Tellurides and bismuth sulfosalts in gold occurrences of Greece: mineralogical and genetic considerations

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Summary: Pre-Tertiary to Tertiary gold deposits in Greece occur in a wide range of genetic types including volcanic massive sulfides, orogenic, intrusion-hosted, skarn, manto-, porphyry- and epithermal-type ores. Almost all of the gold mineralization hosts various Bi-tellurides and Bi-sulfosalts, which in addition to Au-Ag-tellurides, are indicators of specific physicochemical conditions of ore formation. The Bi-bearing mineralization can be subdivided into three groups regarding their spatial relationship to gold: (a) mineralization which lacks tellurides but includes Bi-sulfosalts and native gold, (b) mineralization where Bi-tellurides of the reduced-type (joseite-A, joseite-B, pilsenite) accompany Bi-sulfosalts, native bismuth and native gold, (c) deposits/prospects where Au-Ag-tellurides are abundant and Bi-tellurides and Bi-sulfosalts are absent. Bi-telluride and -sulfosalt mineralization in Greece underwent several stages of remobilization during successive accretionary episodes in active continental margins and arc terranes during the Carboniferous to Pleistocene.

Key Words: Gold ores, tellurides, gold, silver, sulfosalts, bismuth, Greece

1. INTRODUCTION

Primary gold deposits in Greece occur in a wide range of genetic types, comprising VMS deposits of pre-Tertiary age, those related to metamorphic or deformation processes, ores related to Mesozoic-Miocene shearing, and deposits related to Tertiary-Quaternary magmatic activity (skarn and manto-type, porphyry-type, vein-type, epithermal-type) (Eliopoulos and Kalogeropoulos, 1985; Bitsios, 1989; Skarpelis, 2002; Arvanitidis, 2003). The Rhodope metallogenic ore province (Melfos et al., 2002) including the Servomacedonian-Rhodope massifs (Mposkos and Krohe, 2001, Reischmann and Kostopoulos, 2007) host porphyry Cu-Mo-Au and epithermal Au deposits, Pb-Zn (Ag-Au) carbonate replacement, skarn and vein deposits of Late Eocene to Oligocene age, epithermal Au-Ag deposits and shear zone hosted mesothermal to low sulfidation epithermal gold-silver deposits not related to intrusive activity (e.g. Ada Tepe in Bulgaria) (Ashworth et al., 1988; Nesbitt et al., 1988; Arvanitidis and Constantinides, 1989; Hellingwerf et al., 1994; Arvanitidis et al., 1996; Marchev et al., 2005; Eliopoulos and Chryssoulis, 2005). Both the shear zone Au-Ag deposits and the low sulfidation epithermal Au-Ag deposits lie close to or within metamorphic domes and may be related to post collision orogenic collapse and exhumation of these structures (Marchev et al., 2005). The Attico-Cycladic metamorphic belt comprises a number of types of mineralization, which are in part genetically related to arc-related magmatic rocks, and in part controlled by exhumation structures of high-pressure units in a back-arc setting (Skarpelis, 2002; Neubauer, 2005). The Attico-Cycladic Massif (Okrusch andd Bröcker, 1990) contains base- and precious-metal skarn, intrusion-related, orogenic and epithermal mineralization in south Evia, Sifnos, Mykonos, Tinos and Kythnos Islands (Alexouli-Livaditi, 1978; Vavelidis, 1997; Tombros et al., 2004; Neubauer, 2005; Spry et al., 2006). At Sifnos Island, orogenic ore formation occurred during extensional kinematic conditions, when the Sifnos metamorphic core complex reached a near-surface level (Neubauer, 2005).

Despite several mineralogical studies of gold mineralization in Greece, an integrated documentation of telluride and Bi-sulfosalt bearing ores has yet to be done. The aim of the present study is to review all mineralogical data concering tellurides and Bi-sulfosalts from various gold deposits/prospects in Greece (Fig. 1), to present new information concerning the location of bismuth sulfosalts and tellurides, and to discuss their origin.

Fig. 1. Location map of tellurides and Bi-sulfosalts occurrences in Greece

2. Bi SULFOSALTS AND NATIVE BISMUTH

Bismuth sulfosalts are widespread in almost all the gold deposits in Greece. In the Oligocene porphyry-type mineralization of Pagoni Rachi/W. Thrace, Bi-sulfosalts spatially associated with tellurides are enriched in late-stage quartz-carbonate veins, which overprinted chalcopyrite-molybdenite-bearing quartz stockworks (Voudouris and Arikas, 2003; Voudouris and Melfos, 2006). The bismuthinite derivatives (aikinite to lindströmite) and lillianite homologues are closely related to hessite and stützite and occur as tiny inclusions within chalcopyrite, galena, sphalerite and tennantite. New observations in Pagoni Rachi indicate intergrowths between wittichenite $(Cu_{2.92}Ag_{0.06})_{2.97}Bi_{0.99}S_{3.02}$, a berryite-like sulfosalt, $(Cu_{1.33}Ag_{0.66})_{1.99}Bi_{2.29}Pb_{0.96}S_{5.41}$, hessite, bornite and galena (Figs. 2a,b). Bismuthinite was reported by Melfos et al. (1996, 2002) from the adjacent Miocene(?) microgranite-hosted porphyry Cu-Mo system of Maronia/W. Thrace. Intrusion-hosted sheeted quartz veins and stockworks, rich in molybdenite and scheelite from the Kimmeria Oligocene granodioritic body, which outcrop east of Xanthi/Rhodope massif, contain minor emplectite, wittichenite and aikinite (Walenta and Pantzartzis, 1969). The Stypsi prospect, a newly discovered porphyry Cu±Mo mineralization on Lesvos Island contains bismuthinite (Voudouris and Alfieris, 2005). Bismuthinite, galenobismutite, cosalite and aikinite were reported from the Oligocene skarn-carbonate replacement polymetallic Madem Lakkos sulfide deposit, Chalkidiki/northern Greece, in marbles of the Servomacedonian massif (Nebel et al., 1991; Gilg 1993). In a similar style of mineralization, at Kamariza mines in the Lavrion area (Attika), a close spatial association exists among native gold, native bismuth and bismuthinite (Fig. 2c) (Voudouris, 2005). Several other Bi-Cu-Pb-Ag sulfosalts, including bismuthinite, lillianite homologues, Cu-matildite, aikinite, Ag-aikinite, mummeite, emplectite and wittichenite occur as inclusions in chalcopyrite (Voudouris and Economou-Eliopoulos, 2003). In the high-intermediate sulfidation epithermal assemblages of St Philippos, St Demetrios and Pefka in W. Thrace/Greece, the Bi-sulfosalts kirkiite, bismuthinite, cosalite, aikinite and hammarite have been reported (Dimou, 1993; Voudouris et al., 2006; Voudouris, 2006). New paragenetic relationships from the Perama Hill high-intermediate sulfidation epithermal system (Voudouris et al., 2007, this volume) demonstrates the presence of bismuthinite related to enargite and covellite, as well as of lillianite homologues associated with tennantite, galena and precious metal tellurides. Bismuth sulfosalts spatially associated with Bi-tellurides, native bismuth and native gold have been reported from several pre-Tertiary to Tertiary shear-zone-controlled (orogenic according to Groves et al., 1998) and/or metamorphosed gold deposits/mineralization in northern Greece. The ore occurs in both folded/deformed and undeformed varieties as replacement-type, irregular pods or as lenses developed in marble-gneiss contacts, in gneisses, amphibolites, marbles, and as quartz veins crosscutting the above lithologies of the Rhodope-Servomacedonian massifs (Mposkos, 1983; Kalogeropoulos et al., 1986, 1991; Vavelidis, 1994; Hellingwerf et al., 1994; Thymiatis, 1995; Chatzikirkou, 2003; Eliopoulos and Chryssoulis, 2005). Folded and deformed massive to disseminated mineralization is associated with quartz segregations and pegmatoid bodies crosscutting at low angles, or are parallel, to the schistosity of mica gneisses, schists and amphibolites, at Paliomilos, Chalkoma and Paliopyrgos/Stanos areas in Chalkidiki, at Koronouda, Paliomylos and Laodikino in Kilkis. In the Stanos/Chalkidiki area, the Paliomilos and Chalkoma mineralization contains bismuthinite, cosalite, gladite, pavonite and emplectite associated with native gold, as well as native bismuth, native gold and an intergrowth of cosalite/joseite-A included in late chalcopyrite (Kalogeropoulos et al., 1991). Investigations during the present study from Chalkoma mineralization indicated a close spatial relationship between electrum, native bismuth and gustavite ($Ag_{0.92}Cu_{0.05}Pb_{1.20}Bi_{2.82}S_{5.34}$) in chalcopyrite veinlets crosscutting pyrite (Fig. 2i). Our study of mineralization at Paliomilos indicates complex intergrowths among lillianite homologues ($Ag_{0.69}Pb_{1.70}Bi_{2.60}S_{5.26}$), galena and native bismuth (Fig. 2g). Electrum is associated with lillianite homologues (Fig. 2h). In Au-Ag bearing mineralization in the Paliopyrgos area, aikinite is intergrown with native gold, friedrichite, and galena, the latter two minerals being included in chalcopyrite (Vavelidis and Tarkian, 1995). In the Koronouda and Paliomylos areas, quartz segregations and pegmatoid bodies, hosted within gneisses and schists of the Vertiskos series, contain bismuthinite, aikinite and native bismuth in addition to Bi-tellurides and Au-Ag-tellurides as inclusions in chalcopyrite (Vavelidis 1994; Vavelidis et al., 1996; Melfos et al., 2001). During the present study at Koronouda mineralization, tiny inclusions of native Bi occur in galena and hessite and along their grain boundaries (Fig. 2k, l). Bi-sulfosalts are also common constituents in several gold-bearing post-metamorphic quartz veins throughout the Servomacedonian-Rhodope massif: bismuthinite, cosalite and native gold included in chalcopyrite were reported at Palea Kavala (Vavelidis et al., 1997), bismuthinite intergrown with tetradymite and isolated grains of hessite are included in chalcopyrite from a Bi-Te-Ag-bearing Cu prospect in the Panagia area (Thasos island) (Vavelidis et al., 1995) and bismuthinite, native bismuth, cosalite, joseite $Bi_4(S,Te)_3$, and lillianite from Angistron Mt (W. Rhodope massif) (Katirtzoglou et al., 1994). Recently studied samples from Palea Kavala (Chalkero locality), revealed a close spatial association among tetradymite, pyrite and bismuthinite ($Bi_{2.06}Sb_{0.04}S_{2.87}Te_{0.02}Se_{0.01}$) in milky quartz from detachment-related mineralization. Finally, bismuthinite, emplectite, aikinite and wittichenite accompanying tetradymite and hessite, are described from shear zone-hosted ores at the Eptadendron (Aberdeen)-Rachi areas in eastern Rhodope massif (Chatzikirkou, 2003; Chatzikirkou and Michailidis, 2004).

3. Bi TELLURIDES

Bi-tellurides are common in the pre-Tertiary to Tertiary orogenic and metamorphosed gold deposits/mineralization in northern Greece. Joseite-A is intergrown with cosalite from Paliomilos and Chalkoma Stanos/Chalkidiki (Kalogeropoulos et al., 1991), pilsenite-galena-hessite intergrowths and tellurobismuthinite, werhlite are included in chalcopyrite in the Koronouda area (Mposkos, 1983; Vavelidis 1994, Vavelidis et al., 1996), tetradymite is intergrown with bismuthinite and hessite at Panagia area (Thasos island) (Vavelidis et al., 1995), joseite $Bi_4(S,Te)_3$ occurs at Mt. Angistron (Katirtzoglou et al. 1994) and tetradymite occurs in the Eptadendron-Rachi areas in eastern Rhodope Massif (Chatzikirkou, 2003; Chatzikirkou and Michailidis, 2004). In addition, the following Bi-tellurides were identified in the present study: tetradymite ($Bi_{2.06}Te_{1.88}S_{1.03}$) in the Chalkero/Palea Kavala (Fig. 2j), pilsenite ($Bi_{3.97}Te_{3.03}$) included in chalcopyrite (Fig. 2e) in Laodikino, joseite-B ($Bi_{3.92}Au_{0.05}Te_{1.93}Se_{0.04}S_{0.98}$) associated with pilsenite ($Bi_{3.80}Te_{2.85}S_{0.35}$) and tellurobismuthite ($Bi_{2.05}Te_{2.78}S_{0.17}$) surrounding hessite in the Koronouda, area (Fig. 2k).

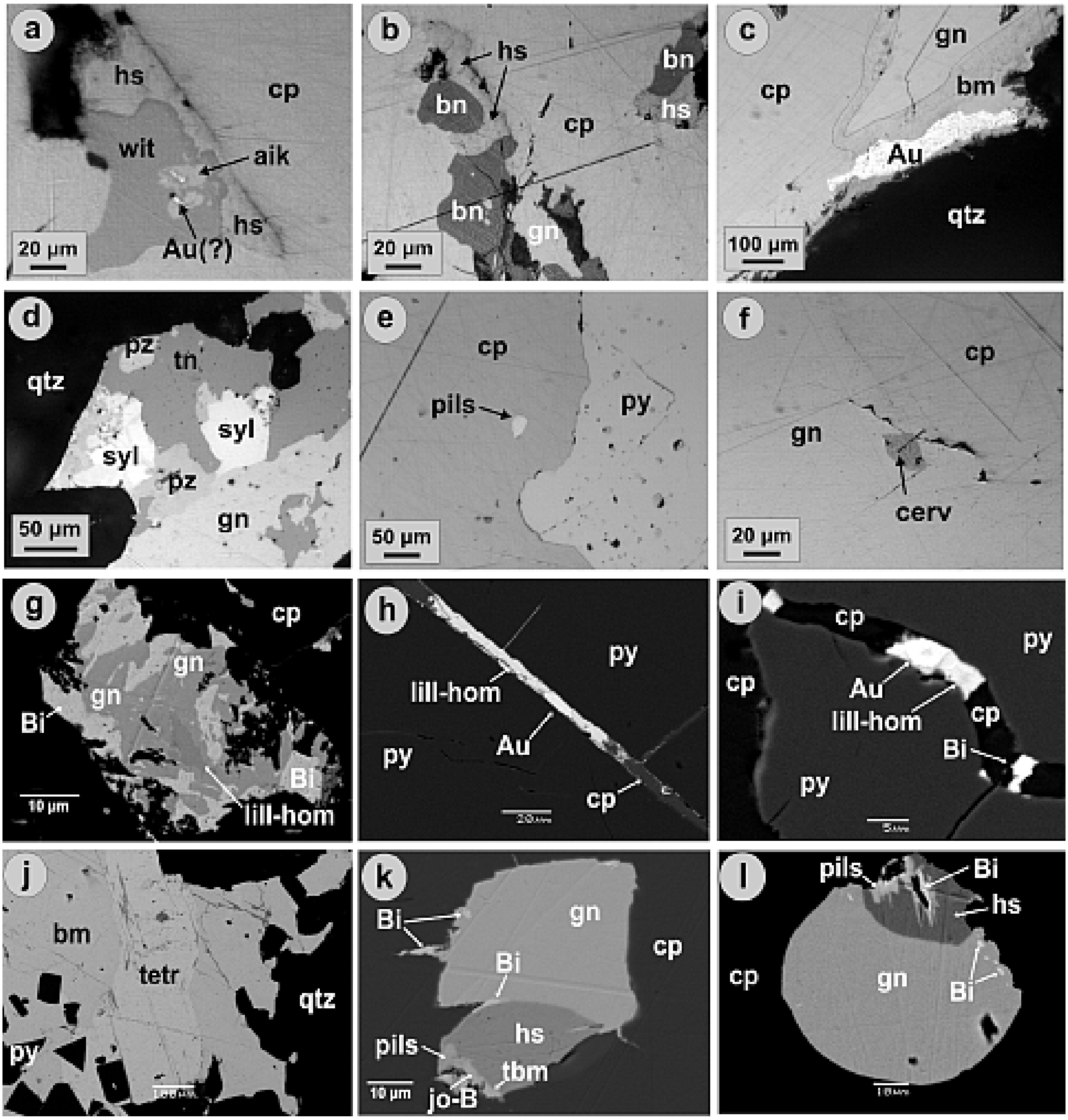

Fig. 2. Photomicrographs demonstrating the ore paragenesis of various telluride-Bi sulfosalt bearing mineralization in Greece: (a, b) Intregrowth of wittichenite (wit), aikinite (aik), hessite (hs), bornite (bn), galena (gn) and native gold (Au?) included in chalcopyrite (cp), Pagoni Rachi porphyry Cu-Mo; (c) native gold (Au) associated with bismuthinite (bm), galena (gn) and chalcopyrite (cp) from the Kamariza/Lavrion carbonate replacement deposit; (d) petzite (pz), sylvanite (syl) intergrowths with tennantite (tn) and galena (gn), Perama Hill epithermal deposit; (e) Pilsenite (pils) included in chalcopyrite (cp); pyrite (py) is also present, Laodikino metamorphosed VMS-orogenic deposit; (f) Cervelleite (cerv) included in galena (gn) at the grain boundaries to chalcopyrite (cp), Kallianou orogenic gold deposit; (g) lillianite homologue (lill-hom), galena (gn) and native bismuth (Bi) included in chalcopyrite (cp), Paliomylos metamorphosed VMS-orogenic deposit; (h) lillianite homologue (lill-hom), native gold (Au) and chalcopyrite (cp) filling a fissure in pyrite (py), Paliomylos metamorphosed VMS-orogenic deposit; (i) lillianite homologue (lill-hom), native gold (Au), native bismuth (Bi) and chalcopyrite (cp) filling a fissure in pyrite (py), Chalkoma metamorphosed VMS-orogenic deposit; (j) tetradymite (tetr) and bismuthinite (bm) surrounding pyrite (py) and included in quartz (qtz), Chalkero orogenic mineralization; (k,l) Blebs of galena (gn) within chalcopyrite (cp) spatially associated with hessite (hs), native bismuth (Bi), pilsenite (pils), joseite-B (jos-B) and tellurobismuthite (tbm), Koronouda metamorphosed VMS-orogenic deposit (a to f: reflected light, g to l: BSE images)

Tellurobismuthite has previously been described in the syenite-hosted porphyry Au-PGE system at Skouries, in addition to gold-silver tellurides and the Pd-telluride merenskyite (Eliopoulos and Economou-Eliopoulos, 1991; Tarkian et al., 1991; Economou-Eliopoulos and Eliopoulos, 2005). Tellurobismuthite and tetradymite occur in association with bismuth sulfosalts and Au-Ag-tellurides in the porphyry-epithermal assemblages of St Demetrios and Pefka prospects in W. Thrace, Greece (Dimou et al., 1994; Voudouris and Arikas, 1994; Voudouris et al., 2006; Voudouris, 2006). Finally, Leleu et al. (1973) reported the presence of tetradymite and native Bi from the Plaka skarn deposit at Lavrion (Attika).

4. Au-Ag TELLURIDES AND Ag SULFO-TELLURIDES

The potassic alteration core of the Skouries porphyry deposit in NE Chalkidiki contains the tellurides hessite, sylvanite and calaverite in association with native gold at grain boundaries between chalcopyrite and bornite (Tarkian at al., 1991). Hessite and stützite in the Pagoni Rachi/W. Thrace, porphyry-Cu-Mo system, occur in contact with Bi-sulfosalts, bornite and galena (Fig. 2a, b) in late-stage veins (Voudouris and Arikas, 2003; Voudouris and Melfos, 2006, this study). In the Fakos porphyry Cu±Mo prospect, on Limnos island, electrum, hessite, petzite and an unnamed Ag-sulfotelluride, occur in late quartz-carbonate veins overprinting tourmaline-sericite veins, as well as quartz stockworks hosted within a Miocene monzonitic body (Voudouris and Alfieris, 2005; Voudouris 2006). Hessite, altaite and electrum were identified as inclusions in galena from the Thermes ore-field, which represents the southward extension of the Madan deposit (Bulgaria) and comprises both fault-controlled vein-type Pb-Zn and stratabound mineralization within marbles of the Rhodope metamorphic complex (Arvanitidis and Dimou, 1990). Au-Ag-tellurides occur throughout the Tertiary precious metal high-intermediate sulfidation epithermal assemblages in W. Thrace, Greece (Dimou et al., 1994; Voudouris and Arikas, 1994; Michael et al., 1995; Arikas and Voudouris, 1998; Border et al., 1999; Mc Alister et al., 1999; Skarpelis et al., 1999; Lescuyer et al., 2003; Melfos et al., 2003; Voudouris et al., 2006; Voudouris, 2006). Hessite, petzite, sylvanite and stützite in association with altaite, coloradoite, native tellurium and goldfieldite, accompany Bi-tellurides and Bi-sulfosalts in the St Demetrios, Pefka, Mavrokoryfi and Kassiteres prospects/deposits. Calaverite was reported, but not quantitatively documented, from the Viper deposit/Sappes area (Shawh and Constantinides, 2001). New paragenetic relationships from the Perama Hill high-intermediate sulfidation epithermal system (Voudouris et al., 2007, this volume) suggest the abundance of hessite, petzite, stützite, sylvanite, krennerite and altaite (Fig. 2d) with late-stage barite-tennantite-galena veins crosscutting earlier high sulfidation enargite-pyrite ores. In the orogenic Au-Ag bearing mineralization in the Paliopyrgos area, Vavelidis and Tarkian (1995) identified hessite intergrown with gold and aikinite as small inclusions in chalcopyrite. The mineralization at Koronouda area contains galena-hessite-pilsenite intergrowths and hessite, petzite, sylvanite and native gold included in chalcopyrite (Mposkos, 1983; Vavelidis, 1994, Vavelidis et al., 1996). However, during the current study only hessite, showing curvilinear boundaries to galena, was detected (Fig. 2k, l). Similarly, in the Paliomylos/Kilkis area, hessite is reported in association with bismuthinite derivatives and with galena in chalcopyrite (Melfos et al., 2001). Isolated grains of hessite included in chalcopyrite occur in the Bi-Te-Ag-bearing Cu mineralization at Panagia (Thasos island) (Vavelidis et al., 1995). Chatzikirkou (2003) and Chatzikirkou and Michailidis (2004), reported on the presence of hessite intergrown with tetradymite and included in chalcopyrite from the Eptadendron-Rachi areas in eastern Rhodope massif.

In the submarine epithermal mineralization of Profitis Ilias (Milos island) at the active south Aegean volcanic arc, Alfieris and Voudouris (2006) and Alfieris (2006) documented the presence of a hessite-petzite-altaite association with native gold/electrum within veinlets crosscutting the Upper Pliocene Profitis Ilias rhyodacitic cryptodome. An extremely rich suite of tellurides, was reported by Tombros (2001) and

Spry et al. (2006), from the orogenic Au-Ag-Te mineralization at Panormos Bay (Tinos island) The marble-hosted Panormos Bay vein system, is characterized by hessite, sylvanite, altaite, native tellurium, stützite, Cu-bearing cervelleite, melonite, rickardite, vulcanite, weissite, native tellurium, rickardite, kostovite, krennerite, petzite, and calaverite. A similar style of Te-rich mineralization is present further to the north in orogenic quartz veins hosted in metamorphic rocks of the Blueschist Unit of the Kallianou area (S. Evia Island). Alexouli-Livaditi (1978) was the first person to describe the presence of sylvanite as small inclusions in pyrite from that area, and new investigations (Voudouris and Spry in prep.) indicate the presence of silver sulfotellurides in association with hessite. These are minerals of the cervelleite-group as well as unnamed minerals with composition approximating the chemical formula Ag_2CuTeS and $(Ag,Cu)_2TeS$. The sulfotellurides are enclosed in galena and occur in proximity to electrum.

5. DISCUSSION AND CONCLUSIONS

Bismuth sulfosalts and tellurides are abundant in several types ore deposits in Greece. They can be considered as pathfinder minerals for gold since they are intimately associated with gold bearing ores. The mineralization studied can be subdivided into three groups based on their association with gold: (a) deposits which lack tellurides but include Bi-sulfosalts and native gold. In this category, belong the Lavrion (manto-type), Maronia (porphyry-Cu-Mo), Kimmeria (intrusion-hosted Cu-Mo), Stypsi/Lesvos (porphyry-Cu-Mo) and some orogenic mineralization in northern Greece. (b) The second category includes mineralization where Bi-tellurides of the reduced-type (joseite-A, joseite-B, pilsenite, where Bi:X>1, X=Te, S, Se) accompany Bi-sulfosalts, native bismuth and native gold. Examples are the VMS-orogenic mineralization in Serbo-Macedonian Massif (Koronouda and Laodikino at Kilkis, Paliomilos and Chalkoma at Stanos/Chalkidiki) whereas at the Agerdeen prospect in the Rhodope massif and at the orogenic Chalkero/Palea Kavala mineralization, the tetradymite (with Bi:X<1) is present. The calcalkaline-alkaline-hosted porphyry and epithermal deposits/prospects in the Chalkidiki and W. Thrace areas contain Bi-tellurides with Bi:X<1 (tetradymite, tellurobismuthite) in association with Bi-sulfosalts and Au-Ag-tellurides, consistent with oxidizing conditions during ore formation (see Cook and Ciobanu 2002, 2004).
(c) The third category includes deposits/prospects where Au-Ag-tellurides are abundant but Bi-tellurides and Bi-sulfosalts are absent. Such deposits include the orogenic Panormos, Kallianou and the epithermal Profitis Ilias mineralization in southern Greece. However a Bi signature in Kallianou is recorded by the elevated Bi-content of electrum (up to 0.5 wt%).

The hypothesis of gold that is scavenged by a Bi-melt may be important in mineralization where formation conditions exceeded those necessary for the generation of such melts (Ciobanu et al. 2005, 2006a,b). This may the case for mineralization at Kamariza (Lavrion district), the Pagoni Rachi prospect, and the VMS-orogenic mineralization in northern Greece. The last occurrence is considered to have formed during metamorphism and/or to be post-metamorphic, which are conditions favourable for the formation of metallic melts (Tomkins et al., 2007). According to Kalogeropoulos et al. (1991) at Paliomilos and Chalkoma/Chalkidiki, the introduction of Au-Bi-Te-Cu-bearing fluids was contemporaneous with retrograde greenschist metamorphism. Based on arsenopyrite and sphalerite geothermometry-geobarometry and fluid inclusion data, Kalogeropoulos et al. (1991), Thymiatis (1995) and Vavelidis et al. (1995, 1997) concluded that the mineralization formed at 460-510 ºC/5.6 ± 0.8kb (Paliomilos and Chalkoma/Stanos), at 500 to 600 ºC/5kb (Koronouda), at 360-450 ºC/4.5-5.7kb (Laodikino). In all of the above occurrences, early VMS sulfide mineralization was affected by deformation and retrograde greenschist metamorphism. A multiple-stage model for the ore occurrences of the Eptadendron-Rachi areas in the Rhodope massif has been proposed by Chatzikirkou (2003) and Chatzikirkou and Michailidis (2004). For the Kallianou mineralization, Theophilopoulos and Vakondios (1978) proposed a remobilization of metals from earlier volcano-sedimentary ores during retrograde greenschist facies metamorphism. The abundance of sulfotellurides in the Kallianou veins may suggest a deficiency in fTe_2 necessary for the deposition of most common Au-Ag-tellurides. The Panormos system started in much the same way as Kallianou but evolved through time towards more Te and Au rich minerals.

There is higher proportion of silver tellurides relative to gold-bearing tellurides in Greece. Exceptions represent the porphyry-epithermal Viper and Perama Hill deposits in Thrace and the Panormos system in Attico-Cycladic belt. The recent discovery of tellurides in the deeper levels of Profitis Ilias deposit may suggest that arc related rhyodacitic magmas in south Aegean area are enriched in tellurium from their source regions. In general, Greece is highly enriched in tellurides, which contrasts to their paucity (but not complete absence) in neighbouring Turkey and southern Bulgaria where similar Tertiary magma generating and metallogenetic processes occurred. However tellurides and Bi-sulfosalts are abundant in the Late Cretaceous Srednogorie-Pontides belt, which host significant porphyry and VMS-epithermal ores (Bogdanov et al., 2004; Kouzmanov et al., 2005; Bogdanov and Filipov, 2006). Telluride mineralization in Greece underwent several stages of remobilization from earlier pre-Tertiary to Tertiary events during successive accretion episodes in active continental margins and arc terranes from the Carboniferous to the Pleistocene.

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