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AN: **V51B-1484**
TI: Thermodynamic modeling of methane production in Early Archean crust by serpentinization: implications for atmospheric methane.

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AB: Abiotic methanogenesis during the hydration of ultramafic minerals by a CO2-bearing fluid may have played an important role in the evolution of Earth's prebiotic atmosphere. If this process was volumetrically significant in the early Earth, it would have provided a robust CH4 source of atmospheric CH4 in the Archean. Abiotic methanogenesis would likely have accompanied metamorphic fluid-rock interaction in olivine- and pyroxene-rich rocks in a range of environments, including shallow crustal hydrothermal systems, contact metamorphic systems, and low-grade regional metamorphism. To quantitatively evaluate the conditions necessary for and productivity of abiotic methanogenesis during Archean crustal metamorphism, we are developing a simple, internally consistent thermodynamic model of the fO2-buffering ability and speciation generated by ultramafic hydration equilibria in the system Mg-Fe-Si-C-O-H at 300-1300 K and 0.3-20 kbar. In our model, we assume that Fe is rejected by brucite and low temperature hydrous Mg-phyllosilicates. This permits computation of upper limits on fCH4 and lower bounds on fO2 for relevant equilibria. Assuming an ideal solid solution in olivine and orthopyroxene, we calculate fO2 vs. T for a fixed bulk-rock XMg using THERMOCALC (Holland and Powell, 1998) and SUPERFLUID (Belonoshko et al, 1992). As an example, our results show that at P=0.3 kb and olivine XMg =0.9, the serpentinization equilibrium (Mg0.9Fe0.1)2SiO4 + 1.3 H2O + 0.033 O2 ↔ 0.029 Mg48Si34O85(OH)62 + 0.388 Mg(OH)2 + 0.067 Fe3O4 (1) crosses the iron-magnetite buffer at about 375 K (± 10). Also, at ~525K at the same P and XMg, the fO2 buffered by reaction (1) is two log units above iron-wüstite and four log units below QFM. Using this fO2, our speciation calculation for a graphite-saturated C-H-O fluid shows that up to ~700K, CH4 is the dominant carbon species in a fluid in equilibrium with reaction (1). At ~500 K, 0.3 kb, CH4/CO2 ~ 1010. As with Phanerozoic serpentinization (e.g., Frost, 1984), our modeling demonstrates that interaction of CO2-bearing metamorphic fluids with Archean olivine-rich volcanic rocks (e.g., komatiites) has the potential to act as a major CH4 source. By analogy with CO2 in the Phanerozoic, the Archean CH4 cycle may have been strongly influenced by crustal sourcing of CH4. Belonoshko, AB, et al. (1992) Comp. Geosci. 18, 1267. Frost, BR (1985) J. Pet. 26, 31.

Holland, TJB, and Powell, R (1998) J. Metamorphic Geol. 16, 309.
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