

High H₂ production in sediment-hosted hydrothermal fluids at an ultraslow spreading mid ocean ridge

Corresponding Author: Dr Alexander Diehl

This file contains all editorial decision letters in order by version, followed by all author rebuttals in order by version.

Version 0:

Decision Letter:

**** Please ensure you delete the link to your author home page in this e-mail if you wish to forward it to your coauthors ****

Dear Dr Diehl,

Your manuscript titled "High H₂ production in sediment-hosted hydrothermal fluids at an ultraslow spreading ridge" has now been seen by 2 reviewers, and we include their comments at the end of this message. They find your work of interest, but some important points are raised. We are interested in the possibility of publishing your study in Communications Earth & Environment, but would like to consider your responses to these concerns and assess a revised manuscript before we make a final decision on publication. Specifically, we ask you to:

1. Provide clear geophysical evidence to support the dominance of detachment faulting in the northern region.
2. Fully explain the geophysical process and origin of the continuous organic matter assumed in the thermodynamic modeling.
3. Compare the findings with other sedimented ridge systems and briefly outline the source of the sediments.

We therefore invite you to revise and resubmit your manuscript, along with a point-by-point response that takes into account the points raised. Please highlight all changes in the manuscript text file.

Please submit your point-by-point responses as a separate file, distinct from your cover letter where you can add responses to the Editors' comments that you do not want to be made available to the reviewers. Word files are preferred. We recommend that any figures, tables or graphs that are included in the response to reviewers are also included in the main article or Supplementary Information.

We are committed to providing a fair and constructive peer-review process. Please don't hesitate to contact us if you wish to discuss the revision in more detail.

Please use the following link to submit your revised manuscript, point-by-point response to the referees' comments (which should be in a separate document to any cover letter), a tracked-changes version of the manuscript (as a PDF file) and the completed checklist:

Link Redacted

**** This url links to your confidential home page and associated information about manuscripts you may have submitted or be reviewing for us. If you wish to forward this email to co-authors, please delete the link to your homepage first ****

We hope to receive your revised paper within six weeks; please let us know if you aren't able to submit it within this time so that we can discuss how best to proceed. If we don't hear from you, and the revision process takes significantly longer, we may close your file. In this event, we will still be happy to reconsider your paper at a later date, as long as nothing similar has been accepted for publication at Communications Earth & Environment or published elsewhere in the meantime.

Please do not hesitate to contact us if you have any questions or would like to discuss these revisions further. We look forward to seeing the revised manuscript and thank you for the opportunity to review your work.

Best regards,

Renbiao Tao, PhD
Editorial Board Member
Communications Earth & Environment
orcid.org/0000-0003-4797-5211

Alireza Bahadori, PhD
Associate Editor
Communications Earth & Environment
Consulting Editor
Communications Sustainability

EDITORIAL POLICIES AND FORMATTING

We ask that you ensure your manuscript complies with our editorial policies. Please ensure that the following formatting requirements are met, and any checklist relevant to your research is completed and uploaded as a Related Manuscript file type with the revised article.

For Manuscripts that fall into the following fields:

- Behavioural and social science
- Ecological, evolutionary & environmental sciences
- Life sciences

An updated and completed version of our Reporting Summary must be uploaded with the revised manuscript

You can download the form here:

<https://www.nature.com/documents/nr-reporting-summary.pdf>

Furthermore, please align your manuscript with our format requirements, which are summarized on the following checklist:

<https://www.nature.com/documents/commsj-phys-style-formatting-checklist-article.pdf> Communications Earth & Environment formatting checklist

and also in our style and formatting guide <https://www.nature.com/documents/commsj-phys-style-formatting-guide-accept.pdf> Communications Earth & Environment formatting guide.

*** DATA: Communications Earth & Environment endorses the principles of the Enabling FAIR data project (<http://www.copdess.org/enabling-fair-data-project/>). We ask authors to make the data that support their conclusions available in permanent, publically accessible data repositories. (Please contact the editor if you are unable to make your data available).

All Communications Earth & Environment manuscripts must include a section titled "Data Availability" at the end of the Methods section or main text (if no Methods). More information on this policy, is available at <http://www.nature.com/authors/policies/data/data-availability-statements-data-citations.pdf>.

In particular, the Data availability statement should include:

- Unique identifiers (such as DOIs and hyperlinks for datasets in public repositories)
- Accession codes where appropriate
- If applicable, a statement regarding data available with restrictions
- If a dataset has a Digital Object Identifier (DOI) as its unique identifier, we strongly encourage including this in the Reference list and citing the dataset in the Data Availability Statement.

DATA SOURCES: All new data associated with the paper should be placed in a persistent repository where they can be freely and enduringly accessed. We recommend submitting the data to discipline-specific, community-recognized repositories, where possible and a list of recommended repositories is provided at <http://www.nature.com/sdata/policies/repositories>.

If a community resource is unavailable, data can be submitted to generalist repositories such as <https://figshare.com/> or <http://datadryad.org/> Dryad Digital Repository. Please provide a unique identifier for the data (for example a DOI or a permanent URL) in the data availability statement, if possible. If the repository does not provide identifiers, we encourage authors to supply the search terms that will return the data. For data that have been obtained from publically available sources, please provide a URL and the specific data product name in the data availability statement. Data with a DOI should be further cited in the methods reference section.

Please refer to our data policies at <http://www.nature.com/authors/policies/availability.html>.

REVIEWER COMMENTS:

Reviewer #1 (Remarks to the Author):

I evaluate the manuscript well-written. Observation has been conducted by appropriate manner. Interpretation is also appropriate while seems biased from a viewpoint of equilibrium, not kinetics. One serious problem is the apparent mention only for Western-side sites while there are numerous sediment-associated sites studied in the other regions. It may be unfair and discriminatory for the efforts of people in such region. I recommend publication after minor revision.

L91-93 Numerous sediment-associated sites have been studied also in the Okinawa Trough, including JApan-Deutschland, JADE site (Sakai et al 1990 Science; Ishibashi et al. 1995 GCA). Some of them discuss the origin of hydrocarbons as well as this study.

L159-161 I cannot follow the meaning of this sentence. Fluid of sediment-associated site unavoidably contains "mix of abiotic and thermogenic". Does it mean no contribution of biogenic one?

L185 I cannot follow the meaning "due to slower isotope exchange kinetics" here. I read here "Slower isotope exchange leads to higher temperature". Is it correct? What is the assumption of the authors?

L190 Why cooling leads to enrichment in CH₄?

L191 I cannot follow the meaning "using CH₄/CO₂ ratios and H₂ concentration". What is the constant constrained by database? Please clarify.

L199-201 Almost all the hydrothermal fluids observed to date exhibits non-equilibrium composition for the Sabatier reaction (see McCollom 2008 AGU book and calculate the data taken from the vent sites). Consistent interpretation for the observations, not experimental works, between previous and present studies is necessary here.

L239 Low temperature fluid-sediment interaction, particularly microbial metabolisms in warm sediment, is not considered. It could reduce sulfate to sulfide, generate hydrogen and methane, and release some elements in sedimentary organic matter like B.

L261 Again, only Western-side works are followed.

Reviewer #2 (Remarks to the Author):

On the whole, this is an interesting contribution that expands our knowledge about processes that can contribute to hydrothermal vent fluid compositions in sedimented ridge settings. This paper presents new chemical data and thermodynamic modelling of vent fluids from the Jøtul hydrothermal field along the ultraslow-spreading Knipovich Ridge (Arctic Mid-Ocean Ridge system). Based on pH values, high methane, hydrogen, and ammonium concentrations and low metal contents, the Jøtul vent field is interpreted as a sediment-hosted vent system.

The authors invoke a detachment fault tectonic setting to hypothesize fluid pathways and reaction zones. Although they mention that detachment faulting is dominant in the northern parts of the Knipovich Ridge, the authors do not clearly present geophysical evidence (other than a change in slope and one soapstone sample) to validate this assumption. Is there any seismic data for this area? The evidence for detachment faulting could possibly be summarized more succinctly in the text. With such high estimated temperatures of venting (Lines 181 – 218), one would expect magmatic activity in the subsurface. However, it seems the authors argue that the high temperatures are controlled solely by the greater water depths of ~3000 mbsl. An additional magmatic heat source is not discussed at all in the paper. Where could a magmatic source be in Fig. 1d?

The authors postulate that within this detachment fault setting, alteration of ultramafic rocks could be considered to contribute to fluid compositions (also inferred in Figure 1d). Alternatively, the authors argue on the basis of thermodynamic modelling that thermal degradation of organic matter accompanied by abiotic methane oxidation in the reaction zone is the dominant control on fluid chemistries and volatile production. This is a viable mechanism in this tectonic setting of an ultraslow ridge close to continental detritus, although it is uncertain how common such settings are on a global scale. The authors bounce back and forth between discussions of these two mechanisms to explain the fluid compositions and high CH₄, CO₂ and H₂ concentrations and low H₂S; however, this back-and-forth strategy makes the arguments for a preference of organic matter degradation more difficult to follow. Primarily on the basis of the thermodynamic reaction path modelling, the authors rule out the possibility that both processes contribute to the distinct fluid compositions. In this modelling, they also assume continuous addition of organic matter, but there is no discussion as to how this occurs geologically and what the source of the organic matter is. One would think that productivity is unlikely to be high at these latitudes, therefore one must assume that the organic matter is primarily detrital, but how it is continually added to the system is unclear. In addition, it would be helpful to have a brief statement about the source of the sediments.

I would have liked to have seen more comparison to other sedimented ridge systems that have been previously investigated. In particular, Loki's Castle, located at the southern part of the ultraslow-spreading Knipovich Ridge can also be considered a sedimented ridge (or sediment-influenced) vent system with similar chemical and isotopic compositions to those reported in this paper by Diehl et al. (see Baumberger et al., 2016 - *Geochimica et Cosmochimica Acta* 187 (2016) 156–178). In the description of the Jøtul fluid compositions (Lines 142-180) and comparisons to other vent fluids (in text and Fig. 2), it might be worth considering using specific symbols for the Loki's Castle fluids in the figures. Since this is also a vent field that is also on the Knipovich Ridge and the first to report sediment influence on fluid compositions in an ultraslow-spreading setting, it would make the similarities along the Knipovich Ridge (and the greater influence of sediments towards the north?) easier to see. In addition, the authors might want to consider including a summary of average Loki's Castle EM compositions in Table 1, and/or provide a separate table (possibly as supplementary material) that compares compositions of vent fluids in other sedimented and ultramafic vent sites. Such comparisons might be of more interest to a general audience than the modelling shown in Figure 5 (which could be included in the supplementary material without compromising the conclusions

of the study). In particular, Figure 5(b) may have less significance to substantiate the conclusions and be harder to understand for non-petrologists.

The authors infer that the Jøtul system is more organic carbon rich than other sedimented ridge vent sites. If this is true, it would have been good to have an indication of the organic carbon concentrations (TOC) in these sediments or the dissolved organic carbon (DOC) concentrations in the fluids. As stated above, where does the organic material come from – is it solely detrital?

In summary, this study presents new data that expand the range of endmember compositions of fluids emanating from sedimented ridges and concludes that hydrothermal circulation at greater depths and higher temperatures than other sedimented ridge vent sites can lead to high H₂ concentrations that can be greater than those produced by water-rock reactions in ultramafic rocks. Such high temperature, sedimented ridge settings can thus be a further source of carbon and hydrogen to the oceans. The question however arises if, or how much, such relatively rare sedimented vent systems contribute to global carbon and hydrogen budgets?

Other minor comments:

Figure 1d: misspelling in label below GeoB20526-2 in the figure. It should be soapstone not saopstone. It is spelled correctly in the figure caption and text.

Introduction, Line 78: You might consider citing the more recent Nature Review and compilation table (supplementary material) of Fröh-Green et al., 2022:

Fröh-Green G.L., Kelley D.S., Lilley M.D., Cannat M., Chavagnac V., Baross J.A. (2022) Diversity of magmatism, hydrothermal processes and microbial interactions at mid-ocean ridges. *Nature Reviews Earth & Environment*. <https://doi.org/10.1038/s43017-022-00364-y>.

Lines 61 and 152 refers to the pH of the fluids as being high pH. This is perhaps a bit misleading, since high pH more commonly infers values above seawater pH, as are typical of ultramafic vent fluids, which can be highly alkaline. It may be appropriate to re-write these sentences to state that the pH values of the fluids are distinctly higher than average basalt-hosted vent fluids and are typical of sedimented ridges.

Line 87 – The authors could include reference to Aquino et al. (*Geochimica et Cosmochimica Acta* 332 (2022) 239–262) which also documents variably high H₂ and low H₂S at Lost City.

Line 106: Mentioning the Logatchev Volcanic Center here is confusing since the paragraph is about the Knipovich Ridge. I suggest adding (central MAR) to be clearer: i.e., Logatchev Volcanic Center (central MAR).

Lines 142-180: This is a very long paragraph - it could probably be split into two paragraphs.

Lines 155-156: “CO₂ concentrations (36.0–41.6 mM) far exceed those from the diluted, and partly degassed KIPS samples recovered 2022” – It is unnecessary to state this, since it is stated earlier in the text and it is well known that samples that are not taken in gas-tight bottles degas upon recovery and provide only minimum concentrations.

Lines -176-178: “These concentrations of dissolved volatiles make the Jøtul fluids particularly enriched in inorganic carbon species with higher than expected H₂ concentrations, which could point to exceptional subsurface conditions and suggest that ultramafic rocks are accountable for the high H₂ and low H₂S concentrations.”

It is not clear to me how the authors come to the conclusion that all of the carbon is inorganic. In addition, “exceptional subsurface conditions” is very vague. The origin/controls producing high CO₂ concentrations could be better elucidated and, as mentioned above, magmatic input could be discussed in more detail.

Minor comment: I suggest changing “associated to” to “associated with”. Lines 124 and 229

Line 189: What is TA?

** Visit Nature Portfolio's author and referees' website at www.nature.com/authors for information about policies, services and author benefits**

Communications Earth & Environment is committed to improving transparency in authorship. As part of our efforts in this direction, we are now requesting that all authors identified as ‘corresponding author’ create and link their Open Researcher and Contributor Identifier (ORCID) with their account on the Manuscript Tracking System prior to acceptance. ORCID helps the scientific community achieve unambiguous attribution of all scholarly contributions. You can create and link your ORCID from the home page of the Manuscript Tracking System by clicking on ‘Modify my Springer Nature account’ and following the

instructions in the link below. Please also inform all co-authors that they can add their ORCID to their accounts and that they must do so prior to acceptance.

<https://www.springernature.com/gp/researchers/orcid/orcid-for-nature-research>

For more information please visit <http://www.springernature.com/orcid>

If you experience problems in linking your ORCID, please contact the [Platform Support Helpdesk](http://platformsupport.nature.com/).

Version 1:

Decision Letter:

**** Please ensure you delete the link to your author home page in this e-mail if you wish to forward it to your coauthors ****

Dear Dr Diehl,

Your revised manuscript titled "High H₂ production in sediment-hosted hydrothermal fluids at an ultraslow spreading mid-ocean ridge" has now been seen by our reviewers, whose comments appear below. In light of their advice we are delighted to say that we are happy, in principle, to publish a suitably revised version in Communications Earth & Environment.

We therefore invite you to revise your paper one last time to address the remaining concerns of our reviewer 2. At the same time we ask that you edit your manuscript to comply with our format requirements and to maximise the accessibility and therefore the impact of your work.

EDITORIAL REQUESTS:

Please review our specific editorial comments and requests regarding your manuscript in the attached "Editorial Requests Table".

****Please take care to match our formatting and policy requirements. We will check revised manuscript and return manuscripts that do not comply. Such requests will lead to delays. ****

Please outline your response to each request in the right hand column. Please upload the completed table with your manuscript files as a Related Manuscript file.

If you have any questions or concerns about any of our requests, please do not hesitate to contact me.

SUBMISSION INFORMATION:

In order to accept your paper, we require the files listed at the end of the Editorial Requests Table; the list of required files is also available at <https://www.nature.com/documents/commsj-file-checklist.pdf>.

OPEN ACCESS:

Communications Earth & Environment is a fully open access journal. Articles are made freely accessible on publication. For further information about article processing charges, open access funding, and advice and support from Nature Portfolio, please visit <https://www.nature.com/commsenv/open-access>

At acceptance, you will be provided with instructions for completing the open access licence agreement on behalf of all authors. This grants us the necessary permissions to publish your paper. Additionally, you will be asked to declare that all required third party permissions have been obtained, and to provide billing information in order to pay the article-processing charge (APC).

Please use the following link to submit the above items:

Link Redacted

**** This url links to your confidential home page and associated information about manuscripts you may have submitted or be reviewing for us. If you wish to forward this email to co-authors, please delete the link to your homepage first ****

We hope to hear from you within two weeks; please let us know if you need more time.

Best regards,

Renbiao Tao, PhD
Editorial Board Member
Communications Earth & Environment

Alireza Bahadori, PhD
Senior Editor
Communications Earth & Environment
Consulting Editor
Communications Sustainability

REVIEWERS' COMMENTS:

Reviewer #1 (Remarks to the Author):

Nothing to add.

Reviewer #2 (Remarks to the Author):

The authors have addressed the pertinent concerns of the reviewers of the initial manuscript. Putting the study into a broader, and more detailed, geological and geochemical context particularly makes this contribution more attractive for a general audience.

My main comments of the revised manuscript concern the figures, and will largely require the editor's input: In Figures 2 and 4, the symbols of other vent sites are relatively small and it is difficult to distinguish the dark green symbols for Rainbow and Logatchev from the other green symbols of peridotite-hosted sites.

Figure 1a is understandably very minimalistic, but would it be worthwhile to include the location of Loki's Castle, since it is also a sediment-influenced vent site along the Knipovich Ridge? I still find that Figure 1d requires modification. The authors have added two very small magmatic lenses, which may be relatively unrealistic. Why not consider more and larger magmatic (gabbroic) lenses? Even though the authors tend to argue that the high temperatures could be controlled by heat from cooling of ultramafic rocks, an additional magmatic source also seems likely in this environment, and has been postulated in other oceanic core complexes. In fact, the authors state this in the discussion, Lines 609-611 (marked up version). In addition, the Introduction Lines 198-199 state that the Jøtul field is located amidst numerous normal faults, but Figure 1 does not depict these.

Supplement Figure S2: What are the mineral compositions of the hydrothermal deposits?

Additional very minor editorial comments are given below. Line numbers refer to the marked up version of the revised manuscript. In general, the revisions could have benefited from more careful proof-reading and more succinct wording.

Line 53-54: add comma - ..., at the ultraslow-spreading northern Knipovich Ridge, show...

Line 133: Misspelling – constraints instead of constrainst

Line 205, delete comma after (...Hornsund)

Lines 305-306: I realise that the Delta values given here are averages, but is it realistic to report a Delta D value to a decimal point since precision of the measurements is likely to be less accurate? Same in Table S7.

Line 610: detachment not detachments. I assume there is likely only one detachment system in the area.

Lines 616-617: Check reaction depth range of 300-2500 mbsf, and in Line 617, add: (corresponding to a pressure of 330-545 bar). This is quite a large range.

** Visit Nature Portfolio's author and referees' website at www.nature.com/authors for information about policies, services and author benefits**

Open Access This Peer Review File is licensed under a Creative Commons Attribution 4.0 International License, which permits use, sharing, adaptation, distribution and reproduction in any medium or format, as long as you give appropriate credit to the original author(s) and the source, provide a link to the Creative Commons license, and indicate if changes were made.

In cases where reviewers are anonymous, credit should be given to 'Anonymous Referee' and the source.

The images or other third party material in this Peer Review File are included in the article's Creative Commons license, unless indicated otherwise in a credit line to the material. If material is not included in the article's Creative Commons license and your intended use is not permitted by statutory regulation or exceeds the permitted use, you will need to obtain permission directly from the copyright holder.

To view a copy of this license, visit <https://creativecommons.org/licenses/by/4.0/>

Authors: We sincerely thank the reviewers and editors for their time and thoughtful feedback, which has significantly improved our manuscript. We are deeply grateful for the constructive comments provided.

One of the main concerns raised was the lack of supporting evidence for the occurrence of detachment faulting below the vent field. While we only partially agree with this criticism, we remain convinced that the two pieces of evidence we presented (seafloor slope and occurrence of altered ultramafic rocks) are valid indications for hypothesizing detachment faulting. We also ask: Which other geological mechanisms could expose ultramafic rocks at the seafloor in the context of this geological setting? Our study focuses on vent fluid analyses coupled with seafloor investigations. Including geophysical investigations would go beyond the scope of this work, and unfortunately, such data are currently unavailable, as the area is not yet well explored. We hope the reviewers and editors recognize that our findings provide valuable impulses for further research in this region.

The second major concern related to the origin of organic matter that we propose involved in generating fluid composition. We now state more clearly that this organic matter is delivered as detritus, together with sedimentary material. We also clarify that the concentration of organic matter we used in our reaction path model is relatively low compared to reported values for sediments on the Knipovich Ridge and in western Svalbard fjords, which are likely the source region of these sediments.

In addition, we have addressed the reviewers' request to compare our vent fluid endmember with endmembers from other sedimented and ultramafic-hosted vent sites, and we now discuss the potential magmatic heat source in the subsurface. The reviewers rightly noted that the high fluid temperatures suggest such a magmatic heat source. While we previously did not emphasize this point, as our evidence based on reaction zone depth is of indirect nature, we now explicitly discuss the heat source and propose that a shallow magmatic body likely drives the hydrothermal circulation.

We have additionally improved the overall flow of the manuscript by another round of language editing and emphasized particularly to homogenize formal aspects for better readability. We have also added quite a number of citations to support some statements that were lacking such.

Our detailed point-by-point responses are provided below, where we have carefully addressed all concerns raised. Finally, we would like to kindly invite the reviewers, if they wish, to reveal their identity so that we may acknowledge their valuable feedback in the manuscript.

Sincerely yours,

Alexander Diehl (on behalf of the authors)

Note: the below mentioned line numbers refer to the tracked change document.

Editors:

1. Provide clear geophysical evidence to support the dominance of detachment faulting in the northern region.

Authors: We fully understand the request for additional evidence supporting the assumed detachment fault in the study area. Unfortunately, to the best of our knowledge, no such

geophysical data are currently available or have been published. Nevertheless, we believe that the seafloor slope, together with the exposure of ultramafic rocks at the seafloor, provides sound evidence for detachment faulting. These geomorphological characteristics have so far mainly been described along detachment footwalls (Blackman et al., 2002; Smith et al., 2006). Additional constraints arise from numerical studies of detachment faults. These show that continued extensions along major faults (detachments) is known to result in reduction of the angle of the exposed footwall surface due to flexural rollover (Buck, 2005; Tucholke et al., 2008; Mezri et al., 2023). We hope that the hypothesis presented in our manuscript will encourage future geophysical surveys around the Jøtul hydrothermal field and we are confident that independent research will further test, and potentially confirm, our interpretation. We edited the paragraph for more clarity and added a sentence with the mentioned references incorporated (L. 151-165).

2. Fully explain the geophysical process and origin of the continuous organic matter assumed in the thermodynamic modeling.

Authors: It appears that the wording of our model description may have caused some confusion. By “continuous addition” of organic matter, we intended to describe its relation to the progression of the water-rock reaction. In other words, the fluid incorporates more organic matter as it continues to percolate through additional rock. To clarify, our approach was simply to react a hydrothermal vent fluid with an organic matter bearing sediment, which we assume to be of detrital origin. We have rephrased this section for more clarity (L. 329-336).

3. Compare the findings with other sedimented ridge systems and briefly outline the source of the sediments.

Authors: We have added values for comparison in Table 1 and Figures 2 and 4.

We have added a section discussing total organic carbon (TOC) concentrations (1-2wt.%) in fjord sediments from western Svalbard, which likely represent the primary parental material for the sediments delivered to our study area. A terrestrial origin of this sedimentary material is strongly supported by the Sr-isotope signature (~ 0.709) of the hydrothermal vent fluids (Bohrmann et al., 2022; see reference list). We also refer to TOC concentrations (0.2-1 wt.%) in surface sediments on the Knipovich Ridge. Based on these TOC values, we demonstrate that our modelling (assuming 0.22 wt.% of total organic matter) uses a conservative estimate of total organic carbon, when compared with sediment samples from both the Knipovich Ridge and the Svalbard margin (L. 132-143).

Reviewer 1:

I evaluate the manuscript well-written. Observation has been conducted by appropriate manner. Interpretation is also appropriate while seems biased from a viewpoint of equilibrium, not kinetics. One serious problem is the apparent mention only for Western-side sites while there are numerous sediment-associated sites studied in the other regions. It may be unfair and discriminatory for the efforts of people in such region. I recommend publication after minor revision.

L91-93 Numerous sediment-associated sites have been studied also in the Okinawa Trough,

including Japan-Deutschland, JADE site (Sakai et al 1990 Science; Ishibashi et al. 1995 GCA). Some of them discuss the origin of hydrocarbons as well as this study.

Authors: We thank the reviewer for pointing out this potential “bias”. The limited citation of these vent sites was primarily due to the strict word and the reference cap of 50 imposed by Nature Geosciences, the Journal to which this manuscript was initially submitted. For this reason, we prioritized citing studies from the most comparable vent sites-namely the “Western Sites”, which are hosted in mid-oceanic ridges similar to the Jøtul hydrothermal field. In contrast, the Okinawa Through sites mentioned here are hosted in back-arc spreading centers, where magmatic degassing likely plays a stronger role and venting occurs at shallower depths (i.e., lower reaction zone pressures). That said, we did consider these sites by citing Toki et al., 2022 (Minami-Ensei) and Ishibashi et al., 2014 (Izena Hole). We nevertheless fully agree with the reviewers comment and have now included the recommended references, along with a few additional ones and mention some of their main conclusions (L. 97-112).

L159-161 I cannot follow the meaning of this sentence. Fluid of sediment-associated site unavoidably contains "mix of abiotic and thermogenic". Does it mean no contribution of biogenic one?

Authors: Yes, our intention was to emphasize that there is no indication of biogenic production and we have now clarified this explicitly in the text underlined by two key citations (L. 200-203).

L185 I cannot follow the meaning "due to slower isotope exchange kinetics" here. I read here "Slower isotope exchange leads to higher temperature". Is it correct? What is the assumption of the authors?

Authors: Yes, the reviewer are correct in this assumption. We suggest that the slower reaction kinetics of carbon isotopes exchange results in higher isotope thermometer temperatures compared to the hydrogen isotope thermometer, as hydrogen isotopes continue to exchange while carbon isotopes do not. This occurs because the isotope exchange rate becomes slower than the cooling rate. To clarify this point, we have revised the text to read: “despite high temperatures” and added the statement “the isotope exchange slows down upon cooling” (L. 239-241).

L190 Why cooling leads to enrichment in CH₄?

Authors: The Sabatier reaction equilibrium will shift towards higher CH₄ with decreasing temperature (3 molecules are favored over 5 molecules at lower T). The original sentence meant solely to highlight the significant compositional difference compared with the other fluid samples. We did not intend to impose any relation to the previously mentioned cooling. We just wanted to mention that this cooling is the only difference between the fluid samples so they might be related (L 243-246).

L191 I cannot follow the meaning "using CH₄/CO₂ ratios and H₂ concentration". What is the constant constrained by database? Please clarify.

Authors: We now explain that this calculation is based on the rearrangement of the mass action equation and the temperature dependent equilibrium constants constrained from thermodynamic database (L.250-256).

L199-201 Almost all the hydrothermal fluids observed to date exhibit non-equilibrium composition for the Sabatier reaction (see McCollom 2008 AGU book and calculate the data taken from the vent sites). Consistent interpretation for the observations, not experimental works, between previous and present studies is necessary here.

Authors: Indeed, McCollom (2008) reports that the Sabatier equilibrium does not adequately explain CH₄ generation in non-sedimented hydrothermal vent fields. In these systems, the chemistry of inorganic carbon is primarily controlled by magmatic degassing, which leads to an excess of CO₂ over CH₄ relative to the Sabatier reaction equilibrium (i.e., the reaction proceeds from right to left (see Reaction 1 in the MS). In contrast, in sediment-hosted vent sites, CH₄ excess is produced through the thermal decomposition of organic matter, and the reaction proceeds from left to right. Other studies (e.g., Seewald et al., 1994) have suggested that when CH₄ excess is present, the kinetics of the Sabatier reaction are more favorable, allowing the reaction to reach equilibrium. We have now added the reference McCollom 2008 (L. 262-267).

L239 Low temperature fluid-sediment interaction, particularly microbial metabolisms in warm sediment, is not considered. It could reduce sulfate to sulfide, generate hydrogen and methane, and release some elements in sedimentary organic matter like B.

Authors: The reviewer raises a valuable point. However, our reaction path model does not incorporate biogenic processes. A key requirement for reproducing the observed fluid compositions is that the fluid enters the reaction zone with low sulfide content. The incorporation of microbially produced sulfide would therefore complicate the successful modeling of these composition. Previously, we used the C₁/(C₂-C₃) ratio to argue that microbial processes are unlikely to control vent fluid compositions in the Jøtul hydrothermal field. While some microbial production of hydrogen and methane may occur, this would not affect our conclusions regarding the high-temperature processes, which would certainly overprint most microbial signatures introduced during cold-warm recharge, prior to entering the high-temperature reaction zone.

L261 Again, only Western-side works are followed.

Authors: We have incorporated some of the Okinawa Trough vent fields with adequate citations (L. 355-358).

Reviewer 2:

On the whole, this is an interesting contribution that expands our knowledge about processes that can contribute to hydrothermal vent fluid compositions in sedimented ridge settings. This paper presents new chemical data and thermodynamic modelling of vent fluids from the Jøtul hydrothermal field along the ultraslow-spreading Knipovich Ridge (Arctic Mid-Ocean Ridge system). Based on pH values, high methane, hydrogen, and ammonium concentrations and low metal contents, the Jøtul vent field is interpreted as a sediment-hosted vent system.

The authors invoke a detachment fault tectonic setting to hypothesize fluid pathways and reaction

zones. Although they mention that detachment faulting is dominant in the northern parts of the Knipovich Ridge, the authors do not clearly present geophysical evidence (other than a change in slope and one soapstone sample) to validate this assumption. Is there any seismic data for this area? The evidence for detachment faulting could possibly be summarized more succinctly in the text. With such high estimated temperatures of venting (Lines 181 – 218), one would expect magmatic activity in the subsurface. However, it seems the authors argue that the high temperatures are controlled solely by the greater water depths of ~3000 mbsl. An additional magmatic heat source is not discussed at all in the paper. Where could a magmatic source be in Fig. 1d?

Authors: The Reviewer are absolutely correct in highlighting the possibility of a magmatic heat source. As we lack direct geophysical observations, we have been cautious in interpreting the nature of that heat source. On the one hand, a magmatic heat source appears very likely, given the shallow reaction zone depth inferred from Si, Cl and the reaction zone temperature, as well as the presence of pillow lavas cropping out of the sediments near the vent site. On the other hand, such a heat source is not strictly required to drive hydrothermal circulation, as heat could also be supplied through the exhumation of hot mantle rocks along the presumed detachment fault. Nevertheless, we have added a paragraph that considers the possibility of a shallow magmatic heat source, which likely accounts for the shallow reaction zone depth (L. 286-303).

The authors postulate that within this detachment fault setting, alteration of ultramafic rocks could be considered to contribute to fluid compositions (also inferred in Figure 1d). Alternatively, the authors argue on the basis of thermodynamic modelling that thermal degradation of organic matter accompanied by abiotic methane oxidation in the reaction zone is the dominant control on fluid chemistries and volatile production. This is a viable mechanism in this tectonic setting of an ultraslow ridge close to continental detritus, although it is uncertain how common such settings are on a global scale. The authors bounce back and forth between discussions of these two mechanisms to explain the fluid compositions and high CH₄, CO₂ and H₂ concentrations and low H₂S; however, this back-and-forth strategy makes the arguments for a preference of organic matter degradation more difficult to follow. Primarily on the basis of the thermodynamic reaction path modelling, the authors rule out the possibility that both processes contribute to the distinct fluid compositions. In this modelling, they also assume continuous addition of organic matter, but there is no discussion as to how this occurs geologically and what the source of the organic matter is. One would think that productivity is unlikely to be high at these latitudes, therefore one must assume that the organic matter is primarily detrital, but how it is continually added to the system is unclear. In addition, it would be helpful to have a brief statement about the source of the sediments.

Authors: We believe that our statement “due to continuous addition of organic matter” (L. 334) may have caused some confusion. This wording refers to the reaction path model, in which the vent fluid undergoes progressive fluid rock interaction. With “continuous addition” we referred to fluid uptake of organic matter when percolating through additional rock. We rephrased this particular statement for more clarity (L. 334-335).

In the model, organic matter is introduced as a rock component at a relatively low concentration of 0.22 wt.%, which we consider a conservative assumption for TOC concentrations in the study area. For comparison, TOC concentrations in Svalbard Fjord sediments (1-2wt.% TOC), that likely reflect the parent material reworked and transported toward the Knipovich Ridge, are significantly higher than our assumption of 0.22 wt.%. Measurements of TOC concentrations in surface sediments at the Knipovich Ridge yield concentrations of (0.2-1 wt.% TOC) and align well with our assumption of 0.22

wt.%. We would therefore like to emphasize that the TOC content assumed in our model is not high. To clarify this point, we have added a paragraph discussing the sediment origin and the TOC concentrations applied in the model (L. 132-143 and L. 329-331).

I would have liked to have seen more comparison to other sedimented ridge systems that have been previously investigated. In particular, Loki's Castle, located at the southern part of the ultraslow-spreading Knipovich Ridge can also be considered a sedimented ridge (or sediment-influenced) vent system with similar chemical and isotopic compositions to those reported in this paper by Diehl et al. (see Baumberger et al., 2016 - *Geochimica et Cosmochimica Acta* 187 (2016) 156–178). In the description of the Jøtul fluid compositions (Lines 142-180) and comparisons to other vent fluids (in text and Fig. 2), it might be worth considering using specific symbols for the Loki's Castle fluids in the figures. Since this is also a vent field that is also on the Knipovich Ridge and the first to report sediment influence on fluid compositions in an ultraslow-spreading setting, it would make the similarities along the Knipovich Ridge (and the greater influence of sediments towards the north?) easier to see. In addition, the authors might want to consider including a summary of average Loki's Castle EM compositions in Table 1, and/or provide a separate table (possibly as supplementary material) that compares compositions of vent fluids in other sedimented and ultramafic vent sites. Such comparisons might be of more interest to a general audience than the modelling shown in Figure 5 (which could be included in the supplementary material without compromising the conclusions of the study). In particular, Figure 5(b) may have less significance to substantiate the conclusions and be harder to understand for non-petrologists.

Authors: We are grateful for this comment and have added the endmember compositions of Loki's Castle, Guaymas Basin, Logatchev and Rainbow to Table 1 and now use specific symbols for the mentioned vent sites in Figures 2 and 4. In addition, we have moved figure 5b to the supplementary material.

The authors infer that the Jøtul system is more organic carbon rich than other sedimented ridge vent sites. If this is true, it would have been good to have an indication of the organic carbon concentrations (TOC) in these sediments or the dissolved organic carbon (DOC) concentrations in the fluids. As stated above, where does the organic material come from – is it solely detrital?

Authors: Yes, we assume that the material represents detrital organic carbon. We would like to clarify that we do not suggest that Jøtul sediments are particularly enriched in organic carbon. Rather, we assume a TOC concentration of 0.22 wt.% in the sediments undergoing fluid-rock reactions. This value lies within the global average for marine sediments and at the lower end of the TOC concentrations reported for Knipovich Ridge sediments (0.2-1 wt.%) sampled about 45 km south of our study area. In comparison, this assumed concentration is about an order of magnitude lower than the TOC content of Fjord sediments from western Svalbard, which likely represent the parent material delivered to the Knipovich Ridge. (see the detailed discussion as reply to the prior comments; L. 132-143)

In summary, this study presents new data that expand the range of endmember compositions of fluids emanating from sedimented ridges and concludes that hydrothermal circulation at greater

depths and higher temperatures than other sedimented ridge vent sites can lead to high H₂ concentrations that can be greater than those produced by water-rock reactions in ultramafic rocks. Such high temperature, sedimented ridge settings can thus be a further source of carbon and hydrogen to the oceans. The question however arises if, or how much, such relatively rare sedimented vent systems contribute to global carbon and hydrogen budgets?

Authors: This is a very interesting comment and should be subject to further research. We are indeed currently working on upscaling these results but this would go beyond the scope of this MS.

Other minor comments:

Figure 1d: misspelling in label below GeoB20526-2 in the figure. It should be soapstone not saopstone. It is spelled correctly in the figure caption and text.

Authors: We thank the reviewer for their close attention. We corrected and updated the figure.

Introduction, Line 78: You might consider citing the more recent Nature Review and compilation table (supplementary material) of Fröh-Green et al., 2022:

Früh-Green G.L., Kelley D.S., Lilley M.D., Cannat M., Chavagnac V., Baross J.A. (2022) Diversity of magmatism, hydrothermal processes and microbial interactions at mid-ocean ridges. Nature Reviews Earth & Environment. <https://doi.org/10.1038/s43017-022-00364-y>.

Authors: Citation was added (L. 88).

Lines 61 and 152 refers to the pH of the fluids as being high pH. This is perhaps a bit misleading, since high pH more commonly infers values above seawater pH, as are typical of ultramafic vent fluids, which can be highly alkaline. It may be appropriate to re-write these sentences to state that the pH values of the fluids are distinctly higher than average basalt-hosted vent fluids and are typical of sedimented ridges.

Authors: We changed “high pH values” to “elevated pH values greater than typical for basalt fluid interaction” (L. 65-66). We changed “high pH” to “elevated pH compared to basalt-hosted systems” (L. 189).

Line 87 – The authors could include reference to Aquino et al. (Geochimica et Cosmochimica Acta 332 (2022) 239–262) which also documents variably high H₂ and low H₂S at Lost City.

Authors: We agree and inserted the citation (L. 93).

Line 106: Mentioning the Logatchev Volcanic Center here is confusing since the paragraph is about the Knipovich Ridge. I suggest adding (central MAR) to be clearer: i.e., Logatchev Volcanic Center (central MAR).

Authors: Indeed, the naming here was somewhat confusing to us as well, as it is reminiscent of the well-known Logatchev hydrothermal field. We would like to clarify that the “Logachev Volcanic Center” refers to a volcanic high located on the Knipovich Ridge with no relation to the “central MAR”. (see the cited literature: Meier, M. et al. Segment-Scale Seismicity of the Ultraslow Spreading Knipovich Ridge. Geochemistry, Geophysics, Geosystems 22 (2021).

<https://doi.org/10.1029/2020gc009375>, Figure 1a). To clarify this, we have added the statement “(the most prominent volcanic center along the Knipovich Ridge)” and we added the citation Meier et al., 2022 that focuses on this volcanic center (L. 123).

Lines 142-180: This is a very long paragraph - it could probably be split into two paragraphs.

Authors: Agreed. We have split into three.

Lines 155-156: “CO₂ concentrations (36.0–41.6 mM) far exceed those from the diluted, and partly degassed KIPS samples recovered 2022” – It is unnecessary to state this, since it is stated earlier in the text and it is well known that samples that are not taken in gas-tight bottles degas upon recovery and provide only minimum concentrations.

Authors: We fully agree with this comment, however we would prefer to retain this sentence, as we consider it important to inform readers that the values reported here should be used in preference to those published in 2022. While for geochemists this fact might be clear, for a broader audience we consider re-mentioning useful.

Lines -176-178: “These concentrations of dissolved volatiles make the Jøtul fluids particularly enriched in inorganic carbon species with higher than expected H₂ concentrations, which could point to exceptional subsurface conditions and suggest that ultramafic rocks are accountable for the high H₂ and low H₂S concentrations.”

It is not clear to me how the authors come to the conclusion that all of the carbon is inorganic. In addition, “exceptional subsurface conditions” is very vague. The origin/controls producing high CO₂ concentrations could be better elucidated and, as mentioned above, magmatic input could be discussed in more detail.

Authors: There is ongoing debate about whether CH₄ should be classified as an organic or inorganic species. In this sentence, we simply referred to CO₂ and CH₄. To avoid implying a specific source of these dissolved species, we have revised the wording to “dissolved carbon species”. Additionally, we have removed the statement “exceptional subsurface conditions”.

Minor comment: I suggest changing “associated to” to “associated with”. Lines 124 and 229

Authors: We changed accordingly.

Line 189: What is TA?

Authors: TA stood for total alkalinity. We changed to “alkalinity”.

Authors: We would like to once again thank the reviewers for their valuable feedback during this final round of revisions. We have carefully addressed all comments (see below) and hope that you will now find our manuscript suitably revised and ready for publication in *Communication Earth & Environment*. If you have any questions regarding the content or format of our manuscript, please do not hesitate to contact us.

With best regards,

Alexander Diehl (on behalf of the authors)

REVIEWERS' COMMENTS:

Reviewer #1 (Remarks to the Author):

Nothing to add.

Authors: We thank again the reviewer for their previous inputs.

Reviewer #2 (Remarks to the Author):

The authors have addressed the pertinent concerns of the reviewers of the initial manuscript. Putting the study into a broader, and more detailed, geological and geochemical context particularly makes this contribution more attractive for a general audience.

Authors: We thank the reviewer for acknowledging our efforts.

My main comments of the revised manuscript concern the figures, and will largely require the editor's input:

In Figures 2 and 4, the symbols of other vent sites are relatively small and it is difficult to distinguish the dark green symbols for Rainbow and Logatchev from the other green symbols of peridotite-hosted sites.

Authors: In response to this comment, we have updated Figures 2 and 4, increasing the symbol sizes for other vent sites as well as the comparison sites (Rainbow, Logatchev, Loki's Castle, Guaymas Basin) to improve visual clarity. In addition, we have enhanced the contrast between peridotite-hosted vents and the Rainbow and Logatchev sites. Please note that the figures have been removed from the main text and are now provided only as separate files.

Figure 1a is understandably very minimalistic, but would it be worthwhile to include the location of Loki's Castle, since it is also a sediment-influenced vent site along the Knipovich Ridge? I still find that Figure 1d requires modification. The authors have added two very small magmatic lenses, which may be relatively unrealistic. Why not consider more and larger magmatic (gabbroic) lenses? Even though the authors tend to argue that the high temperatures could be controlled by heat from cooling of ultramafic rocks, an additional magmatic source also seems likely in this environment, and has been postulated in other oceanic core complexes. In fact, the authors state this in the discussion,

Lines 609-611 (marked up version). In addition, the Introduction Lines 198-199 state that the Jøtul field is located amidst numerous normal faults, but Figure 1 does not depict these.

Authors: In Figure 1a, we have added the location of Loki's Castle. Since the Jøtul area was previously indicated only by a rectangle, we have now also included a symbol for the Jøtul hydrothermal field to help maintain the reader's focus on our study area. Regarding Figure 1d, the reviewer correctly noted that we had intentionally chosen a minimalistic design. Nevertheless, we followed the suggestion and made two adjustments: we added the normal faults within the sediment and enlarged as well as included additional gabbroic intrusions, which we agree is geologically more plausible. Thank you for this helpful advice. Please note that the figures have been removed from the main text and are now provided only as separate files.

Supplement Figure S2: What are the mineral compositions of the hydrothermal deposits?

Authors: Sulfides were sampled on several occasions, and the study of their mineralization will be addressed in a separate project. We have added the most typical mineral parageneses to the Figure caption of Supplementary Figure S2 and cited Bohrmann et al. 2024, where these minerals are described in more detail.

Additional very minor editorial comments are given below. Line numbers refer to the marked up version of the revised manuscript. In general, the revisions could have benefited from more careful proof-reading and more succinct wording.

Line 53-54: add comma - ..., at the ultraslow-spreading northern Knipovich Ridge, show...

Authors: corrected

Line 133: Misspelling – constraints instead of constrainst

Authors: corrected

Line 205, delete comma after (...Hornsund)

Authors: corrected

Lines 305-306: I realise that the Delta values given here are averages, but is it realistic to report a Delta D value to a decimal point since precision of the measurements is likely to be less accurate? Same in Table S7.

Authors: We note that the standard deviation is smaller 1.0‰ for the measurements of $\delta^2\text{H-CH}_4$ and $\delta^2\text{H-H}_2$. Removing the decimal point and rounding to the nearest integer would therefore introduce unnecessary uncertainty. In this case, we have chosen to retain the reported number of digits.

Line 610: detachment not detachments. I assume there is likely only one detachment system in the area.

Authors: The only usage of “detachments” was found line 269. We have changed to “detachment”.

Lines 616-617: Check reaction depth range of 300-2500 mbsf, and in Line 617, add: (corresponding to a pressure of 330-545 bar). This is quite a large range.

Authors: We corrected the text and added “to” as suggested. We acknowledge that the range is broad. However, it encompasses the range we estimated based on the vent fluid compositions. The fluid compositional approach provides only an indirect estimate of the depth of the heat source, which limits our ability to further constrain it. Nevertheless, since heat sources in some detachment settings have been reported to occur at depths greater than 5-10 km below the seafloor, this remains a useful information.