Rebuttal

Anonymous Referee #1

We thank the reviewer for the constructive feedback and many valuable comments. As we describe below, the comments have helped us to significantly improve both the paper and our numerical model in general. Here we provide a very short, general answer to the comments, and provide an additional document with detailed answers to each comment raised by the reviewer. As the reviewer's comments mostly did not comprise line-by-line comments, but rather comments on sections and general procedure, we refer to the revised sections as a whole or to subparagraphs within them.

The key criticism was our choice of host rock permeability for the models, which the reviewer found not representative (too high) of the field study area, as the main host rock is shale. This criticism was fair and correct. As requested, we provide additional runs with much lower host rock permeability (~10-18 m2 at 3 km depth), that better resemble Neuquén basin conditions. This, however, necessitated a change in the hydrofracturing implementation to include transient fracture porosity. This is essential to get realistic results in terms of fluid pressure, and in our view the numerical model has thereby improved drastically and delivers plausible results for low-permeability host rocks. In the revised manuscript, these results are discussed in detail in addition to the medium/high-permeability cases, which we also re-calculated to ensure comparability. As requested by the reviewer, we also show more parameters in the figures (particularly pore fluid pressure and permeability-porosity plots) to give a better foundation for our process interpretations.

To limit the overall length of our contribution, we deleted the parts about the depth-sill thickness-parameter study and focused instead on the role of host rock and sill permeability for a single sill and depth that represents the conditions of our study area.

Additionally, we also accepted most of the smaller suggestions in terms of language and descriptions (those provided in the annotated pdf). We try to use a much more specific, quantitative language throughout the manuscript, which was missing as the reviewer rightfully pointed out.

For answers to the more specific questions raised, please refer to the table-style documented that we provide, with reviewer comments on the left, and author responses on the right (references to sections and subparagraphs refer to the revised manuscript).

Note: Reviewer's comments are grey, responses in black.

1) Permeability model of the host rock

I am not convinced that the described porosity-dependent permeability model for the host rock is reasonable to represent a low-permeability shale. Shales are commonly described as a low-permeability rock; e.g., Goral et al., 2020 (doi.org/10.1038/s41598-019-56885-y) state a maximum permeability typically less than 1000 nD (~9.8E-19 m2), which would not allow for significant advection (e.g., Ingebritsen et al., 2010; doi.org/10.1029/2009RG000287).

If there is no field evidence for these highly permeable shales in the Neuquén Basin,the presented models are not appropriate to interpret the igneous petroleum system and hydrocarbon transport in the Río Grande Valley. In that case, I would like to request the authors to either (1) refer to the host rock as a more permeable rock type (e.g., sandstone) that matches a permeability of ~1E-16 – 1E-14 m2, or (2) to test/proof if simulations with host rock permeabilities of < ~1E-18 m2 would give similar results as presented and discussed in this contribution.

In the prior case, the changed host rock type should be considered in the discussion. E.g., What are the effects on organic matter transformation to methane compared to shale? Due to the change in host rock, the presented flow simulations would further not represent the field locality such that an interpretation of the implications for igneous petroleum systems in the Río Grande Valley may not be feasible.

We agree with the reviewer and accept this criticism. Therefore, we provide two new runs (with impermeable and permeable sill) with a significantly lower host rock permeability. The values are at ~10^{-18} m^2 at sill emplacement depth. We add this to our simulation results (section 3.4.2) and discussion (section 4.2), which we revised overall to incorporate all comments. Thus, our contribution now provides a comparison between high and low permeability host rocks.

The overall results and conclusions remain largely similar, with some added complexity for the low-permeability case. This is because extensive hydrofracturing compensates much of the reduced permeability and thereby facilitates flow. For even lower permeabilities, this compensation effect should be even stronger. As the shales in the study area show extensive networks of (hydrothermal) veins, we think that the new runs show quite plausible behaviour. The hydrofracturing process itself adds a new layer of complexity, which necessitates a detailed description. Please refer to the overhauled result and discussion sections.

The new results allow us to maintain most of our interpretation for low-permeability systems such as the Neuquén case, while also providing insight into systems with higher or intermediate permeability (results from original manuscript).

We did not change the organic matter content in the host rock in order to limit the number of parameters that are changed between the scenarios. However, we do see a small effect on overall generated methane for the two host rocks, mainly due to stronger heat advection in the high-permeability host.

We thank the reviewer for this comment and the valuable resource, especially as host permeability is also discussed. In fact, a brittle-ductile transition for the host rock ranging from 500-750°C is implemented in the model but was not specifically mentioned in section 3. Note that except for the three modifications (only tensile failure mode, hydrofracturing affecting permeability and porosity, cooling joint formation) our model is identical to the one presented by Galerne and Hasenclever (2019). We have clarified this in section 3.

Due to the thin sill and relatively low emplacement temperature, the effects are very limited in time and space, only 5-10 m of the aureole reach >500°C and only for a few years. Maximum temperature at the contact is ca. 670°C, such that full ductile permeability reduction is never reached. Of course, if the BDT were to start at lower temperature, as suggested in paper mentioned by the reviewer, the impact of this could be more pronounced and could be tested. We may add that in the study area, the evidence of ductile behavior in the high-temperature aureole is limited to the first few decimeters of the contact (e.g., Rabbel et al. 2021).

2) Permeability model of the intrusion

Yes, this is how it is implemented in our model: We parameterize the cooling joint formation as a linear transition between 1000 ℃ (zero porosity, zero permeability) and 900 ℃ (8% porosity, 1E-15 m^2). There is, however, a significant difference between our parameterization of cooling joint formation and the one by Iyer et al. (2013), which we now reference: We not only increase permeability but also porosity, which is more realistic and affects pore pressure evolution as well as methane transport.

We added a sketch to illustrate the simple linear permeability model employed (Figure 5c). We also revised the description of the model (section 3.2 "Adjustments for this study").

See answer above, we added a graph for clarification (Figure 5c).

This is a valid comment, the justification of the permeability choice is not clear enough and has been updated in the revised manuscript (section 3.3, third paragraph). We chose Spacapan as the local reference, because it is the only real-world data we have. Spacapan's maximum value as stated by the reviewer is 5E-15, so our 1E-15 are in fact a bit below (not above) Spacapan's (2020) maximum values. We chose 1E-15 as the permeability to be at the upper end of Spacapan's values to make the sill a permeable medium comparable to the local producing sills. However, this likely still comes short of the real values (which we simply do not know), but they are probably higher still, because the values derived by Spacapan are core measurements. Macroscopic cooling joints or hydrofractures will provide much more efficient flow paths – but explicit fracture flow is not possible in the already complex hydrothermal model. Thus, we believe that 1E-15 is a sufficiently high value to term the sills "permeable".

The manuscript explores exactly this difference by comparing the permeable sills to non/very low-permeable sills, where we set permeability to 1E-20, so essentially 0, as a lower boundary. As shown in by the reference runs (Figure 6 and 7), fluid overpressure cannot be released through the impermeable sills upwards in such a case, and the fluids would need to take the route via the sill tip. This is the "standard" assumption in most sill modelling work and therefore the reference.

The value of 1E-16m^2 is the limit below which heat advection vanishes and heat conduction dominates for bottom-heated magmatic hydrothermal systems. In such systems hydrothermal circulation is solely driven by the buoyancy differences between cold recharge flow and hot hydrothermal upflow. In the case of sill emplacement in sedimentary rocks, there are additional forces driving fluid flow: clay mineral dehydration and organic matter transformation release fluid mass into the pore space, leading to much higher pore pressures compared to a buoyancy driven hydrothermal convection system. As a result of these strong pressure gradients, significant fluid flow also occurs at permeabilities lower than 1E-16 m^2. If hydrofracturing is considered, the fluid flow is further enhanced even in rocks with permeabilities as low as 1E-18 m^2.

3) Model description

The authors refer to Galerne and Hasenclever (2019) when describing the model. However, a more detailed description of the model including the governing equations would be beneficial, given that this is the focus of the manuscript. This section could also be provided as appendix.

We understand the desire to present the model in more detail. Yet, since we use the identical model with the same parameters – except for the changes that we describe in section 3.2 – providing the entire model description in our study again would mean to copy & paste several pages of (Galerne and Hasenclever, 2019) into an appendix. We believe this is not necessary, because the study by Galerne and Hasenclever (2019) is a free-access publication in a well-known international journal. We have extended the description of the modifications made for this study.

In addition, simplifications and assumptions of the model setup could be introduced and justified here (Section 3.1). The model considers a single-phase flow of a compressible fluid following Darcy's law (L 251-252). From Galerne and Hasenclever (2019), I learned: "Throughout the calculations, pore pressures are above the critical point of pure water so that the fluid remains in a single-phase state. Our single-phase hydrothermal model requires this assumption, because in the system H2O–NaCl–CO2–CH4 phase transitions would be possible even at higher pressures.". This assumption should be included and explained in the presented manuscript, and potential effects on the modelling results should be considered and discussed later in the manuscript. Given the shallow emplacement depth of 1-3 km, pore fluid boiling is plausible. Would this phase transition change fluid flow patterns and the transport and accumulation of the hydrocarbons?

We now only consider deeper sills at 3 km depth because they are more representative for our case study. Whether phase transitions could occur near the sill depends on the salinity of the fluid. Pure water would be above its critical point hence would remain in the super-critical single-phase state. Saltwater, on the other hand, could potentially undergo phase transitions. However, we find strong overpressures developing in the contact aureole (up to ~90 MPa, where

cold hydrostatic pressure is 30 MPa), so that even salt water may remain in the high-P, high-T single-phase region. Assuming a typical seawater salinity of 3.2 wt.% NaCl, the phase diagram below (calculated using the equation-of-state by Driesner & Heinrich, 2007) shows that boiling at 30 MPa starts at ~410 ℃, while at 90 MPa it starts at ~640 ℃. But all this is speculative, because salinities will change once phase transition and phase separation occur, and a multi-phase hydrothermal model would be required to further elaborate on this question, which is beyond the scope of our study.
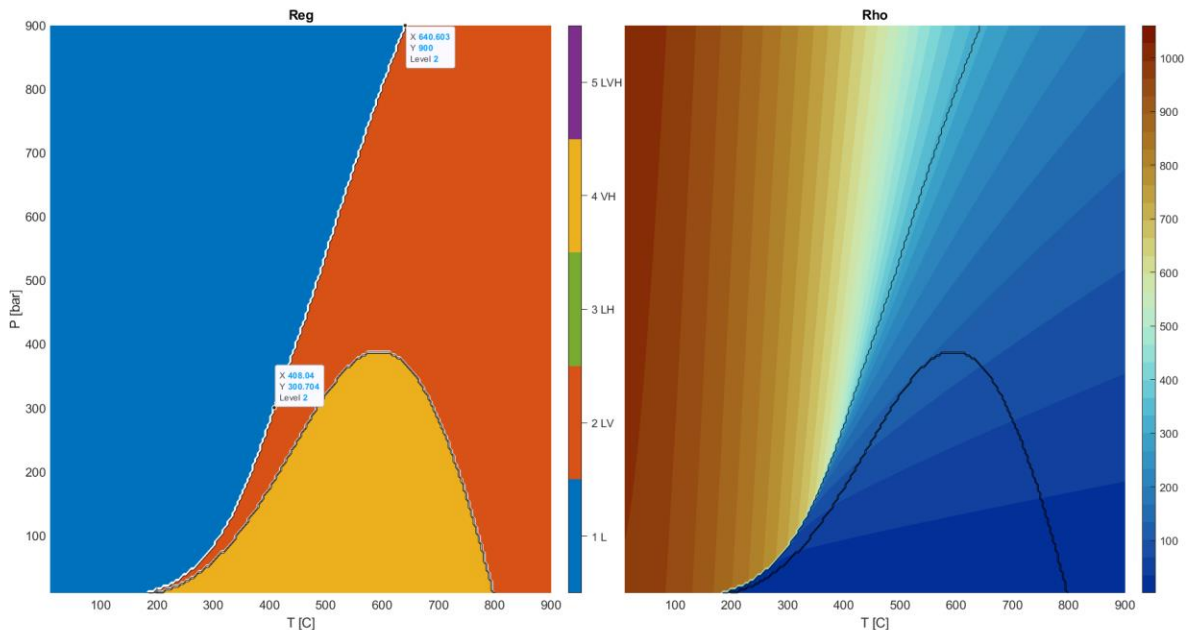

Figure 1: Phase regions (left) and bulk density (right) of seawater (H_2O with 3.2 wt.% NaCl) based on the equation-of-state by Driesner & Heinrich, (2007). Phase regions are: L – single-phase, LV – liquid+vapor coexistence, LH – liquid+halite coexistence, VH – vapor+halite coexistence, LVH – liquid+vapor+halite coexistence. The phase boundary L/LV is pressure dependent and would change by several hundred degrees during the pore pressure build-up after sill emplacement.

Buoyancy effects caused by methane in the fluid are not considered in the models (L 262-264). Is this because the effects are too minor to significantly affect fluid flow or would buoyancy change the modelling results?

We have not tested whether additional buoyancy effects of methane may change our modelling results. Although the lack of buoyancy effects certainly represents a model limitation, we point out that Iyer et al. (2013) found no significant changes in methane discharge in their model comparison with either weak or strong CH_4-related density reductions. A reliable equation-of-state for H2O–NaCl–CO2–CH4 is missing and prescribing an ad hoc density change depending on CH_4 concentrations bears the danger of oversimplifying the actual fluid density evolution. We

therefore decided to not implement supposed density changes but to use the equation-of-state of pure water (cf. Galerne & Hasenclever, 2019)

We improved figure 5 and provide the information in the text.

We now show the mesh resolution in figure 5.

4) Results and Figures

We thank the reviewer for this comment, the pore fluid pressure (distribution) is certainly at the heart of the process interpretation. We changed the displayed data in the Figures 6,7,8,9 (half-sill view for low/high-permeability hosts with impermeable/permeable sills). The updated panels are as follows:

1. Row: Temperature and maturity
2. Pore pressure and flow vectors
3. Permeability and transient fracture porosity (to indicate distribution and intensity of hydrofracturing)
4. Methane distribution as Fluid fraction with flow vectors

The new close-up figure 10 displays pore fluid pressure and permeability/fracture porosity in the same style. We also adjusted the colorbars according to the recommendations made by the reviewer in the annotated pdf, i.e., we use fewer color levels and provide contour labels where we deemed it useful.

The updated figures are also referred to in the updated results (sections 3.4.1, 3.4.2) and discussion (section 4).

I would also like to invite the authors to provide videos of a representative simulation of both a permeable and impermeable sill as supplemental material. These videos could visualise fluid pressure, temperature, CH4 concentration, and velocity vectors, which would allow the reader to see how the fluid flow evolves over time and how the described phases of fluid flow form.

This is a good suggestion; several videos are provided as supplements and are referenced in the text. This gives the reader a much better idea of the dynamics of the phases over time and space.

5) Discussion

This section is a good summary of the distinct fluid flow phases observed in the models, but it misses some explanation on which mechanisms or physical parameters control each phase. For example, it is not clear, which mechanism causes a contact-parallel flow in Phase 1. In Section 4.4, the authors suggest that impermeable sills favour fluid pressure build-up and contact-parallel flow toward the sill tips. This explanation should be extended and data to support this interpretation should be included in the manuscript.

We hope that the added data (see above response) and improved description in the result (section 3.4.1) and discussion sections make the interpretation clearer. We would like to add that pore pressure build-up around sills (particularly those emplaced in organic-rich sedimentary rocks), is a well-documented phenomenon (e.g., Aarnes et al., 2012, Iyer et al. 2013, 2017, Galerne and Hasenclever 2019). In the lower aureole, the fluids cannot escape upwards as long as the sill remains (partly) impermeable. This leads to flow towards the edges of the sill. This contact-parallel flow of fluids from below the sill toward the tips is at the center of the theory for formation of (explosive) hydrothermal vents, so our interpretation of Phase 1 is more a repetition of previous work (e.g., Iyer et al., 2017).

In Phase 2, pore fluid pressure (overpressure and rapid pressure dissipation after the formation of cooling joints) is used to explain the upward-directed fluid flow and hydrothermal "flushing". As mentioned above, I would like to request the authors to provide data to support this interpretation.

See replies above. We acknowledge the need for this data to support our interpretation and now provide data on pore fluid pressure in both overview (half-sill view) and close-up figures (6-10).

We extended the discussion of the vortex slightly, but want to avoid too much focus on this phenomenon, as it is not clear how pronounced it would be in a more realistic scenario, e.g., with lithology variations. In section 4.2, we therefore also renamed the third phase as "stabilization and fading hydrothermal flow" to put less emphasis on the vortex feature.

The focus of this work remains to highlight the possibility for, and potential importance of, permeable pathways through intrusions shortly after solidification. Once our model includes further complexities (e.g., multiphase flow, mineral precipitation, BDT, or realistic geology), it would be interesting to investigate if this feature persists

Discussion - Impact of permeable sills on hydrothermal flow

Again, we thank the reviewer for this comment, and realized that another simulation with lower permeability was necessary to understand the implications of permeable sills on hydrothermal flow. Figures 8 and 10 show the flow patterns for a low-permeability host with a permeable intrusion, and the flow phases are described in section 3.4.2 and discussed in section 4.2.
For a permeable sill in a low-permeable host rock (<< 1E-16, see Figure 5b, 8, 10), flow phases 1 and 2 are quite similar to the higher-permeable host rock, while more complex due to hydrofracturing. A key observation is that there is still fluid flow in a lower permeability host, because hydraulic fracturing is more pronounced and elevates permeability (and porosity). In our model, the permeability increase has a limit to 2 orders of magnitude above reference permeability, somewhat limiting the flow overall. Nevertheless, this shows that hydrofracturing around sills can generate create sufficient permeability even in low permeable rocks like shale. In fact, the outcrops of sills emplaced in shale which we visited in Neuquén show networks of hydrothermal veins in the shale (possibly hydrothermal, compare Spacapan et al., 2019, and Rabbel et al., 2021). One might speculate that at least part of these veins are remnants of this process.

For much lower sill permeabilities refer to the comments above – we present a case for an impermeable sill in Figure 6. Here, we do not see the same flow phases, and the main difference at lower host rock permeability is that hydraulic fracturing is sustained for longer, which in turns increases permeability. Iyer et al (2017) investigated such a case for a large-scale sill complex offshore Norway.

Discussion - Implications for igneous petroleum systems in the Río Grande Valley

Based on the permeabilities used by the authors, I am not convinced that it is feasible to discuss implications for the described field location (please see comments 1 and 2).

Again, this is a valid point and the authors are grateful to take this opportunity to provide new simulations to facilitate the interpretation with respect to Rio Grande Valley. In our view, the new simulations provide scenarios that are suitable to discuss implications for the sills of Rio Grande Valley. Overall, we reduced the discussion for Rio Grande Valley (section 4.4) to the impact of hydrocarbon "flushing", as we believe that this is the main point worth discussing in this context.

Further comments from annotated pdf.

Here, we only reply to comments that are not covered by the ones presented above.

We also exclude smaller corrects regarding typos, or more precise language. We accepted all those comments and corrected them. Line references refer to the original manuscript.

L.54:

"Hydrothermal flow in response to the intrusion of magma into sedimentary host rocks has been investigated for decades." Please provide references

This is just the introductory statement, and the following sentences contain >12 references spanning over 40 years of research. We think that adding the references here is unnecessary repetition. However, we did add Ingebrigtsen et al. (2010) in this paragraph.

Section 2.2 could be shortened and focused on methods and data used in this study.

We shortened section 2.2 and deleted 2 unnecessary sentences.

L. 179: Given this texture is used to interpret graphitisation, I would like to encourage the authors to provide a photograph and an explanation why this fabric suggests graphitisation of the bitumen. Please also include references to support this interpretation.

This was an unfortunate formulation from our side, as the texture itself merely made us hypothesize about thermal grade. We now write:

"The fibrous texture of the observed bitumen within the sills is intriguing and we hypothesized that it may be much higher-grade."

L. 203: Please indicate the sample locations. Were these veins cooling joints or overprinting veins? Do they crosscut the whole intrusion? How thick was the intrusion sampled? The thickness could be of interest when discussing fluid temperatures within the intrusions and the identified flow phases.

We specify: "The D3 band between the peaks is nearly absent in the sample from Sierra de la Cara Cura (from vein in Figure 3b), while it is visible at low intensity in the presented sample from El Manzano (from vein in Figure 3a). Note that both vein samples stem from the intrusion-host contact, and each veins penetrate about 10 m into around 20 m thick sills."

L. 204:
Please indicate the sample location each ratio relates to.
Figure 4 suggests that a ratio of 0.9 describes the sample at Cara Cura (?)

Sample location, see above response.
Correct, there was an error in the description. The correct ratios are 0.9-1, we changed the description.

L. 571 I would suggest to delete this sentence given that the permeability model used in this study also is a simplified linear approximation and hydro-fracturing as well as other processes such as the expulsion of magmatic fluids are not considered.

In the revision process, we deleted this sentence.

L. 607 This section currently does not discuss the limitations of the study and could also be extended by the limitations/simplifications of the model setup used. E.g., single-phase flow, no BDT within the host rock, no hydro-fracturing within the intrusion.

We were a bit surprised here, as we did mention at least two features that could be important but are not considered in the model and thus our study. Anyway, we expanded the paragraph with further neglected effects as suggested by the reviewer(s), and hope that the reader gets an honest view of what the study does and does not address.

References that are not in the manuscript:

Driesner, T., Heinrich, C.A., 2007. The system H2O–NaCl. Part I: Correlation formulae for phase relations in temperature–pressure–composition space from 0 to 1000°C, 0 to 5000bar, and 0 to 1 XNaCl. Geochim. Cosmochim. Acta 71, 4880–4901. doi:10.1016/j.gca.2006.01.033

Anonymous Referee #2

We thank the reviewer for the constructive criticism. Here, we first provide a brief response to the general comments made, and then give our answer to the three specific questions.

The key criticism in general is that we do present a quantitative evaluation of the models and that the general approach is too open-ended, rather than testing a hypothesis. On the one hand, we therefore focused on better presenting our working hypothesis (see revised introduction, "Opening of cooling joints may trigger an early hydrocarbon migration pulse into the sill"). This had been suggested by geological studies, but not quantitatively investigated in a numerical model. Additionally, we now quantify hydrocarbon (methane) accumulation over time to estimate how much of the generated methane may possibly be transformed to graphite (Fig. 11). Putting a number on this process is indeed value, both from methane storage perspective and from a petroleum system perspective. The quantification was added to the extensively revised result (section 3.4.2) and discussion section 4.3 (first paragraph).

On the other hand, it is a key goal of this work to understand the complex coupled hydrothermal-mechanical-chemical processes around sills, and we believe that there is value in using a quantitative numerical value to qualitatively describe the physical process dynamics. We would argue that some exploratory effort here is acceptable, and hope that our description paves the way to answer some of the reviewer's questions in future studies, where the process dynamics themselves then do not have to be described in detail anymore. In this context, we highlight the following question of the second reviewer:

"*Under what conditions (combinations of sill thickness, depth, initial temperatures, source rock chemistries) do we expect the development of through-going bitumen deposits in the sill?*"

This question is beyond the scope of this study, but it is the natural continuation of the presented work, because it would require a physical model for when cooling joints are forming, i.e., under which conditions sills become permeable and porous in the first place. We therefore added this to our future recommendations in section 4.5.

Overall, we believe that the added quantitative analysis in the revised manuscripts and clearer presentation of the working hypothesis improves our paper.

The temperature-permeability relationships are a key component in this study, and evolving permeability the of cooling joint networks is indeed poorly constrained. We added a sentence to the model description that points out this problem as well as the fracture flow issue mentioned in the next commment (section 3.2, third paragraph). We tested a porosity opening of the sill over a lower temperature range (900 ℃ – 800 ℃) and do not see a significant change in the methane distribution. Of course, the hydrocarbons enter the sill at lower temperature in this case but as long as temperatures are above ~350 – 500 ℃ the conditions are sufficient for graphitization and thus consistent with the field observations.

Given the good agreement of field observations and our models, we think that our model captures the essence of the processes. But we must admit that especially the linear permeability increase due to cooling joints is a crude approximation. As mentioned in the general answer, a better understanding of cooling joint formation would certainly lead to more accurate results and allow improved quantification, e.g., of hydrocarbon/methane accumulation. Due to the complexity of the model, it is difficult to say how exactly the results would be influenced by different, perhaps more physical sounds relationships.

We think that our revised model with dynamic fracture-related permeability and porosity qualitatively captures at least some of the effects of fracturing correctly. It is clear, however, that porous flow is not the ideal model in a fractured sill, and we seek to improve this in future work.

Cooling joints propagate with the temperature front, i.e. they develop perpendicular to the isotherms. Over the wide lateral extent of the sill this should lead to network of fractures around vertically oriented columns of intact rock. Near the tips we would expect a much more complex

fracture pattern. Since the network around the columns is inherently three-dimensional it will also allow for fluid flow perpendicular to the columns because they are surrounded by interconnected fractures, which often show polygonal patterns (e.g., Hetényi et al., 2012). Hence both, vertical and horizontal flow directions are possible in the sill. Outside of the sill, one would ideally have to consider stress orientation to predict the orientation of hydrofractures. Currently, this is not possible in our model. Our numerical model allows considering anisotropic permeability but we have not explored the effects arising from, for example, a higher vertical than horizontal permeability within the sill. This would be necessary to reliably answer the question regarding backflow without too much speculation.

References not in the manuscript:

Hetényi, G., Taisne, B., Garel, F., Médard, É., Bosshard, S., & Mattsson, H. B. (2012). Scales of columnar jointing in igneous rocks: field measurements and controlling factors. *Bulletin of Volcanology*, *74*, 457-482.