the most important regions on the border with the Near East during the Bronze Age due to its rich raw material deposits (Bobokhyan et al., 2017: 505-508; Pizchelauri and Pizchelauri, 2002). It is therefore surprising that their importance for the beginnings of metallurgy has hardly been explored and that little information is available so far (Stöllner et al., 2010: 105-107). For the Bronze Age in Armenia (c. 3,500–1,200BCE), first studies could already provide evidence for complex societies with structured settlements, which were mostly grouped around nearby raw materials (gold, copper ore, obsidian), thus proving a direct connection (Kunze et al., 2011, 2013, 2017).

Gold as a raw material is considered to be one of the most important materials of ancient societies. Besides its less important use as a metallic material, it represents an indicator of value and prestige, which is particularly evident in Eastern Armenia on the basis of Urartian expansions (Biscione et al., 2002: 10-11; Salvini, 2008: 257; Wolf and Kunze, 2014: 112-114).

Within the framework of an Armenian-German joint project funded by the *German Research Foundation* (Project number 410373002) and the *Science Committee of the Republic of Armenia* (project number 20RF-141), this paper aims at interdisciplinary research on gold as a raw material. The focus is on the area of present-day Armenia during the Bronze Age, which is geographically and culturally closely connected with other regions of the Caucasus, eastern Turkey, Mesopotamia, and north-western Iran and reflects cultural developments without which the significance of gold cannot be understood.

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https://doi.org/10.1016/j.jasrep.2023.104267
Received 24 July 2023; Received in revised form 6 October 2023; Accepted 19 October 2023
Available online 4 November 2023

2. Geographical and geological conditions

In the Near East and the Black Sea region, gold has been used regularly since the Bronze Age. In both regions, this precious metal was in great demand and played a prominent role in the value systems (Avilova et al., 1999: 57; Picchelauri, 1997: 6). According to the current state of research, however, a paradoxical situation arises for the Caucasian cultural area: While many gold artefacts from the Early Bronze Age (Maikop culture) are known in the North Caucasus, only a few gold objects from the Middle Bronze Age (North Caucasian culture) have been documented so far. In the South Caucasus, on the other hand, only a few gold artefacts from the Early Bronze Age Kura Araxes horizon have been found (Palmieri and Di Nocera, 2000: 180; Frangipane, 2004; Khanzadyan in: Kalantaryan, 2007: 72 pl. 11.5; Simonyan, 2020). However, their share increases sharply with the transition phase (Kurgan or Martkopi-Bedeni horizon) and with the Middle Bronze Age (Trialeti-Vanadzor horizon) (Gambaschidze et al., 2001: 103; Khanzadyan in: Kalantaryan, 2007: 72; Bobokhyan, 2008: 76). During the Late Bronze Age and Early Iron Age, gold is known and widely used in the whole Caucasus. This fact is in apparent contradiction with the Early Bronze Age gold mining at Sakdrisi (Kachagiani) in southeastern Georgia, near the Armenian border (e.g. Jansen, 2019; Gambashidze and Stöllner, 2016; Stöllner et al., 2010). Presumably, this is due to the fact that gold was also mined in the Early Bronze Age South Caucasus, but it served primarily as a trade commodity rather than a personal luxury item.

The special interest of the Near East in the Early Bronze Age cultures of the Caucasus (Munchaev, 1975: 406) has been linked by several scholars to the gold-rich regions of the South Caucasus and a possible important role for the export of the metal to the Near East (Klima, 1964: 146; Petzel, 1987: 13; Chernykh, 1992: 151; Gevorgyan and Zalibekyan in: Kalantaryan, 2007: 25) and the active trade between Assur and Kaneš (e.g. Avetisyan in: Kalantaryan, 2007, 51). Despite the lack so far of clear evidence for the origin of gold from ancient Near Eastern civilisations, such as the Sumerians with the famous royal tombs of Ur from the 3rd millennium B.C. (Jansen, 2019; Schmidt, 2005: 60; Moesta, 1986: 104-105), some Ur III and ancient Assyrian cuneiform sources suggest an origin of part of the Sumerian gold from the South Caucasian region (Maxwell-Hyslop, 1977: 83; Schmidt, 2005: 60).

Mesopotamian societies or artisans imported unprocessed natural gold in the form of 'gold dust' (Borger, 1956: 84, 88; Limet, 1960: 90) from the lands of Ḫarali[1] and Tukriš in the 3rd millennium B.C., acting as middlemen.[2] Despite controversial discussions about the location of Ḫarali,[3] F. Haldar suggests that the gold-rich area is located in the "[…] mountainous regions of Armenia" (Haldar, 1971: 73).

The so-called 'Ushkiani gold project', which has already provided evidence of a rich natural concentration of gold in eastern Armenia (Wolf et al., 2011; Wolf and Kunze, 2014), confirms the importance of the Caucasus region for prehistoric gold mining based on historical and archaeological sources (de Morgan, 1889: 19; Iessen, 1935: 32, 63; Melkumyan, 1972: 119; Madatyan, 1987: 91; Gevorgyan and Zalibekyan in: Kalantaryan, 2007: 17-21).

The eastern region of Gegharkunik (Fig. 1) represents a natural area clearly bounded by mountain ranges to the north and south and Lake Sevan to the west. It is also a geographically important area: the Sotk Pass in the east is the only direct link between the southern and eastern Caucasus, and its strategic importance is evidenced by numerous archaeological sites, some of which have not yet been adequately explored.

However, it is not only the geographical location that makes the region so important from an archaeometallurgical point of view, but above all its proximity to the gold mine, which was known at least in antiquity and the Middle Ages (Eremyan, 1963: 80; Wolf et al., 2013: 38). It is located about 18 km northeast of the city of Vardenis near the present-day village of Sotk at an altitude of 2,100–2,500 m above sea level (Fig. 2). The epithermal vein deposit is still considered one of the largest gold deposits in the Middle East,[4] whose genesis and ore mineralisation (Wolf and Kunze, 2014: 119-121) as well as geomorphology (Henniges, 2016) have been extensively studied. Based on a geological mapping carried out in 1953, there were numerous indications of pre-modern mining activities in the area of today's opencast mine, which have now been destroyed by modern mining (Wolf and Kunze, 2014: 119, Fig. 9). Among others, more than 140 pits were concentrated mainly along the southern and eastern parts of the gold-bearing quartz/carbonate veins. According to S. E. Goginyan and S. A. Esayan, gold was mined from alluvial and diluvial deposits in the entire area during the Bronze Age (Goginyan, 2005:34-35, 75, 96–99; Esayan, 1976: 190-192), as evidenced by the remains of wooden tools and stone mortars (Madatyan, 1987: 93-101). Investigations at the Ushkiani Project have already demonstrated the presence of an unusually gold-rich mining district by the presence of rich gold placers in several anthropogenically unregulated rivers.

A comparison with the Sakdrisi deposit in southeastern Georgia is obvious at this point (cf. Fig. 2), as the distance between the two deposits is only about 180 km. The Sotk and Sakdrisi gold deposits differ only slightly in their geological structure. The formation of the veins, alteration of the surrounding rocks, vein type, mineral associations, and zoning of gold concentration with depth are very similar. Only the size of the two deposits differs significantly, and the gold concentrations of the near-surface mineralised zones differ significantly. The Sotk deposit is the largest gold deposit in the entire Caucasus with an estimated deposit content of 124 tons of gold (Gugushvili, 2010) and (current) gold concentrations averaging 2–4 g/t, but also > 10 g/t and in some deposit areas (gossan structures) individual samples up to > 300 g/t (Elevatorski, 1982: 141). For Sakdrisi, an original deposit content of about 23.5 t gold is assumed, with average gold concentrations of about 1 g/t and > 10 g/t in individual samples (one outlier sample at 50 g/t) (Gugushvili et al., 2002; Hauptmann et al., 2010: 146 Table 1).

3. Brief overview of the archaeological results

The archaeology of the eastern shore of Lake Sevan is still poorly explored, although the region was visited by various scientists and travelers as early as the mid-19th century. These include Gh. Alishan (1893) and E. Lalayan (1931), who mentioned the first sites. In the early 2000 s, an Armenian-Italian expedition led by R. Biscione and S. Hmayakyan conducted a survey of the wider area of the southern Lake Sevan (Biscione et al., 2002), the results of which, together with the sparse written documentation of earlier smaller surveys, served as the basis for our own previous preliminary work on the archaeological

[1] It is the Sumerian name. The Akkadian name is Arallû. Both names denote a so-called 'gold country' or the name of a gold-rich mountain (Reiner, 1956: 147) and qualify the gold as 'collected gold' (Akkadian guškin-arali = liqtu; cf. Komoròczy, 1972: 117, who erroneously sees it as mountain gold).

[2] "The land of Tukriŝ the gold of Harali (and) the [.......] Lazur stone shall deliver unto thee." (Epic 'Enki and Ninhursag', see Komoròczy, 1972: 113). The 'mediator', the land of Tukriŝ, cannot be clearly identified. The respective interpretations of the Hammurabi inscription at Ur and an Assyrian geographical treatise (KAV 92) suggest a location either in the eastern Iranian highlands (Komoròczy, 1972: 114) or in present-day north-western Iran (Moorey, 1995: 444).

[3] Komoròczy suspects that the goods lapis lazuli (from Afghanistan) was delivered to Tukriŝ along with the natural gold, and that Harali must have been on the way from Afghanistan to the Iranian highlands in the north-east of Iran (Komoròczy, 1972: 114).

[4] The current open pit mine is operated by GeoProMining (GPM) with an average annual production of approximately 3.5 tonnes of gold (in 2018). Cf. https://minerals.usgs.gov/minerals/pubs/country/2013/myb3-2013-am.pdf (04.10.2023).

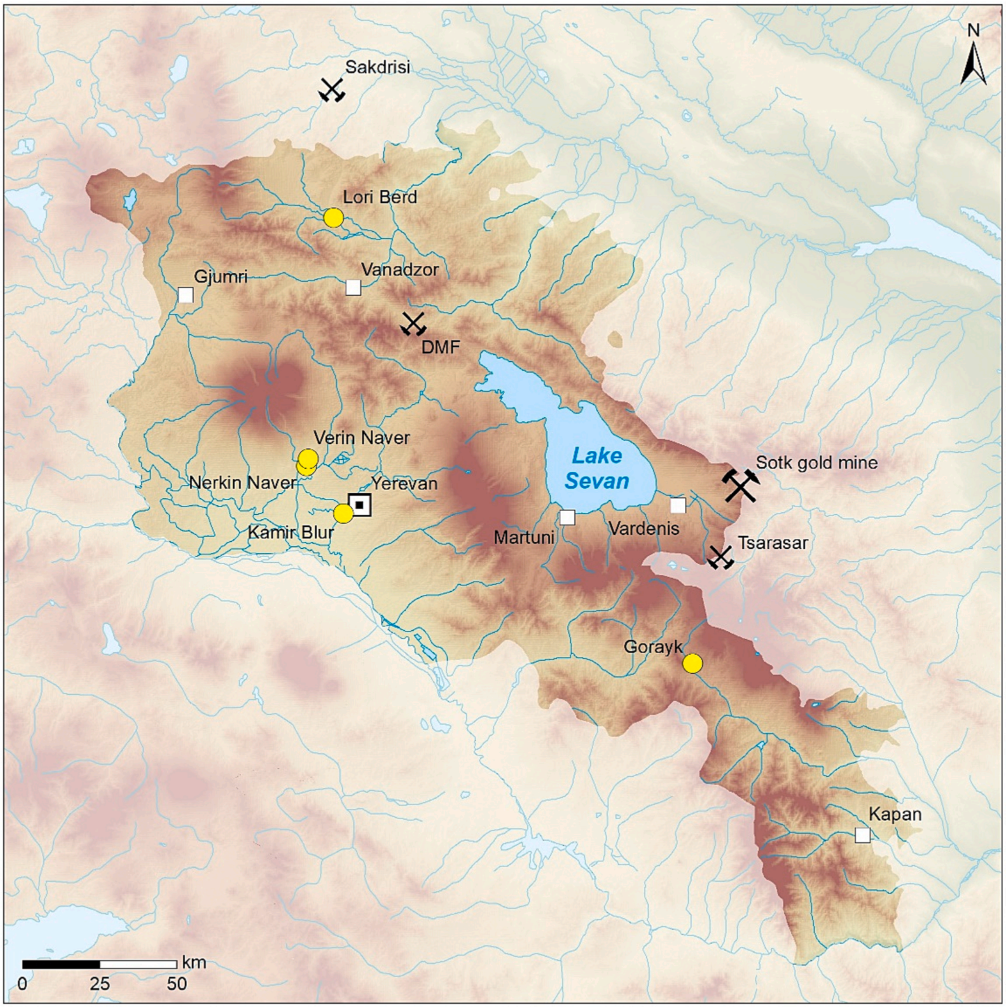

Fig. 1. Location of Armenia with gold deposits discussed in the text (crossed hammers) and locations of analysed gold objects (yellow circles) (Basemap: SRTM ©USGS/NASA; A. Swieder, Halle/Saale). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

investigation of the region as part of the Armenian-German Ushkiani Project (Kunze et al., 2011, 2013; Bobokhyan et al., 2017; Bobokhyan and Kunze, 2021) which was initiated and funded by the State Office for Heritage Management and Archaeology Saxony-Anhalt (Halle/Saale) and the Institute of Archaeology and Ethnography of the Armenian Academy of Sciences (Yerevan).

In the topographically clearly delimited natural area of 'Sotk' (eastern part of the province of Gegharkunik), numerous new sites and were discovered and the knowledge of general patterns of use in the area of the gold deposit and their interrelationships was expanded. To this end, a total of 44 sites were surveyed in the region around the present-day open pit gold mine (Bobokhyan et al., 2017: 510-514; Wolf and Kunze, 2014: 114-116). These are mainly fortified settlements on naturally rounded hills with steep slopes, fortresses on natural rocks with cyclopean ramparts, ground-level settlements, sites with scattered pottery, and cemeteries.

The work carried out so far proves that the region has been inhabited since the Early Bronze Age (no Stone Age sites have been documented in the entire Lake Sevan area). The Early, Middle and Late Bronze Age are known from graves and settlements (Kunze et al., 2011: 33). The Early and Middle Iron Age are generally the most thoroughly studied periods, where the symbiosis between local and Urartian cultures is clearly visible.

The results of the large-scale survey of the research area already allowed the conclusion that the newly explored Bronze Age sites can be classified as an archaeologically closed settlement system grouped in the direction of and around the gold deposit, thus revealing a direct connection. According to Simonyan et al. the densely clustered cyclopean fortresses on the northern slopes of the Geghama Mountains, facing Lake Sevan, were most likely designed to protect the trade route that ran along the southern shore of Lake Sevan (Simonyan et al., 2022: 413-417).

Systematic archaeological investigations were also conducted at two selected sites. The detailed investigation of the sites Sotk 2 (settlement) and Norabak 1 (settlement/cemetery) yielded numerous conclusions for a first understanding of the Bronze Age in the study area (Kunze et al., 2013). At Sotk 2 - a natural mound east of the present-day village of Sotk on the road to the gold mine - occupation layers from all Bronze Age periods were excavated and documented in 2011–2015 (Kunze et al., 2013). It is a multilayered settlement that was inhabited with interruptions from the Early Bronze Age to the Early Iron Age. The Early Bronze Age with the Kura-Araxes culture is represented in the area with disturbances by later settlements. Clay soils and rock pits have been documented here. The rock pits must have had an economic function. Obsidian material, ceramic fragments and archaeobiological material were found here in large quantities. In this context, it is interesting to note that obsidian, like gold, was a widespread commodity in prehistoric times, and thus ancient trade routes and contacts can be sketched on the basis of its distribution (Kunze et al., 2011: 34-35).

The test excavations of the triangular natural promontory Norabak 1 - located about 6 km south of Sotk 2 - showed that it was intermittently inhabited from the Early Bronze Age to the High Middle Ages. The Early

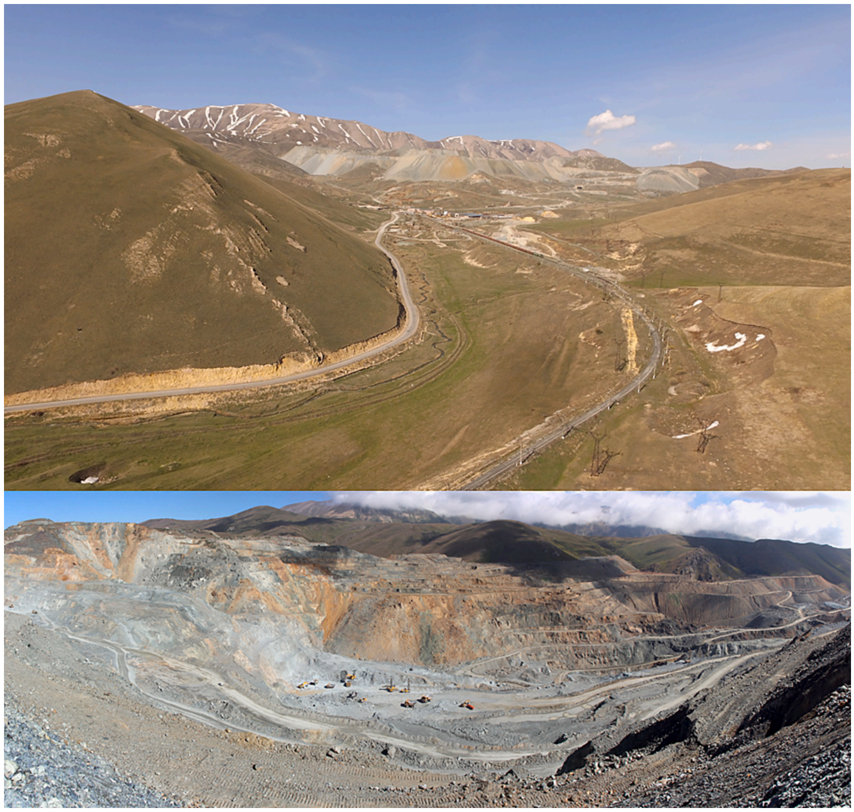

Fig. 2. The Sotk mining area (above) and the interior of todaýs open pit mine (below) (Ushkiani-Team Archive).

Bronze Age is represented here by clay architecture, the pottery collection repeats completely the collection of Sotk 2. An excavation of one of the numerous mounds (kurgans) south of the settlement served primarily to clarify dating issues that could be placed chronologically in the Late Middle Bronze Age – the period of the widespread use of gold in the Near East (Kunze et al., 2013: 61-67).

Despite the lack of direct evidence so far, it can be assumed that the Bronze Age population of this settlement cluster benefited from its strategically favorable location and the metal, ore, and obsidian deposits. For example, obsidian artefacts from the Sotk 2 and Norabak 1 sites, analysed by instrumental neutron activation analysis (INAA), showed a regional origin of the raw material used from the Geghama Mountains south of Lake Sevan (Spitaksar and/or Geghasar volcanoes) and the Syunik Highlands (presumably Sevkar) (Kunze et al., 2013: 76-79). The fact that it must indeed have been part of a regional and supra-regional network is shown, among other things, by several finds of polychrome painted pottery from the Middle Bronze Age layers of Sotk 2 (Bobokhyan and Kunze, 2021). This characteristic pottery (so-called Urmia ware) has counterparts in northwestern Iran (Rubinson, 1976: 235; Boehmer and Kossack, 2000: 10-12; Piller, 2004).

4. Methodology

4.1. Methodological approach

In order to obtain information on the use of raw material sources, on the intended alloys and on the preferred use of these alloys, an integrative evaluation of research results from different disciplines is necessary. For this reason, the results of archaeological and mineralogical prospections, geochemical and archaeometallurgical analyses, as well as the chronological and typological processing of the investigated material will be integratively evaluated within the framework of the project.

We studied the rock and placer gold of the region geochemically (Wolf and Kunze, 2014), then using archaeological (surveys, excavations) and geographical (GIS) analyses to place it within the system of regional and interregional communication.

In fact, we were fully aware that the production and exchange of gold could only be possible in a network provided with an appropriate social system, which could be reflected in the disposition of archaeological sites and studied through the study of unearthed materials and their analysis by scientific methods. The expected result of the project could be a reconstruction of the global significance of the gold 'phenomenon' and its value, as well as an understanding of the process of production, distribution and exchange of this metal within a defined social system.

4.2. Analysis methods

The chemical composition of major, minor and trace elements was determined at the Curt-Engelhorn-Center for Archaeometry (Mannheim, Germany) using mass spectrometry in conjunction with a laser-based sampling unit, the so-called Laser Ablation - Inductively Coupled Plasma Mass Spectrometry (LA-ICP-MS). The samples were previously embedded in epoxy at the Institute of Geosciences and Geography, Department of Economic Geology and Petrology, at the Martin-Luther-University Halle-Wittenberg (Germany) and then metallographically ground and polished. On each sample, material was usually ablated by point analyses < 100 µm Ø. The ablated sample aerosol transported to the mass spectrometer was ionised in the plasma and the ions were subsequently separated and detected in the analyser according to their mass-to-charge ratio. The major element isotopes ^{63}Cu, ^{107}Ag, and ^{197}Au, the platinum group elements (PGE) ^{101}Ru, ^{103}Rh, ^{105}Pd, ^{189}Os, ^{193}Ir, and ^{195}Pt, and the trace elements ^{48}Ti, ^{52}Cr, ^{55}Mn, ^{56}Fe, ^{59}Co, ^{60}Ni, ^{66}Zn, ^{75}As, ^{82}Se, ^{111}Cd, ^{118}Sn, ^{121}Sb, ^{125}Te, ^{208}Pb, ^{209}Bi, and ^{202}Hg were measured.[5]

Six samples from Lori Berd were also examined metallographically using a light microscope and a scanning electron microscope with attached electron beam microanalysis with an energy dispersive X-ray spectrometer.

The primary goal of the chemical analyses is to gain insight into possible chronological changes in gold processing and possible alloying techniques based on a comprehensive material characterisation. Difficulties in the geochemical determination of the origin of gold objects have been discussed in detail by several authors in recent years (most recently Standish et al., 2021) and will not be repeated here, as the possible utilisation of the regional gold deposits described above is considered in this paper, but does not represent a main focus. Furthermore, it must be assumed that precious metals in particular tend to have a very high recycling rate. These aspects were - as with other metals - discussed as unlikely in early archaeometric studies on the origin of gold objects and only considered problematic for later times, for example with the advent of monetarisation. However, this perspective has changed significantly in recent years (for a discussion of different metals, cf: Bray, 2022; Kershaw and Merkel, 2022; Guerra, 2021).

5. Materials and results

5.1. Placer gold

The prehistoric settlement network along an important direct trade route between the southern and eastern Caucasus, presented in chapters 2 and 3, provides numerous indications of a direct link with the extraction and trade of gold. To illustrate this close relationship, the gold content of the running water, alluvium, eluvium and mountain gold veins has been recorded and documented over a wide area. At 76 washing sites, a total of 43 rivers and streams were examined for gold conduction in order to record the spatial distribution of gold placers (for the methodology used, see Wolf and Kunze, 2014: 116).

Placer gold was detected in 25 rivers and streams, in some cases in considerable quantities, and gold concentrations could also be excluded in 18 small rivers and streams (Fig. 3**, a**; Wolf et al., 2013; Wolf and Kunze, 2014).

An important observation of the gold panning experiments was that in gold-bearing rivers, considerable amounts of gold can be recovered in a short time (Fig. 4**, a**), even with the simplest means of pan and channel. This is particular true of the Sotk, Tartar and Masrik Rivers. It should be noted that there has been no anthropogenic regulation of gold-bearing streams and rivers in this region. The status quo is therefore comparable to the situation in the pre-Christian Bronze Age and Iron Age. The natural sediment traps of the gold placers thus continue to receive an unimpeded supply of gold particles from the primary mountain gold deposits. Furthermore, the comparison of the natural gold from the rivers with the primary gold ores showed no geochemical changes during eluvial, alluvial and fluvial transport (Wolf and Kunze, 2014: 128-129).

A total of 133 gold grains were chemically analysed by laser ablation ICP-MS, the results of which were published in Wolf and Kunze, 2014 and are summarised in **Supplementary Material S1**. In addition, a second gold occurrence, southwest of the Sotk deposit, was geochemically characterised for the first time in the small-scale area, in addition to the already known Sotk deposit. This deposit was finally named Tsarasar (Wolf et al., 2013: 27) in reference to a local ridge and volcano (2,500–3,200 m a.s.l.) (**cf.** Fig. 3**, b**). As a result of these investigations, it can be noted that the placer gold of the Sotk and Masrik rivers and their respective tributaries are geochemically distinct (Wolf et al., 2013: 37 Figs. 12-14). The secondary ('placer') gold from the Tsarasar deposit is significantly richer in silver and lower in copper than that from the Sotk deposit (Kunze and Wolf, 2014: 124–125), so that the silver content, with the exception of one micronugget (PA-24), is 21–38 wt-% Ag. In contrast, only one nugget from the Sotk district (PA-28) had a higher silver content, while all other grains contained between 4 and 14 wt-% silver. However, statistically different populations cannot be proven for either Sotk or Tsarasar on the basis of single outliers such as samples PA-24 or PA-28 (Chapman 2021: 1564). Possible geological correlations for the different chemistry have already been discussed in Wolf and Kunze, 2014: 121, 129–130.

The examination of a free gold sample (primary mountain gold) directly from the current open pit of the Sotk gold deposit showed a good correlation with the placer gold grains from the Sotk area in terms of silver and copper contents as well as many trace elements (Wolf and Kunze, 2014: 125 Fig. 16). This can be interpreted as a first indication that little or no secondary alteration has occurred as a result of river transport.

In addition to the major and minor elements silver and copper, the gold grains were analysed for trace elements, i.e. platinum group elements (PGEs), titanium, chromium, manganese, iron, cobalt, nickel, zinc, tin, selenium, cadmium, tellurium, bismuth, antimony, lead and arsenic. PGEs were detected at very low concentrations in 7 of the 133 gold grains analysed. Three samples from the Sotk district (wash sites S08 + S11) were low in platinum (0.5–5 mg/kg), four others from the Tsarasar district were low in palladium (2 grains from wash site PA25; 0.7–1.0 mg/kg) and platinum (2 grains from wash site AA05; both 0.3 mg/kg). Iron was detected as a trace element in about half of the gold flakes, mostly in the range of less than 1000 mg/kg, but in exceptional cases significantly higher up to contents of almost 190,000 mg/kg (S08). At least contents above 1000 mg/kg indicate that iron oxides/hydroxides have also been deposited on the nugget surface or in small pores of the grains, i.e. must be considered as 'contamination'. The elements arsenic, chromium, titanium, selenium, cadmium and lead were not included in the comparisons as they could only be detected analytically in extremely isolated cases. In particular, some elevated lead values are attributed to submicroscopic ore mineral inclusions (Wolf and Kunze, 2014: 125). A comparison of the respective placer gold deposits with regard to the trace elements bismuth, cobalt, nickel, antimony, tin, tellurium and zinc again shows specific differences. The gold of the Tsarasar district differs from that of the Sotk district in four elements in particular. Characteristic for the Tsarasar gold placers is the 'absence' (i. e. below the detection limit) of cobalt and nickel. Tin, in turn, may be a marker for Tsarasar gold. Compared to the Tsarasar gold, the Sotk gold is mainly characterised by bismuth and antimony contents that are several orders of magnitude higher. It is not possible to distinguish between the trace elements tellurium and zinc, but both elements are remarkably abundant in the alloys of both deposits.[6]

In summary, it can be said that the geological conditions of the gold deposits, in addition to the gold richness of the watercourses, favour Bronze Age gold mining in the area, which is therefore highly probable. In addition to the numerous gold flakes in the watercourses of the surrounding area, pits and galleries are of particular interest from a mining

[5] The relative standard error is about 1% for silver, about 10% for copper and gold, and about 40% for the other elements. The isotopes of Ru, Os, Rh, Ir and Hg were measured semi-quantitatively.

[6] The gold flakes assays from 2011 to 2013 were supplemented by additional placer gold samples in subsequent years, resulting in some changes to the trace element comparisons from Wolf and Kunze (2014). From a methodological point of view, the mean values of the replicate analyses, adjusted for the results of 'missed measurements', e.g. due to contamination, are now included in the analysis. Gold flakes from sample S04 from a stream west of the Masrik stream were also attributed to the Tsarasar occurrence, but showed significant differences in composition (Wolf and Kunze, 2014). A new field survey in 2017 showed that the stream does not drain the Tsarasar (volcano), but the north side of the Ayrk volcano to the west, and therefore cannot be assigned to the Tsarasar deposit.

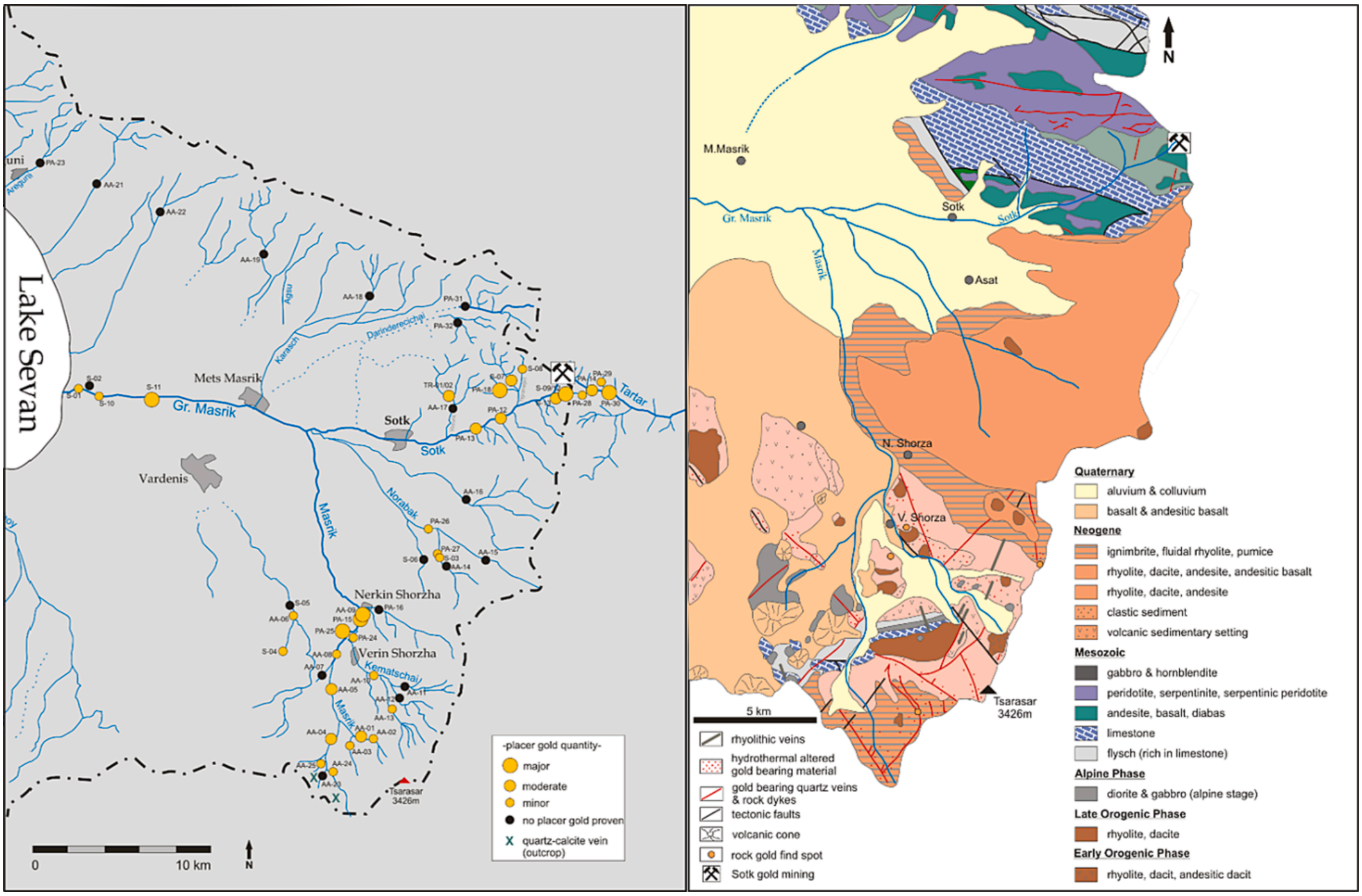

Fig. 3. a) Distribution and relative abundance of placer gold in the study area. The quantities are based on a subjective quantification depending on the amount of washed raw gravel (modified from Wolf and Kunze, 2014). b) Graphical representation of an unpublished geological map of the Sotk region and the prospective gold-bearing areas of the Tsarasar gold occurrence (Geological Survey of the USSR, Geological Report on the Sotk Region 1953: 73; redrawing D. Wolf).

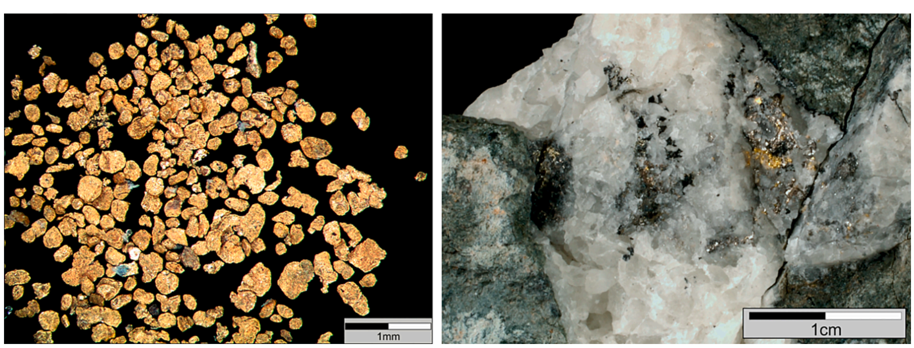

Fig. 4. a) Clearly rolled out placer gold nuggets from the Masrik River (from Wolf et al., 2013: Fig. 5). b) Mountain gold with gangue quartz from the Sotk gold deposit (from Wolf and Kunze, 2014: Fig. 6).

archaeological point of view for early gold mining in the South Caucasus. The construction, structure and nature of the mineralisation also support early mountain gold mining. The strongly quartzified, sulphidic veins striking the surface show macroscopically visible gold due to the weathering of the sulphidic base minerals, thus allowing not only placer gold extraction but also early pit mining (Wolf and Kunze, 2014). It should therefore have been relatively easy in prehistoric times to find easily extractable or even macroscopically visible free gold in the outcropping quartz veins (Fig. 4, **b**). There are also a large number of conspicuous geomorphological negative forms on the surrounding hillsides, which are interpreted as weathered former pits.

As the example of Sakdrisi shows, pits and tunnels initially targeted near-surface oxidised supergene rich zones (Hauptmann et al., 2010; Stöllner et al., 2010). In these areas, the gold had weathered from the originally sulphidic base minerals to form micronuggets of native gold or electrum, and due to its high density, could be extracted by simple mechanical means and relatively easily processed pyrometallurgically and mechanically (Wolf et al., 2013: 39). Mining innovations probably led to the repeated use of mines and tunnels during different mining periods, i.e. existing galleries and pits were widened, dumps were opened up and mining areas were extended laterally. All this meant that older traces of mining were usually destroyed or, more rarely, preserved by backfilling. The gold-bearing veins exposed by the erosion of the adjacent rock, often with dumps in front of them, are evidence that, at least in the early days of gold mining in this region, it was not necessary to mine primary gold at depth, and the metal was easily extracted. Mining close to the surface, and sometimes underground, was facilitated by the fact that some of the hard gold quartz veins and lodes were surrounded by highly altered and conspicuously discoloured rock. This rock material, some of which is highly fractured, soft and easily identifiable by colour, facilitates underground tunnelling and the identification of prospective areas. Several at least pre-modern adit systems in the north of the study area are indicative of such underground mining (Fig. 5).

5.2. Gold and silver objects

5.2.1. Samples of gold artefacts

During the first phase of the project, 43 gold objects were examined (Fig. 6 and **Supplementary Material S2**); 16 from the cemetery of Lori Berd, 15 from Verin Naver and six from the neighbouring site of Nerkin Naver, five from Karmir Blur and one object from Gorayk (**cf. map in** Fig. 1).

The Lori Berd site in northern Armenia is one of the most important Bronze and Iron Age sites in Armenia. It consists of a large necropolis and a settlement that has been little studied (Davtyan et al, forthcoming). Excavations at the cemetery were carried out from 1969 to 2012 by the Institute of Archaeology and Ethnography of the National Academy of Sciences of the Republic of Armenia under the direction of Dr. S. Devejian and have been extensively published (Devejian, 1981; 2006; 2022). A total of 117 graves were excavated at Lori Berd, a large number of which date to the Late Bronze Age (16th-12th centuries B.C.) and the Early Iron Age (12th-9th centuries B.C.). A further 26 graves date from the Middle Bronze Age (22nd-16th century BCE). Of particular importance are more than 30 burials dating from between 1000 and 550BCE, some of which correspond to the period of expansion of the Urartian Empire. A further five tombs date from the Achaemenid period (6th-4th century BCE). In total, about 30 % of the graves examined so far contain gold objects in varying numbers. The 16 gold artefacts examined from Lori Berd consist mainly of small beads, buttons or plates (n = 10). A Neo-Assyrian cylinder seal with gold caps from the 8th/7th century BCE (#3; tomb no. 106, Archaemenid period), a quadruple spiral pendant (#15; tomb no. 3, Late Bronze Age context),[7] an iron pick with a central shaft hole decorated with gold leaf applications (#17; tomb no. 56, Early Iron Age), two gold diadems (#1, #10; tomb no. 2 resp. no. 4; Late Bronze Age or Urartian Period resp. Urartian Period), a gold bracelet with snake head finials (#16; tomb no. 4, Urartian Period) and a pin head with embedded carnelians (#18; tomb no. 114, Urartian Period cenotaph) also point to objects from relatively richly furnished graves dating from the 8th to 6th century B.C.[8] Gold caps on seals have been well documented in Mesopotamia since the Kassite period, and their tradition was continued in the Neo-Assyrian or Neo-Babylonian empires (Collon, 1987: 61, 77, 83). Seal #3 is thought to have been imported into Armenia only during the Achaemenid period (Devedjian and Davtyan, 2021: 225–226). Despite its clear foreign origin, the gold values are consistent with other gold finds at Lori Berd. Stylistically, pinhead #18 stands out from the other Urartian gold jewellery from Lori Berd because of its carnelian inlays (Devedjian in: Kalantaryan, 2007: 135-150). However, despite the dating of the cenotaph in the Urartian context, numerous parallels are known from various monuments in the South Caucasus, which are dated to the Middle Bronze Age, the end of the 3rd and beginning of the 2nd millennium B.C. (Devedjian, 2006, pl. 1–2).

Since 1976, and later since 2002, H. Simonyan, as a representative of the Scientific Research Center for Historical and Cultural Heritage, has been carrying out continuous excavations at the burial sites of Verin and Nerkin Naver. Both sites are located about 25 km northwest of Yerevan in the Aragats region and contain about 500 burial mounds (Simonyan, 2021a). At Verin Naver, five large kurgans were excavated during road construction in 2011–2013 under the programme of the State Scientific Committee for Science. Two burials were found in kurgan No. 1, which measures 50 m in diameter and was mounded about 2 m high. Tomb 1:A (calibrated ^{14}C date 1540–1430BCE)[9] was located in the southern area and Tomb 1:B (calibrated ^{14}C date 1533–1439BCE [2σ; DeA-7033; charcoal]) in the central area (Simonyan 2021a: 100). The 14 investigated gold objects come from Tomb 1:A and are present as solid, perforated jewellery discs (#25–32) as well as tubular (#20–21) and spiral beads (#22–24) or as quadruple spiral pendants (#19). In the 1990 s a number of smaller graves were investigated, including tomb no. 52 – dating from the final phase of the Middle Bronze Age in the 1st half of the 2nd millennium B.C., the so-called Karmir Berdian Culture (Simonyan, 2006) - from which another analysed bead of thin gold sheet (#33) was recovered. Nerkin Naver is located in the immediate vicinity (about 2 km south) of Verin Naver. Numerous kurgan burials from the Middle Bronze Age 'Early Kurgan Culture' have been excavated here (Simonyan and Manaseryan, 2013). In most of the graves investigated, in addition to a human burial with a ritual of cremation of the corpse, a horse burial was also found with numerous grave goods, such as vessels, flint blades and obsidian arrowheads, which were given to the buried person. Of the six gold objects analysed, a hollow bead with hump ornaments from tomb no. 1 (#41; calibrated ^{14}C dates from two animal grave goods: 2136–1960BCE [2σ; MAMS 52758; horse bone] and 2013–1783BCE [2σ; MAMS 52759; ox bone]), a perforated sheet gold disc from tomb no. 9[10] (#34) and four objects from tomb no. 10 (head piece of an amulet pendant and three fragments of further hollow beads with hump ornaments (#42–45; calibrated ^{14}C date of the tomb: 2133–1954BCE [2σ; MAMS 52762; human tooth]).

The site of Karmir Blur lies to the south-west of the present-day capital, Yerevan. Urartian inscriptions testify that the settlement was founded around 680BCE by Rusa II as Teišebai URU. B. Piotrovskii explored the associated palace and parts of the huge lower town surrounding the palace from 1939 onwards (Piotrovskii, 1969; Salvini, 1995: 103-106, 128). Subsequent excavations of the site by H. Simonyan allowed for the additional investigation of several graves, which can be placed in Early Iron Age contexts[11] and contained several gold objects. Five objects were sampled and analysed: Two gold sheet beads (#37–38) from grave no. 12 (calibrated ^{14}C date 1105–925BCE [2σ; MAMS 52764; bone]) and one artefact each from graves no. 9 (sheet gold bead #36;

[7] Tomb no. 3 contains a Late Bronze Age burial (15th/14th century BCE) as well as a secondary burial from the Urartian period (8th century BCE). Due to the typological similarity of the quadruple spiral bead (#15) to an object from Tomb I:A of Verin Naver (#19) with a clearly dated context, it can be assumed that it belongs to the first burial (Late Bronze Age, ca. 15th century BCE). This form of quadruple spiral rings is found not only in the South Caucasus (further examples from Karashamb, Lchashen and Tomb 94 of Lori Berd), but also in the entire Near Eastern region (mapping of finds in Simonian, 2014: 224–225 and Numrich et al., 2023: 9).

[8] All the artefacts come from the National Museum in Yerevan, although some were sampled several years ago by the sités excavator, S. Devedjan (Yerevan).

[9] The laboratory number is unknown, as well as the material under investigated and the standard deviation used. H. Simonyan only refers to 'Project I/1166′ and the laboratory HEKAL AMS, Debrecen (Simonyan, 2021a: 100).

[10] Tomb no. 9 was archaeologically investigated in 2010 and 2011. The dating (^{14}C date 1886–1730BCE [2σ; GrA 54141; human tooth]; Simonyan and Mansaryan, 2013: Pl 1) is viewed critically by the excavator (Simonyan and Manaseryan, 2013: 192). On the basis of the finds, he dates the grave to the end of the 3rd millennium B.C. and thus to the same period as the other burials. Recently analysed material from the immediately adjacent grave no. 10 also gives an earlier date (calibrated ^{14}C date 2133–1954BCE [2σ; MAMS 52762; human bones]), so Simonyańs classification seems possible. Other investigated tombs from Nerkin Naver, such as tomb no. 3, also show comparable dates (calibrated ^{14}C date 2126–1938BCE [2σ; MAMS 52760: cattle bones]).

[11] H. Simonyan (pers. comm.) dates the burials to the 7th century BCE (685–585BCE).

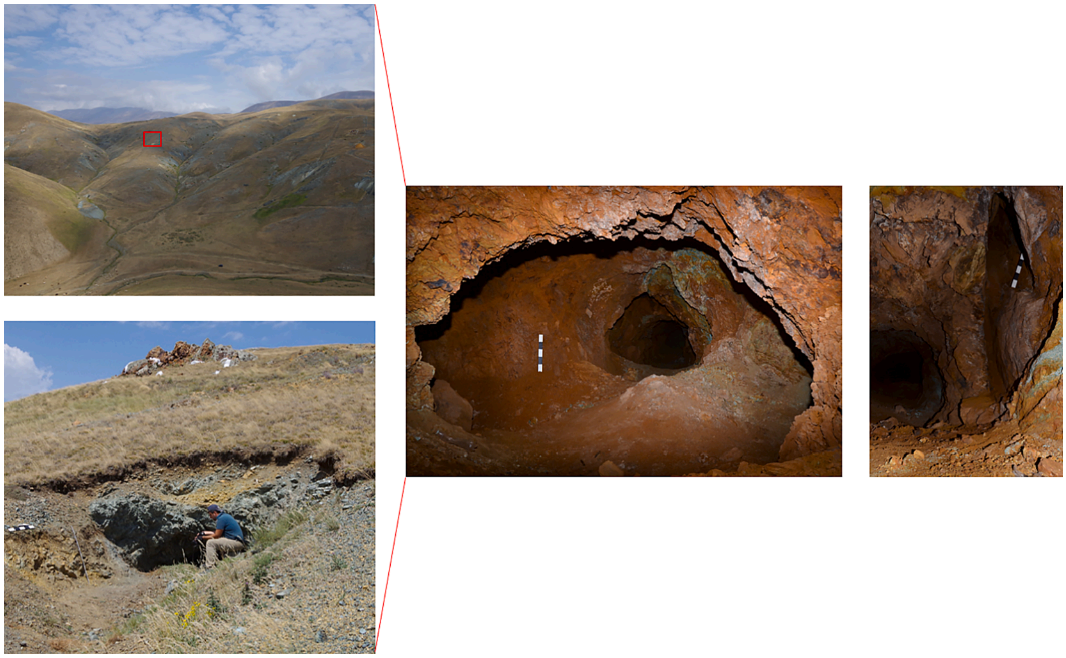

Fig. 5. Traces of ancient mining to the north of the village of Sotk, with cut-outs of gold-quartz veins (Ushkiani-Team Archive).

calibrated ^{14}C date of burial 1108–926BCE [2σ; MAMS 52767; bone]), and no. 240 (spiral bead #39; calibrated ^{14}C date of burial 903–816BCE [2σ; MAMS 52765; bone]) as well as no. 251 (spiral bead #40).

In 2013, H. Simonyan also began an archaeological investigation of another cemetery near the modern village of Gorayk in Syunik province (Simonyan, 2021b). So far, a large burial mound ('Big Kurgan') with an Early Bronze Age context dating to the 1st half of the 3rd millennium B. C. has been excavated in 2020–2021 (calibrated ^{14}C dates are: 2854–2501BCE/2623–2472BCE/2472–2305BCE [2σ; MAMS 49956–58]; charcoal). Among many other artefacts, a golden spiral ring was found (#35). Interestingly, the cemetery is located close to another gold deposit at Amulsar,[12] but the geochemical data of the primary gold mined is not yet known.

5.2.2. *Samples of silver objects*

Four silver objects from Lori Berd and Nerkin Naver have also been analysed, providing additional information on the precious metallurgy of both sites (see also Fig. 6 and **Supplementary Material S2**).

The items from Lori Berd are a vessel from the Urartian period (#2, tomb no. 56) and a spoon from the Archaemenid period (#8, tomb no. 109). The former is made of chased silver sheet with a wall thickness of about 1 mm (Devejian in: Kalantaryan, 2007: pl 47). The diameter is about 8.5 cm and the height is about 11 cm. It is richly decorated with figural motifs. The decoration is punched and engraved. It is reminiscent of the Mesopotamian style (Babylonia), but the workmanship is rather second-rate (Devejian in: Kalantaryan, 2007: 148). The silver spoon is about 13.5 cm long, 3.5 cm wide and about 0.5 cm thick. It is rectangular in cross-section and has geometric incised decorations on the back (Devedjyan et al., 2018: 110).

Two silver objects from Nerkin Naver were also analysed (#46–47). Both objects are elongated grooved beads recovered during the excavations in 2021. Like almost all the gold objects examined, they come from tomb no. 10, which dates to the end of the 3rd millennium or the transition to the 2nd millennium B.C. (calibrated ^{14}C date of bone material from tomb no. 10: cal BCE 2133–1954 [2σ; MAMS 52762]).

[12] Amulsar gold project: see https://www.lydianarmenia.am/eng/pages/arm/pages/amulsar/70/ (24.01.2023).

5.3. *Results*

All results are summarised in **Supplementary Material S3-S8**. Typically, archaeological gold objects are made from ternary gold-silver-copper alloys containing varying amounts of trace elements (Fig. 7). These alloys can either be of natural origin or deliberately produced by adding appropriate amounts of copper and/or silver to a stock of gold (-silver/copper alloy). According to Fig. 8, the silver content varies between 14 and 45 wt-% Ag and the copper content between 1 and 8 wt-% Cu. The majority of the examined objects were made of a so-called electrum alloy, i.e. a gold alloy with $\geq$ 25 wt-% Ag, which makes the production of Sotk placer gold (4–14 wt-% Ag) unlikely (Fig. 9). Only four gold flakes with less than 20 wt-% Ag were identified (**see** Fig. 8): The spiral ring from **Gorayk** (#35) - a single object belonging to the oldest context of this study, a hollow bead (#41) from tomb no. 1 in **Nerkin Naver** and two objects from **Lori Berd** (#17, #18). However, as it is the case with several archaeological gold objects, there is a correlation between copper and silver content, which is particularly evident in the material found at Lori Berd.

The analysed silver objects are chemically very different, while the silver vessel and the silver spoon are of high fineness, the two beads contain higher amounts of copper (16 wt-% and 24 wt-% Cu). Although the two beads are visually very similar, they are chemically very different. However, they share common characteristics: high copper and lead contents (Pb: 5600 and 6900 mg/kg respectively) and high gold and bismuth contents (Bi: 12,600 and 21,100 mg/kg respectively). The cupellation process separates most trace elements from the silver, but gold, bismuth and platinum group elements remain with the silver and are therefore theoretically indicative of the deposit. Remelting and recycling, on the other hand, accumulate these elements in the silver (Pernicka, 2014). While for the Early Bronze Age the use of native silver, e.g. in the Maikop culture, is still discussed (e.g. Hansen and Helwing, 2016: 48-49), in the developed Bronze Age and in the Iron Age the production from silver-bearing lead sulphides is the common technique of silver extraction. This could also be demonstrated by the silver for Lori Berd and Nerkin Naver. The cupellated silver is characterised by high lead and bismuth contents, which allows conclusions to be drawn about possible alloying techniques in gold metallurgy.

A total of 15 gold finds (jewellery discs and (spiral) beads) from

Fig. 6. Items analysed from Lori Berd (1–18), Verin Naver (19–33), Nerkin Naver (34, 41–47), Gorayk (35) and Karmir Blur (36–40). All items made of gold except nos. 2, 8, 46 and 47 (made of silver) (Ushkiani-Team Archive).

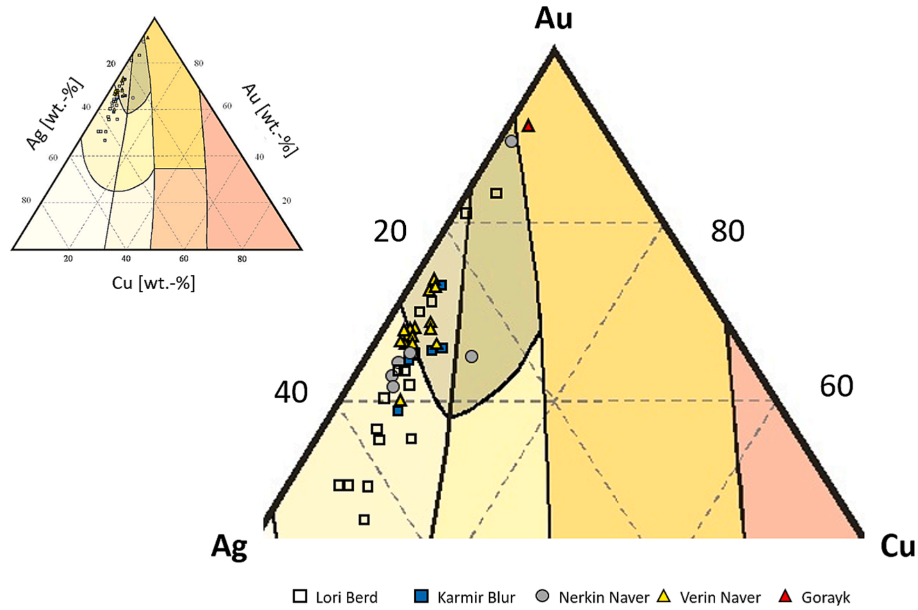

Fig. 7. Plots of gold objects in the ternary system Au-Ag-Cu to visualise the variation in colour due to the different alloy compositions.

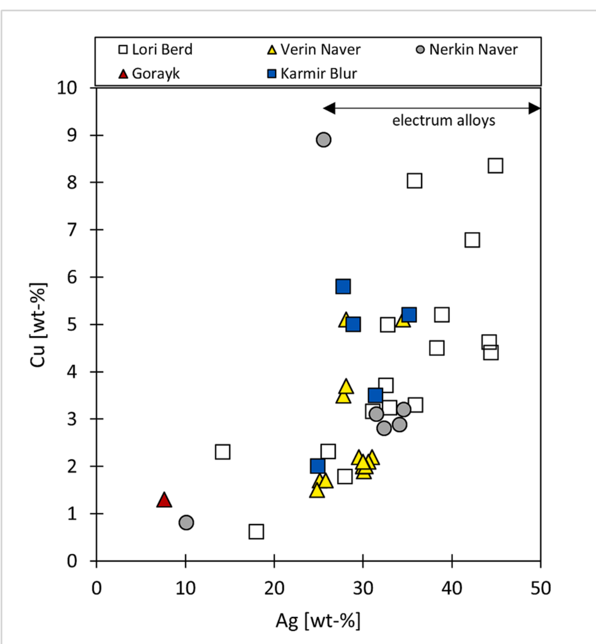

Fig. 8. Bivariate plot of silver and copper. The results of the analysed objects are shown according to their location. The black arrow indicates which objects, by definition, were made of a so-called electrum alloy.

Verin Naver were examined. Compared to the material from Lori Berd, they describe only a narrow compositional range with 25–35 wt-% Ag and 2–5 wt-% Cu and show a distinct grouping of several objects produced by technologically similar techniques (Fig. 10), i.e. the group of tubular beads (#20–21), spiral beads (#22–24) and perforated discs (#25–32). However, another elongated bead made of gold sheet (#33) from the older Tomb 52 differs from the above mentioned objects, but has similarities with a massive quadruple spiral pendant (#19) from the younger Tomb 1:A.

A comparison of the smelting-resistant trace elements shows that the analysed objects of each group are characterised by almost identical trace element patterns (Fig. 11**, a and b**). For example, all objects of within each typological bead group, the tubular beads and the spiral beads, are identical within the analytical error. Accordingly, the beads were most likely made from the same gold raw material, i.e., probably a type of ingot. However, the two types of beads differ slightly, with the tubular beads having higher Cu, Ni, Zn and As contents, but lower Pb and Bi, and systematically lower PGE contents (but the same PGE ratios). It is possible that a very similar starting material was used, diluted with small amounts of copper and higher amounts of Ni, Zn and As.

In the case of the disc beads, a comparison of the trace element pattern clearly shows that only one of the eight beads analysed differs significantly from the others in its composition, but not in its alloy composition (**see** Fig. 10). Different atmospheric conditions during metalworking would explain the lower Sn and Pb contents, but should not affect the PGE, which have the same pattern but significantly lower concentrations. Therefore, production from a different ingot from the same geological source could be a likely explanation. In addition, the two single bead types at this site (#19, #33) are not comparable to each other or to the other beads in terms of trace element composition. Furthermore, the quadruple spiral pendant (#19), which appears as a single point d in Fig. 10, is made of an almost identical alloy as a typologically and chronologically comparable pendant by Lori Berd (#15), but both however, the two beads differ in part in their trace element patterns, which could indicate deliberate alloying with different raw materials.

Comparable results were obtained from the material found in the neighbouring Nerkin Naver. While all analysed objects from Tomb 10 are made of almost identical gold alloys, only a hollow bead with hump ornaments (#41) from Tomb 1 and a perforated jewellery disc (#34) from Tomb 9 were made of different gold stocks. The hollow bead is made of relatively fine gold with only a small amount of copper. Such an alloy is very ductile and malleable, which probably made it technically easier to produce such an elaborate shape. The second object is a round, technically very simple gold ornament, probably an import.

All these finds show that gold objects of different compositions were found in the same graves at the same time. Whether these alloys were actually in circulation at the same time would require a study of their wear. Gold objects that were actively worn during life, and possibly inherited, may have had a long life, while other gold objects seem in some cases to have been made directly for burial.

The objects from Karmir Blur (n = 5) show a wide range of their silver and copper contents (Ag: 25–35 wt-%; Cu: 2–6 wt-%) without any

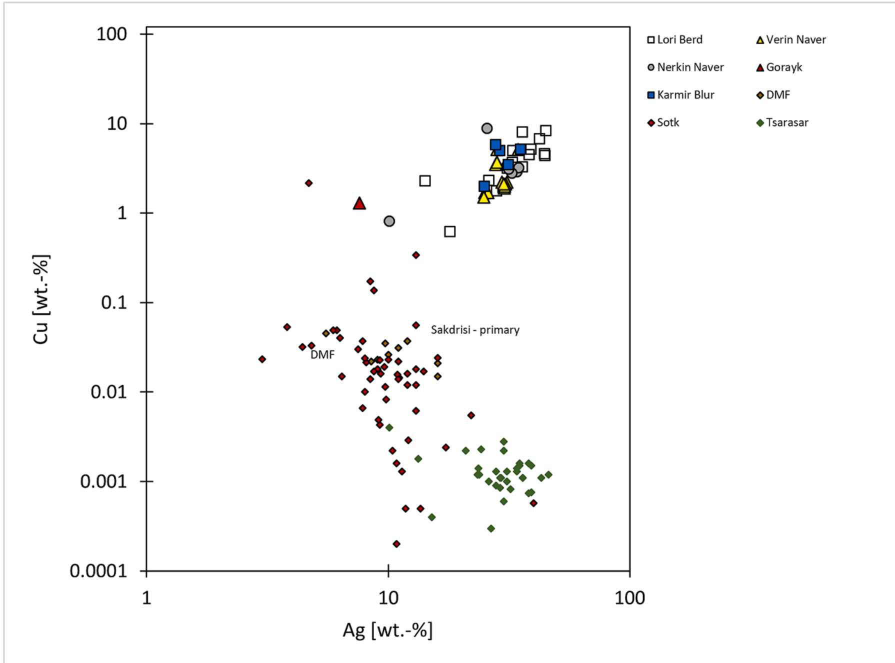

Fig. 9. Bivariate plot of silver and copper from objects and gold grains from Sotk, DMF and Tsarasar, as well as Sakdrisi (from Jansen, 2019) in comparison with gold objects from Armenian sites under investigation.

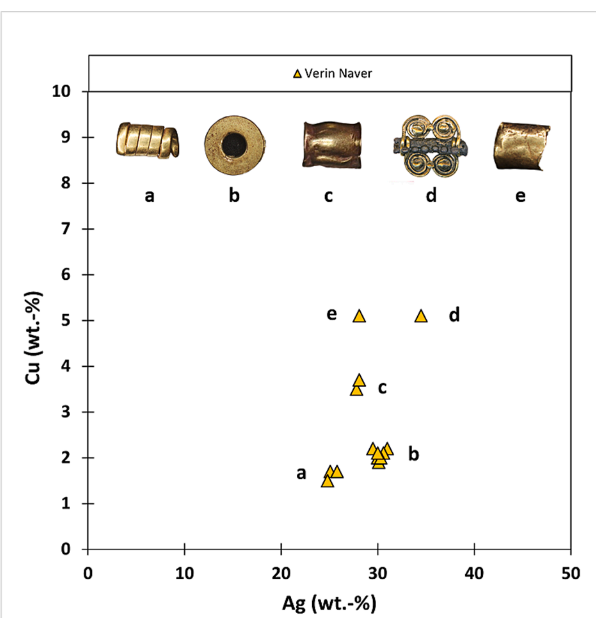

Fig. 10. Bivariate diagram of the silver and copper from the Verin Naver gold finds. Three groups (a-c) consisting of more than one object are recognizable, as well as two single object points. The three groups can be correlated with three different types of beads.

grouping. At Lori Berd, the finds date from the Late Bronze Age to the Archaemenid Period and cover almost the entire range of alloy compositions. These objects therefore follow a trend often observed in gold objects - increasing copper content correlates with increasing silver content. However, there is no direct correlation between age and alloy composition at this site. Typologically, the objects are very diverse and there are only few typological parallels at other sites, such as the above-mentioned quadruple spiral pendant from Tomb 3 (#15), which shows typological, chronological, and also chemical similarities to a bead from Verin Naver (#19).

The gold finds from Lori Berd cover the period from the Middle Bronze Age (4-fold spiral) to the Archaemenid period (cylinder seal and bead), although the dating of some finds is unclear. This long period is also reflected in the great technological diversity of the objects studied. Two finds from the Urartian context of the Middle Iron Age, a round pin head with granulation and carnelian inlay (#18), and the gold inlay of a central shaft hole pick (#17), are made of a gold alloy with a significantly lower silver content than the majority of the finds examined. There are also differences in the trace element composition. At Lori Berd there are a large number of round gold ornaments of various designs, which on the basis of typological considerations are assumed to be imported pieces (Piller, 2012). However, there is a lack of material-analytical studies for comparison. The pick axe, for example, has typological equivalents in the Iron Age Ananinyo culture, which also makes an import also very likely. In addition, the lower copper content of the gold inlay makes the material ductile and well malleable. Although the Mesopotamian rolled seal (#3) found in Tomb 106 was most likely imported, the chemical composition of the gold is comparable to most of the finds, so it is possible that the gold caps and pin were locally made additions. A total of 4 gold objects were analysed from Tomb 2, including a diadem (#1), a round gold ornament (button or bead; #9), and two elongated beads with different manufacturing techniques, one made entirely of granules (#11) and the other of sheet metal with elaborate granule decoration (#12). The grave has two different occupations, a Late Bronze Age inhumation around 1500BCE and a Middle Iron Age posthumation in the 7th/6th century BCE The objects examined cannot be clearly assigned to either inhumation. The assignment of the last two beads with granulation is difficult not only from an archaeological point of view, but also from a material analytical point of view. The gold sheets show a comparable PGE pattern, but differ significantly in the other trace elements, such as As, Sn or Sb. This is most likely due to the alloying with copper, which was probably intentional with 8 wt-% copper. Chemically, the two beads are very different, both from each other and from the diadem and the round ornament. While the gold in the bead made only from granules alone is most likely lode gold, the other bead has a high palladium content. Both

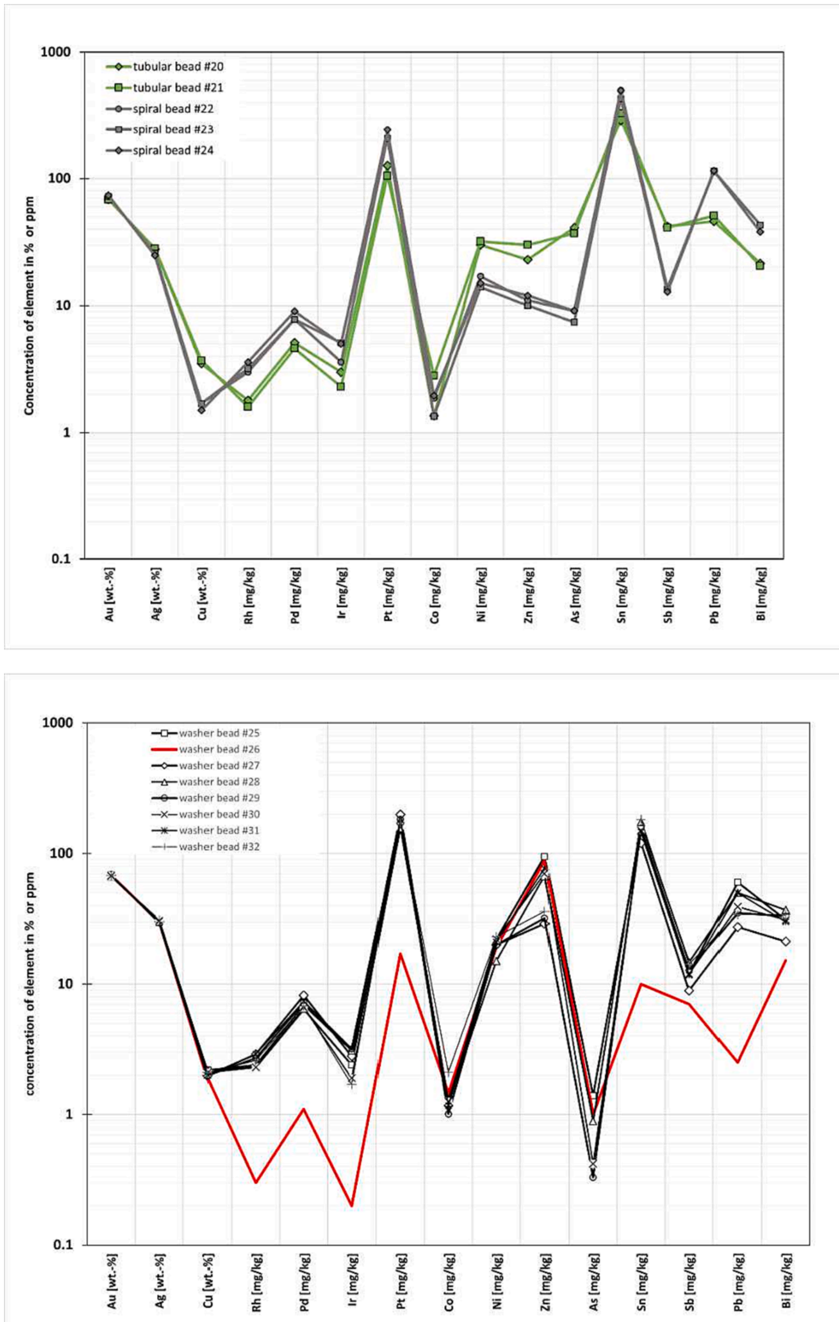

Fig. 11. a and b Spider diagrams of the different types of beads found on Verin Naver. A) Tubular and spiral beads; b) (perforated) jewellery discs.

beads are therefore unique pieces in the present study, both chemically and typologically. While the two high quality sheet metal objects can be assigned to the Middle Iron Age burial, the gold sheet metal fragments from burials 63–2 (#6) and 64 (#7) are similar in both their alloy composition and their trace element patterns, especially the high tin content in the per mill range, suggesting that very similar material was processed here, possibly with a slightly different chemical composition due to the mixing of different batches. For the other finds, data interpretation is more difficult. No clearly defined material groups can be defined. Thus, the gold finds from Lori Berd impressively demonstrate the typological, technical and material-analytical diversity reflected in these elite burials.

Metallographic studies show that the silver is completely dissolved in the solid solution, regardless of the amount of silver in the alloy (Fig. 12**, a and b**). Although copper is completely soluble in the melt, some samples examined, e.g. a granular bead from Lori Berd, Tomb 2, show that a biphasic structure is present from the originally homogeneous alloy (homogeneous solid solution) (Fig. 13).

In the gold object dataset of this study, tin and platinum contents vary by several orders of magnitude, with tin contents ranging from 0.5 to 4800 mg/kg and platinum contents from 0.2 to 790 mg/kg. Similar ranges are observed in other datasets from the Early Bronze Age in the Aegean or in the objects from Ur, Ebla and Georgia (Numrich et al., 2023, Jansen, 2019). While similar tin contents are found in the Central European Bronze Age, they are mainly accompanied by much lower platinum contents, which only increase from the developed Iron Age onwards (Numrich et al., 2023).

High levels of tin and platinum indicate the use of placer gold as a source material for gold deposits, and numerous gold-bearing streams are known in northern and eastern Armenia and neighbouring

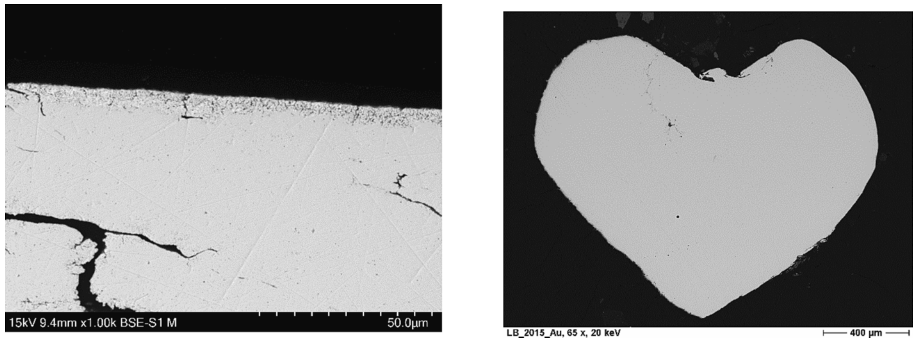

Fig. 12. a and b Scanning electron microscope image of the cross section of two selected objects which have very high (#5) and relatively low (#17) silver content in the respective alloy, approximately 42 wt-% (Fig. 12, a) and 18 wt-% (Fig. 12, b) respectively.

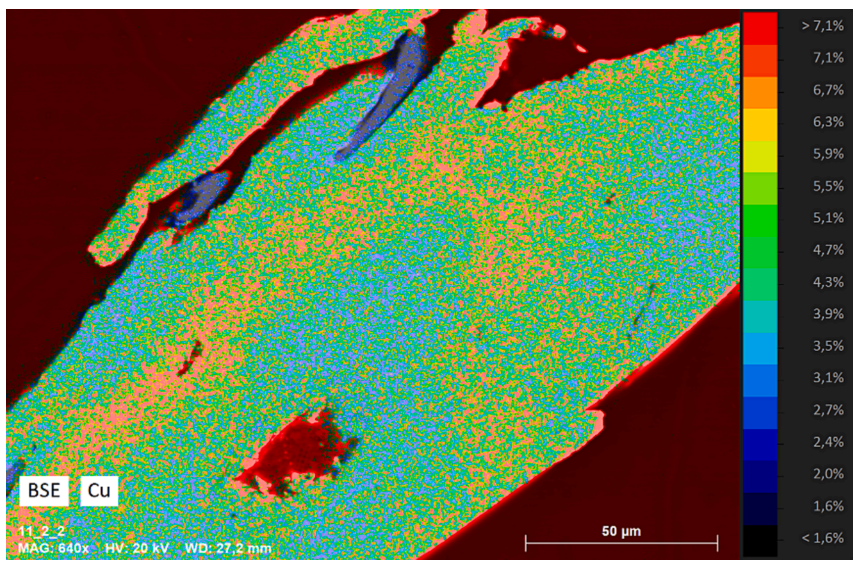

Fig. 13. Copper mapping (BSE image) for a granular bead from Lori Berd, Tomb 2 [#11].

(southern) Georgia (Fig. 14). However, only a few finds have low contents of these two elements and may therefore be lode gold. These are the gold spiral from Gorayk (#35), which can already be clearly distinguished from the other finds by its alloy composition, and another object from Lori Berd (#11; bead of soldered granules). The latter has a tin content of 370 mg/kg, but this may have entered the gold from other sources (such as deliberate or accidental alloying with bronze). The absence of platinum in the gold does not necessarily require the use of lode gold; a placer gold source without PGMs is also a conceivable explanation. However, two gold spirals were found at Hasansu that are typologically comparable to the Gorayk find, and all three are similar in terms of alloy composition and low Pt and Sn concentrations. Stöllner et al., 2018 postulate that the Hasansu rings were produced from primary gold of the Sakdrisi deposit.

Often platinum and palladium are the only elements discussed in the provenance discussion of gold objects (Guerra, 2021; Blet-Lemarquand et al., 2019; Ehser et al., 2011; Jansen, 2019), as other (s)melting-stable elements, such as nickel, cobalt or bismuth, significantly alter the supposed geochemical 'fingerprint' of the gold deposit in the gold object through possible (intentional or accidental) alloys with copper and/or silver. The range of platinum and palladium concentrations can sometimes vary by more than an order of magnitude, so that in many studies, focus is on the platinum-palladium ratio. Reasons for the wide range may include different proportions of gold and PGMs in the wash concentrate. Similarly, large variations have already been observed for tin, as discussed above, which also enters the final gold alloy mainly through with-panned minerals and melting of the wash concentrate in a reducing atmosphere.

Fig. 15 graphically illustrates the Pt and Pd contents of the samples analysed. The platinum content varies, as already described above, between 0.2 and 790 mg/kg with the exception of the object from Gorayk with 0.17 mg/kg, the palladium content is comparatively less concentrated and ranges in the majority of the finds between 1.1 and 20 mg/kg Pd, whereby this range can be extended by a single find from Lori Berd dating from the transition between LBA and EIA up to 67 mg/kg Pd (ellipsoid bead with granulation #12). In general, it can be said that at the sites investigated, the Bronze Age objects tend to have higher platinum and palladium contents than the Iron Age finds. River systems containing platinum-bearing minerals such as Pt-Fe seem to be the most likely source. Ru-Os-Ir minerals typically occur only as inclusions in archaeological gold objects, as they only melt at much higher temperatures than were common in prehistoric gold metallurgy (max. 1300 °C; Armbruster, 2003: 28). However, these have only been found in one case, which was discovered by chance in the time-resolved laser ablation signal.

With the exception of one gold find from Lori Berd (ellipsoid bead #12, dated to the LBA/EIA transition), all of the examined finds have comparable platinum and palladium ratios, comparable to the majority of gold finds from Troia, Poliochni, and Ur, but different from the finds from Varna and the coin finds from western Anatolia (Numrich et al., 2023, Jansen, 2019). While many PGE inclusions were observed in Ur, they appear to be macroscopically absent in the gold finds from Georgia

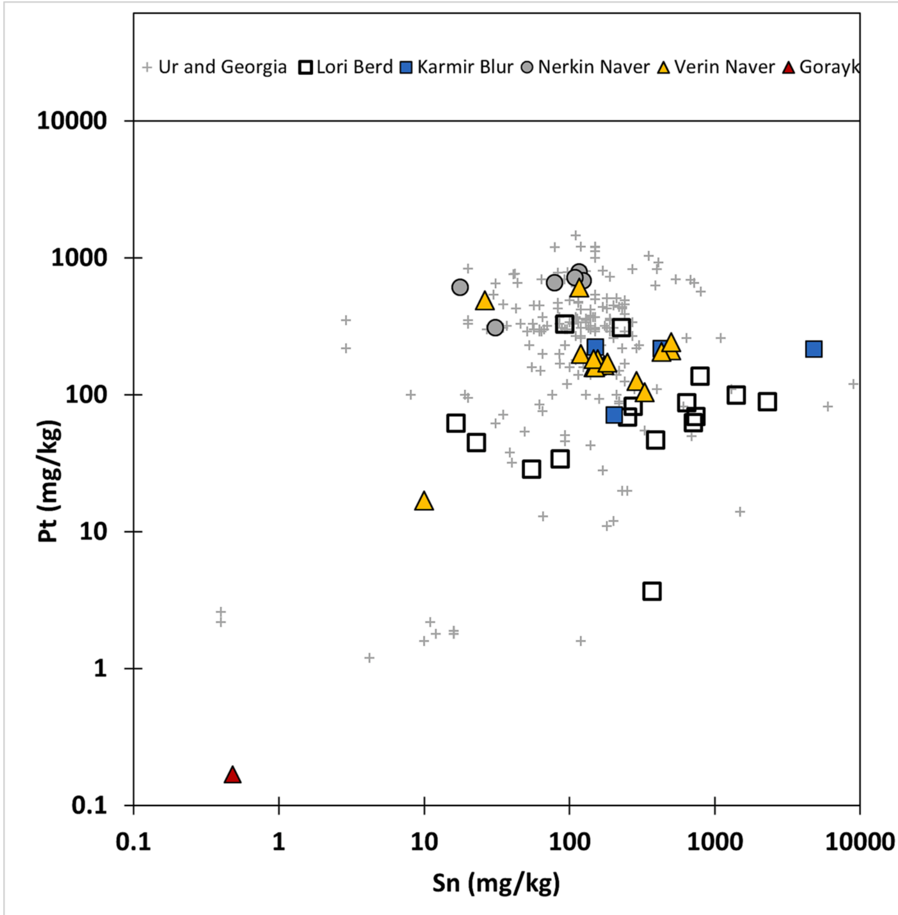

Fig. 14. Bivariate plot of tin and platinum (both in mg/kg) of the investigated gold objects (logarithmic scales). Shaded in grey are published data from Ur and Georgia (Jansen, 2019), which are in a very similar concentration range. The find from Gorayk is significantly different, a production of lode gold is likely.

examined by Jansen, 2019. Even without applying a correction model for Pd concentrations, as presented by Numrich et al., 2023, the similarity of the Pt/Pd ratio to the Armenian finds is evident. However, many of the Iron Age and Late Antique objects from Georgia deviate from this constant Pt/Pd ratio due to higher Pd contents. Since this material is partly contemporaneous with that from Lori Berd, there must have been at least one other source of gold circulating in the area of present-day Georgia at the beginning of the Iron Age.

With the exception of four gold objects, all others were made of a so-called electrum, i.e. a natural gold-silver alloy with $\geq$ 25 wt-% Ag (**cf.** Fig. 8). In archaeometallurgical gold research, values for the identification of intentional alloys - silver or copper - are a controversial and frequently discussed topic (cf. Schlosser, 2012; Leusch, 2019; Jansen, 2019). Nevertheless, it is almost impossible to define general values. As natural gold nuggets contain only small amounts of copper, typically less than 0.1 wt-% (Standish et al., 2021), the mystery of higher copper contents in almost all archaeological gold finds is still a matter of debate with different explanations. For gold alloys with more than 35 wt-% Ag, alloying with silver is usually considered as a possibility, since only a few natural gold deposits with higher average silver concentrations exist (Standish et al., 2021; Rehren et al., 1996), as demonstrated by the studies on gold grains from the Tsarasar placer gold deposit (**see** Fig. 9), which prove that natural gold with high Ag contents of > 40 wt-% Ag occurs in this deposit. For this reason, it is important to discuss other evidence that may answer the question of whether the high silver contents in the electrum-gold finds are of natural or intentional origin. One of the most important clues is the lead content. Natural gold usually contains only small amounts of lead, a few mg/kg of lead (Pernicka and Borg, 2017; Jansen et al., 2021). As archaeological silver was mainly mined from silver-bearing lead ores in pre-Christian times, it typically contains high levels of lead, around 100–1000 mg/kg lead, as found in the objects from Lori Berd and Nerkin Naver, for example. Therefore, intentional alloying with silver should significantly increase the lead content in gold-silver (copper) alloys compared to natural gold-silver alloys. Furthermore, silver-lead contents are expected to correlate positively (cf. Hauptmann et al., 2010; Jansen, 2019), while at the same time anti-correlations of platinum and gold would be expected due to dilution effects (Schlosser et al., 2012; Leusch, 2019). Analyses of contemporary silver objects from the South Caucasus are limited, so that only the four silver artefacts from Lori Berd (7th-5th century BCE) and Nerkin Naver (c. 2000BCE) discussed above (Section 4.3.) can serve as references for lead and bismuth contents. Based on Figs. 16 and 17, there is no positive correlation between silver and lead, and the lead content is in the range of about 100 mg/kg or even less and can therefore be considered as a natural impurity. However, some objects from Lori Berd (#1, 3, 4, 5, 11, 12, 14, 15, 16), but also from Karmir Blur (#38) and Verin Naver (#19) show even higher lead concentrations, but below the theoretically expected level. The addition of silver would also increase the bismuth content of the gold-silver alloy. Accordingly, bismuth via silver is plotted in Fig. 17. Based on the spiral gold ring from Gorayk, around 50 mg/kg Bi could still be of natural origin. Some finds from Lori Berd (#1, 5, 9, 15, 16) as well as one find from Karmir Blur (#40) and one from Verin Naver (#19) are above this assumed natural bismuth concentration, but - as already described for lead - below the theoretically calculated bismuth content. However, silver and bismuth contents correlate well in a number of finds from Lori Berd (#1, 5, 9, and 15). In

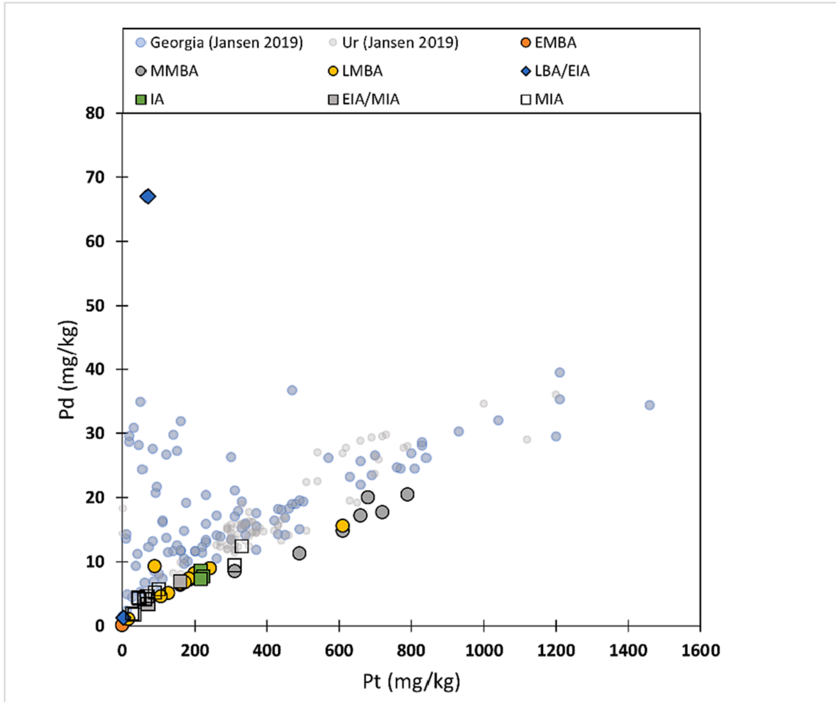

Fig. 15. Bivariate plot of Pt and Pd (in mg/kg) of the analysed objects according to their age. The analysed Georgian finds published in Jansen, 2019 are added in grey. The Late Iron Age objects in Jansen, 2019 show a tendency towards higher Pd contents, a material that is apparently not or rarely distributed in Armenia.

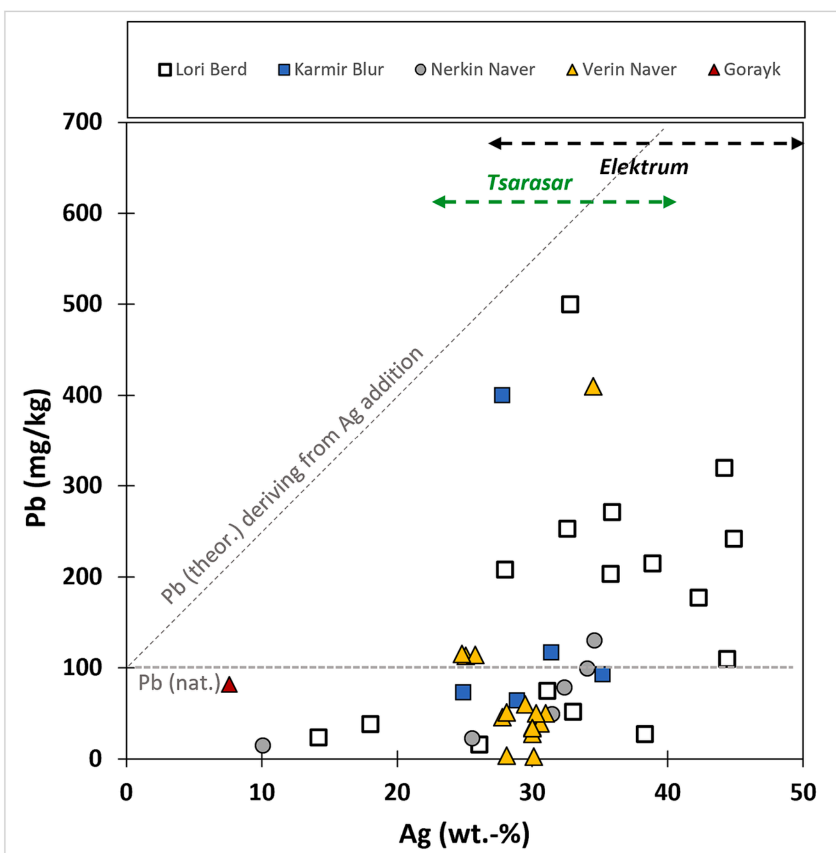

Fig. 16. Bivariate plot of silver (wt.-%) versus lead (mg/kg). The horizontal line at 100 mg/kg of lead marks the range that is introduced into a homogenised gold-silver alloy by natural gold, with-panned minerals or even other impurities during melting, without the intentional addition of silver or copper. The second gray line marks a theoretical amount of lead that would be introduced into the gold by the addition of silver as found in Lori Berd, assuming that the gold has been completely desilvered beforehand.

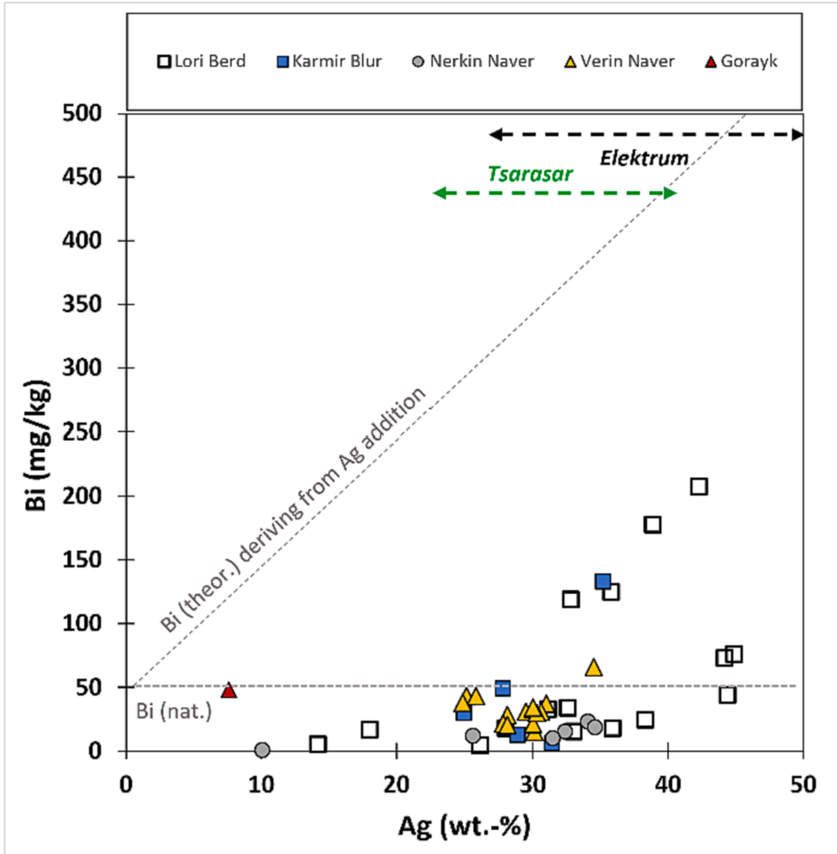

Fig. 17. Bivariate plot of silver (wt.-%) versus bismuth (mg/kg). The horizontal line at 100 mg/kg bismuth marks the range that is introduced into a homogenised gold-silver alloy by natural gold, with-panned minerals or even other impurities during melting, without the intentional addition of silver or copper.

these finds, mixtures with bismuth-bearing silver are likely. It is possible that the original gold was not desilvered, so that only small amounts of silver were added to produce an alloy containing about 40 wt-% Ag. In the case of lead, this correlation may be less obvious as it is less stable than bismuth during melting. However, for the majority of the gold samples examined, these results make it unlikely that silver has been deliberately added. Therefore, as a potential source of raw material, silver-rich natural gold is considered the most likely source.

Therefore, the Dilijan-Margahovit-Fioletovo (DMF) and Sotk deposits investigated at the Ushkiani project can be excluded as the source of the majority of Armenian gold finds (see Section 5.1). At Sotk, the average silver content of 10 wt-% is comparable to that of the primary gold occurrence (9 wt-% Ag). Furthermore, the lode and placer gold of Sakdrisi (southeastern Georgia) can also be excluded (Jansen, 2019, 156: Fig. 5.28). However, the investigated Tsarasar deposit, with its main tributaries Masrik and Kemachai, contains similar silver concentrations (21–38 wt-%) as found in the objects (26–45 wt-% [except for 4 objects with lower silver grades of 8–18 wt-%]) and therefore cannot be excluded as a likely source of raw material.

In addition to the silver content, the copper content also allows important conclusions to be drawn about possible intentional alloys (Fig. 18). The copper contents of the Lori Berd samples vary significantly between 1 and 8 wt-%. At the other sites, the variation in copper content is much smaller, ranging between 2 and 5 wt-%, with the objects from Verin Naver in particular showing a fairly uniform content (2 wt-%). The composition of the objects from different metals could not be determined microscopically. Archaeological gold objects usually contain more copper than would be found in prehistorically relevant natural gold deposits. This 'phenomenon' is discussed in almost all studies and observations of prehistoric gold (cf. Schlosser et al., 2012; Jansen, 2019; Leusch, 2019), also in the neighbouring regions of Armenia, Georgia,

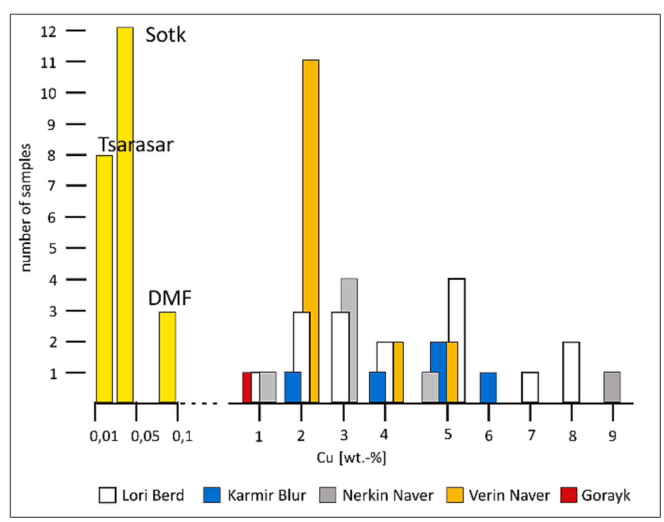

Fig. 18. Histogram of the copper concentrations of flakes from the investigated natural gold occurrences (Sotk, Tsarasar, DMF) and samples from analysed gold objects.

and Mesopotamia. However, similar to silver, however, no reliable values can be defined to distinguish between accidental and intentional alloying. Intentional addition is often suspected for alloys with more than 2 wt-% copper (e.g., Schwab and Pernicka, 2021: 569). To answer the question of the sources of copper in archaeological gold, several hypotheses are presented and discussed. Approaches to this question, for example, via the isotopic composition of copper, were developed by

Jansen, 2019, but could not be pursued in the context of this study.

A frequently discussed accidental with-panning of copper-bearing heavy minerals (e.g. Hauptmann and Klein, 2009; Leusch, 2019; Jansen, 2019) should be excluded for those objects with copper values show > 6 wt-% Cu. In addition to such an accidental contamination, deliberate changes in material properties, such as hardness, melting temperature, or even the rarely cited reflectivity of the gold could have been important (Guerra, 2021). The addition of copper can significantly strengthen gold-silver alloys and thus improve their service properties (Schwab and Pernicka, 2021: 569). The addition of about 5 wt-% Cu increases the Vickers hardness and tensile strength by a factor of about 3; higher Cu contents hardly lead to any further changes. Easier working of the jewellery objects was probably of little importance in most cases, but a deliberate addition of copper could have significantly increased the 'life' of the objects in terms of the resulting increase in hardness and tensile strength. The technique of granulation has been known in the South Caucasus since the first half of the 2nd millennium B.C. and is known, for example, from the Tsalka Plateau on South Georgia (Moesta, 1986: 107). The recycling of granular objects would increase the copper content of the gold alloy over a period of time. A deliberate addition of (metallic) copper to change the colour to a reddish shimmering gold is also regularly discussed (e.g. Pernicka, 2014; Jansen, 2019). The argumentation mainly ends with the assumption that a significant colour change occurs at the earliest at contents of ≥ 6 wt-% (Jansen, 2019). The determined contents of the objects examined are mostly below this, with only three gold beads from Lori Berd and a perforated gold disc from Nerkin Naver above it. However, modern smelting tests by the *Allgemeine Gold- und Silberscheideanstalt AG* (Agosi) have shown that significant colour differences can already be observed "[…] at 2–3 wt-% copper in the alloy" (P. Tews, personal communication). In particular, the high percentage of silver in the (natural) alloy, the addition of copper produces a hue similar to that of higher carat gold. In the Bronze and Early Iron Age of Armenia, elaborate polychrome jewellery objects are known, including from Lori Berd, among other places (Meliksetian et al., 2011). To achieve this colour effect, different copper alloys were combined. For this reason, it is obvious that colour effects in gold jewellery may have been deliberately created, for example by combining beads of different alloys. In a so-called Au-Ag-Cu ternary diagram, the alloy composition can be represented with a colour scale that roughly corresponds to the colour of the respective alloy (Moesta and Franke, 1995: 34), although the colour changes are less strict and more fluid. The colour spectrum (**cf.** Fig. 7) thus ranges from a rich gold (yellow–red) (Gorayk) and a yellow coloration (Nerkin Naver and two objects from Lori Berd) to a green-yellow (all objects from Verin Naver and Karmir Blur, partly from Lori Berd) or pale greenish-yellow (remaining objects from Lori Berd). The ternary diagram shows that the copper content in the present data set is hardly relevant for the colouration, since copper only has an influence on the colour at significantly higher contents. With regard to the silver content, a specific selection of a certain colour cannot generally be excluded, since the analysed specimens are distributed over different colour fields. This is also the case for most natural gold finds, so that it is not a clear argument for the deliberate production of lighter gold alloys.

Last but not least, economic reasons may have been relevant "[…] to manipulate the value of the metal" (Moesta and Franke, 1995: 35). For this reason, a deliberate addition of copper for the purpose of 'material enhancement' or saving of the valuable (placer) gold should not be excluded in principle. As discussed in the previous hypothesis, the addition of copper to the melt - at high silver contents – results in a colour change that appears distinctly golden. Using the ternary phase diagram in Fig. 7 as a basis, an alloy with up to 20 wt-% Cu (with 40 wt-% Au and 40 wt-% Ag), for example, would theoretically appear to be a 'pure' gold object, and thus economic considerations would also come into focus in addition to colour.

A closer look at the silver and copper contents reveals a positive correlation between the two elements, i.e. an increasing silver content in the gold-silver alloy of the raw material is accompanied by an increasing copper concentration. In an intentional copper–gold alloy, one would expect correlations of copper and other chalcophile elements, which are usually present in high concentrations in prehistoric copper alloys. However, only As and Bi show a weak correlation, while elements such as Co, Ni, Sn and Sb are not affected. One explanation could be that the added copper contained only small amounts of other trace elements besides As and Bi. There is also positive correlation between low copper contents and PGEs. This correlation does not occur in the copper-rich group and is difficult to explain.

6. Summary and discussion

The results of LA-ICP-MS analyses of 43 gold and 4 silver objects from 5 different sites in Armenia are presented and discussed with regard to their alloy and trace element composition. They provide information on the use of raw material sources, intentional alloying and the preferred use of these alloys. In addition, they shed light on the serial production of typologically identical bead types, as demonstrated by examples from Verin Naver, but also include beads found at different sites, indicating that these objects were probably traded over long distances. Relatively high concentrations of tin and platinum suggest that placer gold was the main source of raw materials from the Bronze Age onwards. However, the example of the spiral ring found at Gorayk indicates that the use of lode gold cannot be completely excluded. Its chemical characteristics are comparable to two other typological parallels found at Hasansu in the Kura Valley, whose gold probably originated from the Sakdrisi gold deposit, as outlined by Stöllner et al. (2018). Although intensive field work and analyses of natural gold have already been carried out, the explicit geological source of the raw material remains unidentified. However, since during this long period mainly high-silver electrum alloys were in circulation, and intentional addition of silver seems unlikely for most of them, gold deposits in Armenia with comparable low average silver contents around 10 wt-% Ag can be excluded, e.g. Armenian deposits of Sotk and the area around the villages of Dilijan-Margahovit-Fioletovo [DMF] as well as deposits in Georgia [Svaneti and Bolnisi region]). The Tsarasar deposit is therefore of interest for further research. In particular, the electrum finds from Lori Berd show a wide range of alloy compositions as well as a high typological diversity, thus testifying to the extensive trade network, the high status of the elite burials, and the cultural diversity seen, for example, in Neo-Assyrian seals. Although the dataset covers a long period of time, no clear chronological developments or changes in alloy composition have become apparent. Changes in the supply of raw materials could not be identified either. Determining the provenance of gold is complicated by the mixing and recycling of gold from different sources, which is difficult or impossible to reconstruct on the basis of the available data. Gold from the Caucasus may have been mixed with gold from other deposits. In this context, it is interesting to recall the Ur-III and Old Assyrian inscriptions on the subject of gold trade in Mesopotamia mentioned in Chapter 2, in which intermediaries ('Tukriš') are repeatedly mentioned, who received the raw material gold as 'gold dust' from various sources and traded it on.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

Acknowledgments

The studies would not have been possible without the kind support of the German Research Foundation (project number 410373002: 'The Importance of Armenian Gold for Cultural Development in Bronze Age Caucasia ('Ushkiani')' and the Scientific Committee of the Republic of Armenia (20RF-141). The authors would also like to thank the Institutes of Archaeology and Ethnography and Geological Sciences of the Academy of Sciences of the Republic of Armenia, which have always been reliable partners. Sincere thanks also to the two anonymous reviewers for their valuable and additional advice on this paper. Last but not least, the authors would like to thank the State Office for Heritage Management and Archaeology Saxony-Anhalt (Halle/Saale) for initiating the project and all those who actively supported the research in Armenia within the framework of the 'Ushkiani Project'.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.jasrep.2023.104267.

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