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The effect of temperature on sulphur speciation in magmatic fluids and its implications for magma redox and ore genesis

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Sulphur content and speciation in magmatic fluids released from ascending arc magmas affect the climate and control metal solubility, transport, and magmatic-hydrothermal ore deposit formation. To study the effect of temperature on sulphur speciation, we first trapped pre-equilibrated sulphur-bearing aqueous fluids as synthetic fluid inclusions (SFI) at *T* = 750, 875, and 1000 °C and *P* = 200 MPa, and up to eight different oxygen fugacities at each temperature ranging from -1 to +2.5 log units relative to the Ni-NiO buffer, in our prototype externally heated molybdenum-hafnium carbide vessel apparatus with hydrogen membrane simultaneously allowing for rapid quenching and flexible, precise, and accurate redox control [1]. Following the synthesis, we reheated the SFI to their entrapment temperature in a microscope heating stage and analysed them by *in situ* Raman spectroscopy, employing two different laser excitations. During our heating experiments we observed several reactions, including sulphur comproportionation and the formation of sulphur radical species, underlining the unquenchable nature of sulphur speciation in magmatic fluids and the need for *in situ* approaches. Our experimental design allowed for the quantification of different sulphur-bearing aqueous species, including sulphide, sulphate, SO_2 and sulphur radical species, at previously inaccessible temperature and oxygen fugacity conditions and the assessment of their role in ore deposit formation. Our speciation model differs significantly from the results reported in previous studies [2,3]. Furthermore, in contrast to [2], we observed a large temperature-induced shift of the H_2S to SO_2 transition, which has important implications for the redox evolution of magmas and the likelihood of associated ore formation.

[1] Zajacz & Alex (2022) American Mineralogist 107, 692-702.

[2] Binder & Keppler (2011) Earth and Planetary Science Letters 301, 190–198.

[3] Pokrovski & Dubessy (2015) Earth and Planetary Science Letters 411, 298–309.