Ore Formation During Jurassic Subduction of the Tethys Along the Eurasian Margin: Constraints from the Kapan District, Lesser Caucasus, Southern Armenia

MEDERER, Johannes, et al.

Abstract

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Ore Formation During Jurassic Subduction of the Tethys Along the Eurasian Margin: Constraints from the Kapan District, Lesser Caucasus, Southern Armenia

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Abstract
The Kapan mining district in the southernmost Lesser Caucasus is one of the few locations along the central Tethyan metallogenic belt where ore-forming processes were associated with magmatic arc growth during Jurassic Tethyan subduction along the Eurasian margin. Three ore deposits of the Kapan district were investigated in this study: Centralni West, Centralni East, and Shahumyan. The ore deposits are hosted by Middle Jurassic andesitic to dacitic volcanic and volcanoclastic rocks of tholeiitic to transitional affinities below a late Oxfordian unconformity, which is covered by calc-alkaline to transitional Late Jurassic-Early Cretaceous volcanic rocks interlayered with sedimentary rocks.

The mineralization consists of veins, subsidiary stockwork, and partial matrix replacement of breccia host rocks, with chloropyrite, pyrite, tennantite-tetrahedrite, sphalerite, and galena as the main ore minerals. Centralni West is a dominantly Cu deposit, and its host rocks are altered to chlorite, carbonate, epidote, and sericite. At Centralni East, Au is associated with Cu, and the Shahumyan deposit is enriched in Pb and Zn as well as precious metals. Both deposits contain high-sulfidation mineral assemblages with enargite and hizonite. Dickite, sericite, and diaspore prevail in altered host rocks in the Centralni East deposit. At the Shahumyan deposit, phyllic to argillic alteration with sericite, quartz, pyrite, and dickite is dominant with polymetallic veins, and advanced argillic alteration with quartz-alunite ± kaolinite and dickite is locally developed.

Introduction
The Lesser Caucasus is a segment of the Tethys belt (Jankovic, 1977, 1997; Richards, 2015) that records a complex geologic evolution from the Jurassic to the Cenozoic, including several subduction, accretion, and collision events (Golonka, 2004; Adamia et al., 2011; Cowgill et al., 2016; Rolland, 2017). Ore deposits were formed during its entire geodynamic evolution (Moritz et al., 2016a). The Lesser Caucasus is one of the few locations along the Central Tethyan belt that offers the possibility to study ore deposits formed during the early stages of magmatic arc construction related to Neotethys subduction along the Eurasian margin. Indeed, a limited number of ore deposits and prospects are hosted by Middle to Late Jurassic volcanosedimentary sequences of the Lesser Caucasus at the Alaverdi and the Kapan mining districts (Fig. 1; Maghakyan, 1954; Khachaturyan, 1977; Achikgiozyan et al., 1987; Zohrabyan and Melkonyan, 1999; Kekelia et al., 2004; Mederer et al., 2014; Calder et al., 2019).

The Kapan mining district of the southernmost Lesser Caucasus hosts several important past-producing base metal mines at Centralni West (formerly known as 7–10 and Katar mines) and Centralni East (formerly named 1–2, 6, and Kavart mines), and one active mine at the Shahumyan precious and base metal
Late Cretaceous volcanogenic
megar-Chord on 02 May 2019

ATB
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Murgul

Major ore districts, deposits and prospects of the Lesser Caucasus and adjoining tectonic provinces in Turkey and Iran:

- Jurassic polymetallic deposits/prospects
- Late Jurassic-Early Cretaceous porphyry Cu*, epithermal** and skarn*** deposits/prospects
- Late Cretaceous volcanogenic massive sulfide deposits/prospects
- Late Cretaceous precious and base metal epithermal deposits/prospects
- Late Cretaceous minerals and Cenozoic porphyry Cu-Mo*, epithermal** deposits/prospects (uncertain ages)
- Cenozoic volcanogenic massive sulfide deposit
- Cenozoic subduction to post-collision porphyry Cu-Mo*, epithermal**, skarn***, and sediment-hosted (Carlin type?) **** deposits/prospects
- Orogenic gold deposits/prospects

Fig. 1. Geologic map of the central part of the Tethyan belt between eastern Turkey and northern Iran, modified from Mederer et al. (2014) with additional information from Azizi and Moinevaziri (2009), Hänsig et al. (2013a, b), and Zamani and Masson (2014), and location of major ore districts, deposits, and prospects. Location of Figure 2 is centered on the Kapan region in the southernmost Lesser Caucasus. The Lesser Caucasus consists of the Somkheto-Karabagh belt along the Eurasian margin, the ophiolites of the Amasra-Sevan-Aker suture zone, and the South Armenian block. Abbreviations: ABV = Artvin-Bolnisi volcanic arc, ATB = Adjara-Trialeti belt, IAES = Izmir-Ankara-Erzincan suture, KB = Kapan block, KGF = Khustup-Giratagh fault, MOP = Meghri-Ordubad pluton. Sources of location and description of selected ore districts, deposits, and prospects: Eastern Pontides and Maridnay, Turkey: Iran West (2009); Lesser Caucasus from Moritz et al. (2016a); Iranian sources are Simmons et al. (2017) for Sungun, Mazraeh, and Saheb, Nabatian et al. (2017) for Siah Kamar, Mehrabi et al. (2016) for the Glojeh district, Daliran (2008) for the Zarsuran-Aghdarreh district, Richards et al. (2006) for Sari Guney, and Ninomao and et al. (2011) for Kharapeh and the Qoleleh-Karvian district.
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Mining in the Kapan district dates back to the mid-19th century and different state and private companies have been operating the deposits since then. Reliable production records are not available and have to be estimated. Considering all available data, at least 370,000 tonnes (t) of Cu have been mined in the Kapan district since 1953 (Wolfe and Gossage, 2009). Production in the open pit and underground workings of Centralni East ceased in 2005, and the Centralni West underground operation closed in 2008. The underground Shahumyan deposit remains the only active mine in the district (Table 1). In 2016, about 290,000 t of ore was processed, producing a total amount of 13.7 Koz of Au, 0.3 Moz of Ag, 615 t of Cu, and 2,888 t of Zn. The measured, indicated, and inferred resources of the Shahumyan deposit were estimated at 15.0 Mt at 2.7 ppm Au, 48 ppm Ag, and 0.5% Cu by Polymetal International (2017). Because of ongoing exploration and underground exploitation, the Shahumyan polymetallic deposit is more accessible and has been more systematically studied and sampled during this study.

Ore deposits hosted by Middle Jurassic rocks in the Kapan district consist of Cu-enriched massive pyrite bodies and Cu-Au and polymetallic stratiform, vein-type and stockwork-type mineralization, which have been variably interpreted as volcanicogenic massive sulfide (VMS) and porphyry-epithermal ore systems (Schmidt et al., 1985; Moon et al., 2001; Kekelia et al., 2004; Wolfe and Gossage, 2009; Mederer et al., 2014). In this contribution, we provide new constraints to discuss...

![Geologic map of the southern part of the Lesser Caucasus](https://pubs.geoscienceworld.org/segweb/economicgeology/article-pdf/doi/10.5382/econgeo.4640/4683802/4640_tt_mederer+et+al.pdf)

**Fig. 2.** Geologic map of the southern part of the Lesser Caucasus (modified from Mederer et al., 2014), showing the location of the study area and of major ore deposits and prospects of the Kapan zone and the adjacent Cenozoic Meghri-Ordubad and Bargushat plutons of the South Armenian block. A = Agarak Cu-Mo porphyry deposit, B = Bartsravan polymetallic prospect, D = Dastakert porphyry Cu-Mo deposit, K = Kadjaran porphyry Cu-Mo deposit, KGF = Khustup-Giratakh fault, P = Paragachay porphyry Cu-Mo deposit, S = Shikahogh Cu-Au-Mo prospect, SAB = South Armenian block (Gondwana derived).

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**Table 1. Summary of Main Ore Deposit Characteristics in the Kapan District**

<table>
<thead>
<tr>
<th>Deposit type</th>
<th>Age</th>
<th>Status</th>
<th>Ore grade</th>
<th>Ore deposit geometry</th>
<th>Main ore mineralogy</th>
<th>Alteration</th>
<th>Host-rock geology</th>
</tr>
</thead>
<tbody>
<tr>
<td>Centralni West</td>
<td>161.78 ± 0.79 Ma (Ar/Ar muscovite)</td>
<td>Underground operation closed 2008</td>
<td>Centralni deposits (Ar/Ag)</td>
<td>1.16% Cu</td>
<td>Chlorite, carbonate, epidote, and minor sphalerite, tennantite, and galena</td>
<td>Chalcopyrite, pyrite, and minor spilitic, tennantite-tetrahedrite, and gersdorffite</td>
<td>Middle Jurassic andesite and dacite, close to ore and open pit</td>
</tr>
<tr>
<td>Centralni East</td>
<td>144.7 ± 4.2 Ma (Re-Os pyrite isochron)</td>
<td>Underground operation abandoned in 2004</td>
<td>Centralni deposits (Ar/Ag)</td>
<td>1.16% Cu</td>
<td>Pyrite, chalcopyrite, tennantite-tetrahedrite, and minor luzonite</td>
<td>Chalcopyrite, pyrite, and minor tennantite, and galena</td>
<td>Middle Jurassic andesite and dacite, close to ore and open pit</td>
</tr>
</tbody>
</table>
| Shahumyan | 156.14 ± 0.79 Ma (Ar/Ar alunite) | Underground mining | Estimated 30,000 t mined since 1843 (Cu-Au) | 1.8 Mt at 1.53 ppm Au, 29.8 ppm Ag, 0.24% Cu, and 1.52% Zn; estimated resources in early 2017: 15.0 Mt at 2.7 ppm Au, 48 ppm Ag, and 0.5% Cu | Pyrite, chalcopyrite, tennantite-tetrahedrite, and minor sulfides | Phyllic and argillie alteration, advanced argillic alteration, and minor quartz alteration in the northeastern part of the deposit | Middle Jurassic subvolcanic quartz-dacite and dacite porphyry 

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**Note:** The above text and table are excerpts from a scientific article discussing the ore formation and mining activities in the Kapan district of Armenia.
these genetic controversies for the Kapan district. We present new descriptions of the geologic setting and the hydrothermal alteration and mineralization features of the Centralni West, Centralni East, and Shahumyan deposits. These data are discussed with new radiogenic (Pb, Sr) and stable isotope (O, C, S, H) data obtained for various gangue and ore minerals to evaluate the sources of metals and the nature of the fluids involved in the genesis of the different deposits. In the final part of this contribution, we discuss new 40Ar/39Ar and Re-Os dates of hydrothermal gangue and opaque minerals and evaluate them with respect to the regional geologic framework, crosscutting relationships, and previously published U-Pb geochronological data obtained for magmatic rocks (Mederer et al., 2013). The combined dataset strongly suggests that the different deposits were formed within a short time frame or were broadly coeval during Middle Jurassic magmatic construction.

**Geodynamic and Geologic Setting of the Lesser Caucasus**

The Lesser Caucasus is a segment of the Tethyan orogenic belt (Fig. 1) that was formed during N- to NE-verging Jurassic-Cretaceous subduction of the Neotethys beneath Eurasia (Adamia et al., 1977; Kazmin et al., 1986; Rolland et al., 2011), followed by Late Cretaceous collision with the Gondwana-derived South Armenian block (Rolland et al., 2009a). Subsequent E-verging Neotethys subduction during final Cenozoic convergence of Eurasia and Arabia was accompanied by an Eocene magmatic maximum and followed by collisional to postcollisional Oligocene to Pliocene magmatism (Khain,
ORE FORMATION DURING JURASSIC SUBDUCTION OF THE TETHYS ALONG THE EURASIAN MARGIN

by the complex geodynamic processes of the Central Tethys belt (Moritz et al., 2016a). The initial metallogenic evolution of the Lesser Caucasus is related to the Jurassic-Cretaceous subduction of the Tethys along the Eurasian margin (Moritz et al., 2014; Moritz et al., 2016a). Copper-enriched massive pyrite, Cu-Au and polynematic stratiform, vein-type, and stockwork orebodies are hosted by Middle Jurassic volcanic and volcanoclastic rocks in the Alaverdi, Melmana, and Kapan mining districts (Fig. 1). Early Cretaceous porphyry Cu and precious metal epithermal deposits (Moritz et al., 2016a) were emplaced along the Somkhto-Karabakh belt at Teghout (Fig. 1; Amiryan et al., 1985; Calder et al., 2019), Gedabek, Gosh, and Chevdar (Fig. 1; Babazadeh et al., 1990; Hemon et al., 2012), and in the Kapan block at the Shikahogh prospect (Fig. 2; Achikgloyzan et al., 1987). The youngest metallogenic event associated with subduction along the Somkhto-Karabakh belt includes base and precious metal epithermal deposits and prospects with ambiguous VMS and porphyry-epithermal relationships of the Bolnisi district hosted by Late Cretaceous volcanoclastic rocks (Fig. 1; Gugushvili, 2004; Miginevshili, 2005; Popkhazde et al., 2014). The latter district extends to the west into the Turkish Eastern Pontides, which hosts VMS and porphyry-epithermal systems (Fig. 1; Kekelia et al., 2004; Yigit, 2009; Deliba et al., 2016). By contrast, the Mesozoic metallogenic belt of the Lesser Caucasus does not extend to the south into Iran (Moritz et al., 2016a), where the NE-oriented Araks fault constitutes a regional stratigraphic and structural limit between the Iranian Alborz and the Lesser Caucasus (Figs. 1, 2; Sosson et al., 2010).

During the Cenozoic, ore deposit formation was associated with subduction to postcollisional magmatism in the South Armenian block and was spatially associated with the accretionary zone along the Eurasian margin (Moritz et al., 2016a). Important ore mineral centers include the Zod-Sotk (Kozrenko, 2004; Levitan, 2008), Amulsar (Bender et al., 2017), and Meghradzor (Amiryan and Karapetyan, 1964) epithermal gold deposits, and the Hanqavan Cu-Mo prospect (Fig. 1). The most prolific Cenozoic ore deposit cluster is located in the Meghri-Orudbad district of the southernmost Lesser Caucasus (Figs. 1, 2), where porphyry Cu-Mo deposits and subsidiary epithermal prospects are hosted by the composite Meghri-Orudbad and Bargushat plutons (Karyanyan, 1978; Amiryan, 1984; Babazadeh et al., 1990; Moritz et al., 2016b; Rezeau et al., 2016). The Cenozoic Lesser Caucasiaore deposit belt extends to the south into the Alborz and Urmieh-Dokhtar belts of Iran (Fig. 1; e.g., Richards et al., 2006; Darlara, 2008; Aghazadeh et al., 2015; Hassanpour et al., 2015; Mehrabi et al., 2016; Simmonds et al., 2017).

Geologic Setting of the Kapan Block

The Kapan district (39°13′N, 46°24′E) is situated in south Armenia, ~200 km southeast of Yerevan, at an altitude of 800 to 1,500 m (Figs. 2, 3). It forms part of the tectonomagmatic Kapan zone, which extends about 70 km in a north-northeast–south-southeast direction from southern Armenia into northern Iran. Four geologic units are distinguished in the Kapan district: the Middle Jurassic, Late Jurassic–Early Cretaceous, and Paleogene magnatic complexes, overlain by Quaternary basanite flows (Fig. 4). Each complex consists of coherent lava flows, intrusive rocks, and autochthonic, hyaloclastic, pyroclastic, and the Kapan district (39°13′N, 46°24′E) is situated in south Armenia, ~200 km southeast of Yerevan, at an altitude of 800 to 1,500 m (Figs. 2, 3). It forms part of the tectonomagmatic Kapan zone, which extends about 70 km in a north-northeast–south-southeast direction from southern Armenia into northern Iran. Four geologic units are distinguished in the Kapan district: the Middle Jurassic, Late Jurassic–Early Cretaceous, and Paleogene magmatic complexes, overlain by Quaternary basanite flows (Fig. 4). Each complex consists of coherent lava flows, intrusive rocks, and autochthonic, hyaloclastic, pyroclastic, and the Kapan district (39°13′N, 46°24′E) is situated in south Armenia, ~200 km southeast of Yerevan, at an altitude of 800 to 1,500 m (Figs. 2, 3). It forms part of the tectonomagmatic Kapan zone, which extends about 70 km in a north-northeast–south-southeast direction from southern Armenia into northern Iran. Four geologic units are distinguished in the Kapan district: the Middle Jurassic, Late Jurassic–Early Cretaceous, and Paleogene magmatic complexes, overlain by Quaternary basanite flows (Fig. 4). Each complex consists of coherent lava flows, intrusive rocks, and autochthonic, hyaloclastic, pyroclastic,
Cu-Au-Mo (SHI) prospects in Late Jurassic to Early Cretaceous rocks are also shown. The Kapan district and of the Bartsravan polymetallic (BAR) and Shikahogh myan (SHA) and Centralni (CEN) deposits within Middle Jurassic rocks of Wood et al. (2008), and Mederer et al. (2013). The positions of the Shahu-

![Fig. 4. Simplified synthetic lithostratigraphic column of the Kapan district, based on a compilation from Achikgiozyan et al. (1987); Zohrabyan (2005), Wood et al. (2008), and Mederer et al. (2013). The positions of the Shahumyan (SHA) and Centralni (CEN) deposits within Middle Jurassic rocks of the Kapan district and of the Bartsravan polymetallic (BAR) and Shikalagh Cu-Au-Mo (SHI) prospects in Late Jurassic to Early Cretaceous rocks are also shown.](https://pubs.geoscienceworld.org/segweb/economicgeology/article-pdf/doi/10.5382/econgeo.4640/4683802/4640_It_mederer-etal.pdf)

and sedimentary deposits with lateral facies variation (Achikgiozyan et al., 1987; Mederer et al., 2013). Together, they constitute an almost 6,500-m-thick pile of volcanic and volcaniclastic rocks, calcareous sedimentary rocks, and limestone (Fig. 4). A similar stratigraphic division was proposed as a working model for mineral exploration (Wood et al., 2008), in which Late Cretaceous limestone and sequences A, B, and C are distinguished. Sequences B and C comprise rocks from the Middle Jurassic magmatic complex of Achikgiozyan et al. (1987), whereas sequence A refers to the covering rocks from the Late Jurassic-Early Cretaceous magmatic complex. An absolute stratigraphic reference is missing in this model; therefore, we prefer the better-documented stratigraphic division by Achikgiozyan et al. (1987) outlined above.

The oldest rocks in the Kapan district consist of an approximately 1,000-m-thick sequence of Bajocian to Bathonian volcanogenic and volcanosedimentary rocks, with a dominantly andesitic to dacitic and subsidiary basalts to rhyolithic composition (Akopyan, 1962; Achikgiozyan et al., 1987; Mederer et al., 2013). No older basement rocks are cropping out in the Kapan district. The dominant lithofacies in the lower part of the Middle Jurassic sequence are lava flows, brecciated lava, hyaloclastite, ignimbrite, and tuff. Widespread amygdaloidal and porphyritic textures and subsidiary lava structures are described by Achikgiozyan et al. (1987). District-wide epidote alteration is characteristic for the base of the Jurassic section and becomes less intensive toward the upper part of the Middle Jurassic magmatic complex. Both subaqueous and subaerial deposited tuff, ignimbrite and lava flows as well as hyaloclastite with andesitic to dacitic composition are characteristic for the upper part of the Middle Jurassic magmatic complex (Cholahyan and Sarkisyan, 1972; Achikgiozyan et al., 1987). A dominantly subvolcanic quartz-dacite that yielded a K-Ar age of 162 ± 5 Ma (Sarkisyan, 1970), referred to as the Barabatoom Formation (Fig. 3), cuts across Bathonian units and also contains hyaloclastite and lava flows interlayered with ash fall deposits (Zohrabyan, 2005). Callovian tuff, limestone, and calcareous rocks unconformably cover Bathonian and Bajocian rocks (Achikgiozyan et al., 1987; Zohrabyan, 2005). Gabbro-diorite bodies were intersected by drill holes at a depth of 390 m below the town of Kapan (Tumanyan, 1992). Rounded tonalite fragments from polymict pebble dikes hosted by Middle Jurassic rocks have a U-Pb zircon age of 165.6 ± 1.4 Ma (Mederer et al., 2013). Based on their geochemical composition, the Middle Jurassic magmatic rocks were generated in a subduction environment (Mederer et al., 2013) and have a predominantly tholeiitic to transitional affinity (Fig. 5).

The Middle Jurassic magmatic complex was partially eroded and unconformably covered by rocks of the Late Jurassic-Early Cretaceous magmatic complex (Fig. 4). Late Oxfordian to late Aptian basaltic, andesitic, and dacitic tuff, hyaloclastite, brecciated lava, and lava flows are interlayered with fossil-bearing calcareous sandstone and limestone units (Akopyan, 1962; Achikgiozyan et al., 1987) and were intruded by Early Cretaceous granodiorite, quartz-monzodiorite, gabbro, diorite, granodiorite, monzonite, and granite, with U-Pb zircon ages ranging between 139 and 128 Ma (Mederer et al., 2013; Melkonyan et al., 2016). Abundant steeply dipping N- and SE-oriented diabase dikes crosscut Middle Jurassic rocks and are traced across the unconformity between the Middle Jurassic and
Late Jurassic-Early Cretaceous magmatic complexes. Therefore, they are attributed to the Late Jurassic-Early Cretaceous magmatic complex. The thickness of dikes generally ranges between 0.5 and 3 m but can reach up to 20 m. Plagioclase and clinopyroxene phenocrysts occur within the fine-grained matrix of diabase dikes. The Late Jurassic to Early Cretaceous magmatic rocks were also linked to subduction (Mederer et al., 2013), but, in contrast to the Middle Jurassic rocks, they have a mostly calc-alkaline to partly transitional composition (Fig. 5), likely reflecting magmatic arc growth. The Late Jurassic to Early Cretaceous magmatic rocks have more primitive Sr and Nd isotope compositions compared to the Late Jurassic magmatic rocks, which was interpreted as asthenospheric mantle upwelling as a consequence of slab rollback during the younger magmatic arc evolution (Mederer et al., 2013).

Paleogene andesitic to rhyolitic brecciated lava, lava flows, tuff, and ignimbrite with interlayered Eocene limestone dominate in the western part of the Kapan zone, where they unconformably overlie the Late Jurassic-Early Cretaceous magmatic complex. Subsidiary gabbroic and monzogabbroic intrusions are emplaced within the Paleogene magmatic complex. One gabbro has a U-Pb zircon age of 50.8 ± 0.5 Ma (Mederer et al., 2013) and is coeval with the beginning of Eocene magmatic activity recorded in the composite Meghri-Ordubad pluton of the adjacent South Armenian block (Fig. 2; Moritz et al., 2016b; Rezeau et al., 2016). The youngest magmatic event of the Kapan zone consists of columnar-jointed Quaternary basanite flows (Fig. 3).

Recent structural studies in the Kapan district (Davis, 2006; Wood et al., 2008) revealed a complex deformational history with repeated reactivation of preexisting faults. Bedding is well preserved in the volcanosedimentary and volcanic sequence of the Late Jurassic-Early Cretaceous magmatic complex but is difficult to trace in the Middle Jurassic magmatic complex, where lateral facies variation impedes the definition of stratigraphic marker horizons. Where observable, the flat-lying beds dip preferentially toward the north-northeast, whereas in the southwestern part of the district bedding preferentially dips toward the west. Steeply dipping, E-W–striking extensional faults are the predominant structures in the Middle Jurassic magmatic complex. They formed early during the deformational history and are the host to mineralized veins in the Kapan district. Kinematic indicators on moderately northeastward-dipping thrust faults indicate a top-to-the-west displacement. Thrusting typically occurred along the contact between the Middle and Late Jurassic-Early Cretaceous magmatic complexes, but also within the Middle Jurassic sequence (Fig. 6a). These faults define the large-scale duplex-style geometry and control stratigraphic repetition in the district (Wood et al., 2008). Late N-S–oriented and steeply dipping normal faults and NW-oriented strike-slip faults crosscut the thrust faults and the mineralized E-W–striking extensional faults.

**Mineralization in the Kapan District**

Economic mineralization in the Kapan district consists of Cu, Cu ± Au, polymetallic, and precious metal vein- and stockwork-type deposits hosted by the Middle Jurassic magmatic complex. The most important ore deposits are the Centralni West Cu deposit, the Centralni East Cu-Au deposit, and the Shahumyan polymetallic Cu-Au-Ag-Zn ± Pb deposit (Figs. 3, 4, 6a, b; Table 1). Several other ore occurrences and minor deposits have been exploited during the last two centuries in the Kapan district, but many of the old underground workings are not accessible anymore.

Local hydrothermal alteration and sulfide veining are also hosted by the Late Jurassic-Early Cretaceous and Paleogene magmatic complexes. Polymetallic vein-type mineralization at the Barsravan exploration project, 25 km northwest of Kapan, is hosted by volcanic and subvolcanic rocks (Figs. 2, 4; Zohrabian et al., 2003), and stockwork-type Cu-Au-Mo mineralization at the Shikahogh prospect, 20 km south of Kapan, occurs at the contact of the Early Cretaceous Tsav intrusion within Late Jurassic and Early Cretaceous rocks (Figs. 2, 4; Achkigyozyan et al., 1987).

**The Centralni West Cu deposit**

The Centralni West Cu deposit is situated in the western part of the Kapan district (Fig. 3). Mineralization is hosted by Middle Jurassic brecciated lava, bedded hyaloclastite, and lava flows of basaltic to andesitic composition (Fig. 4). E-W–oriented and steeply S dipping veins (60°–80°)
are the dominant mineralization style in the deposit (Fig. 7a). The matrix of the brecciated lava host rock consists in places of ore and gangue minerals (Fig. 7b). Hydraulic breccia textures with fragmented pyrite are typical in individual veins, which generally show an extensional character with unstrained margins and sulfide and gangue mineral growth perpendicular to the host rock walls. Wood et al. (2008) described one vein with a 10° to 20° dip toward the south, with shear fabrics and kinematic thrust fault indicators along its margins. The main ore minerals are an intermediate-sulfidation assemblage comprising chalcopyrite and pyrite, with minor sphalerite, tennantite-tetrahedrite, and galena (Fig. 7c). Trace minerals include marcasite, tellurobismuthite (Bi₂Te₃), hessite (Ag₂Te), petzite (Ag₃AuTe₂), teddymnite (Bi₂Te₂S), wittichenite (Cu₃BiS₃), emplectite (CuBiS₂), and native gold (Achikgiozyan et al., 1987). The host rock adjacent to mineralization in the Centralni West deposit is pervasively altered to chlorite, carbonate, and epidote (Fig. 7d), and fine-grained muscovite occurs in proximity to mineralization and in the shallow parts of the deposit. High-grade veins with up to 10 wt % Cu and replacement-type mineralization in brecciated host rocks are characterized by similar ore and gangue minerals.

**The Centralni East Cu-Au deposit**

The Centralni East Cu-Au deposit is situated in the central part of the Kapan district (Fig. 3). Toward the west and south, the deposit is limited by gyspum-bearing faults, whereas an andesitic dike limits mineralization toward the east (Beaumont, 2006). The dominant mineralization style in the upper part of the Centralni East deposit is of stockwork type (Fig. 8a) that changes with depth to roughly E-W oriented veins, which dip between 65° and 85° to the south. Silicification, residual quartz alteration, and phyllic alteration with sericite, dickite, and diaspore as typical alteration minerals affect the andesitic to dacitic host rocks. Vein-type orebodies dominate over stockwork-style mineralization with increasing depth (Achikgiozyan et al., 1987). An intermediate- to high-sulfidation mineral paragenesis is characteristic for the Centralni East deposit, including pyrite, colusite (Cu₁₂V₃S₁₆), tennantite-tetrahedrite, chalcopyrite, and specular hematite as main ore minerals, and minor luzonite and galena (Fig. 8b, c). Enargite, bornite, sphalerite, covellite, renierite ((Cu,Zn)₁₁(Ge,As)₂Fe₄S₁₆), germanite (Cu₂₆Ge₄Fe₄S₃₂) and minor native silver and tellurides have been described (Achikgiozyan et al., 1987). Quartz is the dominant gangue mineral with minor barite and gypsum. Clast-supported breccia with angular fragments of argillic altered and silicified dacite within a matrix of fine-grained quartz, gyspum, pyrite, hematite, minor sphalerite, chalcopyrite, and native gold (up to 20 µm) have been reported from waste dump samples of the Centralni East deposit (Beaumont, 2006).

**The Shahumyan polymetallic Cu-Au-Ag-Zn ± Pb deposit**

The Shahumyan polymetallic Cu-Au-Ag-Zn ± Pb deposit is situated in the eastern part of the Kapan district (Fig. 3). Mineralization is hosted by the dominantly porphyritic subvolcanic quartz-dacite of the Middle Jurassic Barabatoom Formation that also consists of hyaloclastite and lava flows interlayered with ash fall deposits. The quartz-dacite contains...
Fig. 7. Mineralization style, textures, and mineral associations at the Centralni West deposit. 
a. E-W-oriented and 75° S-dipping quartz-chalcopyrite-pyrite vein (photograph kindly provided by Alan Turner, Deno Gold). 
b. Pyrite, chalcopyrite, quartz, and carbonate constitute the matrix of Middle Jurassic brecciated lava. 
c. Minor galena in chalcopyrite- and pyrite-dominated vein from high-grade (~10 wt % Cu) copper ore (plane-polarized reflected light microscopy); the sulfides occur within a quartz-carbonate gangue (not shown in the figure). 
d. Chalcopyrite-pyrite vein in chlorite-carbonate-epidote-altered andesite hyaloclastite host rock. 
Abbreviations: ccp = chalcopyrite, gn = galena, py = pyrite.

Fig. 8. Mineralization style, textures, and mineral associations at the Centralni East deposit. 
b. Colusite replacing pyrite. 
c. Galena associated with chalcopyrite and tennantite. 
Abbreviations: ccp = chalcopyrite, col = colusite, gn = galena, py = pyrite, tnt = tennantite-tetrahedrite.
abundant, up to 5-cm-sized plagioclase, amphibole, and bipyramidal quartz phenocrysts. More than 100 steeply N and S dipping (75°–85°), E-W–oriented veins (Fig. 6b) occur within an area of about 1.5 × 2.5 km². Individual veins can be traced for up to several hundred meters along strike with a vertical extent between 100 and 400 m. Veins are generally characterized by unstrained margins with gangue and ore mineral orientation perpendicular to the vein walls. They crop out at surface and are cut by the Late Jurassic-Early Cretaceous magmatic complex that overlies Middle Jurassic rocks. The veins are zoned with an early barren quartz-pyrite stage, rimmed by a polymetallic stage, and filled by a late carbonate stage (Fig. 9a). The host rock is affected by hydrothermal brecciation with clasts cemented by ore and gangue minerals (Fig. 9b). In other places, pseudobreccia is developed with alteration along microfractures (Fig. 9b). The thickness of the veins varies from about 1 cm to several meters with an average width between 0.7 and 1.5 m.

The mineral assemblage in the Shahumyan deposit is typical of an intermediate-sulfidation state, with pyrite, chalcopyrite, sphalerite, tennantite-tetrahedrite, and galena being the most abundant ore minerals (Fig. 9c). Minor arsenopyrite is

Fig. 9. Mineralization style, textures, and mineral associations at the Shahumyan deposit. a. Polymetallic vein hosted by altered quartz-dacite of the Barabatoom Formation. b. Clast-rotated monomict breccia in a high-grade ore vein of the Shahumyan deposit (see hammer for scale in the bottom left part of the photograph). Bottom part between the orange lines: quartz-dacite clasts are rimmed by cockade bands of sphalerite, quartz, and carbonate. Top part, above the orange line: the adjacent wall rock is irregularly altered along cracks, resulting in a pseudobreccia. c. Galena and tennantite-tetrahedrite associated with chalcopyrite. d. Inclusions of enargite and chalcopyrite in pyrite. e. Telluride droplet-shaped inclusions along fractures in quartz I with sericite and quartz II, and associated krennerite and altaite (plane-polarized transmitted light microscopy). f. Altaite, hessite, and petzite associated with chalcopyrite and sphalerite in a multiphase inclusion in pyrite. Abbreviations: ccp = chalcopyrite, enr = enargite, gn = galena, py = pyrite, qz = quartz, ser = sericite, sl = sphalerite, tut = tennantite-tetrahedrite.
ORE FORMATION DURING JURASSIC SUBDUCTION OF THE TETHYS ALONG THE EURASIAN Margin

Minerals of a higher-sulfidation fluid composition, such as enargite, digenite, bornite, and chalcocite, occur as up to 40-µm-sized inclusions in pyrite (Fig. 9d). The occurrence of gold and silver is controlled by Au-Ag tellurides, including calaverite (AuTe₂), krenmerite ((Au₉₋₈Ag₀.₂)Te₂), sylvanite (AuAgTe₄), petzite (Ag₃AuTe₂), and hesite (Ag₂Te). Altaite (PbTe) is common, and native tellurium can be found as inclusions in pyrite. Coloradoite (HgTe) was described by Matveev et al. (2006), and Achkigiozyan et al. (1987) reported native gold. Tellurides are observed as droplet-shaped inclusions in virtually sulfide-free quartz veins, where telluride deposition is associated with sericitic alteration and a second generation of quartz (Fig. 9e). Telluride assemblages also occur as polyphase inclusions in pyrite (Fig. 9f), along grain boundaries between sulfide minerals, as elongated aggregates along healed microcracks or, to a minor extent, disseminated within the late carbonate stage. Quartz is the most common gangue mineral, and carbonate minerals (including calcite, rhodochrosite, and kutnohorite (CaMn[CO₃]₂)) formed late in the paragenesis (Fig. 10). Hydrothermal apatite crystallized in clusters toward the end of the polymetallic stage and as disseminated crystals in the late carbonate stage. Hydrothermal fluorite occurs in places in Shahumyan. Distal propylitic alteration surrounds the deposit, where chlorite,

<table>
<thead>
<tr>
<th>a) Centralni West deposit</th>
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<tbody>
<tr>
<td>quartz</td>
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<td></td>
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<tr>
<td>b) Centralni East deposit</td>
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<td>---------------------------</td>
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<tr>
<td>quartz</td>
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<td></td>
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<tr>
<td></td>
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<tr>
<td>c) Shahumyan deposit - polymetallic veins</td>
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<tr>
<td>quart-pyrite stage</td>
</tr>
<tr>
<td>quartz</td>
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<tr>
<td></td>
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<tr>
<td>d) Shahumyan deposit - northeastern flank</td>
</tr>
<tr>
<td>magmatic-hydrothermal alunite (disseminated)</td>
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</table>

Fig. 10. Mineral paragenetic succession for (a) Centralni West, (b) Centralni East, (c) polymetallic veins from the Shahumyan deposit, and (d) the northeastern flank of the Shahumyan deposit. Thick bars indicate increased mineral abundance and dashed lines subsidiary mineral abundance.
epidote, carbonate, and pyrite are typical alteration minerals. In proximity to the orebodies, phyllic alteration with sericite, quartz, and pyrite prevails. With decreasing depth, the phyllic alteration grades into an argillic alteration assemblage with dickite, quartz, and pyrite ± sericite.

Advanced argillic alteration and poorly developed residual quartz within Middle Jurassic quartz-dacite can be found at surface in the northeastern part of the Shahumyan deposit (Fig. 3), where two different types of alunite can be distinguished. Nearly monomineralic, E-W–striking, and steeply dipping alunite veins are composed of coarse-grained (up to 500 µm) bladed pink alunite, associated with minor amounts of hematite, pyrite, and quartz (sample 5-77; Fig. 11a). Up to 150-µm-sized, disseminated, transparent to pinkish bladed alunite crystals are associated with advanced argillic and residual quartz alteration. Alunite, together with kaolinite and dickite, replaces plagioclase phenocrysts of the quartz-dacite host rock (sample 3-63-1; Fig. 11b). Achikgiozyan et al. (1987) described, with increasing distance from the most altered rocks, alunite, diaspore, dickite, and sericite occurring with minor pyrite, hematite, and supergene limonite. Advanced argillic altered quartz-dacite with disseminated alunite (sample 3-63-1; Fig. 11b) contains 0.24 ppm Au and 103 ppm As (Mederer, 2013), which is significantly enriched compared with an average of 3.6 ppm As in silicic volcanic rocks (Onishi and Sandell, 1955). By contrast, alunite from banded veins and its direct host rock (sample 5-77; Fig. 11a) contain only 0.02 ppm and 0.048 ppm Au, respectively, whereas the As content in vein alunite and host rock is 22 ppm and 12 ppm, respectively (Mederer, 2013).

Crosscutting relationships between dikes and mineralization in the Kapan deposits

Abundant dikes crosscut mineralized veins in the deposits of the Kapan district. The relative timing of dike emplacement with respect to mineralization in the Kapan deposits has been widely discussed, and both pre- and postmineralization scenarios have been proposed (see Achikgiozyan et al., 1987, and references therein). E-W–oriented and steeply dipping felsic dikes in the Centralni deposits are offset by sinistral strikeslip faults, whereas mineralized veins subparallel to the felsic dikes remain unaffected by the faults (Achikgiozyan et al., 1987). This relationship was used to attribute a premineralization age to the felsic dikes (Achikgiozyan et al., 1987).

By contrast, diabase dikes postdate the mineralized veins of the Kapan district. The relative timing is clearly documented by dikes crosscutting banded polymetallic veins (Fig. 12a), ductile deformation affecting mineralized veins along the contact with crosscutting dike (Fig. 12b), and well-developed continuous chilled margins of the crosscutting dike along the contacts with adjacent Middle Jurassic host rock and a mineralized vein (Fig. 12c). In addition, diabase dikes crosscut the unconformity between the Middle Jurassic and Late Jurassic–Early Cretaceous magmatic complexes, whereas the mineralized veins are restricted to Middle Jurassic host rocks.

Fig. 11. Textures and mineral associations of samples used for 40Ar/39Ar dating. a. Hand specimen of banded magmatic-steam alunite in contact with altered quartz-dacite. b. Magmatic-hydrothermal alunite, which occurs in vugs and in the matrix of altered quartz-dacite. c. Hydrothermal muscovite and pyrite in contact with the altered host rock. d. Zoned igneous plagioclase. b to d: Cross-polarized transmitted light photographs. Abbreviations: alu = alunite, hem = hematite, ms = muscovite, pl = plagioclase, py = pyrite, qz = quartz.
Analytical Techniques

Radiogenic and stable isotope analyses

The Pb isotope composition of pyrite, galena, chalcopyrite, and alunite was determined at the University of Geneva, Switzerland. A few milligrams of pyrite and chalcopyrite samples were digested in a mixture of 6N HCl and 14N HNO₃ (2:1), and alunite in a mixture of concentrated HF and 14N HNO₃ (3:1), whereas galena was digested in 14N HNO₃. The samples were converted to bromide form, and Pb was separated by ion exchange chromatography. Lead was loaded on single Re filaments using the silica gel technique (Gerstenberger and Haase, 1997). The samples and SRM981 standards were analyzed at a pyrometer-controlled temperature of 1,220°C on a Thermo TRITON mass spectrometer on Faraday cups in static mode. Lead isotope ratios were corrected for instrumental fractionation by a factor of 0.1% per amu based on more than 100 measurements of the SRM981 standard using the values of Todt et al. (1996). External reproducibilities (1σ) of the standard ratios are 0.05% for 206Pb/204Pb, 0.08% for 207Pb/204Pb, and 0.10% for 208Pb/204Pb.

The strontium isotope compositions of calcite samples were obtained at the University of Geneva, Switzerland. About 60 mg of calcite powder was dissolved in 2N HCl, and Sr was separated by ion chromatography in hydrochloric medium. Strontium was loaded on single Re filaments using a Ta oxide solution and measured at a temperature of 1,490°C on a Thermo TRITON mass spectrometer on Faraday cups in static mode. ⁸⁷Sr/⁸⁶Sr ratios were internally corrected for fractionation using an ⁸⁸Sr/⁸⁶Sr ratio of 8.375209. Raw values were further corrected for external fractionation by a value of +0.03‰, determined by repeated measurements of the SRM987 standard (⁸⁷Sr /⁸⁶Sr = 0.710250). External reproducibility (1σ) of the SRM987 standard is 6 ppm.

For stable isotope analyses, monomineralic separates were obtained by careful handpicking under the binocular microscope. Isotopic compositions are expressed as δ values, in per mil (%) deviations relative to the Vienna-Standard Mean Ocean Water (V-SMOW) for O and H, the Canyon Diablo Troilite (CDT) for S, and the Vienna-PeeDee Belemnite (V-PDB) for C. The sulfur isotope composition of sulfides and sulfates was measured using a Carlo Erba 1108 elemental analyzer connected to a Finnigan MAT Delta-S mass spectrometer at the University of Lausanne, Switzerland, following the analytical protocol described by Giesemann et al. (1994). Powdered sulfide (150–200 µg) or sulfate samples (250 µg) were wrapped in tin capsules. Samples were combusted at 1,020°C with vanadium oxide serving as a catalyst/reactant.
and all gases produced during combustion were transported in a stream of helium to the reduction furnace (at 650°C). Water was subsequently removed in a water trap. Other gases like NO₂ and CO₂ were separated in a gas chromatography column (heated at 50°C) from SO₂, which was analyzed in the mass spectrometer. A reference gas SO₂ for isotopic calibration was introduced into the carrier gas stream via a CON-FLO III. The standards during runs of sulfides included NBS 123 sphalerite (δ³⁴S = +17.4‰), Py-E pyrite (δ³⁴S = −7‰), IAEA-S1 silver sulfide (δ³⁴S = −0.3‰), and barium sulfate (δ³⁴S = +20.3‰). External precision for the isotopic compositions was better than ±0.2‰.

The dissolution-reprecipitation method (Wasserman et al., 1992) was applied to analyze the oxygen isotope composition in the tetrahedral site of alunite. As not enough sample material was available from disseminated alunite, only vein alunite was treated by this method. About 120 mg of alunite was dissolved in 0.5N NaOH solution and heated to 50°C (±10°C) for 3 h. The solution was filtered and titrated by 10N HCl to a pH of 2.85 to avoid the precipitation of Al(OH)₃ at intermediate pH. By the addition of 5 ml of 0.5N BaCl₂ to the solution, the dissolved (SO₄)²⁻ anions were precipitated as barite (BaSO₄). This barite precipitate was subsequently analyzed for its oxygen isotope composition together with naturally occurring barite from the Centralni East deposit. Three hundred fifty to 450 µg of sulfate sample was analyzed using an He carrier gas with a high-temperature conversion elemental analyzer (TC-EA) coupled to a DELTAplus XL isotope-ratio mass spectrometer (IRMS) from Thermo Finnigan according to a method similar to that described in Vennemann and O’Neil (1993). The method is based on reduction by graphite and a buffer of glassy carbon at temperatures higher than 1,450°C. The accuracy of analysis was periodically checked by analyses of the international reference barium sulfates NBS-127 (+9.3‰), IAEA SO-5 (+13.05‰), and SO-6 (−10.83‰). δ¹⁸O values are precise within 0.3‰.

The hydrogen isotope composition of alunite was measured using a high-temperature (1,450°C) conversion elemental analyzer with continuous flow reactor linked to a Thermo Finnigan DELTAplus XL IRMS, applying the methods of Sharp et al. (2001). The in-house kaolinite (Kaol-17, δD = −125.3‰) and biotite standards (G1, δD = −66‰) were used to assess measurement reproducibility. The accuracy of the TC/EA/IRMS analysis was checked periodically by analyses of the international reference standard biotite NBS-30 (δD = −65.3‰). δD values are precise to within 2‰.

The oxygen isotope composition of quartz was obtained using a CO₂ laser fluorination system (Kasemann et al., 2001). Between 1 and 2 mg of sample was loaded on a Pt sample holder and evacuated to a vacuum of about 10⁻⁶ mbars. After preflourination of the samples overnight, they were heated using a CO₂ laser in the presence of about 50 mbar of pure F₂, which was generated by a potassium nickel fluoride salt that was heated at >250°C. The hot mineral reacted quickly with F₂, releasing O₂ from silicate or oxide minerals. Excess F₂ was separated from O₂ by exchange with KCl at 150°C. The extracted O₂ was collected on a molecular sieve (13X) and subsequently expanded into the inlet of a Finnigan MAT 253 isotope mass ratio spectrometer. The in-house standard LS-1 (δ¹⁸O = +18.1‰) was used for the normalization of the results. Replicate oxygen isotope analyses generally had an average precision of ±0.2‰.

The carbon and oxygen isotope composition of calcite from the Shahumyan and Centralni West deposits was determined according to the methods of Spötl and Vennemann (2003). CO₂ was released by the reaction of calcite with H₃PO₄, which was then transported by He after several cleaning steps into a continuous-flow GasBench and analyzed by a Thermo Finnigan DELTAplus XL mass spectrometer. Samples were normalized to an in-house calcite standard (CM-STD: δ¹³Cₐ = +1.95 ± 0.06‰; δ¹⁸Oᵥ-FDB = −1.70 ± 0.05‰) that has been calibrated relative to the international standard material NBS-19 (δ¹³Cᵥ-FDB = +1.95‰; δ¹⁸Oᵥ-FDB = −2.20 ± 0.05‰). Precision for the analyses is better than ±0.1 and ±0.15‰ for δ¹³C and δ¹⁸O, respectively.

⁴⁰Ar/³⁹Ar geochronology

⁴⁰Ar/³⁹Ar incremental-heating analyses of one muscovite and two alunite samples (Fig. 11a-c), respectively, from the Centralni West and Shahumyan deposits were conducted. One plagioclase separate from the subvolcanic Barabatoom quartz-dacite was also analyzed (Fig. 11d), and 6.3 mg of magmatic-hydrothermal alunite (100 µm in size) was extracted from cavities within sample 3-63-1 and handpicked under a binocular microscope. Magmatic-steam alunite from the massive vein sample 5-77 was crushed, and 7 mg of alunite (2 mm in size) was separated. Sample JM016 was crushed by a hydraulic press and sieved before muscovite (9.2 mg, 10–100 µm) was handpicked using the binocular microscope. Sample 5-16 (Fig. 11d), which contains plagioclase phenocrysts of up to 1 cm in size, was crushed and sieved, and 6.7 mg of plagioclase was gravitationally separated from quartz using sodium polytungstate and a centrifuge. The analyzed plagioclase had a grain size of ~2 mm, and only the least altered fragments from the originally larger phenocrysts were selected. The four mineral separates were cleaned for 10 min in deionized water in an ultrasonic bath. Sample separates were packed in Cu foil, mounted in a glass silica tube, and subsequently irradiated for 30 h in the CLICIT facility of the TRIGA reactor at Oregon State University, USA. J-values were interpolated from samples of the 28.02 ± 0.28 Ma Fish Canyon Tuff sanidine (Renne et al., 1998), which was used as a flux monitor, and were separated by distances of <1 cm in the columnar irradiation package. Step-wise heating was performed using a 55-W IR-CO₂ laser and a stainless-steel extraction line coupled with a multicollector Argus mass spectrometer (GV Instruments) at the University of Geneva, Switzerland. The instrument was equipped with four high-gain (10¹⁶ Œ) Faraday cups for the measurement of ³⁶Ar, ³⁷Ar, ³⁸Ar, and ³⁹Ar, and a single 10¹⁰ Œ Faraday cup for ⁴⁰Ar measurements. One SAES AP10 getter and one water-cooled SAES GP50-ST101 getter are integrated in the automated ultrahigh vacuum (UHV) stainless steel gas extraction line. The isotopic composition of the gas was measured on the Faraday collectors and time-zero regressions were fitted to data collected from 12 cycles. Peak heights and backgrounds were corrected for baselines, mass discrimination, isotopic decay of ³⁶Ar and ³⁸Ar, and interfering nucleogenic Ca- K-, and Cl-derived isotopes. Error calculations include the errors on mass discrimination measurement and the J value. ³⁶Ar, ³⁷Ar, ³⁸Ar, ³⁹Ar, and ³⁹Ar blanks were calculated before every new sample and after every three
heating steps. $^{40}$Ar blanks were between 6.5E-16 and 1.0E-15 moles. Blank values for m/e 39 to 36 were all less than 6.5E-17 moles. Age plateaus were determined using the criteria of Dalrymple and Lanphere (1974) unless otherwise indicated, and data reduction utilized ArArCALC (Koppers, 2002).

Re-Os isochron dating of pyrite

Re-Os isotope analyses were obtained on pyrite samples from Centralni West and Centralni East. Pure pyrite separates of about 400 to 600 mg were separated at the University of Geneva by handpicking under the binocular microscope, cleaned ultrasonically in ethanol for 10 min, and crushed to 5 mm. The Re and Os isotope analyses were carried out at Durham University, UK, as described by Selby et al. (2009, and references therein). The accurately weighted samples were loaded into a Carius tube with a known amount of mixed Re-Os tracer solution containing $^{185}$Re and $^{190}$Os. The sample and tracer solution were digested and equilibrated using a mix of 11N HCl (3 ml) and 15.5N HNO$_3$ (8 ml) at 220°C during 48 h. Rhenium and Os were isolated by solvent extraction (CHCl$_3$), microdistillation, and anion column and single-bead chromatography methods. The purified Re and Os solutions were loaded onto Ni and Pt filaments, respectively, and analyzed by negative thermal ionization mass spectrometry on a Thermo Scientific TRITON mass spectrometer. The measured isotopic compositions were corrected for oxide and blank contribution and fractionation. Total procedural blanks were 3.3 ± 0.3 pg/g (Re) and 0.27 ± 0.21 pg/g (Os), with an average $^{187}$Os/$^{188}$Os ratio of 0.25 (n = 3). All uncertainties are determined by propagation of uncertainties in the mass spectrometer measurements, blank abundances and isotopic compositions, spike calibrations, weighing of sample and spike, and the results from analyses of an Re-Os standard.

Results

Radiogenic isotopes

The Pb isotope compositions of sulfides and alunite from the Kapan district range between 18.17 and 18.32 for $^{206}$Pb/$^{204}$Pb, 15.57 and 15.61 for $^{207}$Pb/$^{204}$Pb, and 38.17 and 38.41 for $^{208}$Pb/$^{204}$Pb (Table 2; Fig. 13). The $^{87}$Sr/$^{86}$Sr ratios of hydrothermal calcite samples from the Centralni West and Shahumyan deposits fall in a narrow range between 0.70537 and 0.70586 (Table 3; Fig. 14).

Table 2. Lead Isotope Data for Sulfides and Sulfates from the Centralni West, Centralni East, and Shahumyan Deposits

<table>
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<tr>
<th>Sample no.</th>
<th>Mineral</th>
<th>$^{206}$Pb/$^{204}$Pb</th>
<th>$^{207}$Pb/$^{204}$Pb</th>
<th>$^{208}$Pb/$^{204}$Pb</th>
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<td>py</td>
<td>18.296</td>
<td>15.602</td>
<td>38.377</td>
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<td>15.604</td>
<td>38.377</td>
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<td>py</td>
<td>18.297</td>
<td>15.598</td>
<td>38.366</td>
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<td>DC-2-123-1</td>
<td>ccp</td>
<td>18.308</td>
<td>15.604</td>
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<td>py</td>
<td>18.317</td>
<td>15.613</td>
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<td>15.583</td>
<td>38.315</td>
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</table>

Abbreviations: alu = alunite, ccp = chalcopyrite, gn = galena, py = pyrite

Fig. 13. Lead isotope compositions of sulfides and alunite from the Kapan mining district. The initial Pb isotope ratios of igneous rocks from the Kapan district are plotted for reference (age corrected for 161–145 Ma, Mederer et al., 2013). The Pb isotope evolution curves for the mantle (MA), orogen (OR), and upper crust (UC) are from Zartman and Doe (1981).
Table 3. Oxygen, Carbon, Strontium, and Hydrogen Isotope Composition of Quartz, Calcite, Barite, and Alunite

### Part 1: δ¹⁸O in igneous and hydrothermal quartz

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<th>Sample no.</th>
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<th>300°C</th>
<th>200°C</th>
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<td>Middle Jurassic igneous quartz (Barabatoom)</td>
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<td>Shahumyan deposit</td>
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<td>Premineralization</td>
<td>qz</td>
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<td>3-35</td>
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<td>qz</td>
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### Part 2: δ¹⁸O, δ¹³C, and ⁸⁷Sr/⁸⁶Sr in hydrothermal calcite

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<tr>
<th>Sample no.</th>
<th>Stage</th>
<th>Mineral</th>
<th>δ¹⁸O (‰)</th>
<th>δ¹³C (‰)</th>
<th>⁸⁷Sr/⁸⁶Sr</th>
<th>300°C</th>
<th>250°C</th>
<th>200°C</th>
<th>150°C</th>
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<tr>
<td>DC-2-124-1</td>
<td>Postmineralization</td>
<td>cal</td>
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<td>DC-2-125</td>
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<td>cal</td>
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<td>-3.0</td>
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<td>3-142</td>
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<td>cal</td>
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### Part 3: δ¹⁸O and δD in sulfates

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<th>δ¹⁸O_H₂O (‰)</th>
<th>δD (‰)</th>
<th>δ¹⁸O_H₂O</th>
<th>δD_H₂O</th>
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<tr>
<td>OP-3-98-1</td>
<td>Main stage</td>
<td>brt</td>
<td>10.5</td>
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<td>-34 ± 2</td>
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<td>5-77</td>
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<td>alu</td>
<td>10.9</td>
<td>14.4</td>
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<td>7.9</td>
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<td>alu</td>
<td>14.1</td>
<td>-43 ± 2</td>
<td>11.4</td>
<td>9.1</td>
<td>5.2</td>
<td>-25</td>
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Notes: The isotopic composition of the fluid in equilibrium with the minerals was calculated according to the equations of Zheng (1993) in the case of quartz, Zheng (1999) in the case of calcite and barite, and Stoffregen et al. (1994) in the case of alunite; δ¹⁸O and δD are reported relative to Vienna-Standard Mean Ocean Water, and δ¹³C relative to Vienna-Pee Dee Belemnite. Abbreviations: alu = alunite, brt = barite, cal = calcite, qz = quartz.
Stable isotopes

The 32 analyzed sulfide samples from the Kapan district have sulfur isotope compositions that fall in a narrow range between 2.0 and 6.5‰ (Table 4; Fig. 15). Pyrite has the highest δ34S values at 4.5 to 6.4‰, except for one outlier at 2.5‰. Sphalerite, chalcopyrite, and galena from the Shahumyan deposit have the lowest δ34S values at 3.6 to 5.1‰. The pyrite δ34S values from the Centralni East deposit range between 3.3 and 6.5‰, overlapping those of pyrite from the Shahumyan deposit (Fig. 15). The sulfide δ34S values in the deposits cannot be ascribed to discrete mineralization stages. For example, in the Shahumyan deposit, pyrite from the main and late main stages can be characterized by either a high or low δ34S value (Table 4). Gypsum from gypsum-pyrite veins and fault-related gypsum have δ34S values of 19.5 and 18.0‰, respectively. A barite sample associated with the main-stage mineralization in the Centralni East deposit has a sulfur isotope composition of 17.4‰ and a δ18O value of 10.5‰ (Table 3).

δ34S values of chalcopyrite from the Centralni West deposit range between 2.0 and 4.4‰, whereas pyrite from this deposit is characterized by higher values between 4.9 and 6.1‰. Alunite from the massive alunite ± hematite ± quartz...
veins (Fig. 11a) from the northeastern limb of the Shahumyan deposit (Fig. 3) has a $\delta^{34}S$ value of 7.9‰ and a $\delta^D$ value of 34‰. $\delta^{18}O_{SO_4}$ and $\delta^{18}O_{OH}$ of vein alunite are 14.4 and 10.9‰, respectively (Table 3). Disseminated alunite (Fig. 11b), which is associated with argillic alteration and minor pyrite along the northeastern limb of the Shahumyan deposit (Fig. 3), has a $\delta^{34}S$ value of 27.0‰, a $\delta^D$ value of –43‰, and a $\delta^{18}O_{tot}$ of 14.1‰.

Hydrothermal quartz samples from the Centralni West and Centralni East deposits yield, respectively, $\delta^{18}O$ values of 8.3 to 10.9‰ and 9.1 to 12.7‰ (Table 3, Fig. 16). The $\delta^{18}O$ values of hydrothermal quartz from the Shahumyan deposit range from 10.6 to 16.4‰. In all three deposits, the $\delta^{18}O$ values of hydrothermal quartz increase progressively with later mineralization stages. Premineralization and main-stage quartz is generally more depleted in $^{18}O$ relative to late-stage and postmineralization quartz (Fig. 16). Igneous quartz from quartz-dacite of the Middle Jurassic Barabatoom Formation has a $\delta^{18}O$ value of 7.5‰ (Table 3, Fig. 16).

The isotopic composition of hydrothermal calcite from the Centralni West deposit ranges from 11.7 to 13.4‰ for $\delta^{18}O$ and from –3.4 to –2.1‰ for $\delta^{13}C$ (Table 3, Fig. 17a). The isotopic composition of calcite from the Shahumyan deposit is more variable and ranges from 7.7 to 15.4‰ for $\delta^{18}O$ and from –3.0 to 0.7‰ for $\delta^{13}C$. Positive correlation trends in $\delta^{13}C$ vs. $\delta^{18}O$ space can be observed in both deposits (Fig. 17a). The calcite data from the Shahumyan deposit also yield a positive correlation trend between $^{87}Sr/^{86}Sr$ and $\delta^{18}O$ values, whereas, in the Centralni West deposit, higher $^{87}Sr/^{86}Sr$ ratios are uncorrelated with $\delta^{18}O$ values (Fig. 17b). The correlation

\[\text{Fig. 15. Summary of } \delta^{34}S \text{ values for sulfides and sulfates from the Kapan mining district. The sulfur isotope composition of Jurassic marine sulfates (16 ± 1.5‰; Claypool et al., 1980) is displayed for the Centralni East deposit, where barite and gypsum have been analyzed. Abbreviations: alu = alunite, brt = barite, ccp = chalcopyrite, CDT = Canyon Diablo Troilite, gn = galena, gp = gypsum, py = pyrite, sl = sphalerite.}\]

\[\text{Fig. 16. Oxygen isotope composition of quartz from the Kapan district. Abbreviations: min = mineralization, V-SMOW = Vienna-Standard Mean Ocean Water.}\]
arrays in hydrothermal calcite from the Kapan district are independent of depth or geographical distribution in the Centralni West and Shahumyan deposits.

$^{40}$Ar/$^{39}$Ar geochronology

Hydrothermal muscovite from the Centralni West deposit (sample JM016; Fig. 11c) from the western part of the Kapan district (Fig. 3) yields a $^{40}$Ar/$^{39}$Ar plateau age of 161.78 ± 0.79 Ma for 50% of the released gas, which overlaps with the inverse isochron age of 162.73 ± 0.92 Ma with an MSWD of 0.99 (Fig. 18a, Tables 5, 6). The age spectrum shows a typical argon loss profile, with successively older ages at higher temperature during successive step-heating.

The magmatic-hydrothermal alunite sample (sample 3-63-1; Fig. 11b) from the Shahumyan deposit (Fig. 3) yields a slightly disturbed age spectrum (Fig. 18b), which may be a

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**Fig. 17.** (a) C and O and (b) Sr and O isotope compositions of calcites from the Centralni West and Shahumyan deposits. Abbreviations: V-PDB = Vienna-Pee Dee Belemnite; V-SMOW = Vienna-Standard Mean Ocean Water.

**Fig. 18.** $^{40}$Ar/$^{39}$Ar age spectra for (a) hydrothermal muscovite from Centralni West, (b) magmatic-hydrothermal alunite from Shahumyan, (c) igneous plagioclase from Shahumyan host rock, and (d) magmatic-steam alunite from Shahumyan. MSWD = mean square weighted deviation.
Table 5. Summary of $^{40}$Ar/$^{39}$Ar Step-Heating Ages from the Kapan District

<table>
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<tr>
<th>Sample no.</th>
<th>Mineral</th>
<th>Weighted plateau age (Ma ± 2σ)</th>
<th>% of $^{39}$Ar in plateau age</th>
<th>Total fusion age (Ma ± 2σ)</th>
<th>Inverse isochron age (Ma ± 2σ)</th>
<th>Inverse isochron-derived MSWD</th>
<th>Inverse isochron $^{40}$Ar/$^{39}$Ar ratio of intercept (± 2σ)</th>
</tr>
</thead>
<tbody>
<tr>
<td>JM016</td>
<td>Muscovite</td>
<td>161.78 ± 0.79</td>
<td>50.0</td>
<td>159.35 ± 0.65</td>
<td>162.73 ± 0.92</td>
<td>0.99</td>
<td>-143.8 ± 149.7</td>
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<tr>
<td>5-16</td>
<td>Plagioclase</td>
<td>-</td>
<td>-</td>
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<tr>
<td>3-63-1</td>
<td>Alunite</td>
<td>156.14 ± 0.79</td>
<td>40.4</td>
<td>155.67 ± 0.62</td>
<td>157.87 ± 1.19</td>
<td>0.50</td>
<td>128.8 ± 55.2</td>
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<tr>
<td>5-77</td>
<td>Alunite</td>
<td>-</td>
<td>-</td>
<td>118.05 ± 0.54</td>
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<td>-</td>
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Part 2. Sample location and weight

<table>
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<tr>
<th>Sample no.</th>
<th>Mass (mg)</th>
<th>Sample type and description</th>
<th>UTM (E)</th>
<th>UTM (N)</th>
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<td>Shahumyan deposit, propyltically altered quartz-dacite host rock</td>
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<td>Shahumyan deposit, magmatic-hydrothermal disseminated alunite</td>
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<td>7.0</td>
<td>Shahumyan deposit, magmatic-steam alunite from banded veins</td>
<td>624415</td>
<td>4342219</td>
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</table>

Notes: The plateau ages were calculated according to the criteria of Dalrymple and Lanphere (1974); the age in bold is calculated on the basis of less than 50% total released $^{40}$Ar/$^{39}$Ar; MSWD = mean square weighted deviation

consequence of $^{39}$Ar recoil in the higher temperature steps (Oustott et al., 1995). A weighted mean age of 156.14 ± 0.79 Ma has been calculated for a region of the age spectrum where the ages are indistinguishable, although they only span 40.4% of the $^{39}$Ar released. The weighted mean age is indistinguishable from the inverse isochron age of 157.87 ± 1.19 Ma (MSWD = 0.50). An argon loss profile can also be observed in the age spectrum, with a minimum step $^{40}$Ar/$^{39}$Ar age of ~150 Ma.

The age spectra of plagioclase sample 5-16 (Fig. 11d) is highly disturbed, although it is characterized by ages that increase with increasing temperature of the heating steps (Fig. 18c). There are no plateau regions and the ages of single heating steps range between 132 and 170 Ma, with the highest-temperature heating steps yielding ages of 164.89 ± 2.80 and 165.17 ± 5.27 Ma.

The magmatic-steau alunite sample (5-77; Fig. 11a) reveals a disturbed spectra, and no plateau age can be defined (Fig. 18d). The initial two steps yielded elevated ages, perhaps due to the presence of excess $^{40}$Ar, whereas the remaining steps yielded progressively increasing ages, which may be indicative of Ar loss.

Re-Os geochronology of pyrite

Total Re and Os contents in pyrite from the deposits in the Kapan district are low and range from 1.3 to 3.6 ppb and 2.2 to 11.9 ppt, respectively (Table 7). The $^{187}$Re/$^{188}$Os ratios are very high (4,620–74,850), and the $^{187}$Os/$^{188}$Os compositions are highly radiogenic (13–243; Table 7). The elevated $^{187}$Re/$^{188}$Os ratio indicates the near absence of common Os and therefore allows us to classify the sulfides as low-level highly radiogenic (LHHR; Stein et al., 2000). Traditional $^{187}$Re/$^{188}$Os (x-axis) vs. $^{187}$Os/$^{188}$Os (y-axis) plots show highly correlated uncertainties as a consequence of the poorly determined but extremely low concentration of $^{188}$Os. For that reason, the data are plotted with the associated uncertainty correlation value, named rho (Ludwig, 1980), which allows a conventional best fit of the data (e.g., Stein et al., 2000; Selby et al., 2009). However, due to the virtual absence of common $^{188}$Os and the highly radiogenic character of pyrite samples from the Kapan district, a direct regression of parent isotope $^{187}$Re vs. the daughter isotope $^{187}$Os can give meaningful and precise age information (e.g., Stein et al., 2000; Selby et al., 2009; Kerr and Selby, 2012). As in the case of molybdenite, single-mineral $^{187}$Re-$^{187}$Os model ages can be calculated for each individual LHHR sulfide sample (Stein et al., 2000; Selby et al., 2009).

Regression of the $^{187}$Re/$^{188}$Os vs. $^{187}$Os/$^{188}$Os data of four pyrite separates from the Centralni West deposit yield a model 3 Re-Os age of 215.7 ± 9.2 Ma (MSWD = 6.9; Fig. 19a), with an initial $^{187}$Os/$^{188}$Os ratio of 1.3 ± 3.6. Using the initial $^{187}$Os/$^{188}$Os composition of 1.3 ± 3.6, ~92.5 to 99.4% of the $^{187}$Os in the pyrite samples is radiogenic. A model 1 age of 207 ± 24 Ma is determined from the $^{187}$Re vs. $^{187}$Os data (initial $^{187}$Os = 0.31 ± 0.83; MSWD = 0.21; Fig. 19b). Four calculated single-mineral Re-Os model ages range from ~202 to 221 Ma, with a weighted average of 215.7 ± 4.2 Ma (MSWD = 0.32; Fig. 19c). All three ages are the same within uncertainty, but the more precise weighted average model age is our preferred solution.

The traditional $^{187}$Re/$^{188}$Os vs. $^{187}$Os/$^{188}$Os isochron regression of five pyrite separates from the Centralni East deposit yields a model 1 Re-Os age of 144.7 ± 4.2 Ma with an MSWD of 0.099 (Fig. 19d). An initial $^{187}$Os/$^{188}$Os value of 1.41 ± 0.57 was obtained from the isochron. Using this initial $^{187}$Os/$^{188}$Os composition, ~85.4 to 98.0% of the $^{187}$Os in the pyrite samples is radiogenic. The best fit of $^{187}$Re vs. $^{187}$Os yields a less precise model 1 age of 148 ± 11 Ma (initial $^{187}$Os = –0.03 ± 0.22, MSWD = 0.31; Fig. 19e). Calculated single-mineral model ages from the five pyrite analyses range from 144.5 to 148.4 Ma, with a weighted average of 146.2 ± 3.4 Ma (MSWD = 0.25; Fig. 19f). The three ages are the same within uncertainty; the more precise weighted average model age is our preferred solution.

Discussion

Source of metals

The Pb isotope composition of hydrothermal minerals from the Centralni West, Centralni East, and Shahumyan deposits overlap (Fig. 13), which indicates a common metal source.
Fig. 19. (a) $^{187}$Re/$^{188}$Os vs $^{187}$Os/$^{188}$Os, (b) $^{187}$Re vs $^{187}$Os$^r$, and (c) weighted average model age plots for the Centralni West deposit. (d) $^{187}$Re/$^{188}$Os vs $^{187}$Os/$^{188}$Os, (e) $^{187}$Re vs $^{187}$Os$^r$, (f) and weighted average model age plots for the Centralni East deposit. All diagrams were generated using the Isoplot v. 4.1 Excel macro (Ludwig, 2008), and all uncertainties are shown at 2σ level. See text for discussion and additional information. MSWD = mean square weighted deviation.
<table>
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<th>37Ar/39Ar</th>
<th>36Ar/39Ar</th>
<th>40Ar*/39Ar</th>
<th>40Ar (mol)</th>
<th>40Ar* (%)</th>
<th>39ArK (%)</th>
<th>K/Ca</th>
<th>Weighted plateau age:</th>
<th>Total fusion age:</th>
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<td>24.01326</td>
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<td>134.817</td>
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<td>134.817</td>
<td>152.16</td>
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<td>49.15</td>
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### Table 6.

Incremental step-heating results, 5-16 plagioclase, J = 3.8421E-3 ± 7.7E-6 (2σ)

<table>
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<tr>
<th>Step</th>
<th>40Ar/39Ar ±1σ</th>
<th>37Ar/39Ar ±1σ</th>
<th>36Ar/39Ar ±1σ</th>
<th>39ArK (%)</th>
<th>K/Ca ±2</th>
<th>Age (Ma) ±2σ</th>
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<td>0.05971</td>
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<td>12</td>
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<td>0.07850</td>
<td>0.00137</td>
<td>0.00600</td>
<td>18.247632</td>
</tr>
</tbody>
</table>

Note: The ages for steps marked in bold are the step-heating results included in the weighted plateau age for sample JM016 and the weighted mean age for a region of the age spectrum where the ages broadly tapped at the same time. The overlap of the Pb isotope compositions of sulfide minerals and alunite from the ore deposits with age-corrected (161–145 Ma) Middle Jurassic igneous rocks (Fig. 13) suggests that Pb was leached by hydrothermal fluids from the host Middle Jurassic magmatic rocks or was derived by direct exsolution of a magmatic-hydrothermal fluid from Middle Jurassic intrusions at depth. By extension, this interpretation also applies to the other base and precious metals of the Kapan deposits.

**Nature of hydrothermal fluids involved in ore formation: Evidence from Sr, O, and C isotopes**

As calcite generally does not incorporate Rb in its crystal lattice, its δSr87/86Sr ratio reflects the Sr isotope composition of the precipitating fluid (Ruiz et al., 1984). The Sr isotope composition of hydrothermal calcite from the Shahumyan and Central West deposits is slightly more radiogenic in comparison with age-corrected (161–145 Ma) δSr87/86Sr ratios of Middle Jurassic magmatic rocks (Fig. 14). In the Kapan district, no lithological units with δSr87/86Sr ratios higher than the ones of the Middle Jurassic magmatic complex have been observed (Mederer et al., 2013). The only local reservoir with an elevated radiogenic Sr isotopic composition was Middle Jurassic seawater with 87Sr/86Sr ratios between 0.7067 and 0.7073 at 170 to 140 Ma (Jones et al., 1994). Such a seawater environment is supported by the presence of subaqueous volcanioclastic and volcanic rocks within the Middle Jurassic magmatic complex (Achikgyozan et al., 1987; Mederer et al., 2013). Therefore, we conclude that seawater was involved during the late carbonate stage, and we interpret their Sr isotope composition to reflect mixing of seawater-derived Sr and Sr leached or exsolved from Middle Jurassic igneous rocks.

Temperatures between 140° and 433°C were calculated using oxygen isotope data of four quartz-calcite pairs from the late carbonate stage in the Shahumyan deposit (Table 3), according to the equation of Zheng (1993). The large temperature spread indicates that equilibrium conditions were not attained between the two minerals. No more precise temperature estimates are available for the precipitation of gangue and ore minerals in the Kapan district deposits. Therefore, the oxygen isotope composition of fluids in equilibrium with early-stage hydrothermal quartz and late-stage calcite was calculated using the equations of Zheng (1993, 1999) for the oxygen isotope composition of fluids in equilibrium with Middle Jurassic intrusions at depth. In the Kapan district, the overlap of the Pb isotope composition of sulfide minerals and alunite from the ore deposits with age-corrected (161–145 Ma) Middle Jurassic igneous rocks (Fig. 13) suggests that Pb was leached by hydrothermal fluids from the host Middle Jurassic magmatic rocks or was derived by direct exsolution of a magmatic-hydrothermal fluid from Middle Jurassic intrusions at depth. By extension, this interpretation also applies to the other base and precious metals of the Kapan deposits.

**Note:**

The ages for steps marked in bold are the step-heating results included in the weighted plateau age for sample JM016 and the weighted mean age for a region of the age spectrum where the ages broadly tapped at the same time.
between −5.4 and 5.6‰ (Table 3; Fig. 20a). Such a difference between the isotopic composition of pre- and post-mineralization fluids can be explained by both cooling of a magmatic-derived fluid and progressive mixing with seawater or meteoric water.

The positive correlation of $\delta^{18}$O and $\delta^{13}$C values of calcite from the Centralni West and Shahumyan deposits (Fig. 17a) could be consistent with (1) precipitation during fluid cooling, as the fractionation factors for oxygen between calcite and H$_2$O and carbon between CO$_2$ and calcite are linearly correlated with temperature (Ohmoto and Rye, 1979), (2) CO$_2$ degassing or boiling (Zheng, 1990), and (3) fluid/rock interaction or fluid mixing (Zheng and Hoefs, 1993). Calcite solubility in hydrothermal fluids increases with decreasing temperature (Holland and Malinin, 1979); therefore, precipitation of calcite cannot be explained by simple cooling alone. Solely, boiling also cannot explain increasing $^{87}$Sr/$^{86}$Sr ratios at higher or constant $\delta^{18}$O values in hydrothermal calcite (Fig. 17b), because boiling would have no effect on the $^{87}$Sr/$^{86}$Sr ratio of a hydrothermal fluid. Finally, the $^{87}$Sr/$^{86}$Sr ratios of hydrothermal calcite, which are more radiogenic than those of the immediate Middle Jurassic magmatic rocks (Fig. 14), are inconsistent with fluid-rock interaction as the sole mechanism to explain the positive correlation of $\delta^{18}$O and $\delta^{13}$C values of calcite. Therefore, mixing of a magmatic-derived fluid and seawater can explain both the variability in the Sr

<table>
<thead>
<tr>
<th>Sample no.</th>
<th>Total Re (ppb)</th>
<th>±</th>
<th>$^{187}$Re (ppt)</th>
<th>±</th>
<th>Total Os (ppt)</th>
<th>±</th>
<th>$^{187}$Os (ppt)</th>
<th>±</th>
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<td>5.2</td>
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Table 7. Re-Os Isotope Data for Pyrite from the Kapan Mining District, Armenia

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<th>$^{187}$Re/$^{188}$Os</th>
<th>$^{187}$Os/$^{188}$Os</th>
<th>ρ</th>
<th>Model age Ma</th>
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Part 3. Sample locations and weights

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<th>UTM (N)</th>
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Note: All samples from UTM zone 38S
Ore Formation during Jurassic Subduction of the Tethys along the Eurasian Margin

isothe composition and the correlation in $\delta^{18}O$ and $\delta^{13}C$ space, with the stable isotopes being additionally controlled by temperature-dependent fractionation (Zheng and Hoefs, 1993; Zheng, 1999).

Source of sulfur and implications from sulfur isotopes

The sulfur isotope composition of sulfides from the Kapan deposits ranges from 2.0 to 6.5‰ (Fig. 15), which is compatible with a magmatic source for sulfur, either directly derived from an igneous source or by leaching of sulfur-bearing minerals in igneous rocks (Ohmoto, 1986). Using the sulfur isotope geothermometry equations of Kajiwara and Krouse (1971), a temperature range of 96° to 381°C was calculated for pyrite- sphalerite, sphalerite-chalcopyrite, and pyrite-chalcopyrite pairs from the main-stage mineralization at Shahumyan (Table 4). Temperatures calculated from the isotopic composition of pyrite-chalcopyrite pairs from the Centralni West deposit range between 93° and 592°C (Table 4). Pyrite-barite and pyrite-gypsum pairs from the Centralni East deposit indicate precipitation temperatures of 395° and 357°C (Table 4), respectively, using the fractionation equations of Ohmoto and Lasaga (1982).

The wide range of temperatures raises questions on the reliability of calculated results and suggests disequilibrium between the minerals. Such disequilibrium conditions can be attributed to fluid mixing (Zheng, 1991), which is consistent with the conclusions based on the Sr, O, and C isotopes. The $\delta^{34}S$ values of 17.4‰ of barite and 18.0 to 19.5‰ of gypsum from the Centralni East deposit overlap with or are slightly more enriched relative to the $\delta^{34}S$ value of 14.5 to 17.5‰ of Jurassic seawater sulfate (Fig. 15; Claypool et al., 1980). Therefore, the sulfur isotope data of minerals from the Kapan deposits are also consistent with mixing of a hydrothermal fluid dominated by sulfur of magmatic origin and seawater. In such a fluid-mixing scenario, the isotopic composition of sulfide minerals is inherited from the isotopic composition of sulfur in the hydrothermal fluid of magmatic origin prior to mixing (Zheng, 1991), whereas the $\delta^{34}S$ values of sulfate minerals are similar to or slightly higher than contemporaneous seawater (Çagatay and Eastoe, 1995; Ohmoto and Goldhaber, 1997).

Hypogene alunite at the Shahumyan deposit

Oxygen and hydrogen isotope compositions for the two alunite samples hosted in Middle Jurassic quartz-dacite from the northeastern limb of the Shahumyan deposit (Figs. 3, 11a, b) are given in Figure 20b. Fluid compositions in equilibrium with alunite were calculated from equations of Stoffregen et al. (1994) over a temperature range of 400° to 150°C (Table 3, Fig. 20b), which are typical for temperatures related to magmatic vapor condensation and magmatic steam processes (Rye et al., 1992; Rye, 1993, 2005). The calculated fluid compositions (Fig. 20b) overlap with those of typical felsic magmatic fluids (Taylor, 1988) and the range of water compositions discharged from high-temperature fumaroles, i.e., volcanic vapor (Giggenbach, 1992). This indicates a predominant magmatic component in the source fluids for both alunite samples. However, the rather wide field of alunite fluid compositions in Figure 20b (caused in part due to the poorly defined precipitation temperatures as limited by the paragenesis) suggests mixing of a magmatic fluid with meteoric water or seawater. Finally, an equilibrium temperature of 59°C has been calculated from $\Delta^{34}S_{SO_4,OH}$ from the equation of Stoffregen et al. (1994). This is an unreasonably low depositional temperature for alunite in a magmatic environment and is best explained by retrograde isotopic exchange of hydrogen and oxygen from the OH site in alunite with later meteoric water (Rye, 2005).

Based on its sulfur isotope composition of 27‰ (Table 4), its advanced argillic alteration assemblage (quartz-alunite ± kaolinite and dickite), and its metal-enriched character (with 0.24 ppm Au and 103 ppm As; Mederer, 2013), we interpret the disseminated alunite of sample 3-63-1 (Fig. 11b) as belonging to the magmatic-hydrothermal type with sulfate

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**Fig. 20.** a. Calculated $\delta^{18}O$ values of fluids in equilibrium with hydrothermal quartz and calcite at temperatures of 400° to 150°C. See text for details. b. $\delta D$, $\delta^{18}O$$_{H_2O}$, $\delta^{18}O$$_{SO_4}$, and $\delta^{18}O$$_{OH}$ values for vein and disseminated alunite from the Kapan mining district. Fluid compositions ($\Delta^{18}O_{H_2O}$ and $\Delta^{18}O_{SO_4}$) in equilibrium with alunite are calculated from equations of Stoffregen et al. (1994) at 150° to 400°C. Meteoric water line from Craig (1961), kaolinite line from Savin and Epstein (1970), primary magmatic water (PMW) box from Taylor (1974), felsic magmatic fluids from Taylor (1988), volcanic vapor box, i.e., range of water compositions discharged from high-temperature fumaroles, from Giggenbach (1992), and standard mean ocean water (SMOW; Taylor, 1974; Hoefs, 2009).
derived from disproportionation of magmatic SO₂ to H₂S and H₂SO₄ (Rye et al., 1992; Rye, 1993). The presence of dickite indicates formation temperatures of less than 250° to 200°C (Hemley et al., 1980).

Vein alunite of sample 5-77 (Fig. 11a) is interpreted as magmatic-steam alunite (Rye et al., 1992) based on its coarse-grained nature, its occurrence in an extensional vein, its association with hematite and minor pyrite, and its δ³⁴S value of 7.9‰. Hypogene magmatic-steam alunite typically precipitates during expansion of ascending SO₂-rich vapor under disequilibrium conditions without significant disproportionation of SO₂ and reequilibration with the host rock (Rye et al., 1992). Coarsely banded alunite veins are almost monomineralic with only minor associated hematite and pyrite (e.g., Alumine Ridge, Utah; Cunningham et al., 1984; Rye, 1993), and are interpreted to form during rapid episodic degassing of an underlying magma in an extensional setting.

Timing and cooling ages of hydrothermal events

Figure 21 summarizes the Re-Os and ⁴⁰Ar/³⁹Ar ages obtained in this study, together with dated magmatic events in the Kapan district (Mederer et al., 2013), and the sequence of significant geologic events of the Kapan district. Previously, Bagdasaryan et al. (1969) obtained whole-rock K-Ar ages ranging from 137 to 152 Ma for altered host rocks in contact with mineralization. However, they will not be considered any further, because the accuracy of such whole-rock K-Ar ages, especially in the case of altered samples, is questionable (Kelley, 2002). We discuss the meaning and the reliability of the Re-Os pyrite and the ⁴⁰Ar/³⁹Ar muscovite ages obtained in the course of this study.

The subvolcanic Barabatoom quartz-dacite, which hosts the Shahumyan deposit, cuts early Bathonian strata (~168–166 Ma) and is unconformably overlain by Callovian (~166–163.5 Ma) fossil-bearing limestone (Achikgiozyan et al., 1987; Zohrabyan, 2005). A plagioclase separate from the Barabatoom quartz-dacite yielded a disturbed ⁴⁰Ar/³⁹Ar spectra (sample 5-16; Fig. 18c). Nevertheless, the two oldest heating steps yield ages between 160 and 170 Ma, which support a Middle Jurassic age for the Barabatoom quartz-dacite based on crosscutting relationships. The Ar loss profile in the plagioclase is attributed to thermal disturbance after the rock had crystallized, but at a temperature not surpassing the plagioclase argon partial retention zone (160°–375°C; Chew and Spikings, 2015), as the sample still records a Middle Jurassic age in the most retentive parts of the grains.

The ⁴⁰Ar/³⁹Ar muscovite plateau age of 161.78 ± 0.79 Ma obtained from the Centralni West deposit (sample JM016; Fig. 18a) records cooling after hydrothermal alteration and documents the earliest ore-forming event in the Kapan district. This is the most robust and reliable age of hydrothermal ore formation in the Kapan district obtained in the course of our study, and it is consistent with our geologic observations. The Ar loss profile during the lower-temperature steps of the age spectra may be due to secondary alteration or younger heating events to temperatures within the muscovite partial retention zone (310°–435°C; Chew and Spikings, 2015).

The partially disturbed ⁴⁰Ar/³⁹Ar age spectra of the magmatic hydrothermal alunite from the Shahumyan deposit (sample 3-63-1) must be interpreted with caution, since the longest plateau only consists of 40.4% of the total released gas (Fig. 18b). The partially disturbed age spectra may be a consequence of secondary fluid circulation or thermal events that postdate alunite precipitation. Its weighted mean ⁴⁰Ar/³⁹Ar age of 156.14 ± 0.79 Ma is inconsistent with our geologic field observations, indicating that the mineralized veins at Shahumyan are restricted to the Middle Jurassic magmatic complex and do not extend across the late Oxfordian unconformity (Fig. 4).

The same applies to the Re-Os isochron age of 144.7 ± 4.2 Ma (MSWD = 0.099) and the weighted average age of

Igneous intrusive and hydrothermal events in the Kapan district

165.6±1.4 Ma tonalite clasts from pebble dikes
〜163-167 Ma Barabatoom Formation dominated by subvolcanic quartz-dacite (Achikgiozyan et al., 1987; Zohrabyan, 2005)

161.78±0.79 Ma Centralni West on hydrothermal muscovite
Late Oxfordian erosion (~160-157.3 Ma) major unconformity between Middle and Late Jurassic-Early Cretaceous magmatic complexes

? 156.14±0.79 Ma Shahumyan on alunite

? 146.2±3.4 Ma Centralni East on pyrite

139 to 128 Ma intrusions (granodiorite, quartz-monzodiorite, gabbro, diorite, granodiorite, monzonite and granite)

U-Pb zircon, Mederer et al. (2013)
⁴⁰Ar/³⁹Ar age, this study
Re-Os age, this study
U-Pb zircon, Melkonyan et al. (2016)

Fig. 21. Summary of igneous and hydrothermal events in the Kapan district from Middle Jurassic to Early Cretaceous. The uncertain Re-Os isochron age obtained from pyrite of the Centralni East deposit and the ⁴⁰Ar/³⁹Ar age obtained from alunite of the northeastern limb of the Shahumyan deposit are indicated by question marks. See text for details.
146.2 ± 3.4 Ma yielded by pyrite samples from the Centralni East deposit (Fig. 19d-f). Indeed, the Centralni East orebodies are also confined to Middle Jurassic magmatic rocks below the late Oxfordian unconformity (Fig. 4). Furthermore, the initial $^{187}\text{Os}/^{188}\text{Os}$ ratio of 1.41 ± 0.57 obtained for the sulfide mineralization in the Centralni East deposit is similar to the composition of average continental crust at 145 Ma ($^{187}\text{Os}/^{188}\text{Os} = 1.4 ± 0.3$; Peucker-Ehrenbrink and Jahn, 2001) and is significantly more radiogenic than a primitive mantle source ($^{187}\text{Os}/^{188}\text{Os}$ of chondritic uniform reservoir (CHUR) at 145 Ma = 0.12761 ± 0.00046; Chen et al., 1998). The initial $^{187}\text{Os}/^{188}\text{Os}$ composition is inconsistent with the primitive, mantle-dominated Pb, Sr, and Nd isotope composition reported for Late Jurassic-Early Cretaceous magmatic rocks in the Kapan district (Mederer et al., 2013) and suggests that the Re-Os isotope system in pyrite was disturbed by younger thermal events (e.g., Stein et al., 1998). Therefore, the Re-Os pyrite isochron age from the Centralni East deposit must be treated with great caution.

The Re-Os isochron age of 215.7 ± 9.2 Ma and the weighted average model age of 215.7 ± 4.2 Ma yielded by pyrite sampled at the Centralni West deposit (Fig. 19a-c) are at least 45 m.y. older than the Middle Jurassic host rocks of the deposit (Fig. 4). Based on this contradiction, the high uncertainty associated with the initial $^{187}\text{Os}/^{188}\text{Os}$ ratio of the sulfide ore (1.3 ± 3.6), and the high MSWD of 6.9 for the best fit of the Re-Os data, we conclude that the Re-Os isotope system in pyrite of the Centralni West deposit must have been disturbed after it was precipitated. Considering the independently obtained $^{40}\text{Ar}/^{39}\text{Ar}$ muscovite age of 161.78 ± 0.79 Ma for the Centralni West deposit (sample JM016; Fig. 18a), we interpret the Late Triassic Re-Os age to be geologically meaningless.

Further radiometric dating to determine the timing of ore-forming hydrothermal pulses and related igneous events has not been possible in the Kapan district, because suitable minerals were not available. Volcanic, subvolcanic, and volcanoclastic rocks underlie eruptive centers, interpreted as the heat engines sustaining hydrothermal systems in typical VMS districts (e.g., Galley et al., 2007). Tonalite dikes sampled from subvolcanic polymict pebble dikes yielding a U-Pb zircon age of 165.6 ± 1.4 Ma (Mederer et al., 2013), together with gabbro-diorite intersected by drill holes below Kapan (Tumanyan, 1992), provide evidence of intrusive activity at depth during Middle Jurassic arc construction, predating mineralization at the Centralni West deposit by at least 1.6 m.y. The later intrusive association, together with the tholeiitic to transitional composition of the Middle Jurassic volcanic complex (Fig. 5), is comparable to composite, synvolcanic gabbro-diorite-tonalite clusters underlying eruptive centers, interpreted as the heat engines sustaining hydrothermal systems in typical VMS districts (e.g., Galley, 2003; Galley et al., 2007). Moreover, the district-wide epidote alteration at the base of the Middle Jurassic complex in the Kapan area, reported by Cholahyan and Sarkisyan (1972) and Achikgyozyan et al. (1987), is reminiscent of semiconformable, epidote-dominated hydrothermal alteration zones typically described at depth in many VMS districts immediately at the top of synvolcanic gabbro-diorite-tonalite intrusions (e.g., Galley, 1993; fig. 14 in Galley et al., 2007). The submarine setting is supported by the Sr and stable isotope systematics of different hydrothermal minerals sampled at Centralni West (Figs. 14, 15). We conclude that during the Middle Jurassic to Late Jurassic transition (Fig. 22), the Kapan zone corresponded to a nascent volcanic arc with submarine magmatic-hydrothermal ore formation at Centralni West, and it is comparable to other magmatic-hydrothermal and VMS systems associated with submarine volcanic arcs along subduction settings (de Ronde et al., 2005, 2011; Hannington et al., 2005). Because Centralni West is the westernmost deposit of the Kapan district, closest to the subduction trench
~170 to ~160 Ma: Middle to Late Jurassic transition
- Nascent volcanic arc
- Flat subduction
- Tholeiitic to transitional magmatism

~160 to 157 Ma:
- Major erosion before or during late Oxfordian

~157 to ~140 Ma: Late Jurassic - transition to Early Cretaceous
- Slab rollback
- Transitional to calc-alkaline magmatism
- Asthenospheric mantle upwelling

~140 to ~130 Ma: Early Cretaceous
- Slab rollback
- Transitional to calc-alkaline magmatism
- Asthenospheric mantle upwelling

Fig. 22. Proposed sequences of geologic and ore-forming events in the Kapan mining district from the Middle Jurassic to the Early Cretaceous. a. Broadly coeval Middle Jurassic metallogenic events with volcanogenic massive sulfide ore formation at Centralni West at 161.78 ± 0.79 Ma and formation of porphyry-epithermal systems at Centralni East and Shahumyan before the late Oxfordian, based on the evidence that orebodies are only hosted by Middle Jurassic magmatic rocks and do not extend across the late Oxfordian unconformity. b. Middle Jurassic volcanogenic massive sulfide ore formation at Centralni West at 161.78 ± 0.79 Ma, followed by late Oxfordian erosion and Late Jurassic ore formation during arc thickening and slab rollback at Shahumyan and Centralni East if one accepts the questionable 40Ar/39Ar and Re-Os ages from both deposits (see text for discussion). Both scenarios are followed by Late Cretaceous intrusions, including the Tsav intrusion, and stockwork-type Cu-Au-Mo mineralization at the Shikahokh prospect. Scenario (a) is considered as more likely based on field relationships.
(Fig. 3), it may be speculated that this ore deposit might have formed in a fore-arc environment (Fig. 22a), as documented, for instance, in the Uralides (Herrington et al., 2005a, b) and the Caribbean (Torró et al., 2016). However, further studies are required to ascertain such an interpretation.

The polymetallic veins at Shahumyan and the stockwork-type mineralization at Centralni East have distinct characteristics with respect to the Centralni West deposit. Centralni East and Shahumyan are comparable to sulfide-rich base and precious metal veins associated with epithermal-porphyry systems (e.g., Simmons et al., 2005; Sillitto, 2010), such as the Main Stage veins in Butte, U.S.A. (Rusk et al., 2005), or the polymetallic veins in Morococha, Peru (Catchpole et al., 2011). The Centralni East and Shahumyan deposits contain high-sulfidation mineral assemblages (Figs. 5d, 10d) and advanced argillic alteration including hypogene alunite (Fig. 11a, b), which are typically attributed to disproportionation of magmatic SO2 during mixing with surface waters in subaerial settings (Giggenbach, 1992, 1997; Einaudi et al., 2003), but which are also recognized in submarine hydrothermal systems (e.g., de Roule et al., 2005, 2011; Huston et al., 2011). Based on Sr and stable isotope systematics, a submarine or a transitional subaerial-submarine environment can be envisaged in the Kapan district (Figs. 14, 15).

The orebodies at Shahumyan and Centralni East are confined to Middle Jurassic magmatic rocks and do not extend across the late Oxfordian unconformity (Fig. 22a). Based on this geologic evidence, we conclude that both deposits were formed early during the magmatic arc evolution and before the late Oxfordian (Fig. 22a). In the absence of relative crosscutting relationships, we interpret them as being broadly coeval or having been formed within a short time frame with respect to the Middle Jurassic Centralni West submarine hydrothermal deposit. Such a metamorphic setting would be comparable to similar hybrid or juxtaposed VMS-epithermal porphyry systems described in island-arc settings in the Pacific and in Australia (e.g., Hannington, 1997, 2011; Large et al., 2001).

This interpretation is contradicted by the Re-Os pyrite weighted average date of 146.2 ± 3.4 Ma (Fig. 19d-f) and the weighted mean 40Ar/39Ar age of 156.14 ± 0.79 Ma (Fig. 18b) obtained for pyrite samples from Centralni East and for alunite from Shahumyan, respectively. These ages tell us that both deposits were formed during the Late Jurassic, significantly later than Centralni West and after the late Oxfordian erosion event (Figs. 21, 22b). Several pieces of evidence speak against such a scenario. As discussed above, both radiometric ages are highly questionable, and the Re-Os and Ar systems were likely disturbed during overprinting thermal events. Furthermore, it would be remarkable that a Late Jurassic magmatic-related and epigenetic ore system forming stockwork and steep veins would remain confined to the Middle Jurassic rock complex below the late Oxfordian unconformity (Fig. 22b). Finally, the homogenized Pb isotope composition of hydrothermal minerals supports a coeval and common metal source for the Centralni West, Centralni East, and Shahumyan deposits (Fig. 13), and indicates that Pb and the other metals were solely derived from Middle Jurassic magmatic rocks or magmas. There is no supporting evidence for any metal input from Late Jurassic magmatic rocks or magmas. Therefore, we conclude that the combined field and geochemical dataset supports ore formation within a short time frame at Centralni West, Centralni East, and Shahumyan before the late Oxfordian (Fig. 22a), rather than distinct Middle and Late Jurassic ore-forming pulses separated by 5 to 15 m.y. (Fig. 22b).

The Shikahogh Cu-Au-Mo prospect associated with the Tsav intrusion (Fig. 2) shows that, after the Jurassic, renewed magmatic activity and ore formation affected the Kapan block during the Early Cretaceous (Figs. 21, 22). The 139 to 125 Ma U-Pb ages of the Tsav intrusion and the spatially associated Shikahogh prospect coincide with porphyry and epithermal ore formation in the Gedabek district within the Somkheto-Karabagh arc (Fig. 1) at about 133 Ma (Moritz et al., 2016a).

Conclusions

The Kapan district consists of three major ore deposits: Centralni West, Centralni East, and Shahumyan. The three deposits have different metal endowments, orebody geometries, hydrothermal alteration features, and opaque mineral assemblages. However, they are all hosted by Middle Jurassic magmatic rocks cut by a late Oxfordian unconformity, and they have lead isotope compositions indicating derivation of metals from a common Middle Jurassic magma or from the same Middle Jurassic magmatic host rocks. We conclude that the Cu-dominated Centralni West ore deposit was formed along a submarine volcanic arc in a subduction environment, as evidenced by its volcanic host rocks deposited in a subaqueous setting, and its hydrothermal alteration dominated by chlorite, carbonate, quartz, epidote, pyrite, and sericite. The submarine setting is also supported by the Sr and stable isotope compositions of hydrothermal minerals. A well-defined 40Ar/39Ar plateau age of 161.78 ± 0.79 Ma of hydrothermal muscovite indicates that the Centralni West deposit formed early during Middle Jurassic arc construction dominated by tholeiitic to transitional magmatism.

The Shahumyan polymetallic vein-type and the Centralni East Cu-Au stockwork-type deposits contain intermediate- to high-sulfidation opaque mineral assemblages and are hosted by magmatic rocks affected by advanced argillic alteration. They are comparable to sulfide-rich base and precious metal veins associated with epithermal-porphyry systems. Strontium and stable isotope systematics support a submarine or a transitional subaerial-submarine setting. Since orebodies at Shahumyan and Centralni East are confined to Middle Jurassic magmatic rocks, do not extend across the late Oxfordian unconformity, and are cut by Late Jurassic-Early Cretaceous magmatic rocks, we conclude that both deposits were formed broadly in the same time interval as the Centralni West Cu deposit in a nascent tholeiitic to transitional magmatic arc.

Our metallogenic interpretation for the Kapan district could be challenged by a disturbed 40Ar/39Ar plateau age of 156.14 ± 0.79 Ma (only 40% of total released gas) of magmatic-hydrothermal alunite from Shahumyan and a 146.2 ± 3.4 Ma Re-Os weighted average model age of pyrite samples from the Centralni East deposit. However, both ages are rejected because they contradict geologic field relationships and lead isotope systematics. We conclude that both isotope ages were affected by overprinting events, including Early Cretaceous magmatism within the Kapan block, Cretaceous collision of
the South Armenian and the Kapan blocks, final Cenozoic Arabia-Eurasia collision, and Cenozoic magmatic activity in the western Kapan district and the South Armenian block.

Acknowledgments

This study was supported by the Swiss National Science Foundation (grants 200020-121519, 200020-138130, and 200020-168996), the SCOPES project (IZ73ZO-128324), and Dundee Precious Metals, which provided field and logistical support and access to the mine area. J.M. benefited from travel grants from the Augustin Lombard Foundation of the Société de Physique et d’Histoire Naturelle de Genève and an SEG Student Research Grant from the Hugh E. McKinstry Fund. Tamara Beridze is acknowledged for the translation of selected articles from Russian into English and Arman Vardanyan for his support in the field. Michele Sinn-Gerber and Fabio Capponi are thanked for analytical support and technical assistance at the University of Geneva, and Benita Putlitz and Kerstin Bauer for their assistance in the stable isotope laboratory at the University of Lausanne. D.S. acknowledges the TOTAL endowment fund and a Dida Scholarship from CUG, Wuhan. Thanks to Sergej Zohrabyan and Rafael Melkonyan for fruitful discussions about the geology of the Kapan district. Alan Wainwright and Timothy Baker are thanked for their constructive and very helpful reviews, which allowed us to considerably improve this manuscript.

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