Effect of carbon and sulfur on metal transport by hydrothermal fluids

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Carbon and sulfur are ubiquitous components of hydrothermal fluids on Earth, yet their impact on metal behavior remains poorly understood. In this keynote contribution we will present experimental and theoretical results that challenge our traditional view on the roles played by sulfur and carbon on metal transport and ore deposit formation.

The trisulfur (S_3^-) and disulfur (S_2^-) radical ions, recently discovered in aqueous solution at elevated temperatures and pressures [1, 2], may strongly bind gold and other critical metals (Pt, Pd, Mo, Re) [3]. Compared to traditional sulfide, sulfate, chloride or hydroxide ligands, the radicals greatly enhance the extraction, transport, and focused precipitation of metals by S-rich fluids. These results may account for large, and so far unexplained, enrichments by these metals observed in different geological settings, and help in resource prospecting. However, the sulfur radical ligands have a formidable rival – carbon dioxide, which is the major C-bearing form in fluids. Despite its weak complexing capacities for metals, when CO_2 is present in significant amounts (>10 wt%) it lowers the fluid dielectric constant. This change in solvent properties exerts a contrasting effect on different aqueous species, leading to destabilization of the radical ions themselves, their complexes with metals (e.g. $Au(HS)S_3^-$) and other charged species (e.g. $Au(HS)_2^-$, $FeCl_4^{2-}$, $CuCl_2^-$, $PtCl_3^-$), while favoring uncharged complexes (e.g. $AuHS^0$, $FeCl_2^0$, $CuHS^0$, $Pt(HS)_2^0$) [4]. This effect of CO_2, disregarded so far, yields large changes in mineral solubility and fractionations among metals; it may account for Fe-As-Te-Sb enrichment and Cu-Zn-Pb depletion in metamorphic Au deposits, and trigger metal deposition in porphyry Cu-Au-Mo deposits by CO_2 pulses from magma. Thus, depending on geological context, fluid composition (acidity, salinity, redox, and sulfur content), and the resulting metal speciation, carbon and sulfur may act in unison or against one another on the behavior of metals in hydrothermal fluids. These findings offer new perspectives for hydrothermal ore deposit research.

[1] Pokrovski & Dubrovinsky (2011), *Science* 331, 1052-1054. [2] Pokrovski & Dubessy (2015), *EPSL* 411, 298-309. [3] Pokrovski et al. (2015) *PNAS* 112, 13484-13489. [4] Kokh et al. (2017) *GCA* 197, 433-466.