Isotopic and theoretical constraints on the evolving redox state of Earth's atmosphere

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The chemistry of Earth's ocean-atmosphere system has undergone dramatic evolutionary changes throughout its long history, with important ramifications for prebiotic chemistry, the emergence and stability of atmospheric biosignatures, and the evolution of complex life. Perhaps most strikingly, the redox state of Earth's post-biotic atmosphere has generally undergone a secular evolution between mildly reducing (predominantly N_2-CO_2-H_2O, with variable H_2 and CH_4) to strongly oxidizing (predominantly N_2-O_2-H_2O) through time. Delineating both the empirical trajectory of this evolution and the causal mechanisms underpinning it represents a central problem in the fields of biogeochemistry, geobiology, and planetary science.

Significant new insights into this problem have emerged in recent years through the application of a wide range of stable and radiogenic isotope systems. At the same time, existing and emerging biogeochemical modeling tools are pushing the limits of the quantitative constraints on atmospheric chemistry that can be extracted from these isotopic tracers. Although a basic framework for the long-term evolution of atmospheric redox on Earth has long been appreciated[1], we are only just beginning to understand the textural richness of this evolution and the implications this richness may bear for our understanding of the Earth system and the evolutionary fates of other Earth-like planets.

As an illustrative example, this talk will focus on a major chemical inversion hypothesized for Earth's post-biotic atmosphre – from the high-CH_4 and low-O_2 atmosphere dominant during much of the Archean Eon to the low-CH_4 and high-O_2 atmosphere characteristic of the modern Earth. In particular, it will explore existing and emerging constraints on atmospheric O_2 and CH_4 through time from both stable isotopes and quantitative theoretical models. Areas of both striking convergence and puzzling discrepancy between theoretical and proxy-based understanding will be highlighted, with an eye toward pursuing a number of emerging challenges and opportunities.

[1] Holland, H.D. (1962) *Petrologic Studies: A Volume to Honor A.F. Buddington*, Geological Society of America, 447-477.