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The impact of sulfur on the transfer of platinum group elements by geological fluids.

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Knowledge of the speciation and solubility of platinum group elements (PGE) in magmatic-hydrothermal fluids is a key to interpret PGE geochemical transfers and ore deposit formation. PGE's are suggested to be transported as Cl^- and HS^- complexes [1-3]; however, these data predict too small metal contents in the fluids from S-rich geological settings, calling upon a possible role of the trisulfur ion ($S_3^{\bullet-}$) as a ligand for PGE transport [4]. Here we combined molecular dynamics (MD) and thermodynamic (TD) modeling, solubility measurements, and in-situ X-ray absorption spectroscopy (XAS), to obtain a new coherent dataset to quantify the effect of sulfur on PGE mobility. Our calculations and experiments show that three main complexes transport Pt in the fluid over a wide pH range (4-8) at 300°C and 500 bar: $Pt(HS)_4^{2-}$ (Fig. 1A) and possibly $Pt(HS)_2^0$ in hydrogen sulfide H_2S/HS^- solutions, and $Pt(HS)_2(S_3)_2^{2-}$ (Fig. 1B) in sulfide-sulfate $H_2S/SO_4^{2-}/S_3^{\bullet-}$ solutions, in which Pt concentrations are as high as 10s ppm. Furthermore, MD simulations were conducted to confirm the stability of these complexes, and ab-initio thermodynamic integration [6] was employed to predict, independently from our experiments, the stability of those complexes. Our approach integrates, for the first time, complementary cutting-edge techniques to highlight the important role of sulfur in the transport of PGE by aqueous fluids in the Earth's crust.

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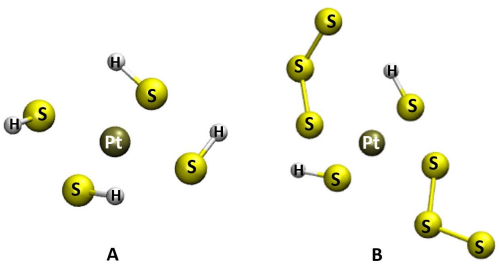

Fig. 1