

Peer Review File

Deep submarine infiltration of altered geothermal groundwater on the south Chilean Margin



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Web links to the author's journal account have been redacted from the decision letters as indicated to maintain confidentiality.

23rd Feb 22

Dear Mr Clementi,

Your manuscript titled "Deep submarine infiltration of altered geothermal groundwater on the south Chilean Margin" has now been seen by 3 reviewers, and I 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. All referees are positive about your study, the main comments come from referee #1 who requests to provide more context and from referee #3 who requests further justification regarding your interpretation of a modern meteoritic groundwater source.

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.

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) 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.

We understand that due to the current global situation, the time required for revision may be longer than usual. We would appreciate it if you could keep us informed about an estimated timescale for resubmission, to facilitate our planning. Of course, if you are unable to estimate, we are happy to accommodate necessary extensions nevertheless.

Please do not hesitate to contact me 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,

Olivier Sulpis, PhD
Editorial Board Member

Communications Earth & Environment
0000-0002-6463-3320

Joe Aslin
Senior Editor
Communications Earth & Environment

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REVIEWER COMMENTS:

Reviewer #1 (Remarks to the Author):

Review comments for 'Deep submarine infiltration of altered geothermal groundwater on the south Chilean Margin', Clementi et al. submitted to Nature Communications

I found this paper an interesting read and clearly reports on some exciting data that do seem to show evidence of the presence of meteoric groundwater underneath the seafloor of the Chilean coast.

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Some other comments:

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Generally, I find the use of the term 'geothermal aquifer's in this paper awkward. These are contrasted against meteoric, and glacially recharged aquifers. Ultimately, geothermal aquifers are also meteorically fed. Mixing these different terms either describing a mechanisms to recharge an aquifer (meteoric versus, glacial), versus deep ('geothermal') in a discussion like the one presented in this paper I find confusing. I think you want to distinguish very deep ('geothermal') and shallow aquifers ('meteoric/glacial'). In the literature I suppose this would usually be 'fossil' versus 'modern'.

I take it that, although the authors do not specifically define the term, that a geothermal aquifer is a 'deep' aquifer in which porewaters are relatively warm (e.g. > 50 °C). Normal geothermal gradients would dictate of these aquifers to be 2-3 km, as the authors confirm.

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Reviewer #2 (Remarks to the Author):

This manuscript provides geochemical data from a core from the south Chilean Margin recording pore water freshening. The authors attribute this freshening to the contribution of meteorically modified geothermal groundwater, and identify the Aysén region as the likely source using geochemical fingerprinting. Additionally, the authors calculate an estimated depth for the geothermal reservoir in the Aysén region. The reasoning behind the authors' interpretation of the pore water freshening as submarine groundwater discharge (rather than diagenesis or gas hydrate decomposition) is well-explained and supported by the geochemical data.

Reports of submarine groundwater discharge at these depths are rare, so there is importance in documenting this, as it could influence how geochemical trends in porewaters are interpreted in other locations where submarine groundwater discharge could be occurring in deep environments. The study uses solid research methodology, which is described in enough detail to replicate, and the mixing model appears to be sound.

This manuscript is well-written, clearly presents the data, and provides well-reasoned conclusions. I recommend that this manuscript be accepted, with very minor revisions.

The following minor revisions are suggested:

Line 26: Rather than 'report that' it would be better to write 'argue that' or 'provide evidence that'.

Lines 132-133: This sentence could use a little more context.

Line 147: Change 'appear' to 'appears'.

Line 148: Though the agreement between the pore fluid and modern meteoric isotopic data

supports a modern origin for the fluids, I'm not sure you can say it rules out paleo-meteoric fluids. It would be good to either use less definitive language or elaborate on this.

Reviewer #3 (Remarks to the Author):

In this paper entitled "Deep submarine infiltration of altered geothermal groundwater on the south Chilean Margin", Clementi et al. report pore fluid composition and isotopic signatures to investigate the processes at play from this tectonically active margin. The primary finding of this paper, as stated in the title, is the meteoric groundwater from a site far away from coast. The fluid was modified, to some extent, by the geothermal activity at depth (ca. 3 km), mixed with the seawater, and was further modified by other diagenetic processes (e.g., dolomitization, gas hydrate dissociation, etc) within the depth range (tens of meters) where the sediment cores were recovered. In general, I find the paper well-written, concise, and logical. The data are of high quality and mostly support the conclusions made in the paper. Their finding is surprising (and novel) and is fully supported by the data. They have considered several alternative interpretations but none of them is able to explain their observations. This documentation of the connection between groundwater and deep marine sediment sequence will likely become an importance case study for those who study large-scale groundwater circulation and land-ocean interaction along the active margins.

Despite the merits I see in this work, I have a few critical comments for some of the interpretations made by the author. First of all, I will be cautious about using the term "submarine groundwater discharge", which implies the movement of water. Not sure if this is applicable to the study site. Based on slightly concave-down shape of the Cl profile, it is likely that there is a minor component of fluid upward advection, which may be associated with the high sedimentation (and thus sediment dewatering) and/or fluid advection encouraged by external processes (salinity- or temperature-driven buoyancy, overpressure, etc). A significant advection has to be first confirmed before calling the phenomenon SGD. Otherwise, a more inclusive term, such as the offshore freshened groundwater (OFG) as used by Micallef et al. (2021), may be more appropriate.

Secondly, based on the overlapping of the water isotope data with the local MWL, the authors conclude the emplacement of groundwater is modern. However, I find it hard to believe that the observed meteoric signals in the pore fluids is modern. For example, as we have shown from the Chukchi Sea, it will be difficult to tell the age of the meteoric water based on its relationship with MWL. From Chukchi Sea (Kim et al., 2021, G3, 10.1029/2021GC009750), modern meteoric water has an end member water isotope of -19 and -144 permil (O and H, respectively), while that for LGM is -33 and -258 permil, or roughly 40% lighter for both isotopes. The data reported in this paper could be very well explained by the mixing of meteoric water of either age. The conclusion of the modern emplacement of the observed meteoric groundwater is thus unsupported. If the author insists on the statement of a modern groundwater infiltration, they have to show that there is any modern mechanism that could sustain a hydrology head gradient that is large enough to drive the fresh groundwater (and a less dense one as compared to seawater) into 3 km below seafloor which is supposed to have a pressure ca. 367 times (including the 670 meters water column) higher than the atmospheric pressure (see Burnett et al., 2003 for some review). Knowing the age of offshore groundwater circulation is indeed difficult, but the current consensus is that for such a large-scale circulation, some geological processes must have involved.

Last, the authors claim dolomitization is responsible for some of the changes observed in the pore fluid composition, especially the low Mg concentration. This statement should be supported by some mineral composition data (XRD for example). Low Mg concentrations from locations with

abundance volcanogenic materials could also be interpreted as reflecting secondary smectite formation (for example, see Berg et al. (2019) <https://doi.org/10.1038/s41467-019-12322-2>, for a review of the different processes). Arguably, it is difficult to tell the actual mechanism that is responsible for the depleted Mg in pore fluid but the authors have to consider other alternatives and support their favorable process with more data.

Response to reviews:

We have revised our submitted paper, “Deep submarine infiltration of altered geothermal groundwater on the south Chilean Margin”. Below, we have addressed the general and specific comments made by the reviewers, which largely focus on the proposed mechanism of SGD, fossil versus modern meteoric water as the source of freshening, and potential alternate controls on pore water chemistry. In response to these reviews, we have provided more context for central concepts, expanded text in certain sections, added a new supplementary figure to complement discussion on pore water strontium systematics, and considered alternate controls on pore water chemistry that reviewers proposed. This latter component includes discussion on fossil versus meteoric waters, as well as whether authigenic smectite formation through ash alteration serves as a control on pore water chemistry.

We believe that the changes made substantially improve the manuscript and will be an important advance in our field that hopefully spurs new efforts to better characterize the Chilean Margin system and examine whether this phenomenon operates elsewhere.

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Review comments for ‘Deep submarine infiltration of altered geothermal groundwater on the south Chilean Margin’, Clementi et al. submitted to Nature Communications

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I feel that a diagram outlining such system would help appreciate the specific finding of this study as one piece, an important one, of this hydrogeological puzzle.

We thank the reviewer for their helpful and constructive review. We acknowledge that the manuscript has a large emphasis on the chemical data, and this was intentional. This is partly because there has been little hydrogeologic evaluation of the Aysén region landward of J1003 in contrast to sectors of Chile in the north and far south in Patagonia, which have long been the focus of such studies (e.g., [Duran-Llacer *et al.*, 2020; Taucare *et al.*, 2020]). Indeed, even studies of geothermal springs in the Aysén region (e.g., Negri *et al.* 2018, from which we use terrestrial chemical data, [Daniele *et al.*, 2020], and [Sanchez *et al.*, 2013]) only provide first order assessments of regional hydroclimate and hydrogeology despite the clear importance of these processes in regulating geothermal groundwater chemistry in the region. Similarly, the geothermal reservoir in the Aysén region has not been extensively studied relative to other systems in Chile, though we draw comparisons when possible (see manuscript discussion in the section “Constraints on the geothermal groundwater reservoir and fluid migration”). That said, we have added more context for the offshore system and how high permeability units in the sediment (e.g., sand beds and sandstone recovered at J1003 and nearby ODP Site 860, respectively, permeability anisotropies, and NNW faults extending offshore) might serve as migration pathways for groundwaters to reach the margin.

A key challenge in our study, as raised by all three reviewers, is determining whether the groundwaters are of modern or fossil origin. As stated by Reviewer 3, dating pore waters is difficult. Instead, we undertake an evaluation of the deuterium excess values at J1003 to better constrain when these groundwaters might have been emplaced (see detailed discussion in response to Reviewer 2). We provide evidence that fresh SGD may have initiated during the last glacial period, when a more voluminous Patagonia Ice Sheet and 120 m drop in sea level would have enhanced the hydraulic gradient between land and sea, providing a mechanism to deliver these groundwaters offshore, potentially through the horizontally permeable units in this region.

We also entertain the possibility of fluids being emplaced during the deglaciation. However, SGD during a glacial termination would have to compete with a diminishing hydraulic gradient as global sea levels rose and the PIS retreated and thinned out. Although this is a far less likely scenario, we leave open the possibility that groundwaters could have been emplaced at this time owing to evidence for onset of hydrothermal activity in central Chile, as determined by radiocarbon dating of sinter deposits [Munoz-Saez *et al.*, 2020], suggesting that reinvigoration of geothermal circulation may have occurred on a large scale in the region at this time.

We provide a more detailed discussion on the possible mechanism(s) that could lead to fresh SGD (Lines 236-268).

Some other comments:

Line 61. Local infiltration, far away recharge? Which is it? Infiltration of what into what, and where?

We have clarified this language. Line 62.

Line 72. Geothermal source? This suggest that geothermal processes themselves produce water, at least that is how you could read this.

We have clarified this language, changing “sources” to “endmembers” to emphasize the multi-component mixing that has led to the observed fjord water composition landward of our study site. Line 78.

Line 100. SGD in itself does not need to be fresh, so to just state SGD is a process of freshening is too simplistic.

We agree with the reviewer that SGD, broadly defined, can include a wide range of groundwaters with diverse geochemical compositions. We have clarified this language to emphasize that SGD emplacement of fresh meteoric waters, and not just SGD (broadly defined), could yield low Cl signals in the pore waters. Line 100.

Generally, I find the use of the term ‘geothermal aquifer’ in this paper awkward. These are contrasted against meteoric, and glacially recharged aquifers. Ultimately, geothermal aquifers are also meteorically fed. Mixing these different terms either describing a mechanisms to recharge an aquifer (meteoric versus, glacial), versus deep (‘geothermal’) in a discussion like the one presented in this paper I find confusing. I think you want to distinguish very deep (‘geothermal’) and shallow aquifers (‘meteoric/glacial’). In the literature I suppose this would usually be ‘fossil’ versus ‘modern’.

I take it that, although the authors do not specifically define the term, that a geothermal aquifer is a ‘deep’ aquifer in which porewaters are relatively warm (e.g. > 50 °C). Normal geothermal gradients would dictate of these aquifers to be 2-3 km, as the authors confirm.

We agree that “aquifer” is an inaccurate way to describe the geothermal groundwater source. Typically, aquifer implies a volume of groundwater that is confined by impermeable strata. Instead, we use “geothermal reservoir” or “geothermal groundwater reservoir” throughout the paper, which is consistent with the literature and more accurately portrays these regimes. We also agree with the reviewer that these are deep pools of geothermal groundwater, often found >2km depth, as confirmed by our Na/K- based estimates.

“Geothermal aquifer” was used once in the paper to contrast with the more common glacial/meteoric groundwater aquifers found throughout Chile. We have corrected this to “deep geothermal reservoir” to be more consistent with the literature and remainder of the paper. Lines 52-53.

Map in Figure seems to be to be a sketch map at most. What do the colors indicate? Presumably topography/bathymetry. What is the elevation and depth like? What kind of fault are the lines drawn on the map? What is the geology of the area? A sketch geological map might be useful? There seem to be en-echelon faults on the map? What is the source of this? Is there any other faults? What is the scale bar for this map? A geological cross-section might be useful. In particular for a discussion on the possible hydrogeological mechanisms for the off shore discharge of terrestrial groundwater.

We have revised Figure 1. New are topographic and bathymetric contours at 250 m steps (and a color bar for reference), directional references for the strike-slip motion of the LOFZ, a more detailed Patagonian Ice Sheet from the Hulton et al. [2002] (already referenced in the paper), and a scale bar. Clarification is made in the figure caption to better describe regional features and processes.

We refrained from adding geological reference map as it is not entirely pertinent to our scientific interpretations; this decision was also made to prevent visually overloading the figure. That said, we have added text in the Introduction section more thoroughly discussing the LOFZ and en echelon fractures (Lines 70-77).

Reviewer #2 (Remarks to the Author):

This manuscript provides geochemical data from a core from the south Chilean Margin recording pore water freshening. The authors attribute this freshening to the contribution of meteorically modified geothermal groundwater, and identify the Aysén region as the likely source using geochemical fingerprinting. Additionally, the authors calculate an estimated depth for the geothermal reservoir in the Aysén region. The reasoning behind the authors' interpretation of the pore water freshening as submarine groundwater discharge (rather than diagenesis or gas hydrate decomposition) is well-explained and supported by the geochemical data.

Reports of submarine groundwater discharge at these depths are rare, so there is importance in documenting this, as it could influence how geochemical trends in porewaters are interpreted in other locations where submarine groundwater discharge could be occurring in deep environments. The study uses solid research methodology, which is described in enough detail to replicate, and the mixing model appears to be sound.

This manuscript is well-written, clearly presents the data, and provides well-reasoned conclusions. I recommend that this manuscript be accepted, with very minor revisions.

We thank the reviewer for their feedback, and for recognizing the novelty of our findings and potential applications elsewhere. We are hopeful that this work will encourage others to pursue field work to determine whether this mechanism operates at other active margins, or potentially re-evaluate existing data considering our new findings.

The following minor revisions are suggested:

Line 26: Rather than 'report that' it would be better to write 'argue that' or 'provide evidence that'.

We have changed this language. Line 27.

Lines 132-133: This sentence could use a little more context.

We have expanded the discussion on d-excess in the manuscript. A more thorough response and detail on these changes can be seen in the response pertaining to Line 148 below. See new Lines 140-159 in the manuscript.

Line 147: Change 'appear' to 'appears'.

We thank the reviewer for catching this typo. We have removed this paragraph in revising the d-excess discussion.

Line 148: Though the agreement between the pore fluid and modern meteoric isotopic data supports a modern origin for the fluids, I'm not sure you can say it rules out paleo-meteoric fluids. It would be good to either use less definitive language or elaborate on this.

The reviewer raises an important point here that was also raised by Reviewer 3 and implied by Reviewer 1. The reviewer is correct that where data fall on a $\delta^{18}\text{O}$ versus δD plot is not necessarily indicative of the age. Although we might expect more isotopically depleted pore waters from groundwaters emplaced during the last glacial period (e.g., [van Geldern *et al.*, 2013]), we currently lack endmember estimates for glacial-age waters from this region. However, model studies suggest the $\delta^{18}\text{O}$ values of meteoric water was $\sim 2\text{--}4\text{‰}$ more depleted during the last glacial period [Jasechko, 2016; Jasechko *et al.*, 2015]. To supplement this lack of data, we revisited the pore water deuterium excess (d-excess) data at J1003 assuming that these data reflect the meteoric component of emplaced groundwater. Meteoric d-excess reflects the evaporation conditions of water vapor, providing diagnostic information about a fluid's source region [Dansgaard, 1964]. In the Aysén region of Chile, d-excess largely reflects the temperate climate conditions ([Sanchez-Murillo *et al.*, 2018]) with modern values of around $+7.7 \pm 5.8\text{‰}$ taken at the nearby Puerto Montt IAEA station. At J1003, the d-excess endmember at Cl⁻ equal to zero is $+11.2 \pm 0.8\text{‰}$. This difference requires meteoric isotopic values of -8.3‰ for $\delta^{18}\text{O}$ and -55.3‰ for δD (for reference, modern meteoric $\delta^{18}\text{O}$ and δD at Puerto Montt - $5.5 \pm 2.7\text{‰}$ and $-36.6 \pm 18.4\text{‰}$, respectively). The inferred 2.8‰ change in $\delta^{18}\text{O}$ is in strong agreement with model estimates for this region, suggesting that the freshening signal at J1003 could be the result of groundwater emplaced during the last glacial period. This would be consistent with widely documented pore water SGD signals in margin sediments along the U.S. Atlantic margin [Person *et al.*, 2003], Peru [Kriete *et al.*, 2004], and Svalbard [W L Hong *et al.*, 2019] from enhanced hydraulic gradients during the last glacial period, when more voluminous

ice sheets (the PIS was ~100x more voluminous than present) and global sea level ~120 m lower than modern.

We have expanded the discussion in the manuscript to include these lines of evidence (Lines 140-159), as well as adding text to detail potential migration pathways for altered geothermal groundwater to reach the Chilean Margin sediments (Lines 236-268).

Reviewer #3 (Remarks to the Author):

In this paper entitled “Deep submarine infiltration of altered geothermal groundwater on the south Chilean Margin”, Clementi et al. report pore fluid composition and isotopic signatures to investigate the processes at play from this tectonically active margin. The primary finding of this paper, as stated in the title, is the meteoric groundwater from a site far away from coast. The fluid was modified, to some extent, by the geothermal activity at depth (ca. 3 km), mixed with the seawater, and was further modified by other diagenetic processes (e.g., dolomitization, gas hydrate dissociation, etc) within the depth range (tens of meters) where the sediment cores were recovered. In general, I find the paper well-written, concise, and logical. The data are of high quality and mostly support the conclusions made in the paper. Their finding is surprising (and novel) and is fully supported by the data. They have considered several alternative interpretations but none of them is able to explain their observations. This documentation of the connection between groundwater and deep marine sediment sequence will likely become an importance case study for those who study large-scale groundwater circulation and land-ocean interaction along the active margins.

Despite the merits I see in this work, I have a few critical comments for some of the interpretations made by the author. First of all, I will be cautious about using the term “submarine groundwater discharge”, which implies the movement of water. Not sure if this is applicable to the study site. Based on slightly concave-down shape of the CI profile, it is likely that there is a minor component of fluid upward advection, which may be associated with the high sedimentation (and thus sediment dewatering) and/or fluid advection encouraged by external processes (salinity- or temperature-driven buoyancy, overpressure, etc). A significant advection has to be first confirmed before calling the phenomenon SGD. Otherwise, a more inclusive term, such as the offshore freshened groundwater (OFG) as used by Micallef et al. (2021), may be more appropriate.

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groundwater is thus unsupported. If the author insists on the statement of a modern groundwater infiltration, they have to show that there is any modern mechanism that could sustain a hydrology head gradient that is large enough to drive the fresh groundwater (and a less dense one as compared to seawater) into 3 km below seafloor which is supposed to have a pressure ca. 367 times (including the 670 meters water column) higher than the atmospheric pressure (see Burnett et al., 2003 for some review). Knowing the age of offshore groundwater circulation is indeed difficult, but the current consensus is that for such a large-scale circulation, some geological processes must have involved.

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(2019) <https://doi.org/10.1038/s41467-019-12322-2>, for a review of the different processes). Arguably, it is difficult to tell the actual mechanism that is responsible for the depleted Mg in pore fluid but the authors have to consider other alternatives and support their favorable process with more data.

We thank the reviewer for their extremely thorough and constructive review, and appreciate their recognition that this study and our findings are novel with broad implications. Several critical points have been made and we summarize them as follows: 1) terminology, 2) paleo versus modern meteoric water, and 3) influences on pore water Mg concentrations. Briefly, we have addressed each of these points in our response and through revisions to the manuscript. Detailed responses are provided below.

Terminology: SGD or OFG?

We thank the reviewer for raising this point. Admittedly, we considered whether to describe the discussed phenomenon as SGD or some other process, including OFG. SGD has been widely used in the literature where pore waters have been used to reveal infiltration of meteoric waters offshore. For some, but not all, offshore SGD instances, groundwater movement has been directly measured through radioisotopes such as radium (e.g., [Burnett et al., 2006]) or use of trace metal isotopes like boron (e.g., [W L Hong et al., 2019]). In many cases, however, it is inferred based on different factors (i.e., location of the source waters, distance of the observation from shore, etc.). At our study site, movement of water from the land source to marine sediments must be occurring to attain the documented geochemical signatures, perhaps most strongly evidenced by the robust linear mixing models shown in Fig. 4.

Site J1003 is located 50 km offshore with horizontally permeable pathways nearby (e.g., NNW lineaments, coarse-grained strata, permeability anisotropies), and our robust constraints on source fluids suggest that the same waters influencing J1003 are also influencing geothermal springs at the land surface in Patagonia. Characterizations of geothermal springs in central Chile indicate that the fluid reservoir is often located directly below the thermal spring cluster and that the spatial extent of springs is a function of lateral groundwater flow through fault systems

in the region [*Daniele et al.*, 2020; *Sanchez et al.*, 2013]. Although detailed seismic characterizations are absent in our study region, these previous studies paint a clear and consistent picture for geothermal groundwater movement throughout Chile. It stands to reason that geothermal groundwaters are moving through marine sediments in this region.

OFG, on the other hand, is an intentionally broad term meant to incorporate freshened offshore fluids from gas hydrate dissociation, diagenetic processes, subglacial injection, paleo meteoric recharge, and active recharge [*Micallef et al.*, 2021]. Although the observations of low-Cl pore fluids at J1003 would certainly fall under this umbrella, we believe that it falls short of capturing the mechanism at play here. For the above reasons, we are confident in our use of SGD to describe the documented phenomenon.

Paleo versus modern meteoric water contributions

This was a point also raised by Reviewer 2. As described above, we have conducted a thorough analysis of deuterium excess values in pore water to further constrain whether emplaced meteoric fluids are of modern or of fossil origin. The endmember d-excess values at J1003 are higher than modern values, which suggest that the meteoric component of infiltrating geothermal groundwaters could have originated in a colder climate since enhanced Rayleigh distillation yields more isotopically depleted O/H values and higher d-excess values. The 2.8‰ difference in $\delta^{18}\text{O}$ required to yield observed d-excess values at Cl=0 is consistent with model-based reconstructions of the change in regional meteoric $\delta^{18}\text{O}$ between the last glacial period and today [*Jasechko*, 2016]. This provides strong evidence that freshened fluids at J1003 may have been emplaced during the last glacial period or shortly after.

We have expanded the discussion in the manuscript to include these lines of evidence (Lines 140-159) and added text to detail potential migration pathways for altered geothermal groundwater to reach the Chilean Margin sediments (Lines 236-268).

Controls on Mg concentrations in pore water

We agree with the reviewer that the pore water system at J1003 is complex, likely with more than one competing influence on pore water chemistry. The non-linear decreases in Mg^{2+} , Ca^{2+} , and Sr^{2+} concentrations follow a similar pattern and suggest a common control. Authigenic carbonate nodules ~5 cm in diameter were found in abundance between 15-40 mbsf. Although shipboard XRD was not completed on these specific nodules, XRD analysis of authigenic carbonates at nearby sites on the margin confirm a dolomitic composition (e.g., Site J1006; [*Bova et al.*, 2019]). The downcore changes in pore water Mg/Ca, Sr/Ca, Mg/Cl, Sr/Cl, and Ca/Cl at J1003 mirror the profiles at sites where dolomites were confirmed: Mg/Ca and Sr/Ca maxima in the core of the dolomite-rich interval, coincident with decreases in pore water Mg, Ca, and Sr concentrations (as evidenced by the ratios normalized to Cl). This paints a consistent picture on the interactions between pore water chemistry and authigenic carbonate formation in this region.

Kastner et al. [1990] outlined a comprehensive analysis for interpreting these IW signals in Peru Margin sediments in their Table 13, which guided our interpretations of pore water profiles at

J1003 in the absence of XRD analysis of the recovered authigenic carbonates. Calcite precipitation ($\text{Ca}^{2+} + 2\text{HCO}_3^- \rightarrow \text{CaCO}_3 + \text{CO}_2 + \text{H}_2\text{O}$) increases Mg/Ca and Sr/Ca ratios in pore water owing to the uptake of Ca^{2+} . Likewise, it also decreases the Ca (and Sr) concentrations from the same pathway. However, Mg concentrations (and Mg/Ca) are unchanged since Mg is not incorporated into the calcite lattice during precipitation. In contrast, dolomitization reactions are what lead to marked changes in Mg concentration in porewater provided sufficient conditions (e.g., alkalinity, low sulfate, adequate magnesium supply [*Baker and Kastner, 1981; Baker and Burns, 1985*]). Dolomitization can occur with or without carbonate precursors and can follow different stoichiometric models, as outlined by Morrow [1982]:

Eq. 1: $2\text{CaCO}_3 + \text{Mg}^{2+} \rightarrow \text{CaMg}(\text{CO}_3)_2 + \text{Ca}^{2+}$. This would decrease pore water Mg/Ca ratios and increase Ca/Cl ratios, neither of which are documented at J1003.

Eq. 2: $\text{CaCO}_3 + \text{Mg}^{2+} + 2\text{HCO}_3^- \rightarrow \text{CaMg}(\text{CO}_3)_2 + 2\text{CO}_2 + 2\text{H}_2\text{O}$. Here, pore water Mg/Ca and Sr/Ca ratios and Mg concentration would decrease, whereas Ca and Sr concentrations would increase. Instead, pore water Mg/Ca and Sr/Ca ratios increase in the authigenic carbonate-rich interval at J1003 and Ca and Sr concentrations decrease with depth.

Eq. 3: $2\text{CaCO}_3 + \text{Mg}^{2+} + \text{SO}_4^{2-} \rightarrow \text{CaMg}(\text{CO}_3)_2 + \text{CaSO}_4$. Again, Mg/Ca ratios and Mg concentrations would decrease, whereas Sr concentrations would increase. None of these features are present at J1003. Similarly, neither anhydrite (CaSO_4) nor gypsum ($\text{CaSO}_4 \cdot \text{H}_2\text{O}$) were documented during shipboard core description and SO_4^{2-} is depleted by 10 mbsf, which is shallower than depths at which authigenic carbonates were first documented.

Eq. 4: $\text{Ca}^{2+} + \text{Mg}^{2+} + 4\text{HCO}_3^- \rightarrow \text{CaMg}(\text{CO}_3)_2 + 2\text{CO}_2 + 2\text{H}_2\text{O}$. In the non-carbonate precursor scenario shown here, pore water Mg/Ca and Sr/Ca ratios increase substantially, whereas Ca, Sr, and Mg concentrations decrease. This is the only authigenic carbonate precipitation model that explains observed pore water profiles at J1003. As we suggest in the paper, organic matter diagenesis increases alkalinity and ammonium concentrations in the pore space. The alkalinity likely induces authigenic carbonate precipitation, and the substitution of NH_4^+ with Mg^{2+} adsorbed onto silicate clay minerals may liberate the Mg needed for direct dolomite precipitation [*Vonbreyman and Suess, 1988*].

Dolomite precipitation is certainly not the only control on pore water Mg concentrations globally, and the reviewer correctly points out that ash diagenesis and the associated formation of authigenic clays (e.g., smectite) is often one dominant control in hemipelagic continental margins sediments [*Gieskes and Lawrence, 1981*]. However, ash diagenesis (and associated smectite formation) releases Ca^{2+} (and Sr^{2+}) to the pore space while consuming Mg^{2+} . The lack of increases in downcore Ca^{2+} and Sr^{2+} concentration contrasts with this expectation. Nevertheless, we consider strontium systematics in the paper to assess secondary influences on the pore water data since even minor ash diagenesis can impart large influence on the strontium isotopic composition of pore water (e.g., [*W-L Hong et al., 2020*]). A new comparison between $^{87}\text{Sr}/^{86}\text{Sr}$ and inverse strontium concentration ($1/\text{Sr}^{2+}$) provides additional support for our assertion that authigenic carbonate formation exerts some control on IW chemistry, as

evidenced by the uptake of Sr^{2+} with only minor changes in $^{87}\text{Sr}/^{86}\text{Sr}$ (see new Supplemental Fig. 4). And as we suggest with the $^{87}\text{Sr}/^{86}\text{Sr}$ vs. $\delta^{18}\text{O}$ mixing model, downcore $^{87}\text{Sr}/^{86}\text{Sr}$ appears to be, at least in part, regulated by influences from non-radiogenic geothermal groundwater. This is not to suggest that ash diagenesis is not occurring in these sediments, but shipboard lithologic description indicates that ash and smectite are both minor components of the bulk sediment composition [Bova *et al.*, 2019], and that influence from alteration imparts a only minor influence on IW chemistry.

Collectively, we are confident that the pore water Mg, Ca, and Sr profiles reflect a common influence from dolomite precipitation that leads to the observed non-linear concentration decreases with depth. We have expanded the discussion to incorporate these caveats (Lines 186-212), revised Supplemental Fig. 3 to include Ca/Cl, Sr/Cl, and Mg/Cl ratios, and made a new Supplemental Figure 4 showing $^{87}\text{Sr}/^{86}\text{Sr}$ versus $1/\text{Sr}^{2+}$.

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21st Jun 22

Dear Mr Clementi,

Your manuscript titled "Deep submarine infiltration of altered geothermal groundwater on the south Chilean Margin" has now been seen by our reviewers, whose comments appear below. In light of their advice I am delighted to say that we are happy, in principle, to publish a suitably revised version in Communications Earth & Environment under the open access CC BY license (Creative Commons Attribution v4.0 International License).

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REVIEWERS' COMMENTS:

Reviewer #1 (Remarks to the Author):

I have reread this revised manuscript and also considered the extensive reviewer rebuttal.

I am impressed how much this manuscript has improved from the hydrogeological viewpoint. A plausible hydrogeological mechanism and discussion is now presented, and this is a substantial improvement over the earlier version of this manuscript. Also, the map in figure 1 is now up to standards, well done.

I would not see a particular reason to object to publication of this manuscript at this point.

Reviewer #3 (Remarks to the Author):

The new version of the paper reads very well now. I appreciate the authors take my suggestions into their consideration. The additional evidence from deuterium-excess really makes the conclusion, SGD during LGM, much stronger now. I also appreciate the authors showing the presence of dolomite in the sediments. Though I am still not 100% convinced about the formation of dolomite, the new evidence presented by the authors makes this a strong case that I cannot argue further. In short, my comments in the first round of review were satisfactorily addressed. I now only have three very minor technical comments:

Line 49: "high discharge rate"...do the authors mean fluid discharge? The unit also reads weird to me or maybe I misunderstood something. Could you clarify a bit?

Line81: replacing “that supply” with “supplying” to avoid repetition in using the word “that” twice.

Line128: Just out of curiosity, ss there any visual observation from the sediment, eg, soupy sediments, to support this inference?

Response to reviews:

We are delighted to hear that our resubmitted manuscript successfully addressed the main points from the three reviewers. Here, we have made final edits to our manuscript, “Deep submarine infiltration of altered geothermal groundwater on the south Chilean Margin”, based on the comments from Reviewer 2. We sincerely appreciate their helpful and constructive feedback on our paper from all three reviewers and believe that it is much stronger than the original version.

REVIEWER COMMENTS:

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I have reread this revised manuscript and also considered the extensive reviewer rebuttal.

I am impressed how much this manuscript has improved from the hydrogeological viewpoint. A plausible hydrogeological mechanism and discussion is now presented, and this is a substantial improvement over the earlier version of this manuscript. Also, the map in figure 1 is now up to standards, well done.

I would not see a particular reason to object to publication of this manuscript at this point.

We thank the Reviewer for their comments and are glad that the revised manuscript has addressed their helpful comments from the initial submission.

Reviewer #3 (Remarks to the Author):

The new version of the paper reads very well now. I appreciate the authors take my suggestions into their consideration. The additional evidence from deuterium-excess really makes the conclusion, SGD during LGM, much stronger now. I also appreciate the authors showing the presence of dolomite in the sediments.

We are grateful for the Reviewer’s critical feedback. We were happy that the deuterium excess exercise provided constraints on potential timing of SGD in this region and thank them for encouraging us to pursue this.

Though I am still not 100% convinced about the formation of dolomite, the new evidence presented by the authors makes this a strong case that I cannot argue further. In short, my comments in the first round of review were satisfactorily addressed. I now only have three very minor technical comments:

Line 49: “high discharge rate”...do the authors mean fluid discharge? The unit also reads weird to me or maybe I misunderstood something. Could you clarify a bit?

We have clarified the language in this sentence to say “high fluid discharge rates” (Line 49). The units used here are adapted from the two references cited (Luijendijk et al. 2020 and Zhou et al. 2019), which provided global estimates of SGD in $\text{m}^2 \text{yr}^{-1}$. Although other studies have used $(\text{cm}) \text{yr}^{-1}$ and $\text{m}^3 \text{yr}^{-1}$, we opted to use the same units as the referenced studies since they are the only ones that provide any estimate of SGD in our study region, albeit a model-based estimate.

Line81: replacing “that supply” with “supplying” to avoid repetition in using the word “that” twice.

Corrected.

Line128: Just out of curiosity, ss there any visual observation from the sediment, eg, soupy sediments, to support this inference?

There is apparent evidence for this assertion in the form of soupy sediments (e.g., J1003A-5H-1, ~36 mbsf) within the interval with higher pore water $\delta^{18}\text{O}$ and δD values.