

Reactivity of aqueous volatiles in Earth's upper mantle: insights from the Deep Earth Water Model 2024 (DEW24)

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Planetary volatiles in contact with ultramafic upper mantles such as in Earth are increasingly recognized as having a wide range of compositions that include not only the traditional volatile elements but also the major rock-forming elements. The character of such planetary fluids is well established from experimental studies of ultramafic rock solubilities and the compositions of fluid inclusions in diamonds [1, 2]. The extremely wide range of compositions, including the development of immiscibility in aqueous fluids [3, 4], constitute a formidable challenge for theoretical models of the reactivity of fluid-rock-melt systems at elevated temperatures and pressures. The DEW24 model originally calibrated with experimental solubilities of mafic and ultramafic rocks in water [5] has now been expanded to include a very wide range of H₂O/CO₂ ratios, as well as the immiscibility of H₂O-H₂ and H₂O-fluid hydrocarbons. DEW24 provides the thermodynamic framework for aqueous speciation/solubility and chemical mass transfer models. As an example [6], prediction of the irreversible reactions of immiscible H₂ with carbonated serpentinites and interstitial aqueous fluid at 400-450°C and 2.0 GPa results in dramatic metasomatism of the host rocks and the development of an immiscible hydrocarbon fluid at elevated temperatures and pressures consistent with field and laboratory studies [6]. Deeper in the upper mantle, at 1,000°C and 5.0 GPa, irreversible mass transfer models of metasomatic reactions between aqueous fluids and upper mantle rocks in Earth during diamond formation indicate that the mineral inclusions preserved in diamonds may reflect the fluids involved, and the extent of fluid-rock reaction, rather than the particular geologic rock environment. Overall, the results provide a tantalizing indication of new complexities during planetary volatile-fluid-rock interactions.

[1] Kessel, R. et al. (2015), *Contrib. Mineralogy and Petrology*, 169, 37.

[2] Weiss et al. (2022), *Reviews in Mineralogy & Geochemistry*, 88, 475.

[3] Abramson et al. (2017), *Amer. J. Science*, 9, 967.

[4] Vlasov et al. (2023), *Contrib. Mineralogy and Petrology*, 178, 36.

[5] Huang and Sverjensky (2019), *Geochim. Cosmochim. Acta*, 254, 192.

[6] Huang et al. (2024) *Lithos*, 468-469, 107498.

[7] Rinaldi et al. (2023), *Geochim. Cosmochim. Acta*, 356, 105.