

Journal of the Geological Institute at the Faculty of Natural and Technical Sciences, University Goce Dell ev - tip, R. Macedonia



Geologica Macedonica	Vol.	33	No	1	pp.	1-84	Štip	2019
Geologica Macedonica	Год.		Број		стр.		Штип	

UDC 55 CODEN – GEOME 2 In print: ISSN 0352–1206 On line: ISSN 1857–8586

GEOLOGICA MACEDONICA

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Geologica Macedonica	Год.	33	Број	1	стр.	1–84	Штип	2019	

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Published by: – Издава:

"Goce Delčev" University in Štip, Faculty of Natural and Technical Sciences, Štip, North Macedonia Универзитет "Гоце Делчев" во Штип, Факултет за природни и технички науки, Штип, Северна Македонија

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400 copies Тираж: 400 Published twice yearly Излегува два пати годишно

Printed by: 2^{ri} Avgust – Štip Печати: 2^{ри} Август – Штип

Price: 10 € The edition is published in June 2019 Цена: 500 ден. Бројот е отпечатен во јуни 2019

Geologica Macedonica	Vol.		No		pp.		Štip	
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Упат	ство за авторите

346 GEOME 2 Manuscript received: December 18, 2018 Accepted: May 2, 2019

Original scientific paper

PLACER GOLD PROSPECTION IN SUŠEVSKA RIVER, SUŠEVO VILLAGE, NORTH MACEDONIA

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A b s t r a c t: This paper will present detailed studies on the chemical composition of gold aggregates found in Strumica Valley, eastern Macedonia, and their morphological characteristics. About a hundred chemical analyses of recovered gold aggregates were done. The chemical composition of gold aggregates shows heterogeneity with an average gold content from 89.85 to 99.72% and a significant mercury (0.44 to 6.27%) and silver content (0.24 to 18.41%). Other impurities are present in low contents: iron, tellurium, copper and arsenic. Although some platness-flattened and elongated shapes are present, the isometric-irregular shape is most frequent. The size of the gold aggregates ranges from 900 μ m to 3 mm.

Key words: schlich prospection; placer gold; morphology; gold grains

1. INTRODUCTION

Microchemical features of placer gold in North Macedonia have been very little studied, although gold occurrences were discovered long ago. In the last decade, there has been an increase in research on gold, especially when the price rises sharply, fuelling global exploration on an unprecedented scale. Many localities have become significant for study, although they are considered uninteresting because of their remote locations or poor outcrop exposure.

The morphology and composition of gold grains from alluvia are widely and increasingly used to determine the source of the primary gold mineralization, the style of mineralization and the potential host rock. Many workers worldwide have used gold grain characterization to link placer Au occurrences to a lode source in regional studies. The morphology of placer Au grains has been analyzed to provide a link to the location of bedrock sources. Compositional studies or complex alloys, together with studies of placer Au grain morphology, are also carried out to find the correlation between the placer and lode signature. (Knight et al., 1999a, 1999b; Townley et al., 2003; Crawford, 2006; Youngson et al., 2002; Chapman et al., 2010a, 2010b; Dumula et al., 2001; Florencia et al., 2004; Knight et al., 1999b; McClenaghan, 2001; Moles et al., 2011; Rasmussen et al., 2006; Omang et al., 2015; Norman et al., 2011; Nakagawa et al., 2005).

Studies of gold in North Macedonia have a long history. About thirty studies show. There are about thirty appear in that set presence of endogenic gold. Most of these appears, which are more or less studied, are economically uninteresting, but newer studies of some of these appears have shown interesting gold contents which might also result in opening new deposits.

There are a few literature data on the chemical composition and morphology of placer gold. The first such studies were carried out in several localities in which previous research had established the presence of endogenous mineralization of gold (Bogoevski et al., 1996, 1998; Kovačev et al., 2006; Stefanova et al., 2007, 2015; Volkov et al., 2010; Kovačev et al., 2007; Stefanova et al., 2016).

The main objective of this research was to study the chemical composition and morphology of gold aggregates collected from Suševska River in Strumica Valley, with the possibility of determining the primary mineralization.

GEOLOGICAL SETTING

The river of Strumica is the main watercourse in the investigated area. It passes through the central part and all the smaller tributaries infiltrate it. Suševska River is one of them and is the object of our investigation. Suševska River is without water during most of the year.

A small amount of old data indicate that research and exploitation of alluvial gold were carried out in this area a long time ago. Some research was conducted along Suševska River in 1990 and also indicated the presence of gold. Our latest investigation confirms the presence of gold aggregates in Suševska River. We examined gold aggregates originating from drainage systems with different types of rocks: alluvial sediments, Pliocene gravel and sands, Precambrian mica, then Palaeozoic granite and rhyolite. The geological setting of the field is presented in Figure 1.

Precambrian micaschist is breakthrough granite-porphyry. Metarhyolites appear at the contact between Serbo-Macedonian massive and the Vardar zone. They are grey-green rocks with porphyritic structure. They are the final differentiations of the granite magma from the Ogražden batholite. Pliocene sediments lie transgressively on the older Precambrian rocks and are represented by gravels, sands, and rarely clays.



Fig. 1. Simplified geological map at study area

METHODOLOGY AND SAMPLING

The fieldwork applied the schlich method. For this method, material was taken from places that were eligible for sampling with the possibility of concentration of heavier minerals. Samples of 15– 20 kg were taken, depending on the availability of material (Figure 2). Then flushing was carried out

and the schlich obtained was subjected to further processing. First, magnetic separation of minerals was performed and both fractions were observed under stereomicroscope. The gold aggregates found were manually separated and subjected to further investigation.



Fig. 2. Location of areas where they are taken schlich samples

To determine the morphological characteristics of gold, a scanning electron microscope (SEM) was used. Analyses were performed in the laboratory of electronic microscopy at the University of Štip using a VEGA3 LMU instrument. The standards were TESCAN. The specific conditions of work were as follows: the tension was 20 keV; the test method was EDS; the type of analysis was quantitative X-act: 10 mm² silicon drift detector; the maximum resolution was 125 EV; and the resolutions of MnK α , FK α and CK α were according to ISO/TS 10798: 2011th.

RESULTS AND DISCUSSION

Our investigation showed the presence of exogenic gold in Suševska River. Schlich prospection was carried out over a length of about 3 km. A total of 12 probes were taken, in which 91 gold aggregates were found, and 55 of them were examined.

The microchemical signature is of great significance for alluvial gold, especially in areas where gold is obtained from stream sediments, while the primary mineralization has not been discovered. This approach allows for the identification of populations of placer gold derived from different sources (Omang et al., 2015).

Gold occurs dominantly as the native metal. Gold aggregates are very often binary Au-Ag alloy or are commonly alloyed with other metals: mercury, copper and palladium. Native gold also occurs within sulfur-rich minerals such as pyrite and arsenopyrite either as submicroscopic inclusions of native gold or as a minor component within the lattice of these minerals (Chapman et al., 2002). According to Norman et al. (2011), the presence of palladium is explained in the context of the environment in which gold is transported and precipitated, but control of mercury and copper remains unclear. The gold has a homogeneous composition, indicating uniform conditions of precipitation.

Chapman et al. (2010) believe that examination of the chemical composition can be used to find the correlation between the chemical composition of primary gold and placer gold and can determine the type of mineralization. These researchers believe that gold aggregates with a narrow range of chemical composition indicate the orogenic origin so that it can overlap the chemical composition of gold from various sources.

The chemical composition of the examined aggregates in Suševska River shows that gold is characterized by great finesse with a gold content of 89.85 to 99.72%. It could be said that the gold aggregates show a uniform distribution of gold (Table 1).

Table 1

Microprobe data for gold grains from Suševska River

Sample no.	Au	Ag	Hg	Sample no.	Au	Ag	Hg
S-3/1	81.59	18.41	_	S-7/6	95.87	_	2.84
S-3/2	98.45	_	1.55	S-7/7	94.28	_	2.90
S-3/3	99.04	0.96	_	S-7/8	98.72	1.28	_
S-3/4	97.94	0.86	1.19	S-7/9	98.27	_	1.32
S-3/5	99.22	0.78	_	S-7/10	99.56	_	0.44
S-3/6	98.92	1.08	_	S-7 /11	98.50	_	1.50
S-4 /1	97.38	_	2.62	S-7/12	98.08	_	1.92
S-4/2	98.26	_	1.74	S-7/13	96.46	_	3.54
S-5 /1	93.73	_	6.27	S-7 /14	98.32	_	1.68
S-5/2	98.87	_	1.13	S- 8/1	98.60	_	1.40
S-5/3	98.96	-	1.04	S8/2	99.17	_	0.83
S-5 /4	98.68	-	1.32	S-9	98.87	_	1.13
S-5/5	97.61	_	2.39	S-10/1	97.43	_	2.57
S-6 /1	98.75	-	1.25	S-10/2	97.60	_	2.40
S-6/2	98.09	_	1.91	S-10/3	99.14	0.86	_
S-6/3	99.25	_	0.75	S-12/1	99.01	_	0.99
S-6 /4	99.72	_	0.28	S-12/2	98.67	_	1.33
S-6/5	98.95	_	1.05	S-12/3	98.69	0.59	0.73
S-6/6	98.69	_	1.31	S-12/4	99.75	_	_
S-6/7	96.73	-	3.27	S-12/5	99.15	0.46	0.39
S-6/8	95.54	_	3.14	S-12/6	99.38	_	0.62
S-6/9	98.89	_	1.11	S-12/7	99.48	_	_
S-6 /10	98.89	-	1.11	S-12/8	98.90	0.24	0.86
S-7 /1	96.47	_	3.53	S-12/9	98.09	_	1.91
S-7/2	98.74	0.29	0.97	S-12/10	99.62	-	_
S-7/3	98.43	1.57	_	S-12/11	99.32	_	0.68
S-7/4	98.58	-	1.42	S-12/12	97.13	_	2.87
S-7/5	97.97	_	2.03				

Only a small number of aggregates show a slight enrichment in gold at the edges of the grain compared with the central parts. This can be interpreted as one of the features of placer gold that is more a result of the extraction of silver than a result of bulking the new grain of gold (Knight et al., 1999b) as is the case for placer gold recovered from Klondike. These over-thickened rims are ascribed to leaching. According to Krupp et al. (1990), this is most likely in order to establish a balance of gold with the existing environment in which it is located due to the long stay in the surface sediments. Hough et al. (2009) show that beside Au-enriched rims there may be another heterogeneity in the composition of gold aggregates, which may be due to the atmospheric effects of Au-Ag alloys. Anyway, the degree to which the grains change is likely to affect the overall composition of the alloys of the sample. Chapman et al. (2010) suggest that the use of fineness is not a particularly useful parameter for the connection of lode and placer compositions when considering the gold from Klondike.

Mineral impurities include a significant content of mercury (0.44 to 6.27) and silver (0.24 to 18.41%). So the test placer gold is Au-Ag followed by Au-Hg and Au-Ag-Hg alloy, where the Au-Hg alloy dominates. Generally, Ag is the main alloying element. Its content ranges from < 1% to > 30% in natural Au alloys (electrum). Sporadic elevated contents of Hg in Au-Ag alloys, which in this case are significant, have not been thoroughly investigated (Moles et al., 2013).

From a total of 55 units studied, mercury occurs as an admixture in 47 units, with a wide range. Based on the content of mercury, three types of units can be distinguished: the first type has a content of < 1%, the second 1–3%, and the third more than 3%. From Table 1 and Figure 3, it can be seen that around 60–70% of grains have a relatively narrow spread of compositions of 1–3% Hg and the remainder have a wider compositional range of higher Hg contents extending to 6%, so the second type of aggregate predominates.



Based on these results, it can be said that mercury has an irregular gold distribution in aggregates. Mercury impurities occur within natural gold at many localities in the world (Boyle, 1979). Mechanisms of Hg transport and deposition in hydrothermal systems were investigated by Barnes (1979) and Krupp and Seward (1990) and, according to them, factors controlling the formation of Au-Ag-Hg alloys are still not well understood. Anyway, the limited understanding of these mechanisms, the presence of Hg is still considered to be a useful variable in geochemical fingerprinting of specific lode sources (Mackenzie and Craw, 2005; Chapman et al., 2010). As can be seen in the investigated gold, the Hg content varies from low (0.44%) to high (6.27%).

Studies by Chapman et al. (2010) in the west of Lone Star Ridge showed that the systematic increase in the number of Hg-bearing Au grains in populations of lode gold grains reflect the period of Au precipitation under conditions conducive to the formation of Hg-bearing alloy rather than indicating multiple episodes of mineralization by Hg-rich and Hg-poor fluids. Peters (1991) suggests that mercury can be transported by hydrothermal fluids, because it is readily mobilized from metamorphic rocks. Also, it is possible that mesothermal fluid systems could contain Hg in a wide range of structural levels. Because of this, the occurrence of Hg may related to the deposition mechanisms rather than the sources (Mackenzie et al., 2005).

Furthermore, the narrower the range of fineness and Hg values, the more likely it is that the signature represents a single source (Knight et al., 1999a,b).

In addition to these studies, analyses of the chemical composition of gold aggregates and polished preparations were done. The results are shown in Table 2.

	1 00	-		0 0				
			Au	Ag	Hg	Cu	As	Al
	2 3	1 Center	64.12	35.12	0.76			
S-5 /1		2 Rim	64.82	35.18	0			
	m ON	3 Rim	65.36	34.64	0			
	2	1 Center	90.45	9.55	0	_	_	_
S-6 /1	3	2 Rim	89.44	10.56	0	_	_	_
		3 Rim	97.36	2.64	0	-	_	-
	p 1	1 Center	90.42	6.92	2.66	_	_	_
S-6/2	· · · · · · · · · · · · · · · · · · ·	2 Rim	97.93	_	_	2.07	_	-
	0	3 Rim	98.51	-	-	-	1.49	-
S-7/1		1 Center	82.18	17.82				
		2 Rim	81.88	18.12				
	2 3	3 Rim	83.31	16.69				
		1 Center	97.29	2.71				
S-7/2	2 martine 2	2 Rim	85.72	14.28				
5 112		3 Rim	86.90	13.10				
		4 Rim	96.30	3.70				
		1 Rim	96.12	2.28	1.60			
S-7/3	2	2 Rim	97.37	2.63				
		1 Center	91.46	6.17	2.37			
S-1 2/1		2 Rim	98.96	1.04				
	We shall be a second							
	2	1 Center	97.54	0.85				1.61
S-12/2	1 . 5	2 Rim	93.18	1.48	5.34			
	3	3 Rim	97.62			0.75		1.63

Table 2

Morphology and microprobe data for gold grains from Suševska River

From the results shown, it can be seen that silver is present in significant contents ranging from 2.28 to 35.18%, while mercury is almost absent. If a comparison of Table 1 and Table 2 is made, it can be seen that in Table 1 mercury is represented as an admixture much more frequently and in a greater percentage in comparison with the analyses carried out on the polished sections. Bearing in mind that in most of the analyszed gold aggregates (Table 1), mercury is present while in the samples of the polished sections it is represented by much lower contents, it can be assumed that the mercury appears in the form of a coating and the reason for this can be sought in anthropogenic influence (Meisser & Brugger, 2000; Florencia et al., 2004). According to these authors, Au amalgams can be formed in river sediments as a result of the reaction between detrital gold and mercury of anthropogenic origin, although gold aggregates can naturally contain elevated mercury content in the central parts as is the case with some of our investigated aggregates (Table 2).

As possible sources of mercury from anthropogenic origin, these authors list certain artificial fertilizers and then fungicides and herbicides used in agriculture, which can be accepted as an explanation in the case of the investigated gold aggregates from Suševska River.

Silver has a non-uniform distribution in gold aggregates. It is represented in only 13 of the units studied, mostly with a low content. According to Hough (2009), such process can be ascribed to the process of decomposition of the primary Au-Ag composition due to diffusion along the grain boundaries. According to Knight et al. (1999a,b), this small difference in the composition may be due to the leaching of silver after gold aggregates are released from the primary source. Low-fineness inhomogeneities reflect the chemical and/or thermal evolution of a hydrothermal fluid (in contrast to a fluid in the surficial environment) and they probably represent a late stage in the lode development (Groen et al., 1990).

Knight et al. (1999a,b) suggest that the existence of differences between the high- and low-fineness areas could be interpreted as indicating either different episodes, changes between episodes, or differences within a single episode of

mineralization. Also, according to this author, the homogeneity and range of fineness values do not fit the characteristics of gold from epithermal sources. Rather they are more in keeping with gold from mesothermal sources.

Among the other impurities in gold aggregates from Suševska River, iron, tellurium, copper and arsenic are present very rarely and in very low contents. This relatively uniform chemical composition of gold aggregates may indicate a single source of alluvial gold (Knight et al., 1999a,b).

In this study we also describe the morphological forms of gold particles in order to determine the potential primary source rocks. The tested gold aggregates from Suševska River are characterized by sizes from a couple of hundred microns to 3 mm. Small gold aggregates from 300 to 500 μ m predominate (Figure 3). The group with the biggest size accounts for 16% of the aggregates. Most of these gold aggregates were found in sample S-7.

The changes in morphological characteristics of placer gold during fluvial transport are in function of transport distance. Many studies have described these characteristics in terms of the relationships between placer and sources, particle origin, and type of source (Tishcenko, 1981; Hérail et al., 1990; DiLabio, 1991; Youngson et al., 1999; Florencia et al., 2004; Rasmussen et al., 2006). Many classification schemes are made, and they include qualitative and quantitative data of parameters such as the shape, outline, size, roundness, flatness, surface texture, or sphericity.

The outline and shape of gold particles are modified during fluvial transport, causing flattening, folding, and rounding. The outline is a two-dimensional descriptor of particles and is easier to use than shape, a three-dimensional descriptor of particles, because there is no classification scheme that can adequately describe a complex indefinite form of undeformed or slightly deformed gold particles that can be found near the primary source. The flatness index and the outline of particles depend directly on changes in shape due to flattening, folding, and rounding (Youngson et al., 1999).

Knight et al. (1994) provided a scheme of the outline of gold particles (Figure 4).



Fig. 4. Gold particle outline classes after Knight et al., 1994 (taken from Youngson et al., 1999)

According to data from Youngson and Craw. (1995), gold particles are characterized by a branched or complex outline in the primary environments. Modification of the outline starts in the first few kilometres when the initial forms undergo transformation toward more complex, equant, and elongated forms (Figure 5). Modification is manifested by rounding and minor infolding (Figure 6: S3/1, S4/1, S9/1, S10/2). As the distance they are

transported downstream increases, flattening, folding, and rounding of particles become more evident. In contrast to the outline, the rounding of gold particles commences as soon as their transportation in the fluvial system commences, and rounding is generally progressive with increased transport distance, mainly as a result of abrasion of particle edges or folding of delicate protrusions and thin edges, as can be seen in Figure 6: S3/5, S5/5, S6/5, S7/7.



Fig. 5. Histograms of outline data from some gold particles, Suševska River

Particle flatness is also important, in particular the flatness index. Investigation shows that the flatness index increases progressively with transport distance. In Suševska River there are gold particles with a high flatness index, as shown in Figure 6 (S5/3, S6/5, S6/9, S6/10, S7/10) most likely pointing to long transport. Some of these flattened particles have been significantly folded (Figure 6: S7/4, S7/7, S12/9).

Gold grain in a natural environment can be deformed during transport by contact with fragments of rock and also with hard minerals. Many investigations show that morphological transformations are a function of the transport distance and environment (Hérail et al., 1990; Knight et al., 1994, 1999). In this way, we can provide information regarding travel distance with respect to the source, which concerns the transport mechanism and sedimentological environment.

Researching the forms of gold grains is one of the many approaches to determining the primary

source of gold aggregates which is used in gold exploration.

Our investigations of gold from Suševska River show that grains are mostly characterized by irregular shape and isometric form (Figure 6). In general, the forms that can be distinguished are isometric and elongated dendritic forms, which can be deemed that grains are delayed close to the roots (Tarabaev, 1990). Then it is turn up irregular shapes, flatness forms likely to delay beyond primary sources and can be transported over considerable distances (Tarabaev, 1990). According to Chapman et al. (2010), the presence of dendritic grains in alluvial sediments indicates nearsness to source bedrock, because dendrite spikes which are folded around the core cannot tolerate long transport.

The studied gold grains showed curvature of the edges and smoothing of the surface (Figure 6) along with simultaneously increasing flatness, which may be due to the significant transport of gold and primarily due to the small fortress and malleability of gold as a mineral.

This appearance also may mean that the grains were subjected to secondary processes, which leads to an increase in the flattening in function of the length of transport as a result of collisions between gold grains and grains of other rock minerals (Knight et al., 1999; Townley et al., 2003; Tishcenko et al., 1981).

However, flatness varies depending on the size of the grains because larger grains (1-2 mm) are exposed more (Knight et al., 1999; Hérail et al., 1990) than smaller grains that range between 8 and 16 mm in size. Grains smaller than 60 μ m are subject to much less flatness.

Circular form, degree of curvature and degree flatness may indicate the type of source and length of transportation (Mudaliar et al., 2007; Knight, 1999a,b). However, according to Heney (1995), measuring the size and shape of the gold aggregates provides little information about the origin of alluvial gold. Angular and irregular shapes indicate that alluvial gold is close to its primary source and are easily dectected under binoculars, while abrasive and circular shapes indicate a longer transport of several kilometres.

Studies performed by Townley et al. (2003) show that gold aggregates at a distance of 300 to 1000 m from the source are characterized by rounded to oval shape and are commonly elongated, with a regular outline and surface topography. Sometimes they can be hammered and have folded edges. Aggregates at a distance of over 1000 m show rounded and oval shapes, with a regular, smooth and polished outline and surface topography, common striations, and also impact marks and a hammered appearance.

Taking into account these data, we can say that gold aggregates from Suševska River are characterized by rounded to oval shape and sometimes elongated, with a regular outline and surface topography (most of the grains from probes S-4, S-6, and S-10). These shapes indicate a distance of 300 to 1000 m.

Most of the gold aggregates from S-5, S-7 and S-12 are characterized by rounded and oval shapes with a regular outline and a smooth and polished surface topography. Also it can be seen that some gold aggregates exhibit striations. These shapes indicate a distance of over 1000 m.

These aggregates do not contain inclusions, which is very characteristic for aggregates up to a distance of 300 m.

Forms such as square, rectangular, irregular star and angular, and grains with irregular angular shapes and regular and irregular outlines and surface indicate distances of 50 to 300 m but were not determined in the samples taken from Suševska River.

The data from Youngson et al. (1999), which were obtained from a river system in Otago, New Zealand, showed that at 15–20 km from primary sources gold aggregates exhibited significant folding. These kinds of morphological forms were observed in Suševska River (Figure 6). According to this author, rounding of free gold usually begins as soon as it begins to be transported, while larger particles (> 3 mm) are typically more rounded than smaller gold particles.

Anyway, individually, none of the abovementioned descriptors such as shape, outline, flatness or folding can adequately quantify or qualify the morphological changes that gold undergoes during the increasing transport distance. But when they are used collectively, they can provide information about the changes in gold particles. Flatness and folding are useful descriptors for defining the relationship between the morphology of gold particles and transport distance. As Youngson et al. (1999) write, at least three parameters (flatness, folding, and roundness) should be included for interpretation of placer gold morphology. In primitive placers, a significant descriptor is roundness. As distance increases, a positive correlation between the flatness index and roundness can be apparent.



S3/5. Porous partially rounded gold grain



S4/2. Subspherical gold grain with rounded edges



S5/5. Gold grain with irregular rounded surface feature



S6/9. Gold grain showing elongated, flattned with rounded surface features



S7/7. Flattned gold grain with folded edges



S6/4. Gold grain with irregular rounded surface feature



S6/10. Complex elongated, flattned, partially rounded gold grain



S7/10. Gold grain with elongated, flattned partially rounded surface features



S5/3. Flattned elongated gold grain with rounded edges



S6/5. Rounded elongated gold grain



S7/4. Iregular gold grain with folded edge



S7/12. Elongated gold grain with rounded edges



S7/13. Flattned gold grain with rounded edges



S12/4. Elongated gold grain showing complex, partialy rounded surface morphology



S9/1. Porous partially rounded gold grain



S12/9. Flattned elongated gold grain with folded edges



S12/3. Gold grain showing complex, partialy rounded surface morphology



S12/11. Gold grain showing complex, elongated, partialy rounded surface morphology

Fig. 6. Morphological forms of matching gold aggregates of Suševska River

CONCLUSION

The prospection organized along Suševska River confirmed the occurrences of placer gold based on the schlich methodology. In individual schlich samples, from 2 to 27 gold grains were discovered. Grain sizes ranged from several hundred microns to about 3 mm.

Gold aggregates are characterized by great fineness of the content, which ranges from 89.85 to

99.72% and belongs to the group of high-grade gold. In gold aggregates from Suševska River, mercury, iron, tellurium, copper and arsenic are present in low contents. The morphology of these grains is characterized by different forms: the flattened form predominates and there are also elongated, irregular, and often dendritic forms.

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Резиме

ПРОСПЕКЦИЈА НА НАНОСНОТО ЗЛАТО ВО СУШЕВСКА РЕКА, С. СУШЕВО, СЕВЕРНА МАКЕДОНИЈА

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Клучни зборови: шлиховска проспекција; наносно злато; морфологија; зрна злато

Во овој труд се презентирани деталните истражувања на хемискиот состав на златните зрнца пронајдени во Струмичката Котлина, источна Македонија, и нивните морфолошки карактеристики. Направени се околу сто хемиски анализи. Хемискиот состав на златните зрна покажа хетерогеност со просечна содржина на злато од 89,85 до 99,72% и значителна содржина на жива (0,44 до 6,27%) и сребро (0,24 до 18,41%). Содржината на други примеси е ниска (железо, телуриум, бакар и арсен). Иако се присутни и плочести и издолжени облици, најчесто е застапена неправилна изометрична форма. Големината на златните зрна се движи од 900 µm до 3 mm.