Ms. Ref. No.: OG-5031
Title: Incorporation of water-derived hydrogen into methane during artificial maturation of kerogen under hydrothermal conditions

Dear Dr. David T. Wang,

Lloyd Snowdon, the Associate Editor, has reviewed your paper. It needs to be revised before proceeding to be sent out for review. The needed revisions are mostly related to format and file inventory. These should not take you long to do. Lloyd also has provided some additional comments that you should consider to be addressed.

Please submit the revision as quickly as possible. Authors are normally expected to provide a revised manuscript within 4 weeks, but I very much doubt that this time will be needed. After 4 weeks, the manuscript will be cleared out of the system and a "revision" after this date would have to be submitted anew and given a new tracking number. Under extenuating circumstances, the Associate Editor should be contacted with a request for an extension. Significantly delayed revisions may become superseded in the literature and require changes beyond those requested by the reviewers and Associate Editor.

For your guidance, reviewers' comments are appended below. Please note that reviewers may have uploaded files as part of the review. If this is the case, you can access the uploaded files on the Editorial Manager.

If you decide to revise the work, please submit a list of changes or a rebuttal against each point raised, a copy of the manuscript with track changes and a clean copy of the manuscript when you submit the revision. Organic Geochemistry also encourages authors to include an acknowledgement of the time and effort provided by the reviewers, whether they are anonymous or have been identified.

Dear Editors,

We have addressed the comments by the AE below. Please see the attached revised manuscript with tracked changes, as well as a clean copy of the Manuscript. Thank you,

David T. Wang on behalf of coauthors.
2/20/2022

Associate Editor (Lloyd Snowdon) comments:

Before this manuscript can be sent out for formal scientific review, the format/style needs to be corrected to adhere to the requirements laid out in the Instructions to Authors for Organic Geochemistry. The manuscript was submitted as a single PDF file which most reviewers find is much more difficult to handle than the required native word processor format. Elsevier provides a hyperlinked version of the references in a submission and this is very useful for reviewers. However, their software cannot handle a PDF document (possibly because of the embedded line numbers?). I also have indicated a number of other format/style issues that need to be addressed. In addition, I have flagged a few science questions that the authors may wish to address before resubmitting the manuscript with the proper formats.

Comments

This submission does not conform to many of the requirements laid out in the Instructions to Authors for Organic Geochemistry. These need to be corrected in order to facilitate the reviewing/editing process.

"It is important that the file be saved in the native format of the word processor used."
Now saved as DOCX instead of PDF.

"In particular, do not use the word processor's options to justify text or to hyphenate words."
Fixed.

"Highlights should be submitted in a separate editable file in the online submission system. Please use 'Highlights' in the file name and include 3 to 5 bullet points (maximum 85 characters, including spaces, per bullet point)."
Now saved as DOCX instead of TXT.

"No capital letters (after the initial one) in titles of journal articles except for proper names. First letter capitals should be used for book titles. No spaces between initials and journal names should be in full."
Formatting of references has been addressed.

Calcite is 12% C. By difference there is 4.77% mineral C (11.0-6.23 in Table 1) and this indicates 39.75% if the carbonate mineral is CaCO3 or 36.57% carbonate if it is dolomite.
It is fine as is. The elemental analysis values shown in Table 1 for EX and DECA represent weight percent of the material that was ingested by the elemental analyzer. The denominator in these values is the residual weights of sample after extraction or decarbonation, not the original weight of rock. This is now clarified in the caption of Table 1.

L95. The specific volume of water at 350 °C is about 1.75 cm3/g. That is, 1 ml of water would occupy 1.75 ml at 350 °C. Thus it is not clear that the phrase "sealed with a small argon-purged headspace (to allow for expansion of the starting fluid at conditions" accurately reflects conditions at high temperature. Even at 250 °C the specific volume is about 1.25 cm3/g.
Clarified as follows:

> ***The reaction cell was loaded with 55.03 g of this starting fluid, sealed with a small argon-purged headspace (to allow for expansion of the starting fluid at conditions), and then pressurized and brought to initial condition (200 °C, 350 bar) rapidly. Several milliliters of fluid were bled during heat-up to purge the exit tube, leaving an estimated 52.6 g of fluid in the cell at the beginning of the experiment (Table*** *Error! Reference source not found.**).***

L126-128 looks more like Methods than Results.
Moved to Methods.

L157ff. "These equi158librium concentrations" …" indicating strong 159 alkane-alkene disequilibrium" does not make much sense to me.
Changed to:

> ***Measured concentrations of alkenes were ~2 orders of magnitude higher than alkane-alkene equilibrium predictions, indicating strong disequilibrium in the relative concentration of alkenes and alkanes.***

This equilibration is quite rapid usually, so it just points to faster generation kinetics than those of hydrogenation-dehydrogenation reactions (as described in the paragraph immediately following).

L175. Under review or submitted citations are not allowed. This must either wait for acceptance or be cited as "unpublished".

Cited now as unpublished. It is expected that the Turner et al. manuscript will be accepted relatively soon since all reviews are in and only minor revisions were required.

L376. Although the water:rock ratio of 5:1 was used, it is probably much more germane to consider the water:organic carbon ratio which would be closer to 80:1 (TOC ~ 6.2%) assuming that the mineral carbon is not participating in the methane generation system such as the "reduction of alkali metal carbonate to methane".

Thank you for the comment. Added:

The water:carbon ratio was concomitantly high, approximately 200:1 given the TOC of 2.5% and ignoring mineral carbon which is assumed to not participate in the generation of thermogenic methane.