# Speciation and sources of aqueous fluids in diamond fluid inclusions

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Fibrous and coated diamonds usually contain fluid inclusions providing information about upper mantle fluids. At least three end-member fluids have been identified based on their chemical composition: silicic, saline (or brine) and carbonatitic[1], suggesting different sources and reaction histories of the fluids involved in diamond formation.

We have taken advantage of recent developments in aqueous solution chemistry at elevated temperatures and pressures to investigate the speciation and possible sources of aqueous fluids in diamonds. We used equilibrium constants linking minerals and aqueous inorganic and organic ions, complexes and neutral species predicted with the Deep Earth Water model[2] to develop aqueous speciation-solubility models at 900 ºC and 4.0 GPa. We calculated the speciation of aqueous fluids in equilibrium with model eclogite and with harzburgitic peridotite in the system $Na_2O$-$K_2O$-CaO-MgO-FeO-$Al_2O_3$-$SiO_2$-$H_2O$-$CO_2$-$H_2$-HCl assuming a log$fO_2$ value of 2 units below the quartz-fayalite-magnetite buffer[3].

We found that the calculated eclogitic and peridotitic fluids match the silicic and saline end-member fluid inclusion compositions, respectively. The calculated pH of the eclogitic fluid is strongly alkaline, resulting in high total dissolved silica in the presence of coesite and the aqueous silica species $Si(OH)_4^0$ (60%), $SiO(OH)_3^-$ (21%) and $Si_2O(OH)_6^0$ (19%). However, the peridotitic fluid has a pH closer to neutral, and low total dissolved silica (mainly $Si(OH)_4^0$) because of enstatite and forsterite. For both fluids, equilibrium with phlogopite was assumed, with Na calculated by charge balance, resulting in low predicted K concentrations in the eclogitic fluid, and high K and K/Na ratio in the peridotitic fluid, consistent with the published fluid inclusion analyses. This result suggests that K-rich aqueous fluids could be a characteristic feature of peridotite in the upper mantle, in contrast to hydrothermal fluids in the shallow crust.

Overall, our results suggest that an aqueous, saline, K-rich fluid could originate from peridotite, and that an aqueous, silicic, low-K fluid could originate from the eclogitic parts of subducting slabs.

[1] Klein-BenDavid *et al* (2007), *GCA*, **71**, 723-744; Tomlinson *et al* (2006), *Earth Plan. Sci. Lett.* , **250**, 581-595. [2] Sverjensky *et al* (2014), *GCA*, **129**, 125-145. [3] Frost & McCammon (2008), *Ann. Rev. Earth Plan. Sci.* **36**, 389–420.