

Trace elements in fluid inclusions of sediment-hosted gold deposits indicate a magmatic-hydrothermal origin of the Carlin ore trend

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ABSTRACT

The Carlin-type deposits in Nevada (western USA) constitute the world's second-largest gold ore province. These structurally and stratigraphically controlled, sediment-hosted ore bodies are characterized by carbonate dissolution attending hydrothermal precipitation of gold-rich arsenian pyrite. The origin of the mineralizing fluids and the source of the gold remain debated. Conceptual models, favoring either sedimentary, metamorphic, or magmatic fluid sources, are based on isotopic tracers, giving ambiguous results. Here we use the trace element compositions of fluid inclusions to separate geochemical signals of the large-scale fluid source from effects of deposit-scale fluid interaction with the sedimentary host rocks. Specifically, we compare the ratios of Rb, K, B, As, Sr, and Ba between clearly magmatic-hydrothermal Cu-Au ores at Copper Canyon in the Battle Mountain–Eureka trend with the Gold Quarry and Chukar Footwall deposits on the Carlin trend that contain high-grade gold in similar sedimentary host rocks. Results indicate that both ore districts can be related to upper crustal hydrous magmatic intrusions, but are now exposed at different levels of erosion and formed at different distances from their magmatic fluid source. Fluid compositions are best explained by separation of a deep magmatic fluid into Rb-K-enriched brine and B-As-Au-enriched vapor, followed by cooling and contraction of the magmatic vapor phase to an epithermal liquid, which reacted with Sr-Ba-bearing sedimentary rocks during ascent and eventual precipitation of Au-rich arsenian pyrite.

INTRODUCTION

The Eocene Au deposits of Nevada (western USA) collectively represent the second-largest Au concentration on Earth (the Archean Witwatersrand Basin, South Africa, is the largest), accounting for ~6% of total

world production (Muntean et al., 2011). Most Carlin-type Au deposits are aligned on three trends in the Basin and Range Province of Nevada (Fig. 1A). Despite their economic attraction, only two comparatively minor districts of similar type have been found outside Nevada (Su et al., 2009; Tucker et al., 2012). Because they are geologically subtle and lack particles of free gold, Carlin-type deposits are difficult to discover without a process-based exploration model.

Carlin-type mineralization forms structurally and stratigraphically controlled orebodies by hydrothermal replacement of organic-rich and variably silty carbonate rocks of early Paleozoic age (Cline et al., 2005). Deposition of gold in solid solution within arsenian pyrite is thought to be controlled by reduction and desulfidation of an Au-As-S-rich fluid attending dissolution of ferroan calcite (Hofstra and Cline, 2000).

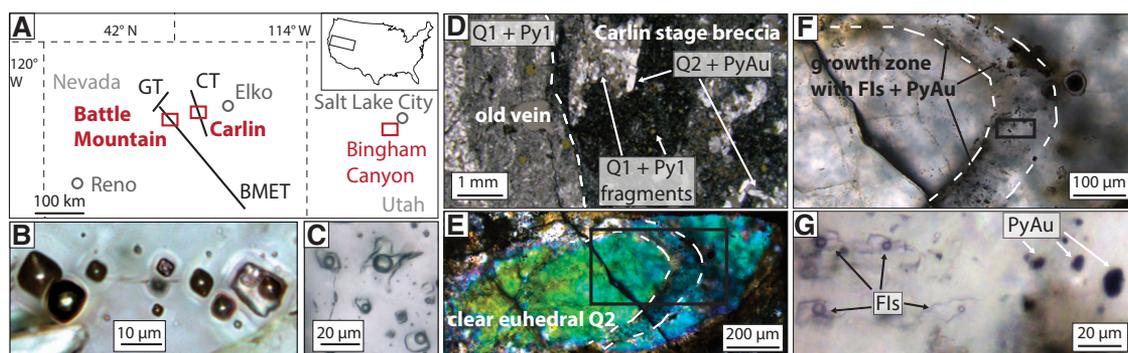
Although the geological characteristics of Carlin-type deposits are well studied, the processes associated with ore formation remain controversial. Three models have been suggested for the source of the Au and its transporting fluids.

1. The sedimentary model favors gold leaching from pre-enriched sediments by circulation of meteoric fluids, driven by elevated heat flow and increased permeability due to extension in the Eocene (Ilchik and Barton, 1997; Seedorff and Barton, 2004; Emsbo et al., 2003).

2. The metamorphic model favors extension initiating prograde metamorphism with lower crustal metamorphic fluids ascending along basement-penetrating rift faults and scavenging gold from pre-enriched metasediments (Cline et al., 2005; Hofstra and Cline, 2000; Large et al., 2011).

3. The magmatic model favors subjacent plutons exsolving magmatic Au-bearing fluids (Sillitoe and Bonham, 1990), separating into an Au-depleted brine and an Au-enriched vapor phase, the latter subsequently contracting to an aqueous ore fluid during cooling (Heinrich, 2005; Muntean et al., 2011).

Figure 1. A: Sampled deposits (red rectangles) in the Au district of Nevada (western USA), consisting of the Battle Mountain–Eureka trend (BMET), Carlin trend (CT), and Getchell trend (GT) (black lines). **B:** A characteristic boiling fluid inclusion assemblage with coexisting brine and vapor phases from Copper Canyon. **C:** Characteristic low-density, aqueous inclusions from Copper Canyon. **D:** Photomicrograph from a high-grade ore zone in the Gold Quarry deposit. An early hydrothermal vein with barren euhedral pyrites (Py1) and an early quartz generation (Q1) is cut by a later breccia rich in Carlin-type Au mineralization containing fragments of the older material cemented by new clear euhedral quartz (Q2) and Carlin-type Au-As-bearing pyrite (PyAu). **E:** Close-up in crossed-polarized light of euhedral quartz with clear growth zoning. **F:** Shadows in the growth zone are ubiquitous fluid inclusions coexisting with Carlin-stage pyrites (small black dots). **G:** Fluid inclusions within the growth zone and coexisting anhedral Carlin-stage arsenian pyrites.



D: Photomicrograph from a high-grade ore zone in the Gold Quarry deposit. An early hydrothermal vein with barren euhedral pyrites (Py1) and an early quartz generation (Q1) is cut by a later breccia rich in Carlin-type Au mineralization containing fragments of the older material cemented by new clear euhedral quartz (Q2) and Carlin-type Au-As-bearing pyrite (PyAu). **E:** Close-up in crossed-polarized light of euhedral quartz with clear growth zoning. **F:** Shadows in the growth zone are ubiquitous fluid inclusions coexisting with Carlin-stage pyrites (small black dots). **G:** Fluid inclusions within the growth zone and coexisting anhedral Carlin-stage arsenian pyrites.

Isotopic tracers give ambiguous results regarding meteoric, sedimentary, or magmatic sources of fluids associated with the main ore stage (e.g., Cline and Hofstra, 2000; Kesler et al., 2005). A possible distal relation to a hidden magmatic fluid source beneath the Carlin trend is indicated by minor but widespread dikes similar in age to the deposits on the Carlin trend (Ressel and Henry, 2006).

Here we compare the first laser ablation–inductively coupled plasma–mass spectrometry (LA-ICP-MS) microanalyses of mineralizing fluids in Carlin-type Au-As deposits on the Carlin trend, where no coeval large magmatic intrusions are exposed, with unambiguously porphyry-related Cu-Au mineralization of the same age at Copper Canyon in the adjacent Battle Mountain–Eureka trend. Our fluid-chemical comparison tests the hypothesis that Copper Canyon exposes a more deeply eroded equivalent of the fluid source beneath the Carlin trend.

GEOLOGY OF TWO CONTRASTING DEPOSITS

The Eocene Copper Canyon Cu-Au-(Pb-Zn-Ag) deposit is partly hosted in a porphyry intrusion cut by numerous quartz veins. High-grade ore zones are lithologically and structurally controlled in carbonate-bearing clastic rocks (Breitt et al., 2010; Theodore et al., 1973; see the GSA Data Repository¹). The Golconda thrust and overlying less reactive rocks are interpreted as having acted as aquitards. Mineralization styles include sulfide minerals in stockwork veins within the Copper Canyon porphyry, sulfide-quartz veins in sedimentary rocks surrounding the central porphyry, and Cu sulfides and native gold in skarn-altered sedimentary rocks (Breitt et al., 2010).

The Gold Quarry and nearby Chukar Footwall deposits are typical Carlin-type deposits of Eocene age in the center of the Carlin trend. They are hosted in Devonian carbonate rocks containing varying amounts of carbonaceous material (Harlan et al., 2002). Prior to the Carlin event, skarn alteration, contact metamorphism, and a few large polymetallic sulfide-quartz veins related to a Jurassic granitoid intrusion affected the host rocks. Carlin-type Au deposition is structurally and stratigraphically controlled by northwest- and northeast-trending faults within a zone of silicification and local kaolinite ± alunite alteration. The faults are interpreted as fluid feeders, including high-angle collapse breccias with high Au grades (Rhys et al., 2015). The ore bodies have sharp contacts against overlying less permeable rocks (Rhys et al., 2015). Alteration styles include decalcification, dolomitization, silicification, argillization, sulfidation, alunization, and later supergene leaching, but only sulfidation is unquestionably synchronous with the Au-mineralization event (Harlan et al., 2002). Typical for Carlin deposits, invisible gold is ionically bound in arsenian pyrite or arsenic-rich pyrite rims overgrowing earlier pyrite or marcasite. The Au-As-rich sulfides mostly occur disseminated in altered sedimentary rocks, but were also found in quartz-bearing veinlets and euhedral quartz crystals, identifying these as part of the ore-forming event (Figs. 1D–1G).

METHODS

At Copper Canyon, samples were collected from porphyry stockwork quartz veins, sulfide-quartz veins in the porphyry and in surrounding mineralized and altered sedimentary rocks, and from mineralized garnet skarn. At the Carlin trend deposits, samples were collected from fault zones interpreted to have acted as feeder zones and from strongly mineralized areas with significant veining or brecciation (Fig. 1D). These samples provide the best chances of finding inclusion assemblages that trapped the ore-forming fluids.

Microthermometry on fluid inclusions was conducted on a Linkam THMSG-600 cooling-heating stage. LA-ICP-MS microanalyses were performed with a Perkin Elmer Elan 6100 ICP quadrupole MS and an Element XR ICP sector field MS connected to a GeoLas 193 nm ArF excimer laser system.

¹GSA Data Repository item 2016341, Figures DR1–DR7 and Tables DR1–DR3 showing data, sample locations, petrographic images, and ternary diagrams, is available online at <http://www.geosociety.org/pubs/ft2016.htm> or on request from editing@geosociety.org.

To date, obtaining chemical data by LA-ICP-MS from fluid inclusions representing the Carlin ore fluids has been hampered by the scarcity and small size (<10 μm) of inclusions in fine-grained ore-stage jasperoid quartz (Cline et al., 2005). For this study we identified ore-stage fluid inclusions hosted in coarser quartz grains in veinlets and breccia zones by relating them to arsenian pyrite grains that are hosted in the same quartz generation (Figs. 1F and 1G). Analyzed arsenian pyrites within these larger quartz crystals are chemically and morphologically identical to typical Carlin-style pyrites in adjacent jasperoid quartz, in contrast to older, commonly euhedral, pyrites with a very different trace element chemistry. Such detailed petrography enables reliable association of fluid inclusion assemblages with the main ore-forming event at the Carlin trend. All inclusions are small (mostly <25 μm; see Fig. 1G), but some are large enough to provide reliable microanalytical data (for more details, see the Data Repository).

RESULTS

Chemical Composition of Fluids at Battle Mountain

At Copper Canyon, three fluid inclusion types were recognized by petrography and microthermometry: (1) vapor inclusions (Fig. 1B) with high vapor/liquid ratios and low salinities (<12.4 wt% NaCl equivalent); (2) brine inclusions (Fig. 1B) with a low vapor/liquid ratio but high salinities (28.5–41 wt% NaCl equivalent), commonly containing halite crystals; and (3) aqueous inclusions with intermediate to low vapor/liquid ratios ranging from 40 to 20 vol% vapor (Fig. 1C) and varying salinities (1–34 wt% NaCl equivalent).

Four groups of fluid compositions can be distinguished by analyses of individual fluid inclusions by LA-ICP-MS. (1) Brine inclusions (Fig. 2A) are enriched in K and Rb. (2) Vapor inclusions (Fig. 2B) are enriched in B and As. The aqueous inclusions can be separated into two groups: (3) a group with low salinities (<12.5 wt% NaCl equivalent) and chemical characteristics similar to those of the vapor inclusions (Fig. 2C) and (4) a group of inclusions with higher salinities (8.8–34 wt% NaCl equivalent) that are enriched in Ba and Sr (Fig. 2D) and only occur in skarn samples.

Fluid inclusion compositions in groups 1 and 2 can be explained by phase separation of a magmatic fluid of intermediate density with some elements partitioning into the brine (e.g., K, Rb), while others partition into the vapor phase (e.g., B, As; Figs. 2A and 2B; Heinrich et al., 1999; Landtwing et al., 2010). Overlapping salinity and element ratios of vapor and low-salinity aqueous inclusions (Figs. 2B and 2C) suggest that fluids of group 3 are the result of isochemical contraction of a magmatic vapor during cooling (Heinrich, 2005). The aqueous inclusions in group 4 are best explained by the interaction of magmatic fluids with the surrounding calcareous sedimentary rocks that contain high contents of soluble Sr and Ba (Fig. 2D).

Chemical Composition of the Carlin Ore Fluids

Fluid inclusions from the Carlin-type deposits have intermediate to low vapor/liquid ratios (Fig. 1G) and salinities mostly between 1 and 7 wt% NaCl equivalent; they homogenize between 120 and 235 °C (T_h). No higher-salinity or vapor-like inclusions were observed. We are confident to have identified ore-stage fluid inclusions in veinlets on the basis of detailed petrography, notably by their intimate association with arsenian pyrite (Figs. 1D–1G). Our microthermometric data agree with previous values reported for ore-stage fluid inclusions in jasperoid quartz (T_h of 180–240 °C and salinities between 2 and 3 wt% equivalent; Cline and Hofstra, 2000; Hofstra and Cline, 2000).

LA-ICP-MS analyses detected B, S, K, As, Rb, Sr, Sb, Cs, Ba, and Tl despite the small inclusion sizes (Table DR1 in the Data Repository). The proportions of the trace elements such as relatively high B and As but low Rb and K in the Carlin fluids overlap with those of magmatic vapor-derived fluids at Battle Mountain (Figs. 3A and 3B), but extend to higher Ba and Sr contents (Figs. 3C and 3D).

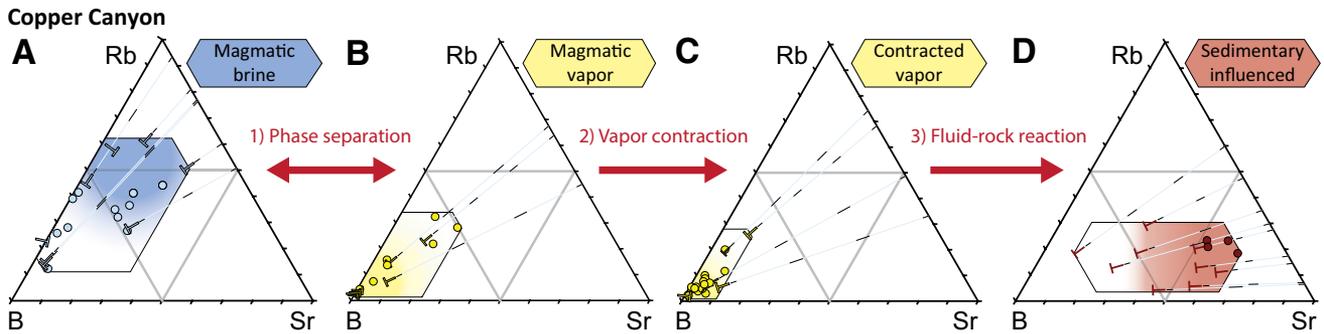


Figure 2. Reconstructed fluid evolution at Copper Canyon (Nevada, USA) from microanalyses of fluid inclusions. Assemblages from Copper Canyon are divided according to petrographic observations and element ratios obtained by laser ablation–inductively coupled plasma–mass spectrometry analyses. **A:** Magmatic brines. **B:** Magmatic vapors. **C:** Contracted vapors. **D:** Sedimentary-influenced fluids. Circles indicate that all three elements are above limit of detection (LOD); T-symbols show that two elements are detected, with T-bar marking LOD of third element; i.e., actual value is located along the dashed line (for details, see the Data Repository [see footnote 1]); Hexagons show range for each fluid type based on maximum and minimum values of measurements and LOD, with intensity of shading emphasizing inferred end-member fluids, brine (blue), vapor (yellow), and sedimentary influenced (red).

Carlin, Copper Canyon and other deposits

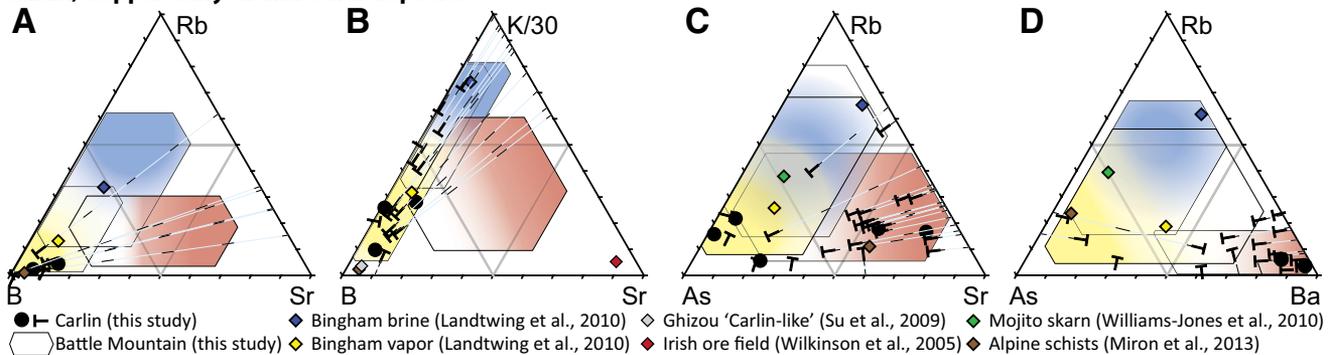


Figure 3. Comparison of fluid chemistry from the Carlin deposits, Nevada, USA (circles, T-symbols, dashed lines) with the end-member fluid fields found at Copper Canyon (hexagons; for data points in K-B-Sr, Rb-As-Sr, and Rb-As-Ba diagrams, see the Data Repository [see footnote 1]) and other deposit types (diamonds). Data support the interpretation that B-As-(S-Au)-rich contracted magmatic vapor is the mineralizing ore fluid for Carlin gold deposits (yellow), locally evolving toward Sr-Ba enrichment by reaction with sedimentary host rocks at the ore deposition site (red).

COMPARISON OF THE CARLIN FLUIDS WITH OTHER FLUID TYPES

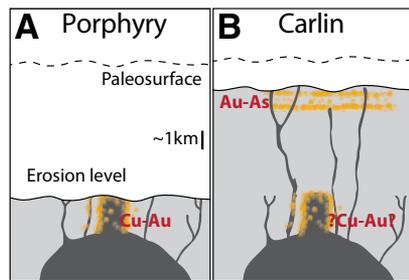
Our distinction of fluid end members based on trace elements is supported by data reported from other hydrothermal ore deposits. Fluid inclusion analyses from Bingham Canyon and other porphyry Cu-Mo-Au deposits confirm selective enrichment of B and As in magmatic vapor, in contrast to enrichment of Rb, Cs, and K in magmatic brines (Fig. 3; Heinrich et al., 1999; Landtwing et al., 2010). Sedimentary basin fluids are enriched in Sr (Sr/Na: 0.024; Kharaka and Hanor, 2014) compared to most Carlin fluids (Sr/Na: 0.0069; data in Table DR2). Trace element data for basin fluids obtained by LA-ICP-MS are sparse, but fluids from the Irish ore field (Wilkinson et al., 2005) are enriched in Sr relative to B and K to an even greater extent than the Carlin fluids and sediment-equilibrated fluids from Battle Mountain (Fig. 3B). Total K concentrations and K/Na ratios in basin fluids are lower than in the Carlin fluids (Table DR2; Wilkinson et al., 2005; Kharaka and Hanor, 2014). Pb-Zn ore-forming fluids from the Mochito skarn deposits in Honduras (Williams-Jones et al., 2010) have higher salinities (0.5–20.8 wt% equivalent) and plot in the magmatic field toward the brine end member, based on their strong Rb enrichment and Ba depletion (Figs. 3C and 3D). However, Sr contents are comparable to Carlin fluids, and the As/Sr ratio from Mochito is between the ratios of the vapors from Battle Mountain and the Carlin fluids. Metamorphic fluids from calc-schists in the Alps (Miron et al., 2013) have similar salinities (~4 wt% equivalent), higher B/Sr ratios, but overall much lower K/Na, As/Na, Sb/Na, and Ba/Na ratios compared to the Carlin ore fluids and the

Battle Mountain magmatic vapor (see Table DR2). Furthermore, their As/Sr ratio is as low as the sediment-equilibrated end member of the Battle Mountain fluids. Fluid inclusion data from the Carlin-like deposits in China (Su et al., 2009) also plot in the magmatic vapor field (Fig. 3B). Although occurring in deposits similar to the Carlin deposits, these inclusions contain significant CO₂ and formed at higher pressure and temperature conditions than the Carlin fluids studied here (Su et al., 2009).

INTERPRETATION AND CONCLUSIONS

Comparing fluid inclusion compositions of typical Carlin-style mineralization at the Gold Quarry and Chukar deposits with unquestionably porphyry-related magmatic-hydrothermal fluids at the Copper Canyon deposit indicates different levels of erosion in otherwise comparable magmatic-hydrothermal systems of crustal extent (Fig. 4). At Copper Canyon, magmatic fluids exsolved from a subjacent magma chamber and phase separated upon ascent through the porphyry intrusion. Hypersaline, K-Rb-Cs-Pb-enriched brines were trapped in porphyry stockwork veins, while B-As-Sb-enriched and presumably S-Au-enriched magmatic vapors cooled at elevated pressure, contracting homogeneously to an aqueous liquid during ascent along fractures and faults through the surrounding sedimentary rocks. Some of the gold precipitated proximally in sediment-hosted ores as a result of partial desulfidation of the contracting vapor due to the precipitation of iron and base-metal sulfides. Interaction with sedimentary host rocks enriched the fluids in Ba and Sr (Figs. 2 and 3), but acid neutralization by carbonate dissolution helped to keep gold in

Figure 4. Schematic interpretation of Eocene Au deposits in Nevada (western USA) forming in vertically extensive magmatic-hydrothermal systems, driven by plutons at depth and exposed today at different levels of formation and subsequent erosion. A: Copper Canyon porphyry-skarn deposit as a proximal and deep deposit. B: Carlin-type deposits as distal and shallow deposits.



solution where gold bisulfide complexes were not exhaustively destabilized by sulfide precipitation (Heinrich, 2005). Base-metal-depleted but still As-S-B-Sb-Au-rich fluids then ascended along permeable pathways until they reached favorable structural sites for Carlin-style Au-As-(Sb-Hg-Tl) ore deposition. At Copper Canyon this higher level has been eroded, but minor Carlin-type deposits occur farther north and south along the Battle Mountain–Eureka trend (Yigit and Hofstra, 2003). Precipitation of gold-enriched arsenian pyrite was dominantly driven by reaction with reduced Fe^{2+} from sedimentary rocks (Hofstra and Cline, 2000; Kesler et al., 2003), either by direct reaction with Fe carbonates or by mixing with sediment-equilibrated FeCl_2 -(BaCl₂-SrCl₂)-rich fluids of marine, meteoric, or magmatic origin. Such redox contrasts associated with Au-As pyrite precipitation are consistent with sharp variations in sulfur isotope signatures of a magmatic-hydrothermal fluid (Kesler et al., 2005).

More generally, this study demonstrates that trace element ratios in fluid inclusions can be used to track fluid source processes and distinguish them from processes of fluid-fluid interaction (phase separation, fluid mixing) and fluid-solid interaction (rock dissolution, alteration, ore mineral precipitation). Trace element proportions help in understanding complex fluid processes in debated ore-forming systems, because they can be directly related to large-scale mass transfer in Earth's crust. Thus, trace elements can be less ambiguous than passive fluid tracers like hydrogen and oxygen isotope ratios (Cline and Hofstra, 2000; Emsbo et al., 2003), which primarily record the origin of the water solvent.

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