Goldschmidt 2022 Abstract
https://doi.org/10.46427/gold2022.12430

Investigating the Complexity in the Marine Oxygen Cycle with Oxygen Isotopologues

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The biogeochemical fluxes that produce and consume O_2 play a critical role in regulating Earth's climate and habitability. The triple oxygen isotope method is one of the most robust tools for directly tracing oxygen cycling and measuring gross primary productivity in the marine environment. This method assumes that photosynthesis, microbial oxygen consumption (in the form of respiration), and gas exchange are the primary influences on dissolved oxygen, and that they have predictable, consistent isotopic end-members. Recent work, however, has highlighted the ubiquitous production of extracellular reactive oxygen species (e.g., superoxide and hydrogen peroxide) by microorganisms as a major sink of dissolved O_2. Additionally, decades of research have demonstrated that abiotic photooxidation of dissolved organic carbon is also a significant sink of O_2 in the surface ocean. Quantifying the impact of these non-respiratory sinks of dissolved O_2 is essential to a complete understanding of the modern oxygen cycle and the trajectory of future ocean deoxygenation. We investigate the triple oxygen isotope fractionation of these seldom considered, yet environmentally widespread reactions. We demonstrate that the triple oxygen isotope mass laws associated with these reactions span virtually the entire range of the mass-dependent domain (λ = 0.499 to λ = 0.535). These results highlight the tremendous spatio-temporal uncertainty in triple-oxygen derived productivity estimates, a method which is often considered the gold-standard for gross-productivity estimates. Lastly, we provide a first look at the clumped oxygen isotope composition of dissolved O_2 in the water column, and explore the ways in which clumped measurements may address outstanding uncertainty in marine oxygen cycling.