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PLATINUM, PALLADIUM AND GOLD CONTENT IN THE PORPHYRY-Cu SYSTEMS OF THE VERTISKOS FORMATION, SERBOMACEDONIAN MASSIF

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ABSTRACT

The studied porphyry copper systems, namely Gerakario, Pontokerasia, Fissoka and Skouries are integral parts of the Vertiskos Formation of the Serbomacedonian massif. Samples of altered mineralized porphyries, amphibolites close to the porphyries and strongly oxidized altered amphibolites from a fault zone, OP-65 (Fissoka), were analyzed for platinum, palladium, gold, tellurium, arsenic and base metals.

Although the Fissoka and Skouries intrusions have some common geochemical features, the former differs in having lower Cu and Pd content, in both porphyry and amphibolite basement, even in the local Au-bearing mineralization. Also, the Gerakario and Pontokerasia, with a relatively weak silicification and small proportion of amphibolites among country rocks, compared to Skouries and Fissoka (OP-65), are accompanied by much lower precious metal content.

Mineralogical and geochemical data from the studied porphyry systems suggest a similar behavior of precious metals during their transportation and deposition and a relationship with silicification. The presence of merenskyite (Pd-telluride) and sylvanite at Skouries, and limited fluid inclusions data from Fissoka may provide evidence for a higher temperature during the deposition of metals in the former than in the latter.

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ΣΥΝΟΨΗ

Τα μελετηθέντα πορφυρικά συστήματα στις περιοχές Γερακαριού, Ποντοκερασίας, Φισώκας και Σκουριών περιλαμβάνονται στον σχηματισμό του Βερτίσκου της Σερβομακεδονικής μάζας. Τα στοιχεία λευκόχρυσος, παλλάδιο, χρυσός, τελλούριο, αρσενικό και βασικά μέταλλα προσδιορίστηκαν σε δείγματα εξαλλοιωμένων μεταλλοφόρων πορφυρών, αμφιβολιτών κοντά στην επαφή τους με τους πορφύρες και έντονα οξειδωμένων αμφιβολιτών από την ρηξιγενή ζώνη στην OP-65 (Φισώκα).

Αν και οι διεισδύσεις Φισώκας και Σκουριών έχουν αρκετά κοινά γεωχημικά χαρακτηριστικά η πρώτη διαφέρει ως προς την μικρότερη περιεκτικότητα Cu και Pd, στον ποφύρη και τους αμφιβολίτες του υποβάθρου, ακόμη και στις περιοχές εμπλουτισμού σε Au. Επίσης, στις περιοχές Γερακαριού και Ποντοκερασίας, όπου η πυριτίωση είναι σχετικά ασθενής και η αναλογία των αμφιβολιτών μικρή σε σχέση με τις Σκουριές και την Φισώκα (OP-65), συνοδεύονται από πολύ μικρότερη περιεκτικότητα ευγενών μετάλλων.

Τα ορυκτολογικά και γεωχημικά δεδομένα από τα μελετηθέντα πορφυρικά συστήματα υποδηλώνουν όμοια συμπεριφορά των ευγενών μετάλλων κατά την διάρκεια της μεταφοράς και απόθεσής τους και μια σχέση με την πυριτίωση. Η παρουσία του μερενσκιίτη (Pd-τελλουρίδιο) και του σιλβανίτη στις Σκουριές, καθώς επίσης περιορισμένα δεδομένα ρευστών εγκλεισμάτων από την περιοχή της Φισώκας ίσως παρέχουν ενδείξεις για υψηλότερη θερμοκρασία κατά την διάρκεια απόθεσης των μετάλλων στην πρώτη περίπτωση σε σχέση με την δεύτερη.

INTRODUCTION

Although a platinum group element (PGE) enrichment can be the result of hydrothermal transport related to a leaching process from mafic - ultramafic host rocks or to alkaline porphyry copper mineralization (Finch et al., 1983; Werle et al., 1984; Mountain and Wood, 1988; Macdonald, 1988; Nyman et al., 1990; Eliopoulos and Economou-Eliopoulos, 1991) very limited data have been published on PGE level in intermediate and felsic rocks.

The Skouries porphyry copper deposit, located in the eastern Chalkidiki peninsula, northern Greece, was chosen for the study of PGE concentrations for the first time in such rock types in Greece (Eliopoulos and Economou-Eliopoulos, 1991; Tarkian et al., 1992). The results of this study are now being applied to other porphyry systems of the Serbomacedonian massif, namely Gerakario, Pontokerasia and Fissoka - more specifically OP-65 - in order to be incorporated into a metallogenic model for the porphyry systems in Yugoslavia, Bulgaria and Greece.

In this paper the precious metal concentrations and other important components of the mineralized porphyries of the Serbomacedonian massif are given and their implication to their metallogenesis are briefly discussed.

GEOTECTONIC SETTING

The Serbomacedonian massif of probable Paleozoic and/or older age, is a highly deformed NW-SE trending belt, which includes geographically most of the Chalkidiki peninsula in Greece and extends partly into Yugoslavia and Bulgaria. It is divided into the Kerdylia and Vertiskos Formations of Kockel et al. (1977); it is comprised of two-mica gneiss, biotite-amphibolite, gneisses, amphibolite, mica schist, marble, and locally some anatexites. The Kerdylia Formation is characterized by the presence of marble and anatexite, whereas rocks of basic to ultrabasic composition are present in the Vertiskos Formation (Fig. 1).

Regarding the Tertiary intrusions suite of the Serbomacedonian massif they are probably related to a subduction zone magmatism (Jancovic et al., 1980). Furthermore, on the basis of combined lead and strontium isotopic data unmineralized plutonites of granitic composition (Ierissos, Sithonia) intruded during the early Eocene, followed by early to middle Oligocene dioritic, monzonitic and granodioritic stocks (Gerakario, Vathi, Straton, Tsikara) some of which are accompanied by sulfide mineralizations. These are distinguished from the Miocene (18 Ma) subvolcanic-porphyrific stocks and dykes, such as the Skouries and Alatina which are related to porphyry-copper type mineralization (Frei, 1990).

DESCRIPTION OF THE STUDIED PORPHYRY SYSTEMS

The studied porphyry copper systems, namely Gerakario, Pontokerasia, Fissoka and Skouries, are integral parts of the Vertiskos Formation of the Serbomacedonian massif.

At the Gerakario area the schist - gneissic host rocks are cut by a small quartz monzonite porphyry stock. Sulfide mineralization consists of chalcopyrite and pyrite in the form of stockworks and disseminations. An oxidation zone including malachite, azurite, goethite and limonite is occasionally present. Alteration is weakly developed and dominated by a K-silicate assemblage with biotite as a replacement of mafics being the main alteration mineral. In places, weak intermediate argillic alteration overprinted the K-silicate assemblage, and is characterized by sericite/illite as a replacement of plagioclase and chloritization of mafics (Apostolou and Stefanidis, 1987).

The Pontokerasia area is characterized by the presence of small domes of acid rhyolite composition, with the exception of a small one of syenitic - granitic composition cutting the host basement, which has a very limited proportion of amphibolites and ultrabasic rocks (Melidonis, 1972; Apostolou and Stefanidis, 1987). Mineralization occurs either in the form of disseminations or veins and stockworks. The first type consists of chalcopyrite, pyrite, pyrrhotite and ilmenite, and oxidation minerals as well, whereas the second type consists of pyrite, galena and sphalerite (Melidonis, 1972). The main characteristic of the area is the development of a wide pyrite - rich sericitic halo, around a central principal por-

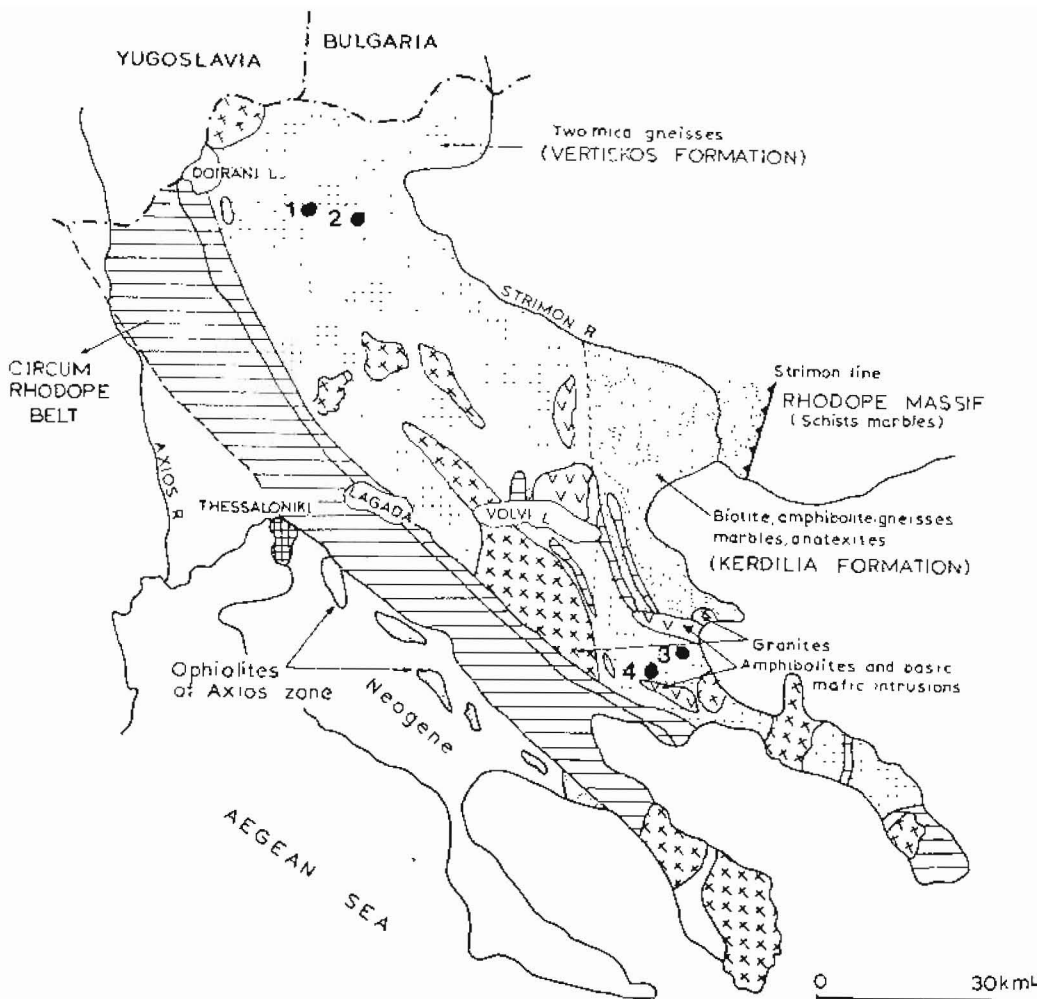


Fig. 1. Simplified geological map of the Chalkidiki peninsula (Kockel et al., 1977). 1: Gerakario; 2: Pantokrator; 3: Fissaka; and 4: Skouries

Εικ. 1. Απλοποιημένος γεωλογικός χάρτης της χερσονήσου της Χαλκιδικής (Kockel et al., 1977). 1: Γερακάριο, 2: Παντοκράτορας, 3: Φισσάκα, 4: Σκούριες

phyry stock which is K-silicate altered. In places, quartz - magnetite stockwork is also well developed.

Small porphyry stocks at the Fissoka area intrude the metamorphic basement which is dominated by amphibolites, especially at the northwest part of the area. A thin zone of supergene chalcosite enrichment developed within the near - surface part of a pyritic porphyry stock has been studied in detail by Perantonis (1988). On the eastern margin of the porphyry copper system the OP-65 prospect is located, in a transition zone between the sericitic alteration and chloritization. This alteration zone as well as the gold-bearing veins of OP-65 are controlled by N.NW striking structures (Veranis and Evangelou, 1990). Mineralization occurs also as disseminations and consists of pyrite, chalcopyrite, galena, sphalerite, arsenopyrite and tetrahydrite. Gold occurs mainly as inclusions in pyrite, hosted in quartz veins (Dimou, 1990).

At the Skouries area the metamorphic basement is intruded by a porphyry stock, the dimensions of which are approximately 180 m north - south, 200 m east - west, and its depth extends to at least 700 m below surface, indicating a pipe like shape (Zachos, 1963; Perantonis, 1982). The Skouries intrusion has been described as syenite or diorite by Papadakis (1971), whereas Perantonis (1982) characterizes that as granite.

Sulfide and oxide mineralization mainly consists of chalcopyrite, pyrite, bornite, chalcocite and magnetite in the form of veins, stockworks and disseminations. Native gold is present commonly as small inclusions in chalcopyrite. The predominant alteration type is silicification, with quartz being the most common vein and vein-controlled alteration mineral. Potassic and phyllic alteration types are also present with phyllic being less common than the potassic one, whereas propylitic is very sparse.

ANALYTICAL METHODS AND RESULTS

Representative samples of mineralized porphyries and altered host rocks (amphibolites) from the areas of Gerakario, Pontokerasia, Fissoka -including samples from OP-65- and Skouries were analyzed initially for major and common trace elements by XRF (Table 1). Subsequently, selected samples were analyzed for palladium, platinum and gold at the X-ray Assay Laboratories, Ontario, Canada, using Lead Fire Assay/ICP method. Other trace elements were analysed using Atomic Absorption Spectroscopy.

Precious and base metal concentrations are given in Table 2. These results provide only a preliminary characterization of the geochemical environment since the present study is in progress. Although the Fissoka and Skouries intrusions have some common geochemical features the former differs in having lower Cu and Pd content in both porphyry and amphibolite basement, even in the local Au-bearing mineralization, which contains unusually high values (more than 180 ppm Au) at OP-65. Also, the Cu-Au mineralization at Gerakario differs from that of Skouries in having low Pd content, while the Pontoker-

Table 1. Chemical analyses of major oxides (wt%) and trace element concentrations (ppm) of porphyry copper intrusions, amphibolites (*) and Au-bearing zone (**)

	Gerakario			Pontokerasia		Fissoka			Skouries			
	1a	1b	1c	2a	2b	3a*	3b*	3e**	4a*	4b	4c	4e
SiO ₂	70.50	66.50	60.50	64.00	62.50	66.00	53.00	62.00	59.50	66.00	72.95	
Al ₂ O ₃	14.70	15.60	14.50	15.90	15.10	22.00	11.10	8.70	11.40	11.00	9.35	
Fe ₂ O ₃	1.40	1.70	8.40	4.25	5.80	8.45	15.80	15.70	9.65	10.30	7.29	
MnO	0.95	1.05	1.70	0.95	0.70	2.60	3.05	1.05	2.00	1.25	0.48	
CaO	1.70	1.90	2.50	1.70	1.70	2.30	3.05	2.20	3.00	2.20	0.34	
MnO	0.02	0.02	0.02	0.03	0.02	0.04	0.03	0.03	0.04	0.07	0.01	
TiO ₂	0.27	0.32	0.28	0.42	0.28	0.47	0.35	0.25	0.37	0.01	0.22	
Na ₂ O	0.57	0.47	2.50	0.36	1.90	0.37	0.30	0.18	2.45	2.95	0.73	
K ₂ O	6.00	5.70	1.70	8.30	5.60	1.95	2.05	2.20	3.90	4.10	3.40	
P ₂ O ₅	0.04	0.01	0.05	0.07	0.11	0.01	0.04	0.03	0.01	0.01	0.06	
L.O.I.	2.40	3.60	3.70	2.60	3.40	6.20	10.30	7.00	1.20	1.00	2.40	
Ba	1030	1520	1980	1380	940	820	370	88	200	1250	1160	
Co	3	3	26	8	13	16	40	30	22	18	15	
Cr	10	11	28	10	16	52	136	190	300	26	14	
Ga	17	18	17	17	16	8	15	130	12	13	13	
Nb	10	11	13	10	11	8	8	6	12	7	4	
Ni	9	12	16	7	23	68	70	32	300	34	20	
Sr	100	130	190	300	290	16	30	88	115	650	250	
Tb	13	13	20	51	44	4	14	130	10	54	45	
U	3	2	6	38	20	2	1	2	3	7	4	
V	45	52	72	68	80	110	180	200	110	80	90	
Zr	50	64	50	62	64	22	33	55	40	13	13	
Zr	Ψηφιακή Βιβλιοθήκη Θεόφραστος - Τμήμα Γεωλογίας, Α.Π.Θ. 200											

Mineralogical composition of the samples

Gerakario

- 1a: quartz, plagioclase, muscovite
- 1b: quartz, orthoclase, muscovite, augite, illite
- 1c: quartz, albite, muscovite, chlorite, augite,

Pontokerassia

- 2a: quartz, plagioclase, muscovite, kaolinite
- 2b: quartz, albite, orthoclase, muscovite, chlorite

Fissoka

- 3a: quartz, amphibole, chlorite, muscovite
- 3b: quartz, albite, chlorite, muscovite, biotite
- 3e: quartz, orthoclase, muscovite, biotite

Skouries

- 4a: quartz, amphibole, chlorite, kaolinite,
- 4b: quartz, orthoclase, muscovite, chlorite
- 4c: quartz, orthoclase, albite, kaolinite, azurite, mala-
chite, hematite
- 4d: quartz, malachite, azurite, hematite, magnetite

Table 2: Precious and base metal concentrations of porphyry copper systems, Chalkidiki peninsula.

	ppb			ppm					
	Au	Pt	Pd	Ag	Te	As	Cu	Pb	Zn
1a. Gerakario	420	<10	2	0.3	<0.02	12	10000	50	15
b. "	1100	<10	4	1.0	0.03	13	21500	38	13
c. "	85	10	2	0.7	<0.02	12	26500	40	100
2a. Pontokerasia	90	10	1	0.6	0.03	12	10000	30	14
b. "	18	10	1	0.5	<0.02	14	19000	60	50
c. "	70	10	2	0.6	0.03	16	12500	70	60
3a. Fissoka	12	<10	<2	0.3	<0.02	14	120	30	20
b. "	45	10	3	0.8	<0.02	18	200	50	150
c. "	13	<10	<1	0.5	<0.02	13	400	40	120
d. "	4100	20	5	0.9	8.1	2200	300	2600	400
e. "	183000	40	50	28.0	8.8	21000	610	3200	200
4a. Skouries	670	<10	10	0.9	0.3	6	500	40	100
b. "	900	<10	70	3.5	0.7	2	5000	910	600
c. "	2000	5	260	1.2	1.5	1	18000	26	11
d. "	7200	10	480	8.7	2.7	2	23000	26	10

asia exhibits both low Au and Pd contents (Table 1).

It is noticeable that Au-rich samples from Fissoka show relatively high Te and Ag content. The strong positive interelement correlation ($r > 0.8$) between Au, Ag, Pd and Te at Skouries reported by Eliopoulos and Economou - Eliopoulos (1991), seems to be supported by the present analytical data (Tables 2 & 3) although any interelement correlation has not been established at Fissoka.

Table 3. Correlation matrix for precious and base metals from the Chalkidiki porphyry intrusions (data from Table 2)

	Au	Pt	Pd	Ag	Te	As	Cu	Pb	Zn
Au	1.00								
Pt	.94	1.00							
Pd	.01	-.11	1.00						
Ag	.93	.94	.21	1.00					
Te	.71	.84	.16	.87	1.00				
As	.99	.96	-.03	.94	.75	1.00			
Cu	-.25	-.37	.41	-.23	-.29	-.29	1.00		
Pb	.75	.90	-.09	.86	.94	.80	-.42	1.00	
Zn	.13	.29	-.15	.27	.39	.17	-.42	.57	1.00

DISCUSSION

Precious metal content and distribution in ores is still a subject of debate. They depend on complex interrelated factors such as their source, mobilization, transport and deposition.

Combined lead and strontium isotopes on porphyry intrusions and related sulfide mineralizations from the Serbomacedonian massif may suggest an extensive chemical interaction of magmas with upper crustal rocks during magma emplacement and hydrothermal solutions may have played an essential role in the mineralization (Frei, 1990). Base and precious metal mineralization could be related either with porphyries, representing a late magmatic phase, and/or wallrock alteration as well. Gold ± copper in porphyry type deposits which accompany quartz stockworks is considered to be introduced with K-silicate alteration, which is commonly overprinted by intermediate algalic assemblages (Sillitoe, 1991). The available structural mineralogical and geochemical data from the studied porphyry intrusions and the presence of a narrow zone of erratic gold values (more than 1000 ppm Au, Veranis and Evangelou, 1990) in a spatial association with shear zone and amphibolites at OP-65, suggest that regional structures resulting from the deformation did provide the necessary permeability for hydrothermal fluids responsible for mineralization. In contrast, the Gerakario and Pontokerasia intrusions with a relatively weak silicification and small proportion of amphibolites among country rocks, compared to Skouries and OP-65 as well,

are accompanied by a much lower precious metal content. Thus, the host basement of the porphyry copper intrusions, in the crystalline basement of the Vertiskos Formation including amphibolites (metabasic rocks) may have contributed precious metals (such as Pd, Pt, Au) to the hydrothermal system.

Besides source, late stage changes in fluid, pH, Eh, salinity and activity of sulphur during evolution of hydrothermal system and by discharge through previously deposited early stage sulfides also may cause remobilization of precious metals to more distal and shallow locations in the whole system. The mineralogical and geochemical data from the Skouries and Fissoka porphyry copper systems indicate an enrichment of mineralized altered rocks in Au, Ag, B, Bi, Te and Se (Dimou, 1990; Eliopoulos and Economou - Eliopoulos, 1991). In addition, a strong positive interelement correlation ($r > 0.83$ between Au, Ag, Pd and Te coupled with their association with silica at Skouries are considered to suggest a similar behavior of these metals during their transportation and deposition and a relationship with silicification. Such an interelement correlation seems to be supported by analytical data from more porphyry intrusions (Tables 1 & 2) with the exception of palladium which may suggest a different behavior during mobilization and/or deposition.

Theoretical - experimental considerations by Hemley et al., (1986) indicate that base metals and silver are mobilized and transported mainly as soluble chlorides over a wide temperature range, in weakly acidic and relatively reduced solutions.

High temperatures ($> 300^{\circ}\text{C}$) and high salinity would favor chloride complexes of gold and copper, whereas low ones ($< 300^{\circ}\text{C}$) would favor chloride complexes of silver, zinc and lead and the bisulfide complex of gold (Barnes, 1979; Seward, 1984; Romberger, 1988). The majority of porphyry copper deposits from around the world contain fluid inclusions suggesting relatively high temperature ($300^{\circ} - 500^{\circ}\text{C}$) and high salinity, whereas cooler and more dilute fluids seem to have been responsible for Au and Ag mineralization around, above and superimposed on porphyry copper deposits (Sillitoe, 1987; Huston and Large, 1989).

Fluid inclusions data on Au-bearing quartz veins from OP-65, Fissoka, provided by Mitsaki - Hafner (1991) and information available from sulfide - platinum group mineral and Au - Ag telluride assemblages (Tarkian et al., 1992) can be used to constrain the physical and chemical conditions at the time of precious metal deposition in the porphyry copper intrusions. The presence of merenskyite as the only Pd-telluride in the studied samples from Skouries may suggest a formation temperature of $335^{\circ} - 490^{\circ}\text{C}$ (Harney and Merkle, 1990; Nyman et al., 1990).

Particularly, Nyman et al., (1990) on the basis of fluid inclusions studies indicate that high salinity inclusions associated with propylitic and phyllic alteration assemblages support PGE transport as chloride complexes at a minimum temperature of 335°C . In addition, the presence of sylvanite, in a close intergrowth with merenskyite may suggest a maximum temperature of formation at $350^{\circ} - 360^{\circ}\text{C}$ (Cabri, 1965; Tarkian et al., 1992).

Mitsaki - Hafner (1991) reported two types of fluid inclusions at OP-65, Fissoka: a) inclusions with a $200^{\circ} - 300^{\circ}\text{C}$ homogenization temperature, salinity ranging from 31 to 39 wt% equiv. NaCl and

corresponding pressure 30 bars, and b) inclusions with 300° - 350° C homogenization temperature, salinity 10 - 23 wt% equiv. NaCl and corresponding pressure 100 bars. Therefore, the presence of merenskyite and sylvanite at Skouries, suggesting a range of temperature between 335° and 490° C, and the fluid inclusions data on Fissoka indicating a homogenization temperature ranging from 200° to 350° C, may provide evidence for a higher temperature during the deposition or re-deposition of sulfides in the former than along the Au-bearing fault zone at OP-65. Therefore, the oxide - silicate mineralogical composition (kaolinite, magnetite/hematite, goethite) provides evidence for the influx of acidic, oxidized fluids during precious metal deposition at Skouries while a chloride gold complex rather than bisulphide seems to be favored. In the case of the OP-65 prospect the fluid inclusions data may suggest that transport of gold was favored as bisulfide complex in more reduced solutions, circulated in the basement which is dominated by amphibolites (metabasic rocks) with a probable participation of carbonaceous sediments (Seward, 1984; Romberger, 1988).

The low content and lack of any correlation between Pd and other precious metals may suggest that the mechanism by which the Pd (and Pt) is incorporated into fluids and deposition may be quite different from that of gold in the case of OP-65. Palladium and Pt may be mobile as chloride, hydroxide or bisulphide complexes, depending on pH, PO₂, temperature and ligand concentrations (Mountain and Wood, 1988). Theoretical models and natural examples support that PGE in association with propylitic and phyllic alteration assemblages, were probably transported by chloride complexes, the minimum deposition temperature being 335° C (Nyman et al., 1990). Therefore, the low Pd content at OP-65 may reflect conditions not favorable for its mobilization.

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