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Coupled U and Fe isotope Records Suggest Onset of Oxygenic Photosynthesis Three Billion Years Ago

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The timing of the onset of oxygenic photosynthesis (OP) is still debated, with current estimates spanning over one billion years. Here we couple new U and existing Fe isotope records to assess the timing of the onset of redox cycling of these elements, which serves as a new estimate for the onset of OP. Both U and Fe are redox sensitive elements, whose mobility and isotope fractionation are primarily controlled by ambient redox states. Therefore, the earliest time when $\delta^{238}\text{U}$ or $\delta^{56}\text{Fe}$ isotope variability changes can be viewed as evidence for shifts in Earth's surface oxidative capacity. Statistical tests on currently available U and Fe isotope data shows significant changes in the variability of $\delta^{238}\text{U}$ and $\delta^{56}\text{Fe}$ at ca. 3.0 Ga, suggesting shifts in the oxygen levels at Earth's surface at this time. Kinetic modeling suggests that oxidation and mobilization of reduced U requires oxygen levels orders of magnitude higher than prebiotic oxygen levels (i.e. 10^{-11} of the present atmospheric level). Therefore, our results suggest that biogenic oxygen production (i.e., oxygenic photosynthesis) should have existed at ca. 3.0 Ga, resulting in local and/or transient, if not pervasive, environmental shifts from predominantly anoxic to relatively oxic conditions, which enabled oxidation of U(IV) and Fe(II). Permanent rise of atmospheric oxygen was delayed until the ca. 2.4 Ga Great Oxidation Event, likely linked to other interplaying factors such as tectonic activity, minimal continental emergence, nutrient limitation, and drawdown of large masses of reducing gases and minerals.