Fluorine systematics in the Yellowstone (USA) hydrothermal system

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In the Yellowstone Plateau Volcanic Field (YPVF) the Cl/F (~0.5 by mass) observed in both glass inclusions trapped in quartz crystals and in erupted rhyolitic glass is significantly lower than the ratio in the rivers draining the Plateau (~8) [1] or in the thermal waters (<140). Potential explanations include: 1) preferential degassing of chlorine (as HCl) compared to fluorine (as HF) from the underlying silicic magmas, 2) an additional crustal source enriched in chlorine that contributes to the thermal waters, or 3) a large sink of fluorine between magma and the ground surface. Within the YPVF, the Cl/F in neutral-alkaline-chloride waters (pH>6) in Norris (80-140) and Gibbon (15-45) Geyser Basins north of the Yellowstone Caldera are substantially higher than the ratios in thermal waters within the Yellowstone Caldera and along the caldera's southern boundary (<15). A crustal source of Cl contributing to thermal waters is unlikely because Cl/Br is notably constant throughout all the geyser basins. Fluorite (CaF_2) is the most common fluorine-rich accessory mineral precipitated in hydrothermal systems. Its solubility is controlled by temperature, salinity, and the calcium and fluoride concentration of the thermal fluids [2, 3]. Fluorite was common in cores from several research holes (<330 m) drilled in Yellowstone's geyser basins during the 1960s. However, calcium concentrations in thermal waters north of the caldera (1-10 mg/L) are typically higher than in thermal waters within the caldera (<1.5 mg/L). Processes in addition to fluorite precipitation are implicated such as temperature-dependent solubilities of calcite and anhydrite, the amount of CO_2 degassing, and the hydrostatic pressure. These additional factors are likely related to fluid flow from the deep subsurface reservoir and the lithology controlling flow within and outside the caldera. Both the physical and chemical constraints appear to give rise to Yellowstone's wide variety of thermal water compositions.

[1] Lowenstern & Hurwitz (2008) *Elements* **4**, 35-40 [2] Nordstrom & Jenne (1977) *GCA* **41**, 175-188 [3] Richardson & Holland (1979) *GCA* **43**, 1327–1335.