4th March 2021

Dear Editor:

I have undertaken a minor revision of the manuscript by following all of the reviewer's suggestions. My point-by-point response is given below (in black), alongside his comments (coloured blue). Section 3.4 is now more complete with a layperson's explanation and should be more accessible.

Best wishes,
Felix Ng

Report by Eric Wolff (dated 03-March-2021)

This is a really impressive and thorough paper. It starts by examining an idea that has certainly caused a lot of interest and concern in the ice core community (cited 87 times), but which seems inconsistent with observed data. Despite this, the mismatch with data has never been explained, and this paper represents a real advance in that it explores the implications of the idea with additional considerations, and starts a discussion of the possible ways of reconciling theory and data. It's quite a tough read for a non-mathematical reader, but with enough simpler explanations that its interesting implications can be understood. I did not review the first version of this paper but I have been asked to look particularly at the new section of text (section 3.4).

Firstly on the inclusion of section 3.4 at all – I think the exchange in the interactive discussion between the author and reviewer 2 was important and it is definitely worthwhile making clear the assumptions that Rempel made about grain size and whether they are reasonable. For this reason I think the first half of section 3.4 should definitely be there, although it may be possible to smooth its edges a little. The second half, where the implications of a non-uniform grain size are explored, is an interesting new angle, and I think is correct. It isn't strictly to the point for this paper and might seem a little distracting, but as it would probably not warrant a separate paper on its own I agree that it should be here, as long as it is clearly explained (which needs some work).

I am therefore suggesting some mainly minor changes (chiefly clarifications) but otherwise I certainly recommend publication.

Thank you for providing this supportive review and sharing your thoughts on the usefulness of Section 3.4 and giving me valuable suggestions below.

In the discussion it was clear that Rempel felt that the justification for neglecting the curvature term in the 2001 paper was clear, while the present author clearly feels it was not. I do not think that the motivation of Rempel et al (2001) on that point is particularly important for readers of this paper and I would suggest some minor wording changes so that this is not a point. In addition this paper should stand without readers needing to look at the discussion, which will just be a distraction. I therefore suggest a rewording of lines 430-434, and that there should be a single and full citation to the discussion without continually referring to it.
Thanks for these reflections. Yes, I am going to follow your advice in the next item to implement those minor wording changes.

Line 431: I suggest "In this connection, in the Interactive Discussions of our manuscript (give proper reference according to TCD style to the necessary discussion comments) it was clarified that Rempel et al. (2001) neglected the Gibbs–Thomson effect from the liquidus relation based on an assumption that the vein radii rv were spatially uniform – the justification for this being an anticorrelation between mean grain size and impurity loading, which has been observed in ice-core records". I would suggest removing "As explained in RC2" in line 434 as it is obvious this is a continuation of discussing what was in the comment.
Your suggested wording improves the tone and efficiency of that passage. I have implemented these changes. Please see Lines 430–433 and 434.

Line 442 and following, where you discuss the need for dg^2 to be proportional to C_B: there is actually another reason why this is unlikely. The actual freezing point depression, and therefore the equilibrium value of C (the vein concentration) is dependent on the entire mix of chemicals in the liquid phase. The evolution of grain size will also somehow be dependent on different chemicals interacting with grain boundaries. It is vanishingly unlikely that the way the different chemicals combine to control freezing point is the same as the way they control grain size. Thus even if the proportionality was true for one mix of chemicals, it would not be true for a different mixture. This is a kind of extension of your reason (iii). I realise this is a detail and I don't insist that you add it but it might be something else to consider.

Thank you for pointing out this extra reason why the inverse-square relationship is unlikely to hold. Yes, item (iii) in the passage is a suitable place to mention it. I have done this, on Lines 454–456.

For lines 460 onwards, you have not (as in previous sections) given a layperson's explanation of what is occurring here, and I found it hard to work out exactly why this creation of peaks is occurring. I think I got it, so I will give an explanation of my own: if I am right you should include something similar so that those not wanting to follow the maths can still understand the mechanism. I think the argument is: <<Smaller grain size implies for a given C_B, more vein length and (by equation 4) lower rv. This in turn implies that the Gibbs-Thomson effect in eq 6 is stronger (more freezing point depression) and therefore for a given ice temperature, the solute effect must be lower, ie c must be smaller in the ice with smaller grain size. This leaves a concentration gradient and causes diffusion that raises C_B in the area with small grains at the expense of the surrounding ice >> It might also be worth spelling out that the effect (again from eq 4) is that rv increases, and presumably diffusion only continues until rv has reached the size it is in the surrounding ice (I think eq 4 then tells us that C_B will reach an asymptotic value related to the square of the ratio of d_g inside and outside the perturbation). It would actually be helpful if Fig 12 included the evolution of rv – could this be added?

Thank you for suggesting adding a layperson's explanation and writing a passage to help me. What you described captures the interactions and matches my understanding. Your description is helped by referring to the curves and the intersection point in Figure 2f, which can be used to explain why fluctuation in the grain size perturbs *c*. In the revision, following the passage where the mathematical result (32) exposing signal formation is first described, I now give the layperson's explanation in the paragraph on Lines 471–479, by adapting your passage and referring to Figure 2f. Several sentences on Lines 464–470 have been adjusted to coordinate with this change and to control the flow.

Your final suggestion is to plot r_v to illustrate the layperson's explanation. This is useful, although not so useful to do in Fig. 12, because that simulation run includes vertical compression and changing temperature (with non-zero temperature gradient), so r_v does not become spatially uniform at large time. Only in the simplified situation of the layperson's explanation would r_v evolve to a constant. Plotting r_v in Fig. 12 and describing its complex evolution probably over-complicates Section 3.4. Therefore I have opted to plot r_v in Movie S6, where the reader can see how r_v evolves --- comprehensively, at all times. In the main text, on Line 488–489, I have added a note in brackets to clarify why r_v doesn't become constant. Similarly, I have added panels in Movie S7 (the EPICA run) to display r_v at all times. Accordingly, the Supplementary File and the Data Repository have been updated with new captions and movies.

Finally in the conclusions, the paper contrasts impurities dissolved in the veins, and impurities in the grains or grain boundaries. Remember that they can also be in the veins but not dissolved. As an example at Dome C at -50 degrees, sulphuric acid would be dissolved (well above the eutectic of -70) but NaCl would almost certainly have precipitated out somewhere around -23. This adds a further complication because in my example, NaCl could dissolve back into the veins at the warmer temperatures at depth.

Thank you for pointing out this. In Section 4, I now signpost this additional complication in two places: Lines 585–587 (inside a paragraph about the distribution of impurities in ice) and Lines 602–603 (in a passage highlighting future modelling challenges).