

5th February 2022

Dear Editor:

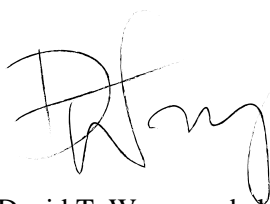
Transmitted is a digital version of our manuscript, “Incorporation of water-derived hydrogen into methane during artificial maturation of kerogen under hydrothermal conditions” by D. T. Wang (MIT/WHOI), J. S. Seewald (WHOI), E. P. Reeves (UiB), S. Ono (MIT), and S. P. Sylva (WHOI), submitted for consideration in *Organic Geochemistry*. All authors have participated in the preparation of the manuscript and agree to its submission.

Understanding the origin of the hydrogen (H) in hydrocarbon gases such as methane (CH₄) is critical for interpretation of hydrogen-isotope signatures (D/H ratios or δD values) in natural gases. Here, we studied the origin of C–H bonds in thermogenic methane by heating kerogen in the presence of D₂O and examining the degree of deuteration in the generated methane. Methane generated at the beginning of the experiment was mostly C¹H₄ but towards the end of the experiment was primarily CD₄. This suggests that competition between rates of kerogen-water isotopic exchange and natural gas generation may govern the D/H ratio of thermogenic gases. Furthermore, it points to possible participation by water in processes that generate petroleum and natural gas hydrocarbons in the subsurface. The results also imply may be more thermogenic natural gas in the world than predicted by basin modeling practices which employ kinetic simulations that do not account for the contribution of H by water.

This paper is our original unpublished work and it is not under consideration at any other journal. We hope you will find this contribution suitable for publication in *OG*.

Recommended referees: Alex Sessions (Caltech), Katherine L. French (USGS), Justin Birdwell (USGS), Arndt Schimmelmann (Indiana University), Michael Lewan (USGS, retired), Everett Shock (ASU).

Sincerely,

A handwritten signature in black ink, appearing to read 'DTW', with a stylized flourish at the end.

David T. Wang, on behalf of co-authors