Geophysical Research Abstracts
Vol. 16, EGU2014-9302, 2014
EGU General Assembly 2014

Supercritical aqueous fluids in subduction zones carrying carbon and sulfur: oxidants for the mantle wedge?

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Much speculation surrounds the nature of aqueous fluids in subduction zones. Aqueous fluids likely trigger partial melting in the mantle wedge, influencing the chemistry of the magmas that erupt in island arcs. They also may play a role in transporting elements that could metasomatize and oxidize the overlying mantle wedge, most importantly C, S and Fe. However, full coupling of aqueous fluid chemistry with the silicate, carbonate, C, sulfide and sulfate minerals has remained limited to pressures of 0.5 GPa because of limitations on the HKF aqueous ion equation of state. Recent progress in developing a Deep Earth Water model (Sverjensky et al., 2014), calibrated with new experimental data, now enables a detailed evaluation of the evolution of aqueous fluid chemistry to a pressure of 6 GPa, well into subduction zone conditions.

We report aqueous speciation models for eclogitic aqueous fluids constrained by model mineral assemblages that give preliminary indications of the solubilities of elements that could contribute to mass transfer and redox changes in the mantle wedge. For example, at 600 °C and 2.5 GPa, an aqueous fluid in equilibrium with jadeite, paragonite, muscovite, quartz, lawsonite, almandine, talc, magnesite and pyrite at QFM oxidation state with 0.1 molal total Cl, contains 5.5 molal C, 0.04 molal S, and 9 micromolal Fe. The fluid has a pH of 4.7, much greater than the neutral pH of 3.3; the predominant species and molalities are CO_2 (5.0), Na+ (0.44), Si(OH)4 (0.36), HCO_3- (0.26), H3SiO4- (0.23), $CaHCO_3$+ (0.18), silica dimer (0.10), Cl- (0.09), K+ (0.08), HCOO- (0.06), H2S (0.03). Calculations for model eclogitic fluids at the higher pressures and temperatures of subarc conditions also show that the solubility of C is much greater than either S or Fe at QFM. However, in subarc eclogitic fluids of higher oxidation state (QFM +3 to +4) in equilibrium with hematite, anhydrite, jadeite, kyanite, phlogopite, coesite, lawsonite, almandine-pyrope, and magnesite, the C/S ratio can vary from 0.2 to 3.5 when temperature varies from 650 to 750 °C at 4 GPa. Fe concentrations remain negligible. These results strongly suggest that aqueous subarc eclogitic fluids that evolve to QFM +3 to +4, perhaps by reaction with metamorphosed Fe-oxide-bearing sediments, could transport significant amounts of C and/or S into the mantle wedge environment depending on the temperature. Hotter subduction should favor high C/S fluids, whereas colder subduction should favor low C/S fluids. Aqueous Fe transport is unlikely to be playing a significant role in oxidizing the mantle wedge.

Sverjensky, D. A., Harrison, B., and Azzolini, D., 2014. Water in the deep Earth: the dielectric constant and the solubilities of quartz and corundum to 60 kb and 1,200°C. Geochim. et Cosmochim. Acta (in press).