

Lipid Biomarkers Record Strong Marine Methane Cycling and an Anoxic Deep Ocean Persisting Through the Ordovician and Silurian Periods

NATHAN MARSHALL, GORDON LOVE, ANDREY BEKKER, MEGAN ROHRSSSEN AND CARINA LEE

University of California Riverside

Presenting Author: nmars008@ucr.edu

Microbial production and consumption of methane represent a critical biogeochemical loop in Earth's carbon cycle which can influence the atmospheric greenhouse gas content and affect the long-term global climate system. In modern aquatic systems, the oxidation of methane largely occurs through the process of anaerobic oxidation of methane (AOM). AOM in modern marine environments is the dominant process of methane consumption trapping >90% of the annually produced methane before it can reach the open water column or atmosphere. During the Ordovician and Silurian Periods, diminished dissolved oxidant availability in the oceans may have induced mutually reinforcing effects that promoted methane oxidation and methanogenesis in marine waters and shallow sediments through diagenesis of organic matter[1]. The rising Devonian global inventory of dissolved oxidants[2,3] likely contributed to a global and systematic change in ocean redox that could be reflected in an increasingly muted marine and sedimentary methane cycle.

We have analyzed lipid biomarker distributions in over 200 thermally well-preserved marine sedimentary rock samples, from multiple formations encompassing both high- and low-latitude marine settings, through the Ordovician-Devonian transition. Rock extracts were analyzed for a suite of branched and polycyclic hydrocarbon biomarkers utilizing the sensitivity and selectivity of Metastable Reaction Monitoring-Gas Chromatography-Mass Spectrometry (MRM-GC-MS). Anomalously high relative and absolute abundances of lipid biomarkers derived from groups of methanotrophic bacteria and archaea involved in methane cycling have been consistently found in Ordovician and Silurian marine sedimentary rocks, spanning at least 80 million years of geologic time. Compound-specific stable carbon isotopic analyses of Ordovician-age hopanes, sampled from numerous formations and locations, using Gas Chromatography-Isotope Ratio Mass Spectrometry (GC-IRMS) reveal significant ¹³C-depletion in comparison with marine hopanes sourced from other Phanerozoic intervals. The persistence of a strong Early Paleozoic marine methane cycle, persisting within the paleotropics through the Late Silurian, suggests that full oxygenation of the deep ocean system was not complete until the Devonian Period.

REFERENCES

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- [2] Beerling D. j. *et al.* (1998), *S. E. Philos. Trans. R. Soc. B.* 53, 113–130.