

Copper- and cobalt-rich, ultrapotassic bittern brines responsible for the formation of the Nkana-Mindola deposits, Zambian Copperbelt

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ABSTRACT

The Central African Copperbelt (CACB) is Earth's largest repository of sediment-hosted copper and cobalt. The criticality of these elements in battery technology and electricity transmission establishes them as fundamental components of the carbon-free energy revolution, yet the nature and origin of the hydrothermal fluids responsible for ore formation in the CACB remain controversial. Here, we present microthermometric, scanning electron microscopy and laser ablation–inductively coupled plasma–mass spectrometry analyses of fluid inclusions from the Nkana-Mindola deposits in Zambia. We find that base metal concentrations vary by one to two orders of magnitude between “barren” and “ore” fluids, with concomitant distinctions in major salt chemistry. Primary fluid inclusions, hosted by pre- to synkinematic mineralized quartz veins, are characterized by high homogenization temperatures (~200–300 °C) and salinities, with K/Na >0.8 and elevated metal concentrations (10² to 10³ ppm Cu and Co). Conversely, barren, post-kinematic vein quartz contains lower homogenization temperature (~110–210 °C) and lower-salinity primary inclusions, characterized by K/Na <0.8 with low metal contents (<10² ppm Cu and Co). We propose a model in which high-temperature, sulfate-deficient, metalliferous, potassic residual brines, formed during advanced evaporation of CaCl₂-rich, mid-Neoproterozoic seawater, were responsible for ore formation. During basin closure, lower-temperature, halite-undersaturated fluids interacted with evaporites and formed structurally controlled, sodic metasomatism. Reconciliation of these fluid chemistries and base metal concentrations with reported alteration assemblages from a majority of Zambian Copperbelt deposits suggests highly evolved, residual brines were critical to the formation of this unique metallogenic province.

INTRODUCTION

Copper and cobalt are critical metals for carbon-free energy generation, yet we do not fully understand how they become concentrated in hydrothermal deposits in Earth's crust. Metal concentrations in the fluids involved provide a key constraint on ore-forming processes, but, although measured in a variety of systems (e.g., Audetat et al., 1999; Wilkinson et al., 2009), such data are largely absent for sediment-hosted Cu ± Co deposits, one of the primary repositories of these metals. Here, we report evidence for ore formation in the Zambian Copperbelt (ZCB),

southern Central African Copperbelt (CACB), from pre- to synorogenic, potassic brines that contain unusually high Cu-Co-(Pb-Zn) concentrations, and we infer an origin via evaporation of CaCl₂-rich Neoproterozoic seawater.

GEOLOGIC SETTING

The Cu ± Co deposits of the ZCB are primarily hosted in Lower Roan Subgroup clastic units of the late Precambrian Katangan Supergroup, fringing Archean to Mesoproterozoic basement inliers within the Katangan Basin (Fig. 1). Katangan sediments (<877 ± 11 Ma; Armstrong et al., 1999) were deposited during two rift phases, with the Lower Roan Subgroup

reflecting the earliest stage of rifting, during periods of terrigenous and shallow-marine clastic deposition. The Zambian portion of the basin was inverted and metamorphosed to greenschist facies during the Lufilian orogeny, which peaked at 545–530 Ma (John et al., 2004).

The Nkana-Mindola deposits (containing >15 Mt Cu and >0.4 Mt Co; Taylor et al., 2013), which represent a typical style of ZCB mineralization, are located on the western flank of the Kafue anticline (Fig. 1) and on the northeastern limb of the Nkana Syncline. The Nkana South and Central orebodies are hosted by locally dolomitic shales, pyritic argillites, and schists, whereas the Mindola orebody is hosted by more arenaceous, dolomitic lithologies. Cu ± Co mineralization formed disseminated sulfides and coarser aggregates within veins, typically comprising bornite and chalcopyrite, with accessory carrollite, and minor chalcocite and pyrite (Brems et al., 2009).

The presence of veins spanning pre-orogenic and Lufilian stages at Nkana-Mindola provides an excellent opportunity to assess the compositional evolution of fluids responsible for some of the vein-forming ± associated disseminated mineralizing events in the ZCB. Using relationships with the prevailing tectonic fabric, the veins can be subdivided into two end-member generations. Bedding-parallel, quartz-carbonate-sulfide veins are most abundant in the carbonaceous shales of the Nkana South and Central orebodies, and within dolomitic lithologies in the Mindola North open pit (Fig. 1B). These <1-cm-wide to ~25-cm-wide veins form planar or folded and boudinaged arrays (Fig. 1C), implying formation prior to peak orogenic conditions. In contrast, massive quartz veins with lesser

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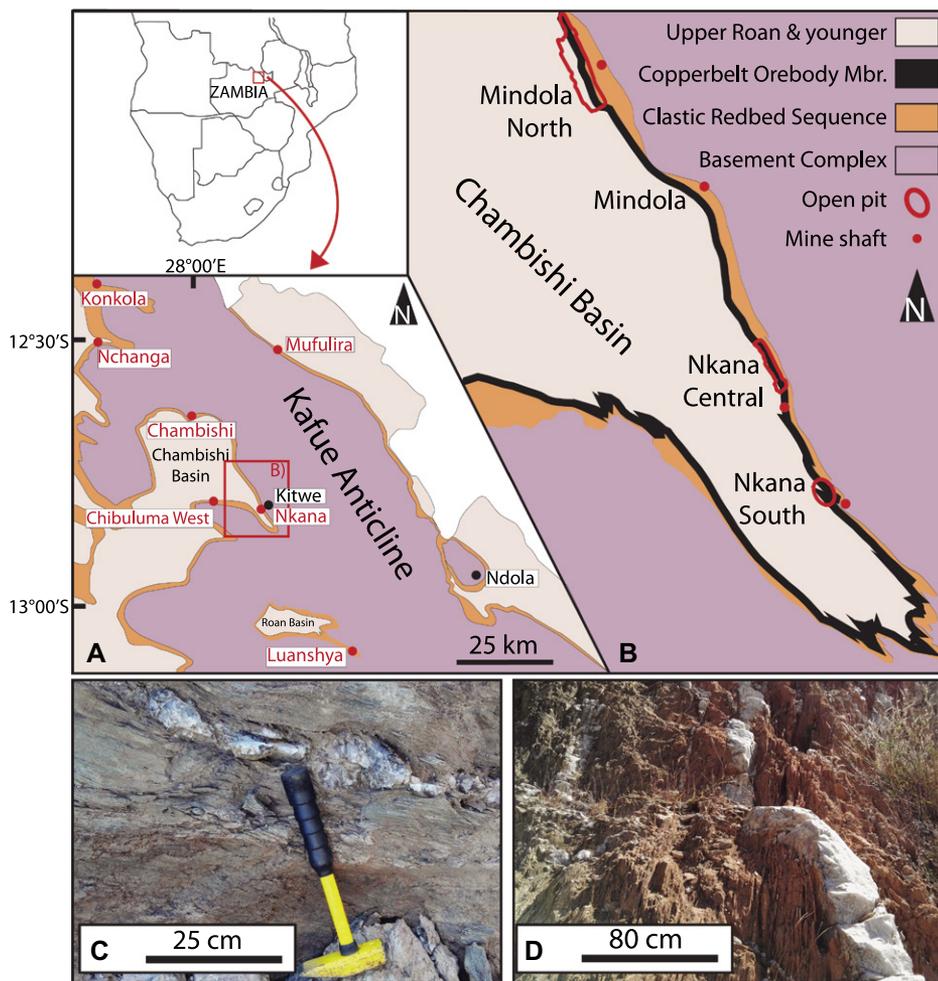


Figure 1. (A) Geological map of the Zambian Copperbelt showing Archean–Mesoproterozoic basement complexes with younger metasedimentary rocks of the Katangan Supergroup flanking the Kafue anticline. (B) Local geology with Nkana–Mindola open pit outlines and mine shafts, adapted from Torremans et al. (2014) and McGowan et al. (2006). (C) Example of shale-hosted, pre- to synkinematic, boudinaged quartz-bornite-chalcocopyrite vein in Nkana South open pit. (D) Example of arkose- and shale-hosted, discordant quartz vein crosscutting Lufilian foliation in Nkana Central open pit.

carbonate and rare pyrite typically crosscut bedding and the Lufilian cleavage fabrics (Fig. 1D), indicating postorogenic timing. Such veins are typically 40–60 cm wide and often transgress both mineralized argillites and hanging-wall siliciclastic rocks.

METHODS

Fluid inclusions in 14 vein samples from Nkana–Mindola were categorized as either primary or secondary using conventional criteria (Roedder, 1984), and their spatial and textural relationships to zones of distinct cathodoluminescence response (see example images in the Supplemental Material¹) were determined.

¹Supplemental Material. Appendices S1 (sample images and descriptions, detailed methods, and example cathodoluminescence images) and S2 (full analytical results). Please visit <https://doi.org/10.1130/GEOL.S.13150871> to access the supplemental material, and contact editing@geosociety.org with any questions.

Microthermometric analyses of ~190 fluid inclusions and 120 laser ablation–inductively coupled plasma–mass spectrometry (LA-ICP-MS) analyses were completed for 23 fluid inclusion assemblages (Figs. 2 and 3). High-resolution element maps of daughter phases within fluid inclusion cavities were acquired using scanning electron microscope imaging. Sample descriptions, images, full analytical data, and methods are provided in the Supplemental Material.

RESULTS

Primary inclusions in vein quartz hosting Cu- and Co-sulfides in pre- to synkinematic veins contain KCl-enriched fluids with homogenization temperatures of ~200–300 °C and salinities of >40 wt% NaCl + KCl equivalent. These inclusions invariably contain chloride daughter minerals, which comprise unusual halite-sylvite intergrowths (Fig. 2). Primary inclusions hosted by barren, postkinematic veins, as well as secondary inclusions hosted in all vein types, trapped

NaCl-dominant solutions (eutectic melting close to –21 °C) with low to moderate homogenization temperature–salinity characteristics of ~110–210 °C and 8–21 wt% NaCl equiv.

LA-ICP-MS analyses of primary fluid inclusion assemblages in pre- to synkinematic veins (K/Na >0.8) contrast with those in postkinematic vein quartz (K/Na <0.8; Fig. 3). Cu, Co, Pb, and Zn concentrations are typically one to two orders of magnitude higher in the K-rich fluids; many of the later fluid inclusion populations with K/Na <0.8 and simpler NaCl-dominant chemistry did not return Cu or Co concentrations above their respective limits of detection. The metal-rich fluids also display a negative correlation between Cu and Co, as well as distinct Ba and Li enrichment, averaging 1.5 ± 0.9 wt% Ba ($n = 26, 1\sigma$) and 1100 ± 900 ppm Li ($n = 26, 1\sigma$).

DISCUSSION

Fluid inclusion homogenization temperatures, major-element chemistry and base metal budgets provide a clear distinction between the pre- to synkinematic fluids associated with mineralization and the postorogenic fluids in largely barren veins. Theoretical data suggesting that high-temperature chloride brines are capable of transporting appreciable quantities of Cu and Co (Liu and McPhail, 2005) are confirmed here with up to 1600 ppm Cu and 770 ppm Co at >250 °C and 40–60 wt% NaCl + KCl. These are the most Cu–Co-rich fluids recorded from a sedimentary basin environment and overlap with Cu concentrations reported from magmatic-hydrothermal systems (Audetat et al., 2008). Our results provide confidence that ore-forming fluids in the ZCB were significantly Cu–Co enriched compared to fluids described from most sedimentary basins (cf. Wilkinson et al., 2009). The negative correlation between Cu and Co in the primary potassic brines at Nkana–Mindola indicates a decoupling of metal budgets and therefore potentially more than one source of Cu–Co being exploited by these fluids.

Potassic Brine Origins

Most sedimentary basin fluids are low- to moderate-temperature, moderate-salinity NaCl (\pm CaCl₂) brines (e.g., Wilkinson, 2010). The distinctive high-temperature and metal-rich, potassic compositions of primary fluid inclusions reported here and hinted at elsewhere in the ZCB (Nowecki, 2014) indicate an atypical brine source or residence history in the basin.

Fluids released from sulfide-hosted inclusions during crush-leach experiments from several CACB deposits exhibited distinct, low Cl/Br and Na/Br ratios, characteristic of bittern brines produced by extensive evaporation in a restricted environment (Selley et al., 2018). Experimental studies of modern seawater evaporation indicate

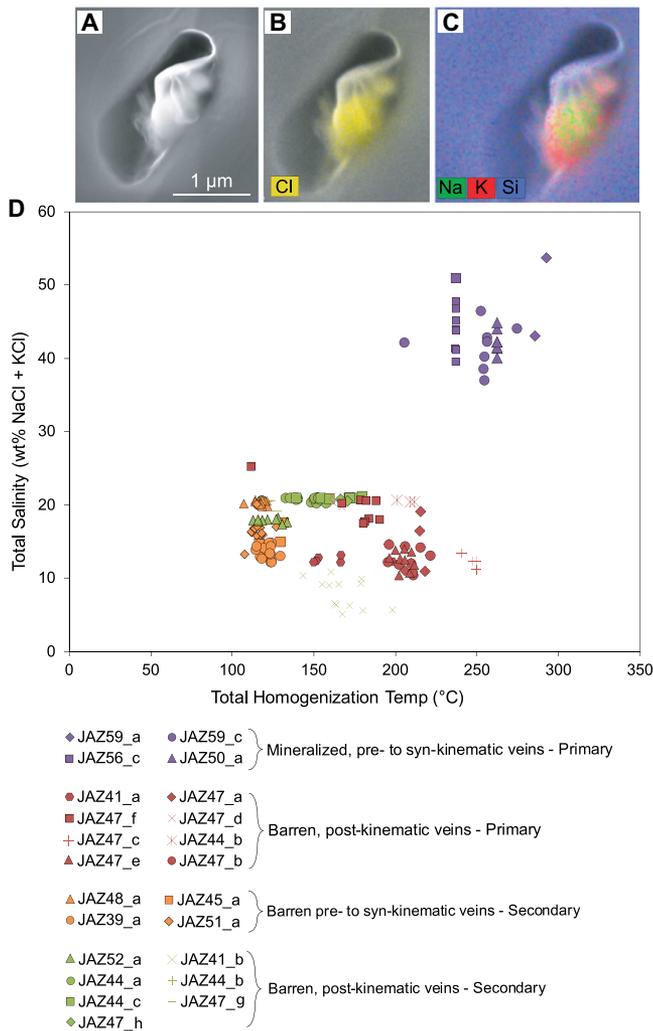


Figure 2. (A–C) Scanning electron microscope images of fluid inclusion cavity: (A) secondary electron image showing anhedral salt mass; (B,C) element maps showing relative response for Cl and intergrown NaCl and KCl salts prior to microthermometric heating. (D) Plot of total homogenization temperature and salinity data derived from microthermometry. Total salinity was estimated from freezing point depression of ice, modeled in NaCl-H₂O system for all two-phase inclusions, and from halite dissolution temperatures and Na/K ratios derived from laser ablation-inductively coupled plasma-mass spectrometry data, modeled in NaCl-KCl-H₂O system for multiphase, saturated inclusions. Primary inclusions associated with mineralization in pre- to synkinematic veins show higher temperature and salinity values than both primary inclusions in discordant postkinematic veins and secondary inclusions in a range of kinematic settings.

that such residual brines develop elevated K/Na ratios only at degrees of evaporation >65–70 (ratio of element concentration per kg H₂O relative to seawater), with K/Na ~2 prior to carnallite and kainite precipitation (McCaffrey et al., 1987; Babel and Schreiber, 2013). Although Neoproterozoic seawater compositions remain poorly constrained, halite-hosted fluid inclusions from the Amadeus and Officer Basins (Australia) and the Otavi Formation (Namibia) indicate a period of CaCl₂-rich and MgSO₄-poor seas for up to 200 m.y. during the middle Neoproterozoic (Kah et al., 2004; Kovalevych et al., 2006; Spear et al., 2014). Theoretical evaporite crystallization sequences of such sulfate-deficient ancient seawaters replicate evaporite sequences observed in the Bonneville Salt Flats (Utah, USA) where sylvite immediately follows halite, with the omission of significant MgSO₄ salts (Valyashko, 1962). This implies that high K/Na ratios can be developed from CaCl₂ seawater at lower degrees of evaporation when an initially low sulfate budget is almost entirely consumed through early gypsum precipitation. The abundance of Ba in the ore fluids from Nkana-Mindola, yet scarcity of barite in the basin successions, can be reconciled if these

brines had low SO₄²⁻ concentrations, consistent with an origin from CaCl₂-rich, MgSO₄-poor seawater.

Potassium Exchange During Subsequent Fluid-Rock Interactions

Burial of the Lower Roan Subgroup clastic sequence with bittern brine pore waters by up to 11 km of Upper Roan Subgroup and Nguaba/Kundelungu Group sediments (Selley et al., 2018) would permit elevated-temperature fluid-rock interactions (Fig. 4). The presence of 20%–30% detrital potassium feldspar in some Lower Roan Subgroup sediments (Selley et al., 2005) provides a potential source to account for further enrichment of K(-Ba) in the brines. Petrographic studies (Sutton and Maynard, 2005) indicate that partial breakdown and replacement of potassic minerals occurred in some Lower Roan Subgroup sequences. Potassium addition ± Na loss during this stage likely played an additional role in the generation of elevated K/Na ratios in the Nkana-Mindola brines.

The intimate spatial and temporal relationships between Cu-Co sulfides and secondary (often Ba-rich) potassium feldspars observed in Ore Shale units in Zambian deposits (Sut-

ton and Maynard, 2005) reflect the importance of K-metasomatism during ore formation. This observation lends support to models invoking mineralization from unusually potassic fluids, as noted by Selley et al. (2005) and Sutton and Maynard (2005), and hinted at by Darnley (1960).

Ore deposit metasomatism in the CACB is partitioned, with Congolese deposits characterized by Mg metasomatism and many Zambian deposits characterized by pervasive K alteration (Hitzman et al., 2012). Extensive Mg metasomatism and the concomitant removal of K (±Ca) from basal clastic sequences in the central Congolese Copperbelt may indicate basin-scale remobilization of K (±Ca) from the depocenter toward basin margins, i.e., the ZCB in the south and Kamao to the north (Selley et al., 2018).

Transition to Halite Dissolution Brines

Primary fluid inclusions from discordant veins and secondary inclusions from all vein types at Nkana-Mindola trapped moderate-temperature, metal-poor, NaCl-dominant fluids. Some fluids from postorogenic ZCB deposits have similar halogen chemistries, consistent with a halite dissolution origin (Heijlen et al., 2008; Selley et al., 2018). Extensive dissolution breccias in the Upper Roan Subgroup (Selley et al., 2018) imply that widespread exploitation of suprasalt levels by halite-undersaturated fluids occurred (Fig. 4). The distinct chemistry of these lower-temperature fluids compared with earlier potassic brines suggests that minimal mixing with deeper basinal fluids occurred prior to removal of salt seals. The progressive penetration of halite-dissolution brines into the Lower Roan Subgroup during the onset of orogenesis is consistent with pervasive, structurally controlled, sodic-calcic (albite ± scapolite) alteration, which locally overprints stratiform mineralization and potassic alteration assemblages (Sweeney and Binda, 1989; Selley et al., 2005).

Controls on Copper-Cobalt Mineralization in Sedimentary Basins

We found that the fluids responsible for mineralization in the ZCB were high-K brines, anomalously enriched in Cu, Co, Pb, and Zn. Consequently, understanding the origin of these fluids is a key factor in the exploration for comparable, basin-hosted resources worldwide. Elevated base metal concentrations are attributed to enhanced leaching efficiencies of high-temperature, potentially sulfate-deficient, bittern brines that had long residence in the deep basin prior to Pan-African orogenesis. In addition to advanced evaporation, the evolution of brines into fertile ore fluids may have been linked to a primary sulfate-deficient chemistry related to Neoproterozoic CaCl₂-rich seawater. Such residual, CaCl₂-rich brines have been proposed as proto-ore fluids

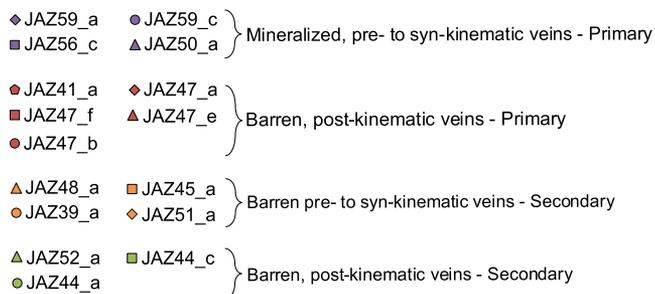
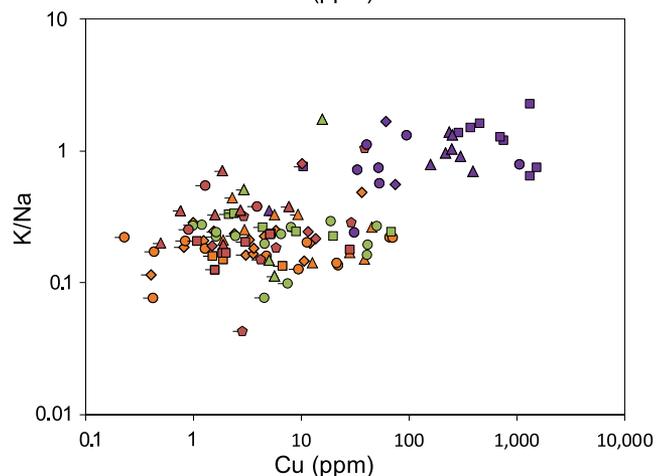
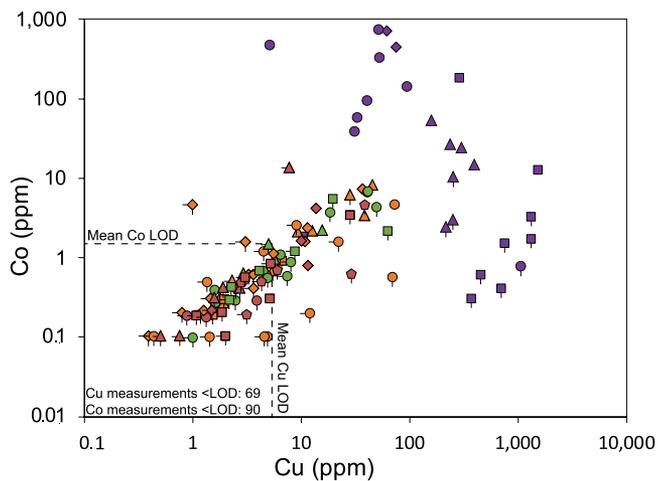


Figure 3. Laser ablation–inductively coupled plasma–mass spectrometry data for individual inclusions and inclusion assemblages. Colors and symbols match those in Figure 2. (A) Co versus Cu, where dashed lines denote mean limit of detection (LOD) values for each element. (B) K/Na versus Cu. Major- and trace-element chemistries of fluid inclusion populations highlight clear distinction between base metal-rich, higher homogenization temperature and salinity ore fluids in pre- to synkinematic veins and several populations of later, metal-poor, lower homogenization temperature and salinity fluids. Bars on symbols indicate that value represents maximum (LOD) value for that element in an inclusion.

in other, lower-temperature, sediment-hosted base metal systems (Wilkinson, 2014), and so there may be a secular control on the development of globally significant basin-margin Cu-Co-(Pb-Zn) deposits. Low fluid pH is an unlikely factor in metal transport, given the carbonate-bearing host rocks and the fact that a dominance of divalent cations over monovalent alkali species is unfavorable for increasing fluid acidity as salinity increases (Yardley, 2005). Oxidized basin sequences are clearly important for limiting reduced sulfur availability, which would otherwise suppress metal solubility (e.g., Cooke et al., 2000). We conclude that the long residence time of sulfate-deficient, high-salinity, high-temperature fluids in deep, potentially thermally anomalous, settings is a critical characteristic for the generation of the most Cu-fertile basins.

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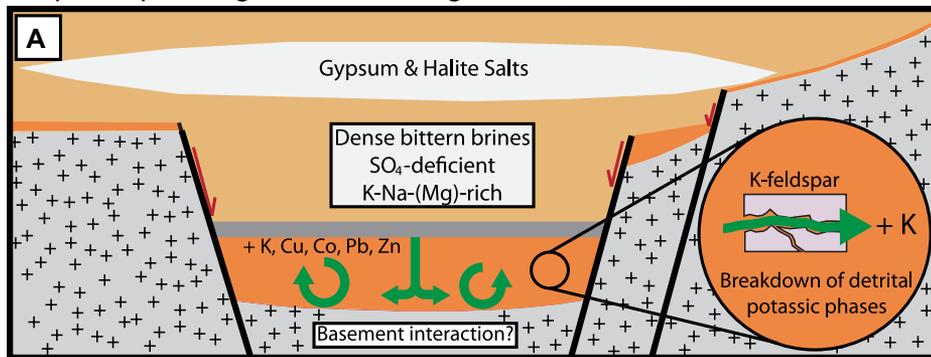
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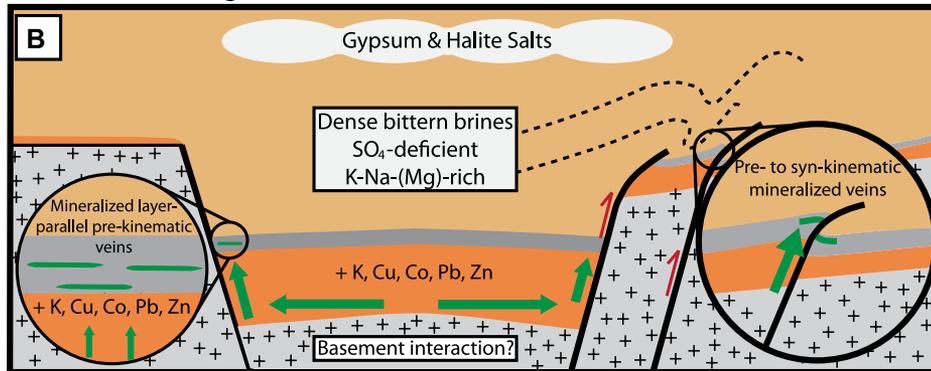
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Simplified pre-orogenic basin configuration (~800 Ma)



Halokinesis through to onset of basin inversion (~745-550 Ma)



Post-peak-orogenic basin configuration (~530-500 Ma)

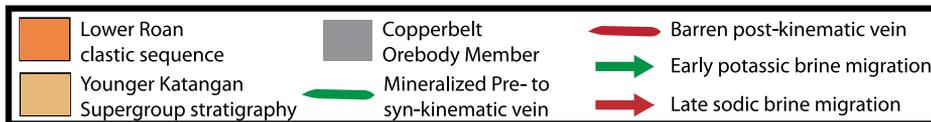
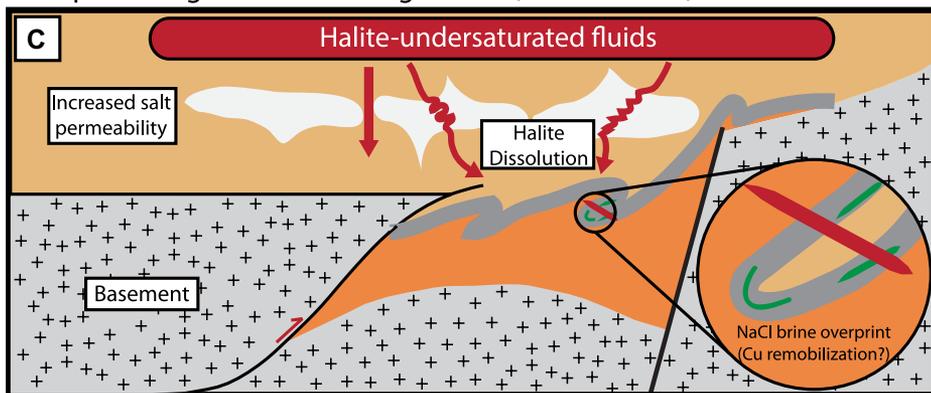


Figure 4. Simplified schematic diagrams illustrating evolving fluid regimes from pre- to post-peak orogenic times in the Zambian Copperbelt. (A) Following evaporation from CaCl_2 -rich, MgSO_4 -poor mid-Neoproterozoic seawater, dense, sulfate-deficient residual brines were resident in basal clastic sequences for extended durations. Fluids acquired high temperatures ($>250^\circ\text{C}$) and elevated K/Na and total salinities. (B) Fluid migration along basin margins prior to (and during) onset of basin inversion focused ore fluids to structural and chemical trap sites, where Cu- and Co-sulfides were precipitated with pervasive potassic metasomatism. (C) During basin inversion, halite-undersaturated NaCl brines at suprasalt levels interacted with remaining salt rafts. Following peak orogenesis, largely barren NaCl-brines penetrated into deep basin, forming discordant veins and associated, structurally controlled sodic metasomatism.

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