Halogen incorporation in forsterite at Earth´s mantle conditions

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The incompatible behavior of halogens gives them the ability to act as tracers of volatile transport processes in the Earth´s mantle. It is crucial to investigate halogen partitioning behavior and incorporation mechanisms in olivine, which is the dominant upper mantle mineral, to better understand halogen behavior during partial melting processes.

Halogen partitioning between forsterite and silicate melt has been measured in experiments at 1.0-2.3 GPa and 1500-1600°C, representing partial melting conditions of the Earth´s upper mantle. Our data show that partitioning into forsterite decreases with increasing anion size: $D_F^{fo/melt} > D_{Cl}^{fo/melt} \geq D_{Br}^{fo/melt}$. Combining our data with results of [1] shows that F and Cl partitioning between forsterite and melt is not pressure dependent, but is strongly temperature dependent, as it increases by a factor of 500 (F) and 2000 (Cl) between 1300°C and 1600°C. Bromine appears to show a similar behavior.

Back scattered electron images and transmission electron microscopy measurements show that halogens are not incorporated in melt inclusions, fluid inclusions or humite type defects in forsterites. The most reasonable incorporation mechanism for halogens is via point defects in the forsterite lattice, where they are inferred to be charged-balanced via oxygen defects. Possible substitution mechanisms are $[MgO_2]^{2-}=[\square F_2]^2$ as suggested by [2] or the replacement of a $[SiO_4]^{4-}$ tetrahedon with a $[halide]^4$ quadruplet. [3] suggested by using first principles calculations that the latter mechanism increases the fluorine solubility in forsterite exponentially with increasing temperature whereas the effect of pressure is almost negligible. This is in agreement with the halogen partitioning behavior observed in our study and might be a sign that this mechanism is predominantly controlling the halogen incorporation in forsterite.

The strong temperature dependence of the partitioning behavior and the modal abundance of olivine in mantle peridotite indicate that olivine is a major host for halogens in the Earth´s upper mantle.

[1] Beyer *et al* (2012) *EPSL* **337-338**, 1-9 [2] Bernini *et al* (2013) CMP **165**, 117-128. [3] Bernini (2011) Ph.D. thesis.